Gd-Loaded Liquid Scintillator (Gd-LS): Past Problems, Current Solutions, & Future Directions

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⁺R&D sponsored by Offices of High Energy Physics and of Nuclear Physics, Office of Science, USDOE

Key Requirements for Gd-LS for Daya Bay

- High light transmission = high optical attenuation length (low optical absorbance).
- High light output in the Liquid Scintillator, LS.
- Long-term chemical stability, since the experiment will go on for at least 3 years.
- Stability of the LS means no development of color; no colloids, particulates, cloudiness, nor precipitation; no gel formation; no changes in optical properties.

Development of Metal-loaded LS, Leading up to Daya Bay Project

- Initial goal at **BNL**: prepare *high* concentrations of Metal-loaded LS for solar-neutrino experiments (LENS). Not easy to achieve.

- During 2002-05, BNL developed *new chemical synthesis methods:* ~10% metal (Yb or In) in pseudocumene, PC (trimethylbenzene).
- **Key steps**: complexed metal as organic carboxylates, purified components, used solvent extraction to dissolve in LS.
- BNL systematically studied carboxylic acids with 1-9 carbon (C) atoms; found that 2-methylvaleric acid (with C=6) is best.

- Starting in 2004, BNL successfully made **~0.1% Gd** (mainly in PC). Hundreds of samples have been made.

Development of Gd-LS, for Daya Bay, continued

- **IHEP** (Beijing) also began R&D on Gd-LS in 2004.
- Different approach from BNL: focusing on precipitating solid organo-Gd complexes that are soluble in LS.

- **JINR** (Dubna) has long experience in the development of plastic scintillators.
- Currently studying the characteristics of LS solvents.

- **The three Labs, BNL, IHEP, and JINR, are intensifying their collaboration in this work.**
- Their current R&D focus is on a new LS, Linear Alkyl Benzene, **LAB**. An attractive alternative to PC (to protect the environment, health, & safety): high flashpoint, biodegradable, tons are commercially produced for detergents.

Past Problems in Reactor Experiments with Gd-LS

CHOOZ, 5 t 0.1% Gd-LS

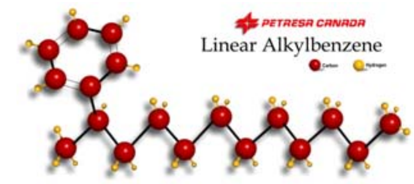
- Used "brute force" to load Gd into LS: dissolved $Gd(NO_3)_3$ in alcohol, which was then put into benzene-like LS.
- The resulting Gd-LS was not stable.
- **Turned yellow a few months after deployment (0.4% degradation per day). Moral:** nitrates plus organics is not a good choice.

