Warm Heavy Liquid Calorimetry: A Proposal to Measure Performance of Candidate Materials

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Abstract

We propose to measure the resolution and linearity of an electromagnetic calorimeter using a warm, short radiation length liquid as a radiator. We have identified a liquid with radiation length and transmission properties similar to lead glass. A liquid has the property that it can be purified or replaced without disassembling the detector. It is also suspected that it will be intrinsically more radiation hard than lead glass or crystalline detectors since the radiation damage associated with the solid state is avoided. If this material proves suitable it will be possible to achieve the energy resolution of lead glass without the difficulties associated with radiation damage and its implied calibration drift. The expense of casting and polishing will be avoided and it will be possible to construct "seamless" calorimeters in nearly any required geometry.

1 Introduction

Using electromagnetic calorimetry to measure the positions and energies of electrons and photons is quite simple in principle but is rife with technical difficulties when used in high rate experiments. After exposure to a sufficient amount of radiation lead glass begins to darken and its performance is degraded. The darkening causes a systematic downward shift in the calibration of the detector. During the run of E621 the amount of radiation collected during one week of routine running was sufficient to damage the type SF2 lead glass used in that experiment.

This damage can be reversed by exposing the glass to ultra-violet light, either from the sun or from artificial sources. Since the experiment is down during this period radiation hard substitutes for lead glass are very desirable. Others are working on various candidates and some serious possibilities have been found.

In a typical application of this type of detector the radiation damage is not uniform but occurs in some localized central portion of it. Suppose a liquid with the same properties as lead glass could be used. Since a liquid is mobile in the detector, the damage is spread over the area of the entire detector. A further advantage is that the damage in lead glass is associated with the solid state of the material. Liquids are expected to have much higher intrinsic radiation hardness, giving another large factor in the useful life of such a detector. This longer lifetime

would allow a future experiment to run without the downtime for curing and at a much higher instantaneous rate.

Such a material was identified by Kusumegi *et al.*[1] and tested in a low energy beam at KEK. The results were promising but they had difficulties obtaining samples of sufficient purity to produce good transmission to the Cerenkov light. The material was a saturated solution of Thallium formate and Thallium malonate in water. The important results they obtained were an energy resolution 50 % worse than lead glass and a demonstration of extreme radiation hardness. (A sample was exposed to a beam long enough to severly darken the glass bottle without measurably degrading the transmission of the solution)

These materials are currently selling for 1 to 3 dollars per gram in 100 gram lots.[2] Thallium compounds are also very toxic and a large scale (several tons) detector made of materials of this toxicity would pose a very significant disaster potential. For these reasons a search for alternate materials was undertaken.

Tetrabromoethane has been identified as an inexpensive candidate material. We propose to expose a test scale electromagnetic calorimeter based on this material to a beam of electrons or positrons to measure the resolution and linearity achievable with this technology. We will also expose small scale modules of lead glass and lead-scintillator sandwich counters to establish benchmarks for comparison.

2 Selection Criteria for Candidate Materials

The physical processes that govern the operation of the lead glass calorimeter determine the properties a liquid replacement must have. The liquid must have a high concentration of a high Z element. It must be transparent to its own Cerenkov radiation, typically in the soft UV . It is also desirable, for a high rate experiment, that the detector respond quickly to the presence of the shower and, it should not fluoresce.

An experiment runs in the presence of humans, therefore the liquid should not be overly toxic, explosive, flammable, corrosive, etc. Many liquids exist that are rich in high Z elements but have been rejected from consideration for the above reasons. Economic realities can also be used to rule out possible candidates.

3 Candidate Materials

Several categories of materials have been identified that satisfy these requirements. A brief discussion of those that satisfy both physical and safety requirements is

given below. Appendix A gives some materials that are physically suitable but probably not useful from a safety or economic point of view. A candidate material, tetrabromoethane, has been identified and the search for others continues.

3.1 Halogenated Hydrocarbons

These are hydrocarbon-like materials with one or more hydrogen atoms replaced by Iodine or Bromine atoms.

