Turk J Phys
26 (2002) , 41 – 47.

Experimental Investigation of Temperature Effect on the Refractive Index of Dye Laser Liquids

Ş. YALTKAYA
University of Akdeniz, Faculty of Science and Arts,
Department of Physics, Antalya-TURKEY
e-mail: seref@pascal.sci.akdeniz.edu.tr
R. AYDIN
Middle East Technical University, Faculty of Science and Arts,
Department of Physics, Ankara-TURKEY

Received 19.03.2001

Abstract

In this work, refractive index of selected dye laser solutions were measured by fiber optic sensor. The fiber optic probe was dipped into the liquids and according to Fresnel's Reflection Law, refractive index values were obtained at the wavelength of the laser utilized. The solutions consisted of Rhodamine 6G, Rhodamine B and Coumarin 481 dissolved in methanol, and ethylene glycol, respectively. Refractive index variations of the dye solutions and their solvents with temperature were measured and the refractive index temperature coefficient dn/dT were calculated at the wavelength of 780 nm.

1. Introduction

The thermal coefficient values of refractive index is in increasing demand in many optical applications. Most of the time there is no data available in the literature for various mixed liquids such as dye laser active material solutions. In recent years, due to the rapid improvement in fiber optic technology and sensing devices, and in addition to the many different types of non fiber optics techniques available for measuring refractive index of liquids [1], [2], [3], fiber optic sensors have became one of the major techniques in measuring refractive index of solutions due to its flexibility, small size, and ease in which optical sensors can be connected to their related electronics [4],[5],[6],[7].

Our experimental apparatus is similar to Pierscionek's setup [8] and, limitedly, to the design given in reference [9], wherein multimode fiber and diode laser are used instead of single mode fiber and gas laser, and the data acquisition was via computer.

The aim of this study is to measure the variation of refractive index of a solution whose temperature variation occurs via continuous heating or cooling processes, using a fiber optic refractive index sensor. Selected laser dye solutions and their solvents were used, for each of which a graph is drawn exhibiting temperature versus refractive index values and the thermal refractive index coefficient for the material as calculated from the slope of the line formulated with the least squares method, at the known temperature interval.

The thermal coefficient of refractive index of materials is a significant optical parameter, especially for the dye laser solution, which is an active medium. If dye solutions exhibit appropriate thermal coefficient refractive index in addition to other required physical properties (e.g., spectral range, viscosity) it can be considered for a laser dye solution.

Since the active medium of the dye laser is in the form of solution, thermal coefficient of refractive index of solutions may become a significant optical parameter. As a result of the pumping process for CW dye laser resonator operation, the thermal coefficient of the refractive index becomes more important than the pulsed case due to elevated temperature at the center of the dye solution in the jet stream. The focal length of the thermally induced lens at the center of the focussed pump beam is estimated from the formula [10] [11]

$$f_{center} = \frac{\sqrt{2\pi}\ v w_p^2}{4PA\varepsilon_1 k_{th}}. \tag{1}$$

Here, $v$is the flow velocity of the jet; w_p the pump beam waist radius; ε_1 the fraction of the absorbed pump power converted to heat; P the pump power; A the fraction of the total pump power absorbed in the jet; and k_{th} is the thermo-optical constant of the solution, given by

$$k_{th} = \frac{1}{\rho\, C_p}\frac{dn}{dT}, \tag{2}$$

where ρ is the density; C_p the specific heat and $\frac{dn}{dT}$ is the thermal coefficient of refractive of the dye laser solution.

By utilizing Fresnel's fundamental reflection law [12], the intensity of the reflected light from the boundary surface was measured at 780 nm from which the refractive index of the second medium was calculated. The optical fiber collects the whole-reflected light from the boundary surface and fulfils the small angle criteria. At small angles ($< 10°$) reflection coefficient for both parallel and perpendicular polarisation state is the same. Determination of the effective refractive index of the fiber, used as a sensor probe, is the most important parameter for further calculations, and was also measured at wavelength of 780 nm.

