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Optical properties of UV transmitting acrylics for use in a heavy water Cerenkov detector

Joanne C. Zwinkels, W. F. Davidson, and C. X. Dodd

The absorption, refraction, and scattering properties of several UV transmitting acrylics have been investigated over the wavelength range 300–700 nm using a combination of near-normal incidence regular transmittance and reflectance and diffuse-only reflectance measurements, followed by a Fresnel and a Kubelka-Munk analysis. The samples were evaluated in the as-cast and thermoformed forms, and both before and after an accelerated aging procedure. The results show significant differences in the optical behavior of the various acrylics in the UV region and stress the importance of carefully characterizing acrylic from different sources for each intended use. In our case, acrylic is the proposed material for a heavy water containment vessel for the detection of solar neutrinos. The significance of our findings to the overall performance of this Cerenkov detector, known as the Sudbury neutrino observatory detector, is discussed.

I. Introduction

A deep underground neutrino observatory, known as the Sudbury neutrino observatory (SNO) detector, has been proposed.¹ It will be located at the Creighton mine of INCO, Ltd., near Sudbury, Ontario. A crucial component of this detector is the 1000 tons of enriched D_2O as a neutrino detecting medium. As shown in Fig. 1, the D_2O will be contained within a transparent cylindrical acrylic vessel of 5-cm wall thickness immersed in a large volume of H_2O . The yield of Cerenkov radiation resulting from the interaction of solar neutrinos with D_2O is low; for a 7-MeV neutrino only ~1140 photons are generated. Successful reconstruction of an event dictates that attenuation of radiation in transit from the initial interaction point to the photomultiplier tubes be kept to a strict minimum. An investigation on attenuation of visible and UV radiation in purified D_2O and H_2O was previously reported.² and it was found that attenuation in these media was sufficiently low so as not to compromise the operation of the detector. In this paper we address the experimental determination of the optical properties of the acrylic vessel.

The material for the containment of the D_2O must satisfy several criteria. First of all, the material must

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have uranium and thorium concentrations at or below the 10^{-12} g/g level; it must have very high UV and near visible transmittance; it must retain its properties essentially unchanged over 5-10 yr exposure to pure water; it must be robust and retain its mechanical integrity; and because of its size (10 m diameter) its components must be easily joined *in situ*. The above considerations led to the selection of acrylic. Acrylic is the plastic equivalent of crown glass with respect to its optical properties. It is relatively inexpensive, tough, and has almost infinite design flexibility. It possesses the requisite low levels of radioactive contamination. Most commercially available acrylics are not suitable for our particular application as they contain UV absorbing stabilizers to prevent tarnishing on exposure to sunlight. There does exist, however, an emerging class of premium high quality UV transmitting acrylics, but detailed information about their optical behavior is not available in the literature.³ The present paper reports our findings on the optical properties of several candidate UV transmitting acrylics, particularly as they relate to the performance of the SNO detector, but also as they have important implications for other technological applications, such as the design of UV optical systems.

The optical properties of $acrylic^{3-6}$ exert a strong influence on how light is propagated from a neutrino interaction point within the D₂O to the PMTs arranged on the periphery of the H₂O shield. The transmittance of the acrylic has to be the highest attainable, especially in the blue and near UV spectral regions where the PMT responsivity is a maximum⁷ and the yield of Cerenkov radiation is increasing. Not only is it necessary to characterize the UV absorption and scattering properties of the virgin acrylic but it is im-

When this paper was written the authors were with the National Research Council of Canada. NRC has undergone a reorganization since this paper was submitted. Joanne C. Zwinkels and C. X. Dodd are now with the Institute for National Measurement Standards and W. F. Davidson is with the Herzberg Institute of Astrophysics. Ottawa, Ontario K1A 0R6, Canada.