Tetrabromoethane (also known as Acetylene tetrabromide), $(CHBr_2)_2$, density 2.964 grams/cc, radiation length 4.0 cm and a liquid at room temperature. A sample of material of practical grade (lowest available) was not visibly discolored as it came out of the bottle. After exposure to metallic zinc for 24 hours and its transmission curve was improved, most notably at short wavelengths. (see appendix B for transmission curves of representative materials) This technique could be used to purify the material in an operating detector which may significantly improve radiation hardness. This material has the added attraction of low cost. When bought in lots larger than 170 Kg its price is \$ 0.049 /(cm^2· radiation length) compared with $\approx$ \$ 0.40 /(cm^2· radiation length) for lead glass.

Diiodomethane (also known as Methelene iodide), formula CH_2I_2 with a density of 3.325 grams/cc. It is considered to be of low hazard for usual industrial handling. The material has a radiation length of 2.6 cm and so is very attractive for this reason. The transmission of the material in usual grade is not acceptable for this application but if the poor transmission is due to free iodine in solution there may be chemical techniques to remove it. It is also suspected that iodine may plate out onto the windows of the detector thereby degrading overall performance. If these problems can be overcome a detector based on this material will be tested.

Bromoform formula $CHBr_3$, density 2.904 grams/cc, radiation length 4.1 cm. A manufacturer, Kodak, provides a transmission curve that indicates this material is suitable for use as a Cerenkov radiator. The curve given was, however, for their "Spectroscopic Grade". This material was prohibitively expensive for large scale use but two other grades are available and have been evaluated for transmission. The lower grades are probably not suitable but chemical purification may improve their performance.

3.2 Water solutions

These materials have the attraction of being potentially very cheap. The heavy liquid already tested by Kusumegi *et al.* is in this category. There are problems associated with this approach independent of the material chosen. It is desirable to have a radiation length as short as possible which implies the solution be as close to saturation as possible. The amount of material that can be dissolved depends on the temperature of the solution (sometimes strongly) so that solutions close to saturated must be temperature controlled to prevent precipitation of the solute. Calculation of the expected radiation length is complicated by the fact that the fraction by weight of solute is not determined by the density of the solution. We have assumed for these calculations that the fraction by weight of solute is 100 % and the density is the density of the solution, where available. Candidates were identified by a search of the Handbook of Chemistry and Physics.[3] A property listed for several of them was "Deliquescent". This is defined as "Tending to absorb water vapor and become liquid"[4]. This property leads us to believe that high density solutions could be made, but this has not yet been verified.

Barium chlorate Mineralogists use a saturated solution of Barium and Mercury chlorides for floatation separation of ores. The density of "Rohrbach solution" is listed as 3.5 grams/cc. Mercury chloride is very toxic so we have obtained a sample of Barium chloride to see what fraction by weight of barium and density can be achieved. Under the assumptions above a radiation length of 3.0 cm may be achievable with Barium chloride. This solution has several advantages: it is very cheap, it is an ionic solution so disassociation by radiation cannot cause radiation damage, and it has a potentially short radiation length. The transmission of a saturated solution was not acceptable for this use. Further investigation is under way.

Lead compounds Several lead compounds, Lead chlorate ($Pb(ClO_3)_2$), lead chlorate hydrate ($Pb(ClO_3)_2 \cdot H_2O$) and, lead peroxydisulfite ($PbS_2O_8 \cdot H_2O$) are listed by reference 3 as deliquescent. A search for samples is under way to evaluate their properties. These materials are potentially as attractive as Barium chloride for the same reasons.

3.3 Other materials

This class consists of materials with low melting point but not in some other category and compounds very soluble in a solvent other than water. Several other materials of this type exist but were ruled out for toxicity or economic reasons.

Iodine Pentafluoride (IF_5) , melting point 9.6° C, density 3.75 grams/cc, radiation length 3.3 cm. An attractive candidate because of the short radiation length. Listed in reference 3 as a colorless liquid but this has not been verified nor has transmission been measured. A search for a sample is under way. It is interesting to note that this material is also hydrogen free giving it a small sensitivity to neutrons.