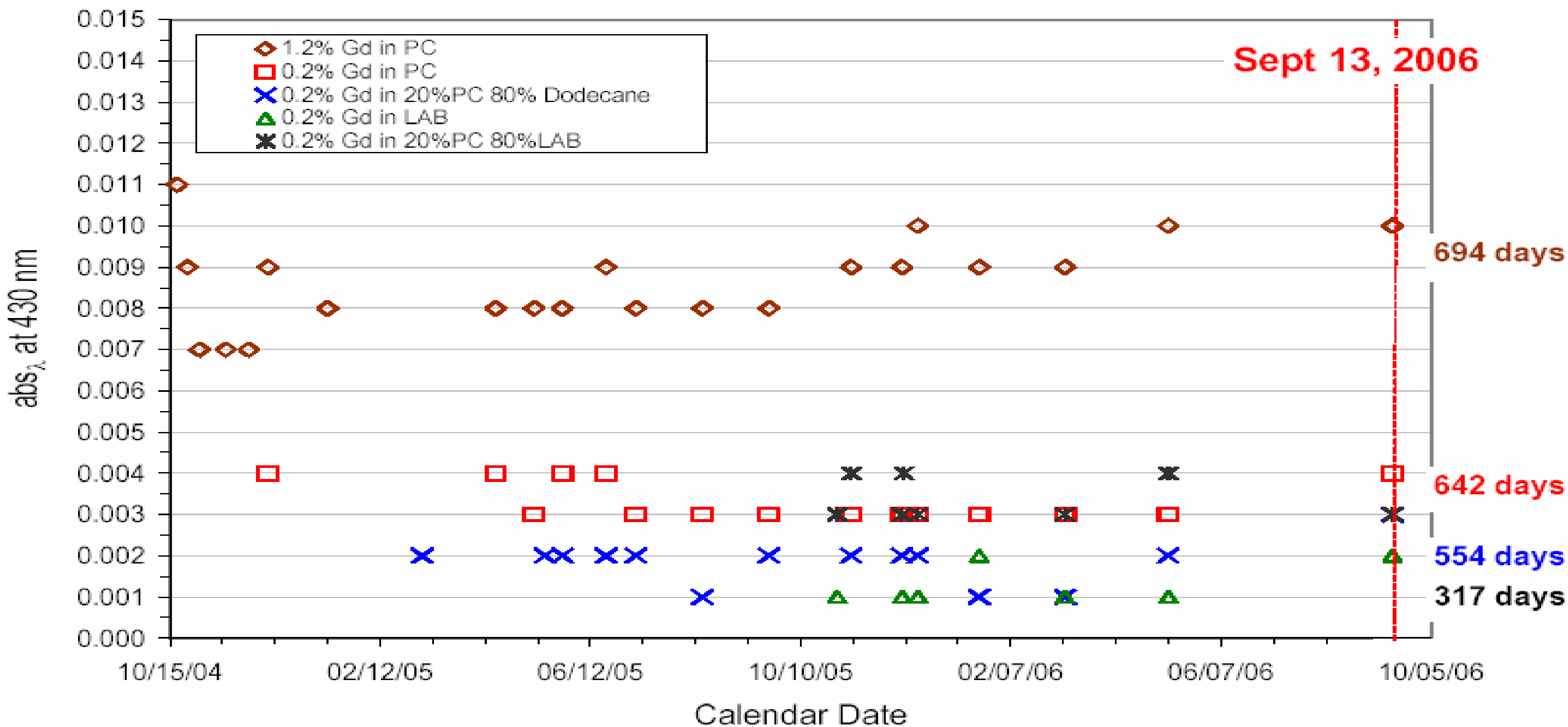
Palo Verde, 12 t 0.1% Gd-LS

- Obtained Gd-LS, BC-521, from Bicron Co.
- Was prepared by making an Gd-organic complex, a carboxylate (of 2-ethylhexanoic acid) that was soluble in pseudocumene, PC.
- Similar approach to that used by BNL.
- Report of **slow deterioration with time (0.03% degradation per day).**
- However, G. Gratta says there was some initial deterioration but then the Gd-LS stabilized. It is still usable today, by A. Bernstein at San Onofre.

BNL Gd-LS Optical Attenuation: Stable So Far ~700 days

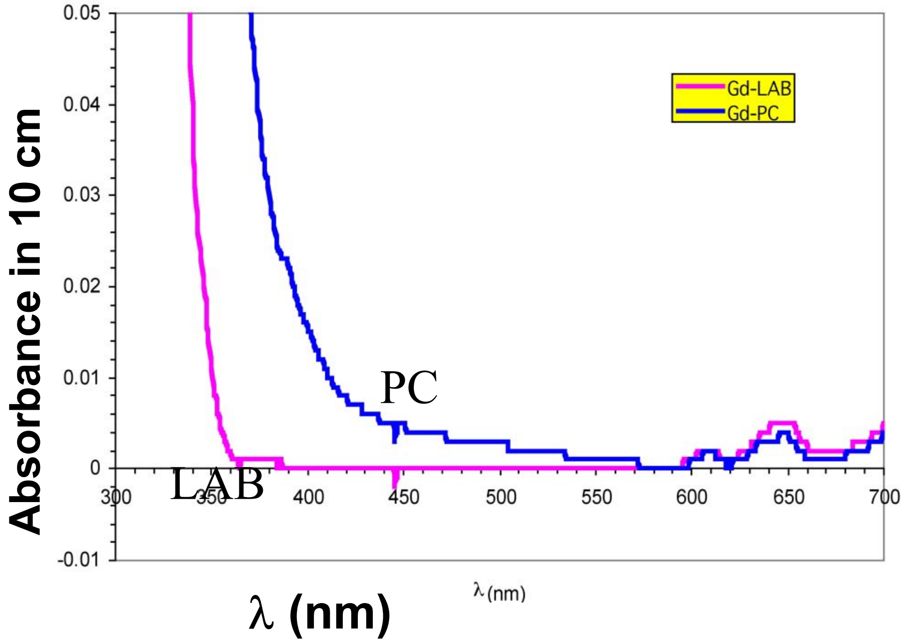
- Gd-carboxylate in PC-based LS stable for ~2 years.
- Attenuation Length >15m (for abs < 0.003).
- Promising data for Linear Alkyl Benzene, LAB
  (LAB use suggested by SNO+ experiment).