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AIR CONDITIONING DUCT ACCESS DRIFT CABLES FROM PMT'S н₂с PHOTOMULTIPLIER VESSEL SUPPORT TUBES H₂O ACRYLIC VESSE (approx 14m x 10m dia.) D₂O STAINLESS STEEL LINER SULFURCRETE CONCRETE PMT SUPPORT FRAMEWORK ROCK

CROSS-SECTION OF NEUTRINO DETECTOR

Fig. 1. Conceptual design of proposed neutrino detector. The heavy water is contained in a cylindrical acrylic vessel and is shielded from activity in the rock by low activity concrete and light water. The Cerenkov radiation is detected by an array of photomultiplier tubes located in the light water. The dimensions of the cavity are 20 m diameter and 30 m height. The detector will be sited at a depth of 2070 m.

portant operationally to determine how these properties are affected by thermoforming and by prolonged immersion of the acrylic in liquids such as D_2O , H_2O , or a weak solution of brine, to which it will be exposed at the observatory. The leaching action of ultrapure water on acrylic can deplete the acrylic surface of lighter molecular weight components leading to significant environmental stress cracking,^{8,9} which increases absorption and scattering. However, experience with large scale acrylic structures in sea water aquaria over the long term indicates minimal deleterious effects with respect to environmental stress cracking.¹⁰

Spectral transmittance and reflectance measurements were selected for the optical characterization of the various candidate UV-grade acrylic specimens. From these measurements a consistent set of spectral optical properties-refractive index, absorption coefficient, and scattering coefficient—was derived. This spectrophotometric method was selected for its simplicity and its ability to provide reasonably accurate (within 10%) optical constant data for weakly absorbing materials over a wide spectral range and with minimum sample preparation.¹¹ Both as-cast and thermoformed acrylic specimens were evaluated as-received, and after being subjected to an accelerated aging process. These data will assist in selecting the optimum acrylic for the SNO facility and are already being used

as input in the Monte Carlo simulations of the neutrino detector response.

Acrylic Samples and Aging И.

Study on As-Cast Samples A, B, and C Α.

Premium UV-grade cast acrylic sheets from three specialist suppliers: Polycast Technology Corporation, CY/RO Industries, and Rohm GmbH (Germany) were selected for this initial study. Precise details of the processing histories, such as stabilization and polymerization, of the various acrylics were kept proprietary by the suppliers. In no particular order, these specimens are denoted here as A, B, and C. The thickness of the sheets was nominally 25 mm. Specimens of dimensions 35×35 mm were cut and the protective paper removed. Apart from careful cleaning of the exposed surfaces with a nonfluorescent soap solution, no optical polishing of the exposed faces was performed so that conditions similar to those that would prevail operationally in dealing with a 30-ton acrylic vessel in the field would be approximated as closely as possible.

For the aging study, the acrylic samples were totally immersed in solutions of ultrapure H_2O , D_2O , and a 2.3% NaCl aqueous solution in quartz beakers, each covered with a quartz lid. The ultrapure H_2O was taken from a double distillation system made of fused silica. The D_2O was of similar high chemical purity. The brine was made up from the ultrapure H_2O with 2.3% reagent grade NaCl. The concentration of NaCl is $10 \times$ that which is envisaged for the SNO detector. The beakers plus contents were placed in a constant temperature oven for 46 days at 66°C, a temperature safely below the onset of complicating phase changes (glass transition) known to occur around 90–105°C in commercial acrylic.¹² Thereafter, all samples remained immersed in their respective beakers except when a particular sample was removed for the purpose of measurement. Assuming that chemical reactivity increases for every 10°C rise in temperature¹³ by between a factor of 2 or 3, we can simulate the operation of the SNO detector at 10°C over 10-15 yr by a 1-2 month exposure under these aging conditions. Acrylic blanks were not subjected to elevated temperature but were kept in air at room temperature.

Study on As-Cast and Thermoformed Samples D, E В.

A second batch of premium UV-grade cast acrylic samples was provided in duplicate by two of the specialist suppliers for tests connected with the thermoforming process. In one case the thickness was 50 mm, and in the other 69 mm. Each supplier furnished us with two slabs of dimensions 90×20 cm, from which our samples were cut. These samples, denoted D and E, were subjected to thermoforming procedures similar to those that will prevail when the cylindrical segments of the acrylic vessel are formed. The thermoforming was carried out over a metallic form in two ways: the bare metal was used in one batch and, in the other, a rubber flocking coated the form. The surfaces



were kept in the as-delivered state, except for mild cleaning with nonfluorescent soap. For the aging study, the acrylic samples were immersed only in the ultrapure H_2O as described in Sec. II.A.

III. Experimental Methods

To predict accurately the path and attenuation of the Cerenkov photons in the acrylic vessel and the losses at the acrylic-water interface, it is necessary to characterize the absorbing, scattering, and refracting properties of the acrylic. The theoretical models used for determining the optical properties are described below, as are the details of the instrumentation and the verification of the experimental and computational procedures.

