Ellipsometric measurements of the refractive indices of linear alkylbenzene and EJ-301 scintillators from 210 to 1000 nm

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Abstract
We report on ellipsometric measurements of the refractive indices of linear alkylbenzene–2,5-diphenyloxazole (LAB–PPO), Nd-doped LAB–PPO and EJ-301 scintillators to the nearest ±0.005, in the wavelength range 210–1000 nm.

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1. Introduction

There is at present a great deal of interest in the use of linear alkylbenzene (LAB) as a scintillator solvent for neutrino experiments (e.g. SNO+, LENA, RENO and Daya Bay [1–4]). First proposed as a scintillator by Chen [1], this organic liquid has the advantages of low toxicity, good light yield, high flash-point and long attenuation length. Furthermore, it is compatible with acrylic, a material frequently used for building the scintillator containment vessel.

To understand the propagation of light and reconstruct events in these experiments, and to accurately calculate the Cherenkov light production, it is imperative to know the refractive index of the liquid scintillator as a function of wavelength. Currently, our knowledge of the refractive index of LAB-based scintillators is limited to the optical range. Recently, the RENO collaboration reported a measurement of the refractive index of LAB at six wavelengths in the interval 400 < λ < 630 nm using the minimum deviation technique [5]. In this paper, we describe a measurement of the real component of the refractive index, n, of two LAB-based scintillators to the nearest ±0.005 at 500 points from 210 to 1000 nm using an ellipsometric method [6, 7]. These two scintillators were LAB with 3 g l⁻¹ of 2,5-diphenyloxazole (PPO) and LAB–PPO loaded with 0.094% Nd by weight.

We also report on a measurement of n for the EJ-301 scintillator [8], which is equivalent to the very commonly used NE-213 and BC-501A scintillators. As far as we know, to date the dispersion of NE-213 has not been measured down to the UV region.

2. Experimental method

The use of ellipsometers to measure the refractive indices of liquids is convenient but not widespread. Because of challenges brought about by surface vibrations, evaporation and difficulties in controlling the temperature of the liquid medium, the ellipsometric method is not expected to be as competitive as the minimum deviation technique for precision refractive index measurements. However, measurements accurate to the third decimal place are possible. In this work, we have adopted the simplest ellipsometric configuration, in which polarized light reflects off an infinitely deep layer of liquid. This is described below.

2.1. Theory

Consider polarized light incident on the interface between two media, both of which are infinite and homogeneous (figure 1(a)). After reflection, there is a change in polarization that is dependent on the incident angle, the refractive indices of the two media and the initial polarization state. The ellipsometer measures the change in polarization in terms of
two parameters, $\Delta$ and $\Psi$. If $R_p$ and $R_s$ are the reflection coefficients of the electric field components parallel ($E_p$) and perpendicular ($E_s$) to the plane of incidence, the ellipsometric parameters are given by

$$\frac{R_p}{R_s} = \tan \Psi e^{i\Delta}. \quad (1)$$

$R_p$ and $R_s$ are expressed in terms of the complex refractive indices of media 1 and 2 ($N_1$ and $N_2$), and the angles $\phi_1$ and $\phi_2$ (figure 1(a)) by Fresnel’s equations. Combining Fresnel’s equations and Snell’s law with equation (1) and separating real and imaginary components, one obtains

$$n_2^2 - k_2^2 = n_1^2 \sin^2 \phi_1 \left[ 1 + \frac{\tan^2 \phi_1 \cos^2 2\Psi - \sin^2 \Delta \sin^2 2\Psi}{(1 + \sin 2\Psi \cos \Delta)^2} \right] = X, \quad (2)$$

$$2n_2k_2 = \frac{n_1^2 \sin^2 \phi_1 \tan^2 \phi_1 \sin 4\Psi \sin \Delta}{(1 + \sin 2\Psi \cos \Delta)^2} = Y, \quad (3)$$

where $n_1$ and $n_2$ are, respectively, the real components of $N_1$ and $N_2$, and $k_2$ is the imaginary component of $N_2$. Given $\Delta$, $\Psi$, $\phi_1$ and $n_1$, one can therefore solve for $n_2$ and $k_2$ separately:

$$n_2^2 = \frac{1}{2} (X + \sqrt{X^2 + Y^2}), \quad k_2^2 = \frac{Y}{2n_2^2}. \quad (4)$$

In our case, medium 1 is air, and medium 2 is the liquid scintillator whose real refractive index component ($n_2$) is to be measured. The sensitivity to $k_2$ is weak, since the measurement is made on the reflected light component and not the transmitted light component, which conveys most of the information on attenuation in medium 2.