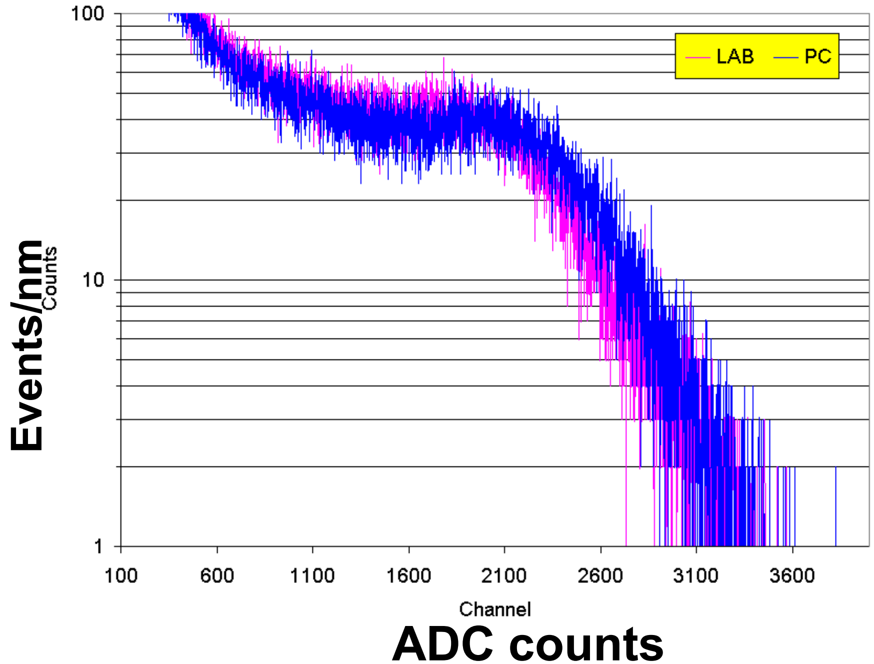


BNL: Details of performance of Gd in PC and LAB

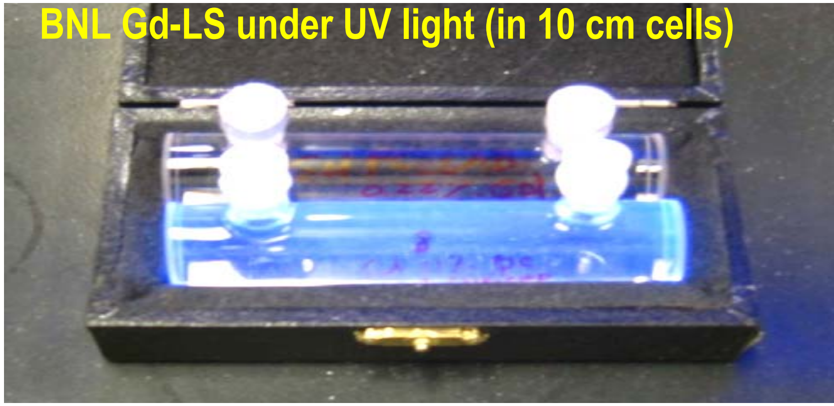
Optical Spectra



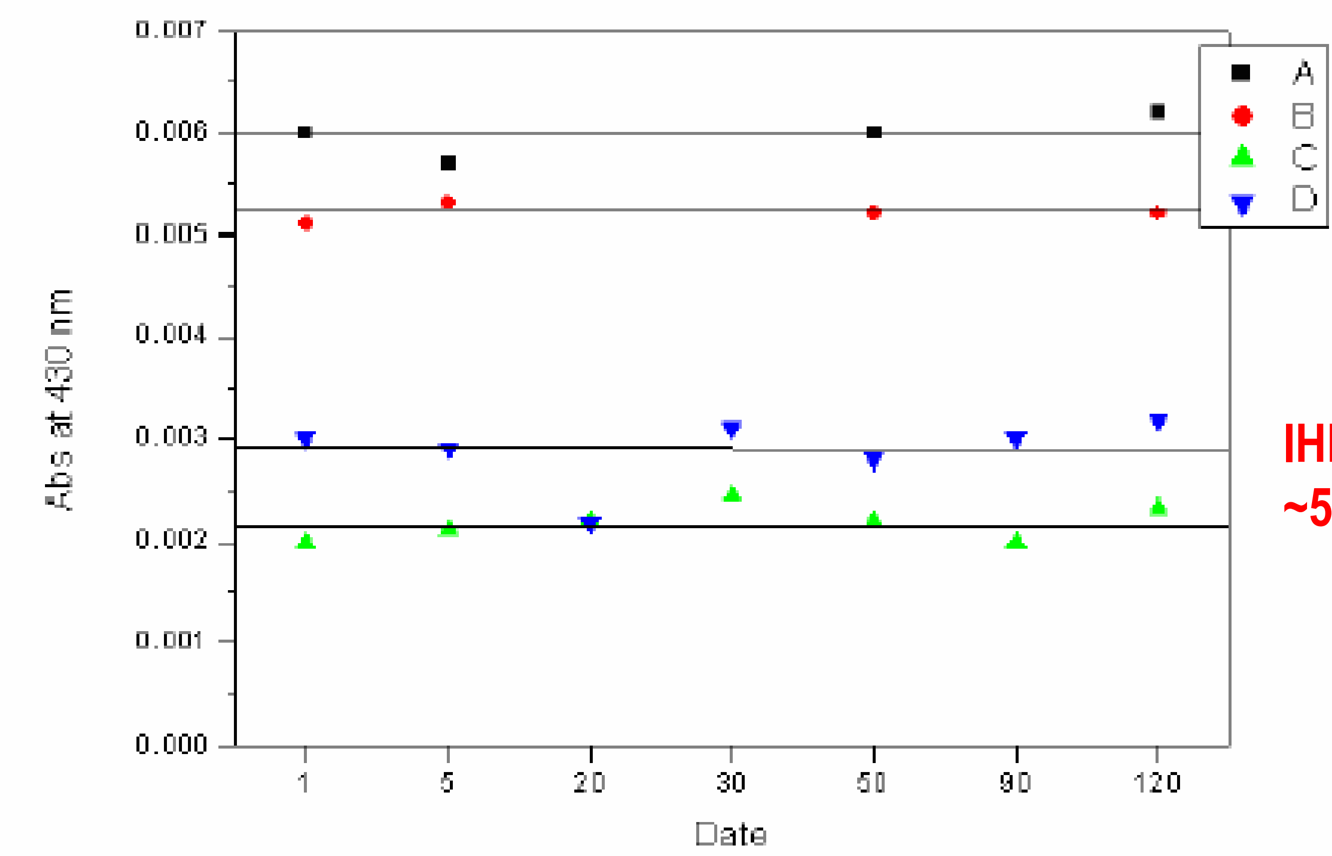
Light Output Spectra



- LAB has lower optical absorption.
- 100% LAB and PC have similar light outputs.
- But LAB has 2X light output of 20% PC + 80% dodecane mixture.
- BNL Gd-PC has ~3X better optical absorption than Bicron BC-521.



Stability of IHEP Gd-LS vs. Time



IHEP Gd-LS light yields ~50% vs. anthracene

Legend: IHEP carboxylate samples (mesitylene is a trimethylbenzene, similar to PC)