2. Experimental procedure

The sensor arrangement shown in Fig.1 is based on highly intensity-stable, single mode diode laser at 780 nm (FWHM) wavelength (Lasermax, 15 mW max.). During the experiment the light intensity of the laser diode was recorded with an ADC (Analog Digital Converter, Keithley Metrabyte; DAS-801) coupled detector (Detector 1 in Fig. 1), and a computer.

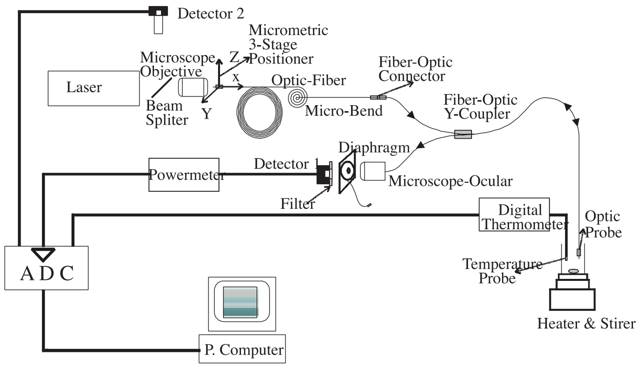

Figure 1. The experimental diagram for determining the refractive index values at various temperatures.

The laser beam was launched into the optical fiber (multi-mode, 50-μm-core diameter) by a fiber coupler (Newport F-916) and a microscope lens (20X, 12:2, Olympos). A mode filter (form of nine turns of fiber wound on a mandrel of about 2 cm diameter) was placed to eliminate unwanted modes and ensure the small angle criteria at boundary surface at the interface of the second media and the fiber core. One arm of the pigtailed Y-coupler (3dB coupler) was connected to the mode filter. The other pigtailed arm was positioned at detector 1 and the remaining arm was used as a sensor probe, as depicted in Fig. 1. The sensor probe, with a ST type connector, is fixed firmly during the experiment. Detector 2 (a silicon photodiode), detector 1 (powermeter, Coherent Radiation P200; 0.1-100 mW) and the temperature sensor were connected to the

ADC's input channels. The data acquisition lasted 15 minutes, and signals coming to the three input channels of the ADC at the same time (max. 1 μs time intervals) were collected with a speed of 8-10 datum per second. To convert analog voltages output of the powermeter and values the necessary calibration equations were derived. Temperature of the dye solutions and the solvents were increased via a magnetic stirrer-heater, with stirring at constant speed to obtain uniform temperature increase. For this experimental setup, the necessary equations to calculate the refractive index of the laser dye solutions and their solvents also can be obtained from reference [8].

The amount of light reflected from the boundary between two mediums (media 1 and media 2) depends not only on the change in refractive index and the angle of incidence θ_i at the interface but also on the polarisation state of the incident light. Therefore, the reflection coefficients, which determine the intensity of the reflected light, are computed as:

$$\Gamma_{\perp}^{b}\big|_{\mu_1=\mu_2} = \frac{\cos\theta_i - \sqrt{\frac{\varepsilon_2}{\varepsilon_1}}\sqrt{1-\left(\frac{\varepsilon_1}{\varepsilon_2}\right)\sin^2\theta_i}}{\cos\theta_i + \sqrt{\frac{\varepsilon_2}{\varepsilon_1}}\sqrt{1-\left(\frac{\varepsilon_1}{\varepsilon_2}\right)\sin^2\theta_i}}, \tag{3a}$$

$$\Gamma_{\shortparallel}^{b}\big|_{\mu_1=\mu_2} = \frac{-\cos\theta_i + \sqrt{\frac{\varepsilon_1}{\varepsilon_2}}\sqrt{1-\left(\frac{\varepsilon_1}{\varepsilon_2}\right)\sin^2\theta_i}}{\cos\theta_i + \sqrt{\frac{\varepsilon_1}{\varepsilon_2}}\sqrt{1-\left(\frac{\varepsilon_1}{\varepsilon_2}\right)\sin^2\theta_i}}, \tag{3b}$$

where ε_1, ε_2 are the permittivity of medium 1 and medium 2, respectively. $\Gamma_{\perp}^{b}$ and $\Gamma_{\shortparallel}^{b}$ are known as the plane wave Fresnel reflection coefficients for perpendicular and parallel polarisation, respectively [13]. For most of the dielectric media (excluding ferromagnetic materials) permeability of the medium is given by $\mu_1 \cong \mu_2 \cong \mu_0$.