Pentabromoethane (Br_3CCHBr_2) , melting point 56° C, density 3.312 grams/cc, radiation length 3.6 cm. This material has a melting point too high to be useful as is but, it is very soluble in ethyl alcohol. The transmission properties of the solution have not been tested. The fraction by weight of bromine in such a solution has not been determined. When a sample is obtained these questions will be answered and the suitability of this material re-evaluated. This material may also be very soluble in bromoform or tetrabromoethane and may be useful for reducing the radiation length of those materials.

Tin (IV) bromide ($SnBr_4$) , melting point 31° C, density 3.34 grams/cc, radiation length 2.9 cm. Listed by reference 3 as a colorless solid or liquid. An attractive candidate because of the short radiation length. It is a skin and eye irritant but not very toxic. [5] The melting point is somewhat high but not excessively so. The material is also very soluble in water and so may be useful in a solution form. A search for a sample is under way.

4 Evaluation procedures

The specific tests required to establish the suitability of these materials are as follows:

- Obtain any reference material available regarding the toxicity, flammability, etc. of the material and any other known physical properties. Many candidate materials can be ruled out from published data on hazards or color information.
- Obtain a small sample ($\simeq$ 50 grams) and convert it to a liquid if it is not already in this form and determine the transmission properties.
- Expose a sample of the material to a large ($\simeq$ 100 Kilo rad) neutron and photon dose in a reactor. If it is in fact radiation hard, or can be cleaned by a simple chemical process full scale tests are in order.

- If the candidate proves suitable, a full scale detector module will be made for use in a test beam. This will require a minimum of 30 kilograms of liquid, possibly more depending on the radiation length and density of the specific material. The resolution achievable will be determined by measuring the pulse height distribution from a test module when it is struck by an electron of known momentum. Safety section will be contacted before any large amounts of material are brought on site. They will advise us on any special precautions required to handle the materials.

5 The Experimental Setup and Run Plan

The apparatus envisioned for these tests (see figure 1.) is very simple. The apparatus was designed to allow us to test several sorts of detectors in the same setup. The test cells will be interchangeable allowing us to compare the performance of various materials and methods directly. At a minimum we will test a tight packed array of 7 lead glass blocks, a sample portion of a segmented detector using tetrabromoethane and, an array of lead-scintillator sandwich counters. We may also test another radiation hard material, lead fluoride, if a sample is available by the time of the run. The search for suitable heavy liquids is ongoing, therefore we may test other candidates should they become available.

6 Specification of the Beam Line

A suitable material will exhibit narrow resolution and good linearity. To establish these properties, the detector must be exposed to an electron (or positron) beam of known energy. The flux must be sufficient to allow collection of good statistics in a reasonable amount of time. To demonstrate linearity and determine the scaling law for resolution of the detector we must be able to expose it to a range of electron energies or, to several discrete energies covering as large a range as possible. If the beam has a large ($\geq \pm 5\%$) momentum band pass it must be momentum analyzed.

The M-bottom beam line, for example, upgraded to include momentum analysis, will be suitable to our needs. We have used the following parameters for a sample calculation:

Energy GeV	Flux Particles/spill	Fraction of e^+	e^+/spill
20 ± 1	1700	99%	1700
100 ± 5	6000	20%	1200
200 ± 10	5000	8%	400

This information was provided to us by Jim Volk. The large momentum band pass of this beam line requires us to use momentum tagging. We intend have the capability to distinguish electrons from hadrons, using an iron absorber behind the test module, but independent particle tagging could be potentially useful.

7 Services Required of the Lab

Again, using the M-bottom beam line as an example, the flux of electrons is high enough to take one data point in a dozen spills or less. We will want to map the response of the detector in energy, and in transverse position of the electron in the cells of the detector. So in practice we could take the data at one energy in one day, or a week of steady beam time in all. The set-up and debugging time will take a bit longer than that. If we are sharing the beam line with other users, the test could take on the order of a month.

We will require data aquisition electronics as follows from PREP:

Type	Number of Channels
CAMAC ADC	16
CAMAC Latch	16
Discriminator	16
4-fold Coincidence	16
Linear Fan-in/Fanout	16
Phototube High Voltage	16
Visual Scaler	16

We will also need a data aquisition computer, perhaps a micro VAX, with disk,tape drive and CAMAC interface and, 1 CAMAC and 1 NIM crate with power supplies. Assistance in configuring the data aquisition software is also requested from the computer department.