A. Refractive Index and Absorption Coefficient

The absorbing and refracting properties of a homogeneous, isotropic material are described by the extinction coefficient κ and the refractive index *n* respectively, which are functions of wavelength. These are termed the optical constants and are intimately connected with the constitutive nature of the material. Several experimental methods exist for the determination of *n* and κ ,¹⁴ with the method of choice usually being determined by the magnitudes of *n* and κ , the desired wavelength range, and the instrumentation available.

In our case, we wanted to characterize the optical constants of a weakly absorbing material—acrylic over a wide wavelength range—from 300 nm to 700 nm. The most commonly used method under these circumstances is the measurement of normal incidence regular transmittance and near-normal incidence regular (specular) reflectance, followed by a Fresnel analysis, often referred to as the (RT) method.¹⁵ Since we have a high precision spectrophotometer with which to perform these photometric measurements, this was the method we adopted.

The Fresnel relationships connecting the front-surface primary reflectance R and the bulk or internal transmittance T for normal incidence, with the optical constants n and κ at each wavelength λ are:

$$R = \frac{(n-1)^2 + \kappa^2}{(n+1)^2 + \kappa^2} \tag{1}$$

$$T = \exp\left(\frac{-4\pi\kappa d}{\lambda}\right),\tag{2}$$

where d is the sample thickness.

In Eq. (2), the term $4\pi\kappa/\lambda$ is often described by the absorption coefficient α giving the familiar Lambert's law:

$$T = \exp(-\alpha d). \tag{3}$$

The measured sample reflectance and transmittance usually differ from the Fresnel quantities in Eqs. (1) and (2) because of multiple internal reflections which augment the primary reflectance and reflection losses, respectively. It is possible to restrict the reflectance measurement to reflectance from the front surface by making the sample wedge-shaped, or grinding the back surface and blackening it, and to reduce the reflection losses in a transmission measurement by immersing the sample in a medium of similar refractive index. However, aside from the obvious inconvenience of these procedures, it was considered advantageous to include the reflectance contribution from the back face, and to measure the samples in air. This enhances the measured reflectance, thereby improving its measurement accuracy, and also provides a more sensitive indicator of the sample's refracting properties.

For a plane-parallel sample, the relationship connecting the measured normal-incidence reflectance R^* and transmittance T^* with the quantities, R and T are:

$$T^* = \frac{(1-R)^2 T}{1-R^2 T^2} \tag{4}$$

$$R^* = R(1 + TT^*). (5)$$

This is the model we assumed for the analysis of the R^*T^* data. Since analytic solutions to n(R,T) and $\kappa(R,T)$ do not exist, we obtained a numerical solution by a successive approximation procedure. The values of n and κ , at each wavelength, were optimized to be consistent with the measured reflectance, transmittance, and thickness data, within their experimental uncertainties. The convergence criterion used was that both the calculated reflectance and transmittance agree with the measured values to better than ± 0.004 and ± 0.008 units, respectively.

B. Scattering Coefficient

The path and attenuation of the Cerenkov photons in the acrylic vessel are largely determined by regular reflections and absorption, i.e., by the optical constants of the acrylic. However, if the acrylic does not have an optically smooth surface or contains inhomogeneities, such as bubbles or impurities or other density fluctuations, these imperfections will act as light scattering centers, perturbing the photon's travel, and hence will contribute to losses. It was therefore important to obtain an estimate of these scattering losses in the candidate acrylics.

A common method of estimating scattering coefficients in turbid media is to measure the diffuse reflectance and to apply a Kubelka-Munk (K-M) analysis. Diffuse reflection comes about through penetration of a portion of the incident flux into the interior of the sample where it undergoes absorption and multiple scattering at uniformly and randomly distributed particles before finally emerging back through the surface through which it entered.¹⁶ According to K-M theory, the diffuse reflectance (R_{∞}) for an infinitely thick (optically opaque) sample depends exclusively on the ratio of the absorption to scattering coefficient called the K-M remission function,^{16,17} $F(R_{\infty})$.

$$F(R_{\infty}) = \frac{\alpha}{S} = \frac{(1 - R_{\infty})^2}{2R_{\infty}}.$$
 (6)