When the incident light is travelling close to the interface surface normal ($\sin\theta \approx \theta$ and $\cos\theta \approx 1$) instead of the permittivity one can use the refractive index of the core, that is n_f can be substituted in to the reflection coefficients (i.e. $\sqrt{\varepsilon_1\mu_1} = n_1 = n_f$), and both Fresnel reflection coefficients reduces to $\frac{n_f-n_2}{n_f+n_2}$. For ($n_f > n_2$) the reflection coefficient is negative. If the incident light intensity is I_0, the reflected light intensity is I and the second media is air, then the reflectance R becomes equal to the square of the reflection coefficient:

$$\frac{I}{I_0} = R = \left(\frac{n_f - n_{air}}{n_f + n_{air}}\right)^2 + S, \tag{4}$$

where S is the spurious light intensity, originating from the experimental set up. To calculate the magnitude of S, we assume that there is a fictitious reflecting surface in the optical fiber setup. When the optical fiber probe is immersed in the sample, whose refractive index n_x will be measured, the reflected light intensity is I_x. Then a new parameter K can be defined as follows:

$$K = \frac{I_x}{I} = \frac{\frac{I_x}{I_0}}{\frac{I}{I_0}} = \frac{\left(\frac{n_f-n_x}{n_f+n_x}\right)^2 + S}{g+S}, \tag{5}$$

where $g = \left(\frac{n_f-1.00029}{n_f+1.00029}\right)^2$ and $n_{air} = 1.00029$. Finally, the refractive index of the sample is

$$n_x = \frac{n_f\left\{1 - [(g+S)K - S]^{\frac{1}{2}}\right\}}{1 + [(g+S)K - S]^{\frac{1}{2}}}. \tag{6}$$

If the fiber-optic probe is immersed in a fluid whose refractive index is equal to that of the fiber probe (core), reflected light intensity I_m can be considered as the intensity of spurious light. If I_0 (incident light intensity at the fiber-sample interface) is measured, S can be calculated easily; however, it is not possible to get the exact value of I_0. For this reason a new parameter S' is defined:

$$S' = \frac{I}{I_m} = \frac{\frac{I}{I_0}}{\frac{I_m}{I_0}} = \frac{S+g}{S} \ . \tag{7}$$

S can be calculated via the following expression:

$$S = \frac{g}{S'-1}. \tag{8}$$

To measure the background (spurious) radiation, trichloroethylene was used since its refractive index matches the refractive index of the fiber core. To obtain the refractive index of trichloroethylene at wavelength 780 nm the Cauchy dispersion formula was applied. To perform this calculation the refractive index of trichloroethylene was measured at different wavelengths by using an Abbe 60 refractometer (Bellingham & Stanley).

The response of the sensor has been tested over various refractive index ranges and compared with the theory (Eq. 4), and is shown in Fig. 2. In order to measure the refractive index of the fiber core, trichloroethylene was used, and as its temperature was raised gradually from 2°C to 32°C, the refractive indexes were measured. The data acquisition time was about 32-second with 100 data per second. Fig. 3 shows the variation of the refractive index of trichloroethylene with temperature, a relationship which can be represented with a second order polynomial. Refractive index of trichloroethylene is calculated, at the specified temperature, from the corresponding minimum point of the curve by using $(dn/dT)_D$ value [14] and is taken to be equal to the refractive index of the fiber core.

3. Results and discussion

In this study, Rhodamine 6G, Rhodamine B and Coumarin were dissolved separately in methanol, ethanol and ethylene glycol in the concentration of around 10^{-3} molar. Data acquisition was initiated when temperature-refractive index linearity was reached.. During the run of the experiment, fluctuations in the intensity of reflected beam from the boundary surface was observed due to the heating-induced haphazard variations of the laser (diode lasers are more suitable than gas lasers in this respect) and small refractive index variations due to the temperature gradient at the tip of the probe.