A. 2 g/L Gd-isonanoate complex in 4: 6 mesitylene/dodecane

B. 2 g/L Gd-ethylhexanoate in 2: 8 mesitylene/dodecane

C. 2 g/L Gd-isonanoate in LAB

D. 2 g/L Gd-ethylhexanoate in 2: 8 mesitylene/LAB

Important to Determine H/C and Gd in LS

BNL has begun New R&D, Combustion Analysis

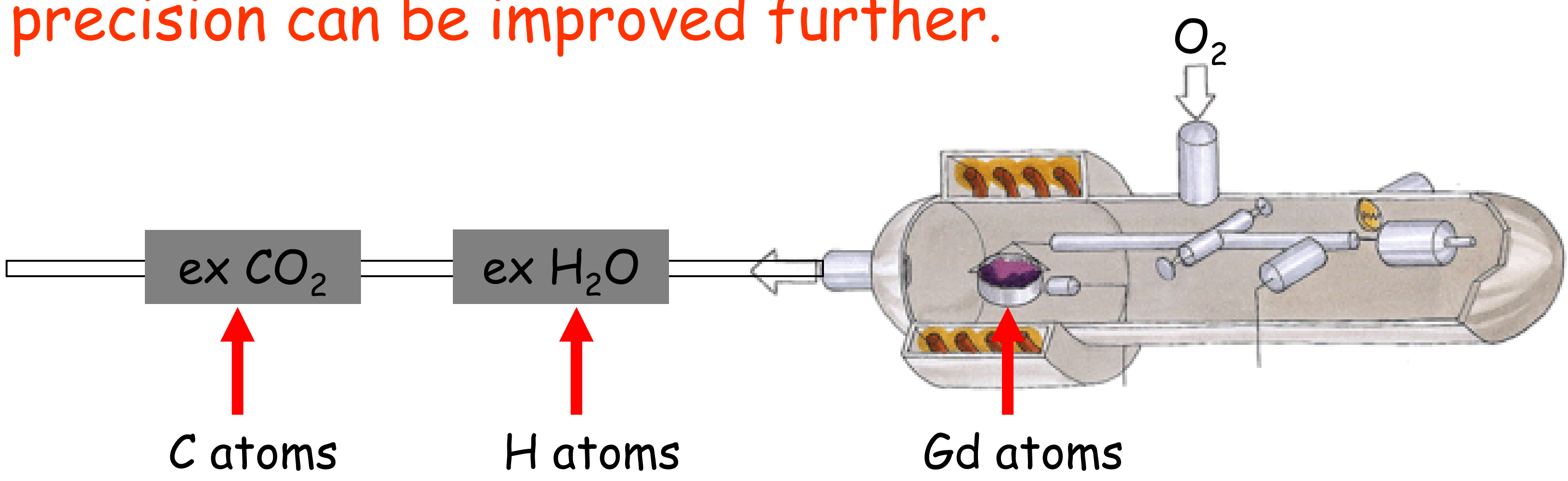
To determine number of Hydrogen antineutrino targets in the LS.

Gd-LS decomposition in O_2:

LS: $C_xH_y + (x + y/4).O_2 \rightarrow x.\ CO_2 + y/2.H_2O$

Gd: $2.Gd + (3/2).O_2 \rightarrow Gd_2O_3$

1. Potential of measuring C, H and Gd simultaneously with good precision.
2. Samples were measured by certified, commercial laboratory; achieved C/H measurements at 0.3%. This precision can be improved further.



Important Directions of Future Work

- To apply expertise in nuclear chemistry to develop methods to assay, reduce, and/or eliminate radioactive contaminants in materials.
- To evaluate chemical compatibility of Gd-LS with acrylic vessel and other construction components.
- To develop mass-production chemical techniques to go from current bench-top scale of tens of kg (tens of Liters) to tons (thousands of Liters).
- Near-term goals, to complete prototype ~1-ton Gd-LS detectors: at IHEP by end of 2006, at HKU by April 2007.

Back up slides

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*R&D sponsored by Offices of High Energy Physics
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EVALUATION OF CLEANERS, WAXES, PROTECTIVE COATINGS, AND GASKETING MATERIALS FOR USE ON ACRYLIC WINDOWS

APPENDIX 19B

SCOPE

1. This method of testing **is intended** to evaluate the possible crazing tendency of cleaners, waxes, protective coatings, and flexible gaskets, for use on acrylic windows.
2. This method of testing **is not intended** to evaluate ease of application, cleaning or polishing efficiency, aging resistance, or other properties of the test material.

MATERIAL

1. Cell cast acrylic sheet of 0.25-in (6-mm) thickness (meets MIL-P-5425 specification).

APPARATUS

1. A stress-applying jig, with light source built in accordance with Figure A.
2. A portable specimen rack constructed in a manner shown in Figure B.
3. A forced-circulation air oven operating at 90 ± 2°C.
4. A constant temperature-constant humidity (23°C *& 50% R.H.) room for conditioning the test specimens and for conducting the test.
5. A micrometer reading from 0.0025 by 0.001".
6. Suitable weight assemblies with weights.
7. A quantity of 1/2" x 1" pieces of No. 1 Whatman filter paper.
8. A 10 ml. pipette.
9. A suitable elapsed-time indicator. For most work an electric clock is accurate enough.
10. A beam balance, capacity 0–21 pounds by 0.01 pound divisions.