Thus the scattering coefficients S may be determined from measurements of R_{∞} if the corresponding α values are known. In our case we have the complication that the acrylic samples are not optically opaque. An approximation to the opaque reflectance R_{∞} can, however, be obtained from one of the K-M solutions,¹⁸ which is:

$$R_{\infty} = a - (a^2 - 1)^{1/2},\tag{7}$$

where *a* is given by

$$a = 0.5 \left[R + \frac{R_0 - R + R_g}{R_o R_g} \right],\tag{8}$$

and R_0 is the reflectance of the translucent sample over an ideal black background; R is the sample reflectance over a white background; and R_g is the reflectance of the white background material itself. This was the procedure used to determine the scattering properties of the candidate acrylics.

C. Instrumentation and Verification of Methods

The spectral transmittance and reflectance data were obtained with a Perkin-Elmer Lambda-9 (PE λ -9) spectrophotometer equipped with the appropriate sampling accessory. Measurements were carried out from 300 to 700 nm at 5-nm intervals, and at ambient temperature.

The regular transmittance spectra were measured at normal incidence in a near-collimated beam (the maximum angle of convergence is 4°) with an irradiated sample area of $\sim 2 \times 10$ mm. Two independent measurements were recorded on each specimen, oriented in two mutually orthogonal directions. This procedure, followed for all the spectrophotometric measurements reported here, checked for sample nonuniformities. Since the instrument possesses a high degree of polarization,¹⁹ this procedure also checked for sample birefringence, which was of particular concern for the thermoformed specimens. The agreement between these two curves was typically 0.003 transmittance units and in the worst case 0.006 transmittance units. The average result was therefore used in the optical constant calculations.

The regular reflectance spectra were measured with a VW accessory in the PE λ -9 sample compartment, which gives a direct measure of the square of the absolute specular reflectance for a 7.5° incidence angle. This assumes that the optical path lengths traversed with and without the sample in the beam are identical. The accuracy of the method is therefore optimum for highly reflecting opaque materials but can produce accurate results for low reflecting transparent materials if suitable precautions, discussed below, are taken. The optical alignment of the VW accessory was checked with a calibrated second-surface aluminum mirror (NIST SRM 2023), which is $\sim 80\%$ reflecting. Two different thicknesses (1.6 and 9.6 mm) of planeparallel Suprasil, which are $\sim 7\%$ reflecting, were used to check the photometric accuracy at low reflectance levels and the effect of sample thickness on the detected fraction of second surface reflection. To eliminate reflections from surfaces behind a transparent specimen, such as Suprasil, it must be backed with an absorbing material. We used a piece of black felt which

is $\leq 0.5\%$ reflecting from 300 to 700 nm, and the acrylic specimens were measured in an identical manner. The average difference between the NIST calibration values for SRM 2023 and those obtained with the VW accessory was ± 0.002 reflectance units. The agreement between the Suprasil reflectance values computed from the Amersil catalog optical constant data, and the measured data for both the thin and thick specimens was better than 0.003 reflectance units. This is the estimated uncertainty of the VW accessory at the $\sim 10\%$ reflectance level, and a second-surface recess of ≤ 10 mm. However this error will increase for samples that are not optically plane and parallel or are further recessed.

The (RT) analysis requires plane-parallel samples and an accurate measure of the sample thickness. Each acrylic specimen was measured at the four corners with a high accuracy micrometer (0.01 mm) and the average result was used in the optical constant calculations. The thickness uniformity varied from $\pm 0.01 \text{ mm}$ to $\pm 0.16 \text{ mm}$ depending upon the sample. This thickness nonuniformity introduces negligible error in the calculated transmittance because of the large sample thicknesses involved here but may have a serious effect on the calculated reflectance because the condition of parallelism is no longer satisfied.

Diffuse reflectance spectra were measured on the PE λ -9 equipped with a 150-mm diam integrating sphere accessory coated with BaSO₄. The measurement geometry was 8° incidence, hemispherical collection with the specular component excluded. The irradiated area of the specimen was $\sim 5 \times 15$ mm and the reference standard was pressed polytetrafluoroethylene (PTFE) powder, whose absolute reflectance is certified with a relative uncertainty of $\pm 0.5\%$. For the measurements of acrylic over a white and black background, a 3-mm thick white Russian opal glass and a piece of black felt were used, respectively. The reproducibility of these measurements was typically better than ± 0.003 reflectance units.