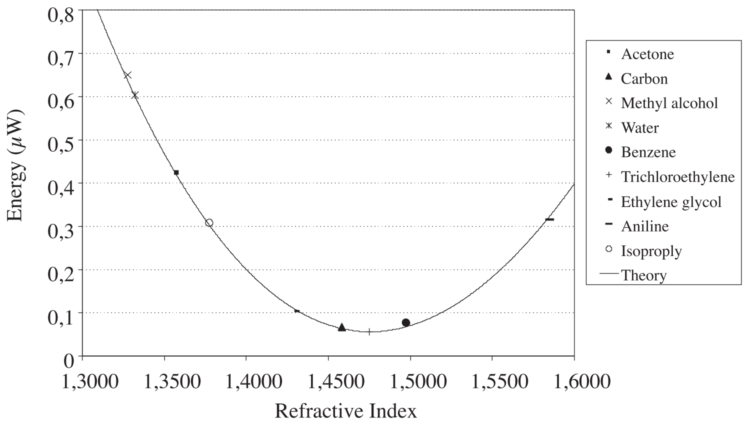

Figure 2. The reflected power distribution from the optic-probe versus refractive index values for several selected substances.

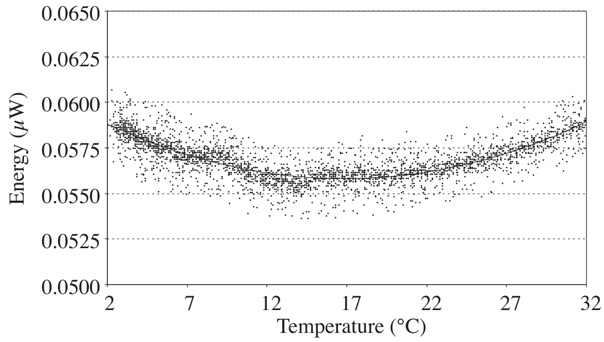

Figure 3. The reflected power distribution from the optic-probe versus various temperatures of trichloroethylene for calculating the refractive index of the fiber core (optic-probe).

Figures 4-7 show the temperature dependence of the refractive index at 780 nm wavelength in temperature interval 5°C - 50°C for pure ethylene glycol (Merck, >99%) and the dye laser solutions. The variation of the refractive index with temperature i.e. $(dn/dT)_{780}$ is assumed almost linear and is fitted to all graphs. The slope of the lines, $(dn/dT)_{780}$ values, and their standard errors are computed and shown in Table. Since the accumulated data is quite large in number, standard errors become rather low. Figures 8-11 show the temperature dependence of the refractive index at 780 nm in the temperature interval 5°C - 40°C for pure methanol (Merck >99%) and the dye laser solutions. It was found that the results obtained for the solvents were in very close agreement with literature values around wavelength of 780 nm that shown in Table 1.

Table 1. dn/dT values and standard errors.

Substance	$-\frac{dn}{dT}(°C^{-1})$ Experimental	$-\frac{dn}{dT}(°C^{-1})$ Literature
Ethylene glycol ($C_2H_6O_2$)	2.958 x10^{-4}± 1.47x10^{-7}	2.6 x10^{-4} (H_α) [14]
($C_2H_6O_2$)+ Rhodamin 6G ($C_{28}H_{31}N_2O_3$)	3.435 x10^{-4}± 2.24x10^{-7}	
($C_2H_6O_2$) + RhodaminB ($C_{28}H_{31}N_2O_3$)	3.335 x10^{-4}± 1.93x10^{-7}	
($C_2H_6O_2$) + Coumarin ($C_9H_6O_2$)	3.205 x10^{-4}± 2.17x10^{-7}	
Methanol (CH_3OH)	4.185 x10^{-4}± 9.30x10^{-7}	4.0 x10^{-4} (H_α) [14]
(CH_3OH)+ Rhodamin 6G ($C_{28}H_{31}N_2O_3$)	4.863 x10^{-4}± 9.58x10^{-7}	
(CH_3OH)+ RhodaminB ($C_{28}H_{31}N_2O_3$)	4.806x10^{-4}± 9.513x10^{-7}	
(CH_3OH)+ Coumarin ($C_9H_6O_2$)	4.9 x10^{-4}.± 2.52 x10^{-5}	