We will require access to the chemistry lab in the basement of Wilson hall to assemble the test modules containing heavy liquids. We also request access to these facilities to do any chemical purification or stabilization that may be

needed. We anticipate that any chemical procedures we undertake will be of a simple nature and will not require large amounts of time. We do not anticipate setting up any sort of apparatus that would require a long term allocation of space.

We also request that facilities to store about 100 kilograms of heavy liquid be provided. Since the density of these materials is high the volume required will be small. ($\approx$ 35 liters) A cabinet suitable for corrosive liquids would be suitable since no flammable materials have been considered.

8 Safety Considerations

We have been consulting with the safety section throughout the development of these ideas. We will require their assistance and advice to determine the methods for the proper handling, containment and, disposal of the heavy liquid materials. A typical test cell will contain approximately 50 kilograms of potentially hazardous liquid. We are actively working with safety section to develop methods to handle these materials on this scale. Since candidate materials of extreme toxicity, volatility, or corrosiveness were rejected from consideration we anticipate no problems in developing suitable procedures.

We have experience with working with radiation and anticipate no special precautions over or above the standard ones appropriate to a beam of the intensity we intend to use.

9 Estimated Budget and Manpower

The major equipment expense for these tests are the candidate liquids and the test cells to contain them. The materials and test cells will be supplied by Rutgers. The electronics needs are very small compared to any other experiment at the lab.

The test stand is of small physical size and can easily share time in the beam with other tests or experiments being set up. We anticipate building it on wheels to facilitate such sharing.

We believe that our manpower is sufficient to complete these tests. The design and assembly of the test stand and modules can begin well in advance of anticipated turn on. During actual operation of the test we will be assisted full time by one or more graduate students and part time by several faculty personnel. Scott Teige is here full time and will continue to be so.

10 Milestones and Scheduling Considerations

The requirements of this test are relatively slight. Several samples have already been obtained for the transmission and reactor tests. Chemical cleaning techniques [6] are currently being evaluated for the halogenated hydrocarbon candidates. We anticipate that the design of the test cells will be very simple and that they can be fabricated quickly by Rutgers.

We are designing a test module for the purpose of conducting tests on the transmission of Cerenkov light in tetrabromoethane. From this we hope to learn how to build the spacers to define cells that go inside a tetrabromoethane tank, as well as gain experience in handling the chemical.

Assembly of the test cell, including installation of the liquid, optical coupling of the photomultiplier and, timing of the electronics can be completed in a matter of a few days. We will have several test cells so the assemblies can be done concurrently. Installation of the test cell in the carrier should take less than an hour.

The data acquisition system is also very simple, only a few channels of ADC and some scalers and latches. Such a simple system should be able to be brought online in short order. Furthermore, work can be begun well in advance of anticipated beam startup.

11 Summary

To summarize, we are requesting one month of beam time in a momentum analyzed electron or positron beam to establish the feasibility of using high density liquids as Cerenkov media for detection of photons and electrons. We have identified several promising materials and evaluated the transmission properties of one of them sufficiently to require testing in a beam.

Detectors based on this technology have the promise of extreme radiation hardness and relatively low cost. The energy resolution achievable with them should be approximately that of lead glass as should the time response giving the possibility of using them as trigger devices. Fabrication of these detectors is facilitated by the liquid state of the material allowing construction of large, nearly homogeneous devices with simple technology. If chemical cleaning techniques prove feasible they could be used in very high radiation environments and purified while in operation.

A further advantage, possible for the future but not exploited here, is the ease of adding "doping" materials to attempt to compensate the response to hadrons. To conclude, a potentially promising new type of detector has been found, and we wish to test a prototype.

References

[a] Spokesman, contact at 312-840-4144

[1] A. Kusumegi and K. Kondo, Nucl. Instr. and Meth. **177**, 605 (1980) A. Kusumegi *et al.* Nucl. Instr. and Meth. **185**, 83 (1981) , A. Kusumegi *et al.* Nucl. Instr. and Meth. **196**, 231 (1982)

[2] The prices in the current KODAK catalog are $112.50 for 100 grams of thallium formate and $363.00 for 100 grams of thallium malonate.