The procedure for determining *n* and α values from combined RT measurements and using the α values to calculate the scattering coefficient from diffuse reflectance measurements is not new and has been applied successfully by Kortum et al.¹⁷ to several colored glass filters. However, in that study, the specular reflection component in the diffuse reflectance measurement was eliminated by grinding the samples to a powder and diluting with a nonabsorbing powder. Since we did not want to alter destructively the physical properties of the acrylic samples, we performed diffuse-only reflectance measurements, relying on the ability of the instrument's gloss trap to exclude the specular component. To verify this approach, we performed an identical set of measurements and optical constant analysis on a ~ 2 mm-thick didymium glass filter. Since this is a translucent specimen, it was necessary to estimate its opaque reflectance in the manner described for the acrylics. This test specimen is perhaps an extreme example since it is highly spectrally selective, exhibiting many sharp minima and maxima. However, the computed refractive index at 590 nm of 1.69 is in



Fig. 2. Absorption coefficients for unaged acrylic samples A, B, and C as determined using the (*RT*) method.

excellent agreement with the Schott catalogue value of 1.69 (567.6 nm) and the logarithm of the absorption coefficient curve (log α) and the logarithm of the remission curve [log $F(R) = \log \alpha - \log S$] are in reasonably good agreement with the corresponding curves reported by Kortum *et al.*¹⁷ The only significant discrepancy occurs at ~580 nm where both the measured regular transmittance and diffuse reflectance are ~0.0001 units.

IV. Results

A. Aging Study on Samples A, B, and C

The absorption coefficients α for as-cast acrylic samples A, B, and C, determined by the (RT) method are shown graphically in Fig. 2 from 300 to 500 nm. While there is essentially very little change in absorption above 400 nm where the curves are relatively featureless, there exist very large variations in the range 300-400 nm, the extent of which is sample dependent. The increased absorption at the shorter wavelengths is largely due to the tail of the UV absorption peak, known as the Urbach edge, which has a wavelength dependence approximated by:

$$\alpha \simeq \alpha^{\circ} \exp\left(\frac{A}{\lambda}\right),\tag{9}$$

where A and α° are constants.²⁰

The corresponding values of refractive index n derived for the acrylic samples by the (RT) method were in poor agreement with the literature values. For example, at 400 nm, the computed n values for the three samples ranged from 1.42 to 1.45 compared with a literature value of $1.507.^{21}$ This result is not surprising, since the (RT) method is known to be particularly sensitive to minor experimental errors in the measured reflectance and transmittance. As discussed in Sec. III.C the accuracy of the VW measurements of absolute regular reflectance requires that the optical path lengths traversed with and without the sample in the

beam be identical. For a thick transparent sample, where the reflectance contribution from the back surface is relatively large, this condition is not satisfied. This situation occurs for those acrylic specimens which are \sim 25-mm thick, although the associated systematic error decreases with decreasing wavelength (due to increasing absorption). To reduce this error in n, the (RT) measurements should be performed on a thinner specimen so that the actual displacement of the back surface is minimal. Unfortunately, this measurement is then a less sensitive indicator of α because of reduced absorption losses. An obvious solution to the dilemma is to determine n from an (RT) analysis on a thin acrylic specimen and to use these values as good initial estimates in a Lambert's law analysis of transmittance measurements on two different sample thicknesses to obtain a consistent set of n and α values for the three independent photometric measurements. This procedure is referred to here as the (RTT') method.

A comparison between the (RTT') method and the (RT) method is shown in Table I for acrylic sample A at selected wavelengths. The n and α values were derived from R and T measurements on a 25-mm thick specimen for the (RT) method and from R and T measurements on a 10-mm thick specimen and T measurements on a 25-mm thick specimen for the (RTT')method. Where available, literature n values have been included for comparison. It can be seen that the (RTT') method provides n values much closer to those obtained by more direct means (e.g., measurements of prism refraction or total internal reflection) than the (RT) method. This is not a serious error for our application, since the *n* values are only needed to compute the reflection losses at the acrylic-water interface which are ~0.4%. The reliability of the α values is a