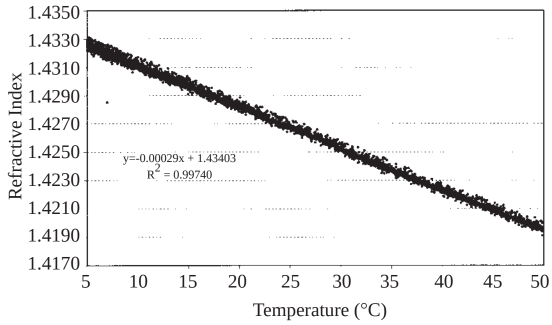

Figure 4. The variation of refractive index of ethylene glycol with temperature.

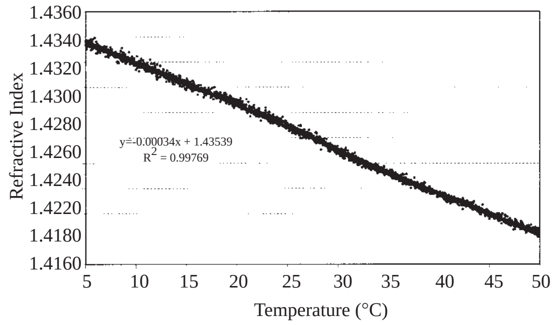

Figure 5. The variation of refractive index of Rhodamin 6G dissolved in ethylene glycol with temperature.

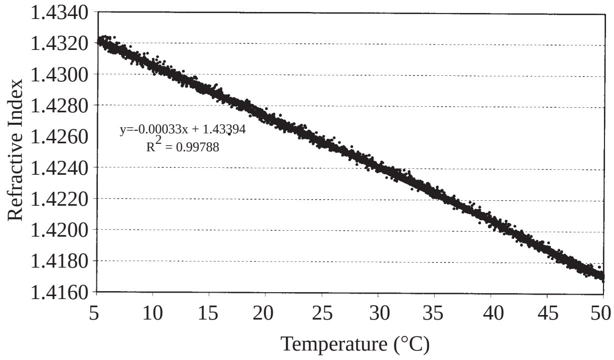

Figure 6. The variation of refractive index of Rhodamin B dissolved in ethylene glycol with temperature.

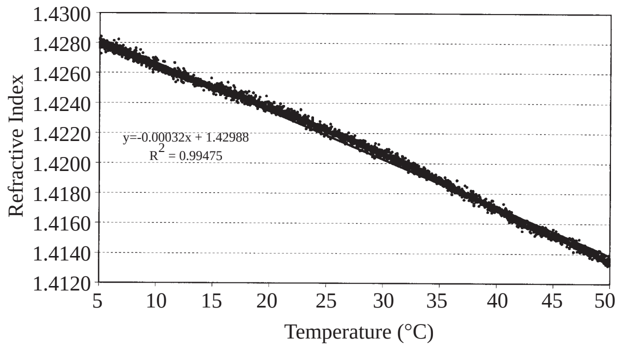

Figure 7. The variation of refractive index of Coumarin dissolved in ethylene glycol with temperature.

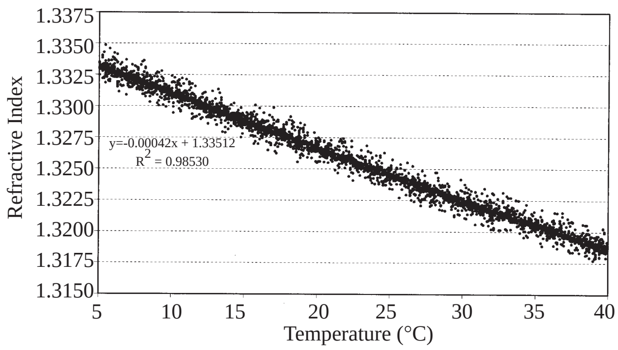

Figure 8. The variation of refractive index of methanol with temperature.