2.2. Setup

For equations (2) and (3) to be valid, one must have: (i) an ideal interface and (ii) an infinitely deep medium 2 (so that no interference occurs with light reflected off the bottom of medium 2). To emulate a deep container with a flat liquid surface, we followed the method of Synowicki et al [6]: a few drops of liquid were allowed to spread into a thin film on a sandblast-frosted microscope glass slide (figure 1(b)). Measurements were then made with the light reflected specularly off the upper surface of the liquid film.
Table 1. The Sellmeier coefficients for LAB–PPO, Nd-doped LAB–PPO and EJ-301. The LAB–PPO and Nd-doped LAB–PPO results are valid for the wavelength range 230–1000 nm, while the EJ-301 results apply for the range 300–1000 nm. The $C_i$ coefficients are given in nm.

| Liquid       | $B_1$    | $C_1$         | $B_2$    | $C_2$         | $B_3$    | $C_3$         | $B_4$    | $C_4$         |
|--------------|----------|---------------|----------|---------------|----------|---------------|----------|---------------|
| LAB–PPO      | 0.821384 | 94.7625       | 0.311375 | 160.751       | 0.0170999| 219.575       | 0.608268 | 9385.54       |
| Nd LAB–PPO   | 0.80559  | 98.7814       | 0.325456 | 157.743       | 0.0175545| 219.323       | 7.53674  $\times 10^{-5}$ | 7301.7   |
| EJ-301       | 0.715235 | 1.29988       | 0.464991 | 203.739       | 0.0302529| 0.484641      | 1.99773  | 147.196       |

Table 2. The coefficients for third-order polynomial fits to LAB–PPO and Nd-doped LAB–PPO refractive indices in the wavelength range 210–230 nm.

| Liquid       | $A_0$    | $A_1$         | $A_2$    | $A_3$         |
|--------------|----------|---------------|----------|---------------|
| LAB–PPO      | −209.56  | 2.81043       | −0.012012| 4283.1        |
| Nd LAB–PPO   | −206.13  | 2.76412       | −0.012220| 3795.91       $\times 10^{-5}$ |

The transmitted component was diffusely reflected off the frosted glass and did not affect the results. The slides, as well as all other glassware used in this work, were first cleaned in an ultrasound bath. Data were recorded at five values of $\phi_1$, namely 55°, 60°, 65°, 70° and 75°. The experiment was performed at room temperature (25°C) with a variable-angle Woollam M2000 ellipsometer fitted with a Xe lamp. Excluding alignment procedures, a spectral scan at one angle typically took around 30 s.

3. Results

3.1. LAB–PPO and Nd-doped LAB–PPO

The results for LAB–PPO are shown in figure 2(a). The measurements at the five different angles agree within about ±0.005 over 200–1000 nm. The solid curve is the average of the five angles, while the shaded area shows the standard deviation. The circle symbols are measurements from RENO [5], obtained using the minimum deviation technique. The diamond symbols are previous measurements taken by the SNO+ collaboration [10] using an Abbe refractometer and other methods. The Nd-doped LAB–PPO results are shown in figure 2(b). The values are rather close to undoped LAB–PPO, the difference being within ±0.5% over the entire wavelength range under study.

For the wavelength range 230–1000 nm, where the refractive indices of both liquids decrease monotonically with wavelength $\lambda$, we parameterize the dispersion with the Sellmeier formula

$$n^2(\lambda) = 1 + \sum_{i=1}^{N} \frac{B_i}{1 - (C_i/\lambda)^2}$$

(5)

with $N = 4$. For the range 210–230 nm, we fit to a third-order polynomial:

$$n = A_0 + A_1 \lambda + A_2 \lambda^2 + A_3 \lambda^3.$$  

(6)

In both equations (5) and (6), $\lambda$ is in nm. The Sellmeier and polynomial coefficients are shown in tables 1 and 2, respectively. The fit residuals are shown in figure 3.

3.2. EJ-301

The EJ-301 results are shown in figure 4(a). There appears to be a region of anomalous dispersion just below 230 nm. The measurement uncertainty increased markedly below 220 nm. To parameterize the EJ-301 refractive index, we fit to the Sellmeier equation (equation (5)), with $N = 4$ in the region 300–1000 nm, and to two separate fourth-order polynomials in the regions 210–240 and 240–300 nm. The Sellmeier coefficients are given in the third row of table 1, while the
Table 3. The coefficients for fourth-order polynomial fits to the EJ-301 refractive index in the wavelength ranges 210–240 and 240–300 nm.

| Wavelength range | $A_0$      | $A_1$      | $A_2$      | $A_3$      | $A_4$      |
|------------------|------------|------------|------------|------------|------------|
| 210–240 nm       | 11784.7388 | −205.807243 | 1.34648820 | −3.9107264 × 10^{-3} | 4.25436903 × 10^{-6} |
| 240–300 nm       | 183.668    | −2.51192   | 0.0129879  | −2.98047 × 10^{-5}  | 2.55974 × 10^{-8}  |

Figure 4. (a) Measured real refractive index components for EJ-301. The value at 589.3 nm is from [8]. (b) Residuals of fits to two fourth-order polynomials in the wavelength regions 210–240 and 240–300 nm and to the Sellmeier equation in the region 300–1000 nm.

fourth-order polynomial coefficients are shown in table 3. The fit residuals are given in figure 4(b).

4. Conclusions

We carried out ellipsometric measurements of the refractive indices of EJ-301, LAB–PPO and Nd-doped LAB–PPO scintillators between 210 and 1000 nm. Our measurements are accurate to ±0.005. Empirical formulae for the index of each scintillator are provided in the paper.

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