[3] R.C. Weast, "Handbook of Chemistry and Physics" (54th ed.; CRC Press, Cleveland Ohio, 1974) All of the materials listed here are given in the section on inorganic compounds beginning on page B-63, the section on organic compounds beginning on page C-75 or the section on organometallic compounds starting on page C-680.

[4] Gessner G. Hawley, "The Condensed Chemical Dictionary" (9th ed.; Van Nostrand Reinhold Company)

[5] N. Irving Sax, "Dangerous Properties of Industrial Materials" (3rd ed.; Van Nostrand Reinhold Company) Many of the compounds discussed here are listed in alphabetical order in this work.

[6] Su Yumin in Tianjin Daxue Xuebao, **2**, 91 (1988), investigates the causes of deterioration of tetrabromoethane and evaluates a stabilizer. It was found that the material could be stabilized by the addition of 0.1 % by weight of BHT and stored for at least 2 years. Our tests indicate that the addition of BHT does not adversely effect the transmission of this material. Similar techniques exist for other halogenated hydrocarbons.

Appendix A: Other materials

Name Formula	Density g/cm^3	Radiation Length,cm	Color	Toxicity
Thallium malonate $CH_2(COOTl)_2$	4.9	1.3	??	very high
Mercury (II) nitrate $Hg_2(NO_3)_4 \cdot H_2O$	deliq. (5.3)	?? (1.7)	colorless (yellowish)	very high
Thallium (III) nitrate $Tl(NO_3) \cdot 3H_2O$	deliq. (5.3)	?? (1.8)	colorless	very high explosive
Mercuric chloride $HgCl_2$	3.5	2.3	??	very high
Thallium ethoxide $(TlOC_2H_5)_4$	3.522 mp=-3	2.2	colorless	very high
Thallium ethylate $TlOC_2H_5$	3.493 mp= -3	2.2	colorless	very high
Dimethyl mercury $Hg(CH_3)_2$	3.069	2.4	colorless	very high
Borohalogens BBr_3, BBr_2I $BBrI_2$, BI_3	varies	2.7 - 4.5	colorless	reactive with water
Tungsten hexafluoride WF_6	3.44 mp=2.5	2.9 bp=17.5	lt. yellow	??
Tribromo germane $GeHBr_3$	≈ 3.3 mp=-24	≈ 3.0	colorless	??
Borotungstic acid $H_5BW_{12}O_{40} \cdot 30H_2O$	3.0 mp= 45-51	3.2	??	prob. slight
Tetramethyl lead $Pb(CH_3)_4$	1.995	4.1	??	very high
Antimony pentafluoride SbF_5	2.336 mp=2.8	4.3	colorless	??
Ethyl iodide	1.94	5.2	colorless	??
Cadmium chlorate $Cd(ClO_3)_2 \cdot 2H_2O$	deliq.	??	??	very high explosive
Thallium acetate $TlC_2H_3O_2$	deliq.	??	colorless	very high
Mercuric barium bromide $HgBr_2 \cdot BaBr_2$	deliq.	??	??	very high

Appendix B: Transmission Curves

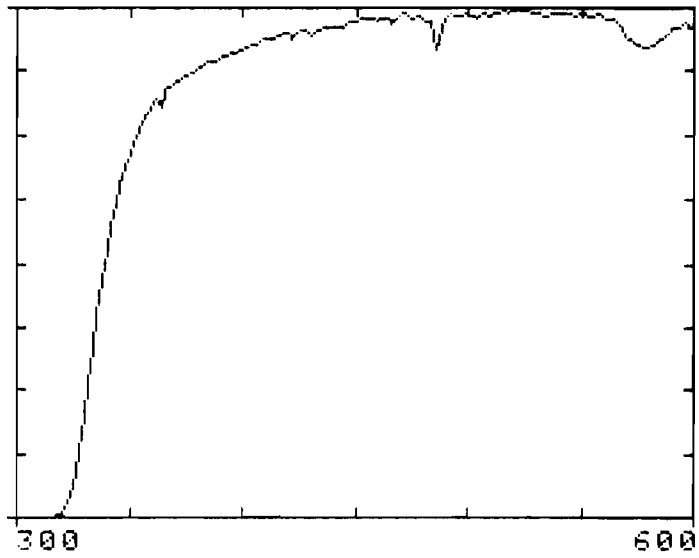

FIG. B-1. The transmission of practical grade tetrabromoethane. horizontal scale is in nanometers, vertical scale is arbitrary but the value in the flat region is believed to be near 100%