 Table I.
 Comparison of UV Optical Constants Obtained for Unaged Acrylic Sample A by (RT) and (RTT') Methods^a

| | $n(\lambda)$ | | | $\alpha(\lambda) \ cm^{-1}$ | |
|----------------|--------------|--------|-------------------|-----------------------------|--------|
| λ (nm) | (RT) | (RTT') | lit. ⁶ | (RT) | (RTT') |
| 300 | 1.524 | 1.492 | | 0.410 | 0.424 |
| 310 | 1.478 | 1.509 | | 0.091 | 0.093 |
| 320 | 1.470 | 1.516 | | 0.061 | 0.059 |
| 335 | 1.463 | 1.514 | | 0.038 | 0.037 |
| 350 | 1.456 | 1.514 | | 0.025 | 0.022 |
| 365 | 1.452 | 1.510 | 1.514 | 0.017 | 0.014 |
| 385 | 1.450 | 1.511 | | 0.010 | 0.007 |
| 405 | 1.448 | 1.505 | 1.507 | 0.009 | 0.006 |
| | 1 | | | 1 | |

a For details of measurements, see text.

b R.M. Altman and J.D. Lytle, Proc. SPIE 237, 380 (1980).



Fig. 3. Absorption coefficients for acrylic sample A following aging in various liquids.

more serious concern since they have a greater impact on the overall detection efficiency. For a 2.5-cm thick acrylic vessel, the relative difference in attenuation predicted by the two sets of α values in Table I is <5%. Consequently, we adopted the set of $n(\lambda)$ values determined for acrylic sample A by the (RTT') method as representative of the various UV transmitting acrylics and used the simpler (RT) method to obtain $\alpha(\lambda)$ data for each of the acrylic specimens, before and after aging.

Figure 3 gives the α values derived for acrylic sample A, after aging in ultrapure H₂O, D₂O, and a brine solution, as described in Sec. II.A. Comparison with the as-received specimen indicates a similar significant increase in absorption irrespective of the particular liquid into which the acrylic was immersed. Thickness measurements showed that the sample had swollen by ~0.1 mm. These observations were generally true for acrylic samples A, B, and C. However, the extent of the increased absorption upon aging was sample dependent, as can be seen in Fig. 4 which compares α curves for as-received and aged samples B and C. The error bars indicate the range of α values measured for the different immersion liquids.

To obtain complementary data about the degree of light scattering from the various candidate acrylics, diffuse reflectance measurements were carried out as discussed in Sec. III.B. Figure 5 gives an illustration of the procedure for estimating the opaque diffuse reflectance R_{∞} of the acrylic by Eq. (7) from measurements of the diffuse reflectance of the acrylic over a black background R_0 , over a white background R, and of the white background itself R_g .

Since K-M theory does not take account of regular reflection, the K-M equations are only valid for measurements of the diffuse component of reflected flux. Here we have assumed that the instrument's gloss trap excludes this regular component. The validity of this basic approach has been checked (Sec. III.C), but even



Fig. 4. Absorption coefficients for acrylic samples B and C following aging in water.



Fig. 5. Opaque diffuse reflectance, R_{∞} , of unaged acrylic sample A from 300 to 700 nm as derived from one of the Kubelka-Munk solutions in which R_0 is sample reflectance over black felt background, R is the sample reflectance over a white opal background, and R_g is reflectance of white opal glass. See text for further details.

a small systematic error can be significant when the sample reflectance is low. To evaluate the magnitude of this error we performed diffuse reflectance measurements on a piece of high optical quality quartz. This test sample should be representative of the error incurred with the acrylics since its regular reflectance is of the same order as that of the acrylic samples ($\sim 7\%$). If the gloss trap was perfect, its diffuse reflectance should measure zero. Instead, we found this systematic error to be measurably small but increasing with decreasing wavelength, being 0.0002-reflectance units at 700 nm and 0.0007 reflectance units at 300 nm. Since the measured diffuse reflectance of the acrylics over black backing is very low ($\leq 1\%$) this error term is significant. We therefore corrected these diffuse reflectance measurements before computing the acrylic's opaque reflectance, R_{∞} .