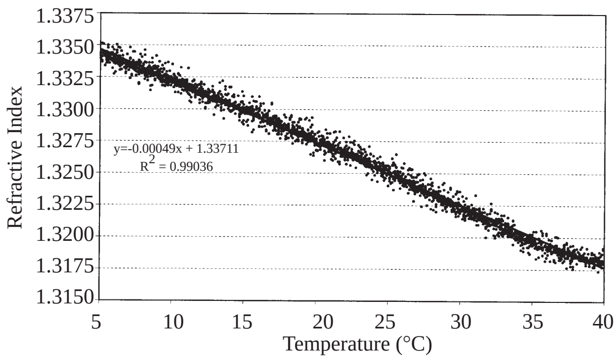

Figure 9. The variation of refractive index of Rhodamin 6G dissolved in methanol with temperature.

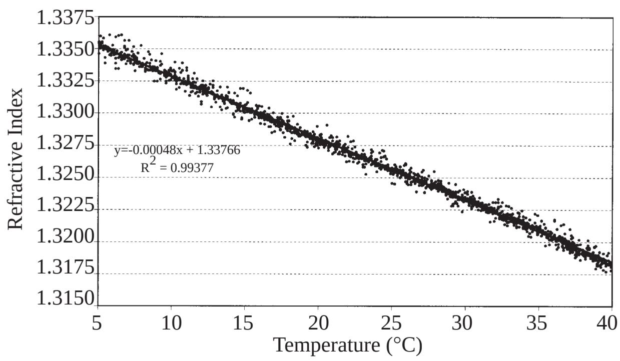

Figure 10. The variation of refractive index of Rhodamin B dissolved in methanol with temperature.

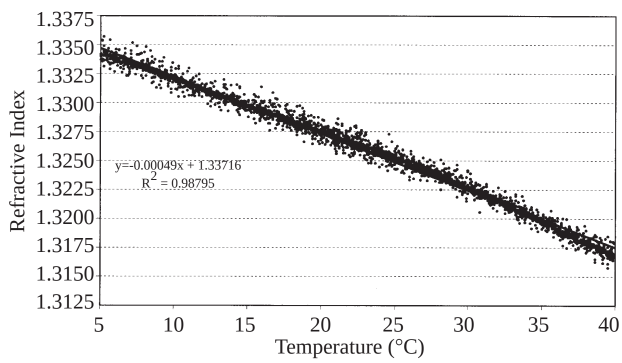

Figure 11. The variation of refractive index of Coumarin dissolved in methanol with temperature.

Acknowledgments

We are indebted to M. S. Kormalı, A. Alaçakır and İ. Aydemir.

References

[1] R.S. Longhurst, Geometrical and Physical Optics, (Wiley, New York 1967).

[2] B.W. Grange, W. H. Stevenson and R. Viskanta, *Appl. Optics,* **15,** (1976) 858.

[3] M de Angelis, S De Nicola, P Ferraro, A Finizio and G. Pierattini, *Pure Appl. Opt.*, **5**, (1996) 761.

[4] B. Culshaw and J. Dakin, Optical Fiber Sensor, Application Analysis and Future Trends (Artech House Pub. 1997).

[5] Takashi Takeo and Hajime Hattori, *Jap. Journal of App. Phy.* **21,10**, (1982) 1592.

[6] T.L. Bergman, F. P. Incropera and W.H. Stevenson, *Rev. Sci. Instrum.,* **56,2**, (1985) 291.

[7] I. K. Ilev, *App. Optics,* **34,10** (1995) 1741.

[8] B. K. Pierscionek, *J. Opt. Soc. Am.*, **A 10, 9**, (1993) 1867.

[9] Farhad Ansari and Qi-Yu Chen, *App. Optics,* **30,28,** (1991) 4056.

[10] B. Welleghausen, H. Welling and R. Beigang, *Appl. Phys.*, **3**, (1974) 387.

[11] P. Anliker and H. P. Weber, *Opt. Commun.,* **19,** (1976) 197.

[12] M. Born and E. Wolf, Principles of Optics (Pergamon Press, 1959).

[13] C. A. Balanis, Advanced Engineering Electromagnetics (John Wiley & Sons, 1989).

[14] J. Timmermans, Physico-Chemical Constants of Pure Organic Compounds (Elsevier Publishing Company, second reprint 1965).