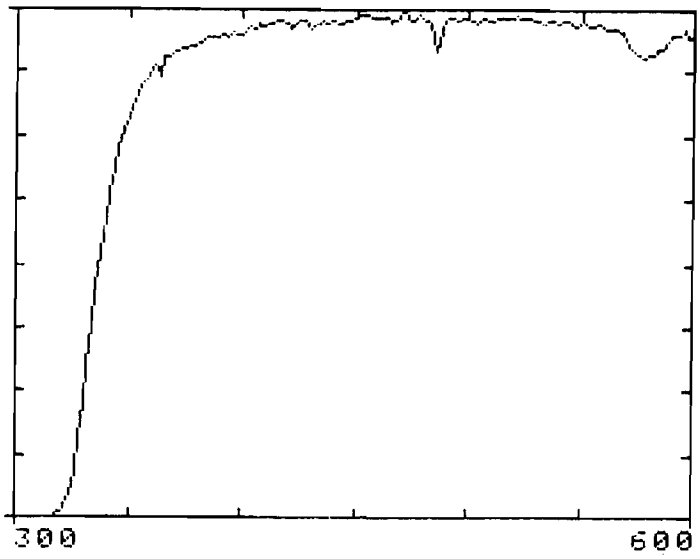

FIG. B-2. The transmission of practical grade tetrabromoethane after being exposed to metallic zinc for 24 hours. We believe that the free bromine in the sample reacted with the zinc to form zinc bromide and so was removed from solution.

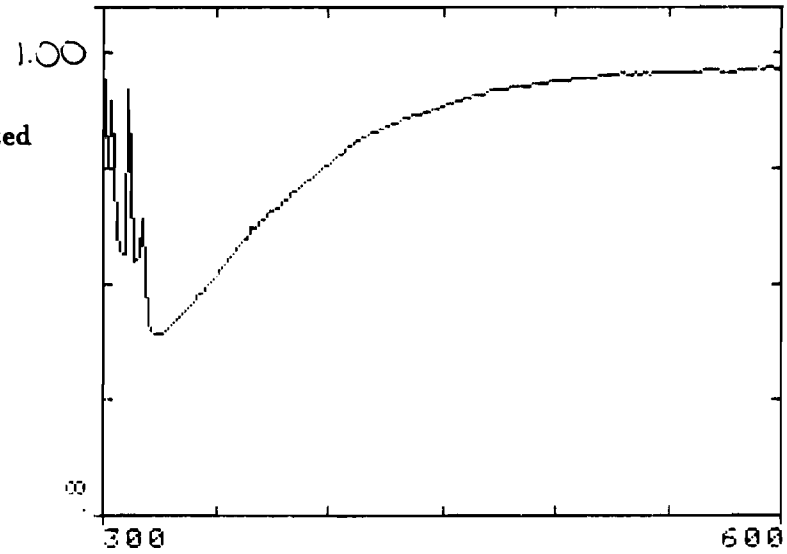

FIG. B-3. The transmission of untreated tetrabromoethane relative to the treated material.