The scattering coefficients S were then calculated directly from Eq. (6), after substitution of the previously determined α values from the (RT) analysis. The computed S curves for acrylic sample A, before and after immersion in the various liquids, are shown in Fig. 6 from 300 to 500 nm. The reproducibility of this procedure was independently checked by subjecting a second acrylic specimen to immersion in H₂O and evaluating its S curve in an identical manner. It can be seen that the agreement of this repeat measurement is extremely good, being within $0.0002 \,\mathrm{cm}^{-1}$ of the first set of measurements. Similar S data were determined for acrylic samples B and C. However, their obviously poorer UV optical transmittance characteristics preclude their further consideration for our purposes. All three evaluated acrylics exhibit a large increase in scattering with decreasing wavelength suggesting a predominance of Rayleigh scattering which has a λ^{-4} dependence, but, unlike the α curves, the extent of the increased scattering in the aged samples depends upon the particular immersion liquid. For example, at 300 nm, sample B exhibits a $2.5 \times$ increase in scattering for all three immersion liquids compared with the blank specimen, whereas, sample A exhibits a $2 \times$ increase for immersion in D_2O and a 3 \times increase for immersion in H_2O and in brine. In the visible region, the computed S values for all three unaged acrylic samples range from $(2-5) \times 10^{-4}$ cm⁻¹, with the lowest value being in excellent accord with that determined by Crist and Marhic⁴ at 570 nm using a light scattering goniometer $(S = 2.3 \times 10^{-4})$. However, these authors also report α values of $\sim 10^{-4}$ cm⁻¹ from 488 to 680 nm which are almost an order of magnitude smaller than those determined in this study. There are two plausible explanations for this discrepancy. First, Crist and Marhic's study was conducted on extremely pure optical grade acrylic and not on commercially available UV-grade acrylic. Second, the inherent accuracy of the (RT)method is insufficient to determine extremely small α values of $\sim 10^{-4}$ cm⁻¹. However, it is adequate at extracting α data (to better than 10%) at the level of 10^{-2} cm⁻¹ assuming relatively small experimental errors in R and T ($\leq 1\%$). This is the situation encountered here in the characterization of UV optical constants of UV transmitting acrylics and, therefore, does not pose a serious impediment to our application.

B. Aging Study on Samples D and E

The values of α derived for as-cast acrylic samples D and E, before and after aging in ultrapure H_2O are shown in Fig. 7 from 300 to 400 nm. It can be seen that these specimens are generally superior to the previous candidates A, B, and C, both in terms of their reduced absorption losses in the UV region, and also their retention of these favorable characteristics after aging. The aged samples D and E also increased by only $\sim 0.1\%$ and $\sim 0.2\%$, respectively, in thickness as compared with an $\sim 0.4\%$ increase with aged samples A, B, and C. The absorption coefficient results for the aging study on the thermoformed versions of samples D and E are shown in Figs. 8 and 9. The designations BM and RF refer to the thermoforming procedure used, i.e., over bare metal or over metal coated with rubber flocking, respectively. It can be seen that the RFprocess produces a slightly superior acrylic, although both thermoformed samples exhibit improved retention of their optical properties compared with the unthermoformed samples. While the optical transmittance of the as-cast and thermoformed versions of sample D are closely similar, those of sample E are



Fig. 6. Scattering coefficients for unaged acrylic sample A computed from Kubelka-Munk remission function and from α -data derived from (*RT*) analysis. See text for further details.



Fig. 7. Absorption coefficients for blank and aged specimens of acrylic samples D and E.



Fig. 8. Absorption coefficients for thermoformed blank and aged specimens of acrylic Sample D. BM and RF refer to whether the thermoforming took place over bare metal or rubber flocked metal,





Fig. 9. Absorption coefficients for thermoformed blank and aged specimens of acrylic sample E. BM and RF refer to whether the thermoforming took place over bare metal or rubber flocked metal, respectively.

profoundly different, with the thermoformed samples exhibiting significantly higher absorption.

V. Discussion

This investigation of the optical properties of UVgrade acrylics has answered a number of questions regarding its use in the proposed SNO detector, but which also have bearing on design of other optical systems. The optical transmittance properties vary significantly, particularly in the UV region, for acrylic samples from different manufacturers and even different formulations or batches from the same manufacturer. For example, the absorption coefficients at 300

nm for as-cast unaged acrylics vary by a factor of 2.5, ranging from 0.29 to 0.72 cm⁻¹. Since it is impractical to evaluate all of the acrylic raw material to be used in the SNO detector, the material and processing variables will have to be carefully controlled by the manufacturer. Regarding the weatherability of the acrylics. it was found generally that prolonged immersion of the as-cast specimens in the various liquids led to an overall deterioration in optical transmittance, and an increase in scattering, the extent depending on the particular acrylic under study, with the greatest differences occurring in the wavelength interval 300-400 nm. However, sample D exhibited minimal deterioration in its optical transmittance upon aging or upon thermoforming. It is important to note that the surfaces of the acrylics did not visibly craze due to long term exposure to ultrapure water, heavy water or brine. It is also interesting that the thermoformed acrylics appeared to be more durable than the corresponding as-cast samples in retaining their optical properties after long-term liquid immersion.