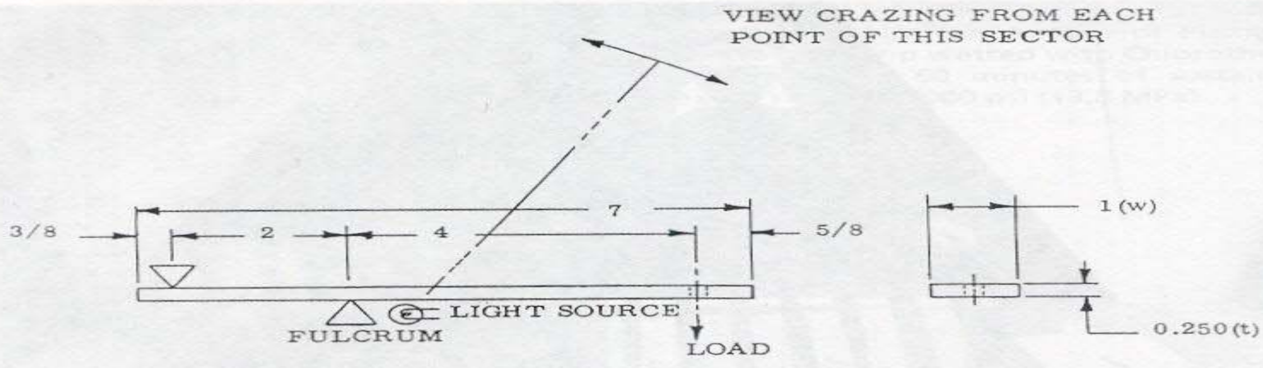
PROCEDURE

1. Without removing the protective masking tape, the acrylic sheet shall be sawed into 1" wide x 7" long test specimens. A minimum of three specimens shall be used for each stress and cleaner condition.

The test specimens may be longer and wider to accommodate larger test racks. Much of the data shown in Appendix A was generated with 2" x 22.5" x 0.25" specimens.

At elevated temperatures (example, 50°C) controls shall be tested at each stress. Precautions shall be taken to avoid inhaling fumes or vapors from the cleaners, waxes, or flexible materials. Controls means material tested at each stress level, as test specimens, except no cleaners, gaskets, etc., are used.

Dr. Jerry D. Stachiw, P.E., B.S., M.S., D.Ed., Fellow ASME, Hon. Member ASME Board on Pressure Technology Codes and Standards Pressure Vessels for Human Occupancy. He wrote a book, HANDBOOK ON ACRYLICS FOR SUBMERSIBLES, HYPERBARIC CHAMBERS AND AQUARIA



LOAD - OUTER FIBER STRESS OF 2,000 PSI

FORMULA: $\text{LOAD (POUNDS)} = \frac{w \times t^2 \times 2{,}000}{24}$

w = Width of panel (measured to nearest 0.001 inch)
t = Thickness of panel (measured to nearest 0.001 inch)

Dimensions in inches. Tolerances on all dimensions ± 0.030 inch, except thickness dimension which shall be ± 0.025 inch.

Figure 19B.1
Loading condition for cantilever beam flexure stress application.

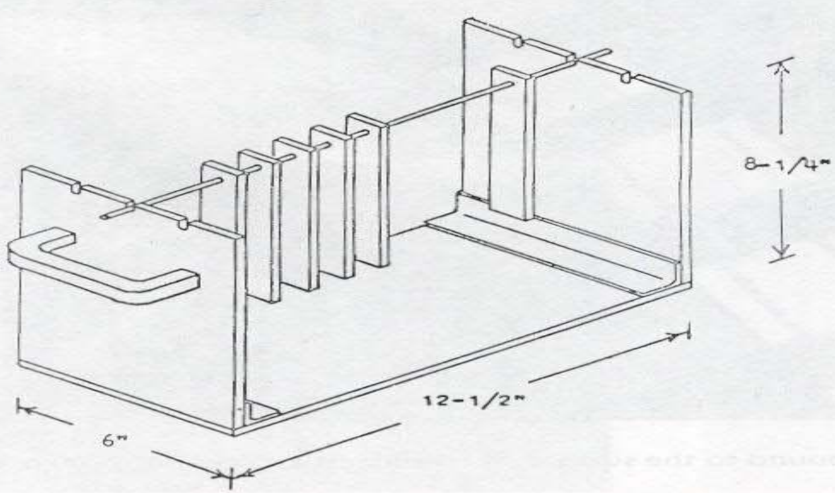


Figure 19B.2
Rack for conditioning of test specimens before flexure testing.

Figure 19B.3
Testing of acrylic for compatibility with cleaning and disinfecting compounds for acrylic plastic.