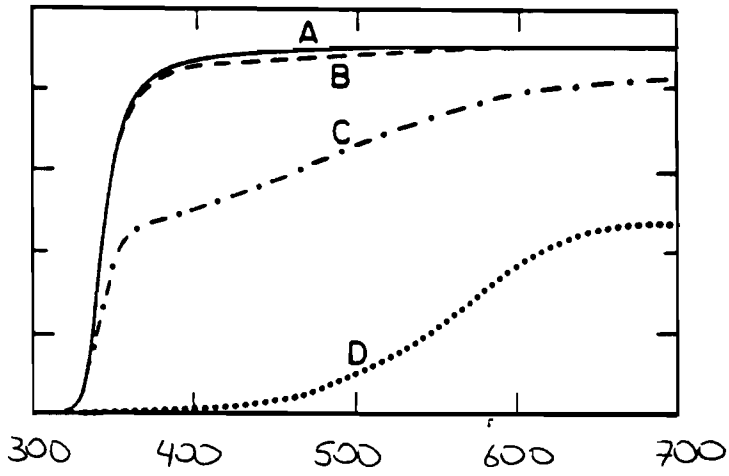

FIG. B-4. Transmission of typical lead glass. Curve A is before exposure to any radiation, B-D are after exposure to 1.2 $\times 10^2$,1.2 $\times 10^3$ and, 1.2 $\times 10^4$ rad of ^{60}Co γ radiation. (Taken from G. T. Bartha *et al* Nucl. Instr. and Meth. **A275**, 59 (1989)

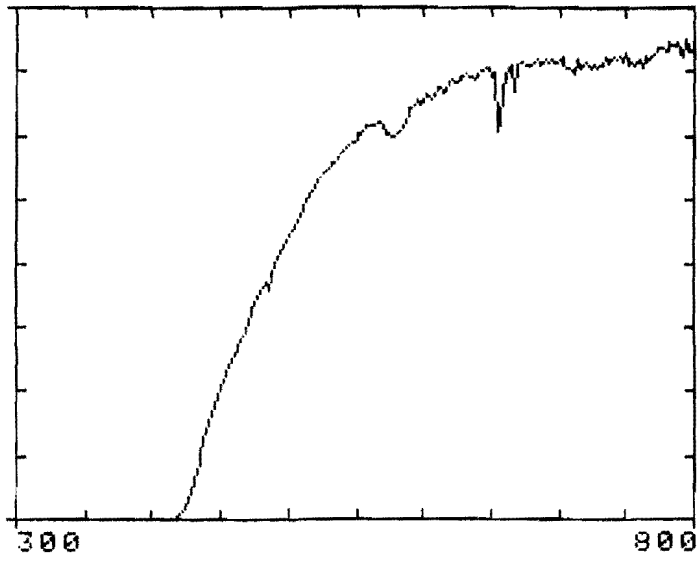

FIG. B-5. The transmission of diiodomethane. Exposure to metallic zinc turned the material dark purple. Other chemical cleaning techniques for this material are being sought.

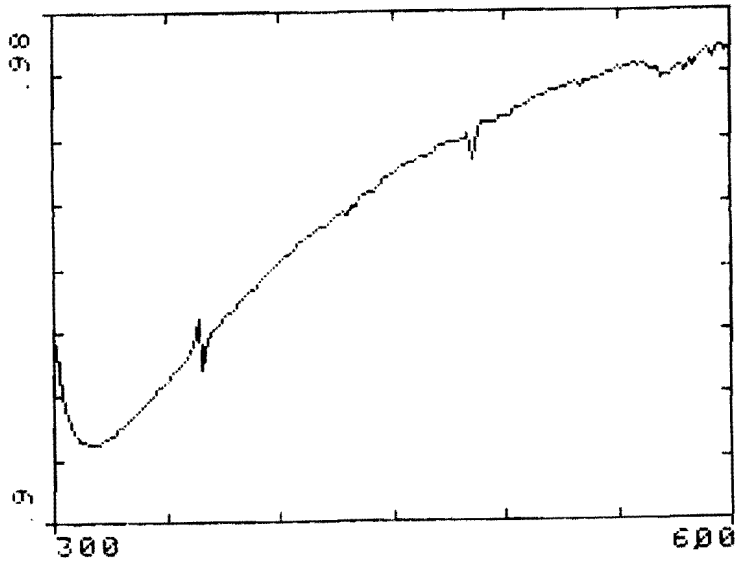

FIG. B-6. The transmission of a saturated solution of barium chloride in water relative to distilled water. It was concluded that this material is unsuitable for this application.

FIG. 1. A schematic of the proposed set-up. S1 and S2 are scintillators forming a beam telescope. The test cell, S1, S2, and the veto counter will be mounted on a cart to raise it to beam height.