The acrylic absorption data obtained in this study, in conjunction with the previously determined D_2O and H_2O absorption data,² has enabled the performance of the proposed SNO neutrino detector to be modeled. Given a hypothetical neutrino event at the center of the D_2O volume, the resultant Cerenkov radiation traverses 5 m of D_2O , 5 cm of acrylic wall, and 2.5m of H₂O before being recorded at the photomultiplier tubes. Using absorption data for thermoformed specimens of samples D and E [Figs. 8 and 9] and normalizing to the photomultiplier quantum efficiency response,⁷ the transmittance of Cerenkov radiation is 0.685 (sample D) and 0.668 (sample E). Since the transmittance for 5-cm thick acrylic alone is 0.843 (sample D) and 0.815 (sample E), absorption losses are shared approximately equally between water and acrylic. While the transmittances for aged and blank unthermoformed versions of sample E are 0.899 and 0.918, respectively, the thermoforming process drastically lowers the transmittances for aged and blank specimens to a value of 0.815 ± 0.003 . In contrast, for sample D, there was essentially little difference in transmittance for thermoformed and unthermoformed specimens and for aged and blank specimens; the value for transmittance was 0.843 ± 0.008 . Since the acrylic vessel will be largely thermoformed, this 3.4% gain in transmitted radiation for thermoformed sample D over thermoformed sample E is important for optimizing the neutrino detector, and is considerably more cost-effective than increasing the photocathode coverage.

The Monte Carlo reconstruction of neutrino events in the SNO detector also requires information on the relative amounts of absorption and scattering of Cerenkov photons. In Table II, the absorption and scattering coefficients, and S/α percentages for blank and immersed (in D₂O) specimens of sample A acrylic are summarized. In current simulations, input parameters of S/α which vary from 0.5% at 300 nm up to ~4.1% in the visible region for immersed acrylic, are used.

 Table II. Absorption and Scattering Coefficients for Sample A

 Blank Specimen

| Wavelength (nm) | α | Scatt. Coeff. | S/α |
|-------------------|------|---------------|------------|
| 300 | .410 | .00119 | 0.3 % |
| 325 | .055 | .00084 | 1.5 % |
| 350 | .025 | .00054 | 2.2 % |
| 400 | .009 | .00023 | 2.6 % |
| 450 | .009 | .00023 | 2.6 % |
| 500 | .007 | .00020 | 2.8 % |

Specimen Immersed in D₂O

| Wavelength (<i>nm</i>) | α Scatt. Coeff. | | S/a |
|--------------------------|-----------------|--------|-------|
| 300 | .488 | .00250 | 0.5 % |
| 325 | .099 | .00186 | 1.9 % |
| 350 | .043 | .00112 | 2.6 % |
| 400 | .013 | .00045 | 3.5 % |
| 450 | .009 | .00037 | 4.1 % |
| 500 | .006 | .00025 | 4.2 % |

Note: α and S in cm^{-1} .

VII. Conclusions

Optical properties of several premium UV transmitting acrylics have been determined as a function of wavelength via spectrophotometric methods. These acrylic specimens are proposed for use in a heavy water Cerenkov detector (SNO). The results suggest that the optical properties, particularly in the UV region, depend on the manufacturer, the fabrication, and the aging history. While immersion aging in H_2O , D_2O , and brine similarly affect their absorption properties, this is not necessarily the case for the scattering properties. Thermoformed specimens appear to display greater integrity of their optical properties to immersion aging compared with unthermoformed specimens. Acrylic samples A and D tested here exhibit the best optical performance for the SNO detector and their measured absorption, scattering and refractive data have been used in Monte Carlo simulations of the detector response with respect to reconstruction of neutrino events. The proposed SNO detector has received (Jan. 1990) final approval with respect to funding, and has moved to project status.

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