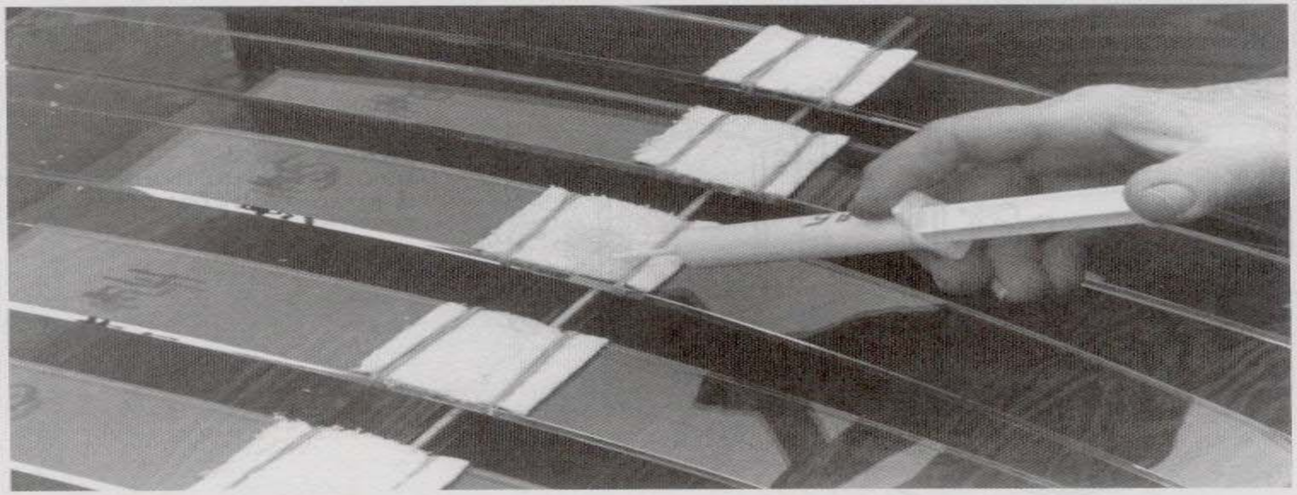


Figure 19B.4
Application of disinfecting compound to the surface of acrylic test strip under 2000 psi (13.8 MPa) sustained flexure.

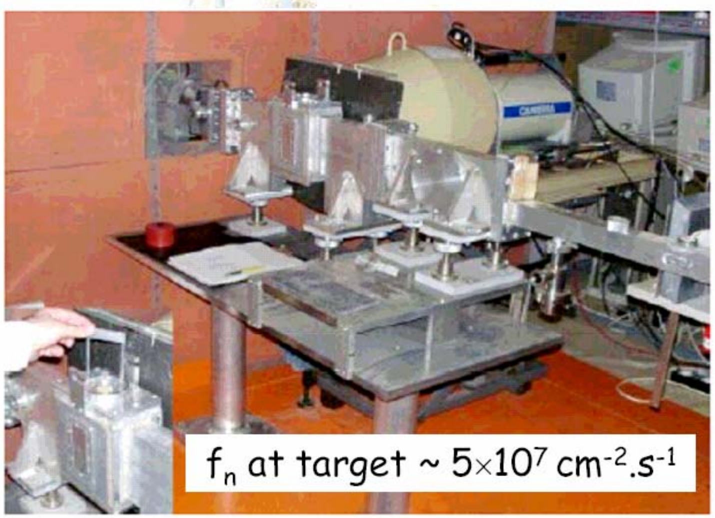
Also

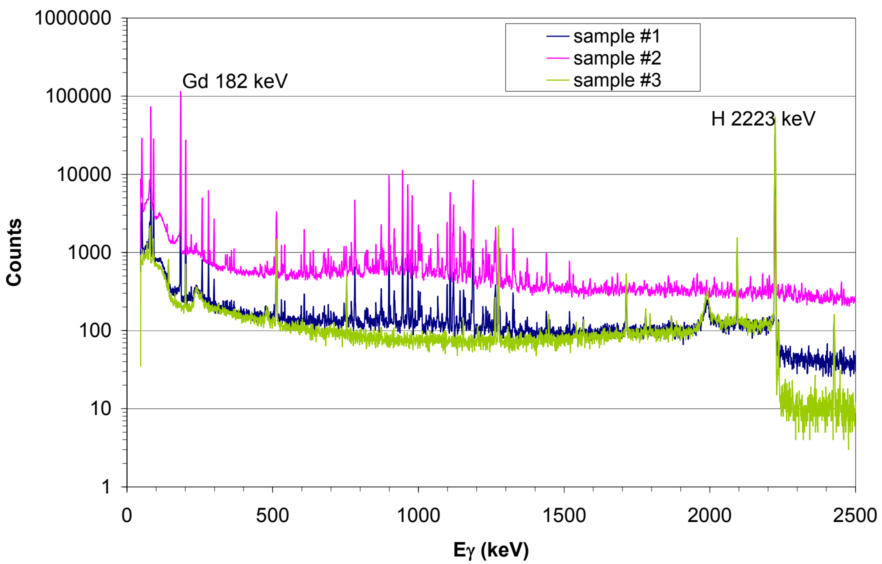
SNO-acrylic cleaning STR-96-070

D. Earle, R.L. Hahn, J. Boger, E. Bonvin

New R&D: Determination of H/C and Gd in LS

By Prompt Gamma Neutron-Activation-Analysis



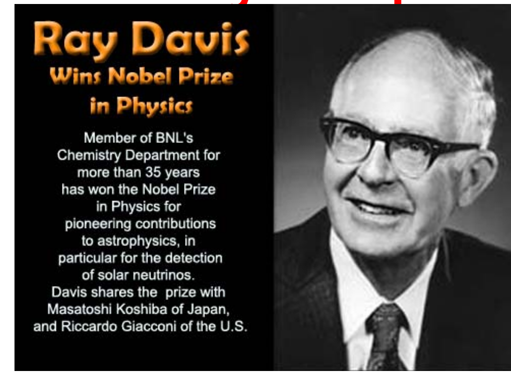


1. Measure 2.2-MeV γ from H; 0.18-MeV and other γ's from Gd after thermal neutron capture.
2. Samples were measured by the Institute of Isotopes, Hungary Academy of Sciences;. Achieved Gd and H measurement at 1%; the precision needs to be improved.

Gd-LS from different institutions

Lab	Solvent	Extractant	Fluors
BNL	1,2,4-trimethylbenzene or Linear Alkyl Benzene	carboxylic acids	0.3 g/L PBD, 15 mg/L bis-MSB
Univ. of Sheffield	α-hydroxytoluene	tri-ethylphosphate	butyl-PBD (2-(1-Naphthyl)-5-phenyl-oxazole)
MPI-K	1-methoxy-benzene (anisole)	β-diketone	PPO (200g/L) bis-MSB (500 mg/L)
Palo Verde	Bicron BC521 40% PC + 60% Oil	2-ethylhexanoic acid	4 g/L PPO, 100 mg/L bis-MSB
CHOOZ	IPB	Hexanol	*p*-PTP, bis-MSB
Bicron	PC or mix of PC with mineral oil	2-ethylhexanoic acid	4 g/L PPO, 100 mg/L bis-MSB
Eljen Technol.	Anthracene	Unknown	3 g/L PPO, 0.3 g/L POPOP

>40 Years of Neutrino R&D @ BNL Chemistry Dep't.



- **Done: HOMESTAKE** Radiochemical Detector
  C_2Cl_4; $^{37}Cl + \nu_e \rightarrow {}^{37}Ar + e^-$ (~40 years)
- **Done: GALLEX** Radiochemical Detector
  Ga; $^{71}Ga + \nu_e \rightarrow {}^{71}Ge + e^-$ (1986 - 1998)
- **Now: SNO** Water Čerenkov Real-time Detector
  Ultra-pure D_2O (1996 - ≥ 2006)

Note: Hahn became Leader of BNL Group in 1986: GALLEX, SNO, θ_{13}

- **New:** ***THETA-13 High-Precision Oscillation Experiments*** *at Daya Bay Nuclear Reactors Real-time Detector* (R&D)
  ~0.1% Gd in Liquid Scintillator, Gd-**LS** (began 2004)
- **New: LENS** Real-time Detector (R&D)

~10% ^{115}In-**LS** (began 2000), Detect pp and ^7Be Solar Neutrinos

- **New: Very Long-Baseline Neutrino Oscillations**
  Neutrino Beam from Accelerator (R&D began 2002)
- **New: SNOLab, SNO+** (R&D) with **LS** (began 2005)
- **New: SNOLab, SNO++** (R&D) with ^{150}Nd-**LS** (began 2005)

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