

Optical transmission property of electro-optic dye-doped polymer waveguide

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Optical transmission of electro-optic effect polymers from dye-doped acrylate copolymer and guest–host polysulfones was evaluated with varying electric fields at room temperature. The dye-doped copolymer behaved differently from the guest–host polymer system upon the application of the electric field. Time-dependent phenomena were obtained due to relaxation. The dye hyperpolarizability affected the manner in which the dye is poled.

Keywords: doped-acrylate copolymer with DR-1, optical transmission, electro-optic effect.

1. Introduction

Interest in linear and nonlinear optics has grown tremendously in recent years due to the telecommunication needs for high bandwidth optical switching and processing devices. In optical telecommunications, there is a great demand for materials whose optical propagation properties can be influenced by an electric field. For slow switching rates, liquid crystals that have very high electro-optic coefficients are used due to the mobility of the whole molecule. For higher switching rates, materials with electro-optic properties, such as the Pockels effect and the Kerr effect, can be used. Doped polymeric materials offer high optical quality and potentially large, bulk nonlinear susceptibilities if the molecular hyperpolarizability of the dopant is large [1]. Various studies have been performed, namely, by SMALL *et al.* [2], who found the electro-optic effect of organic dyes in PMMA using extremely strong electric fields (from 30 V/ μm), by KURIHARA *et al.* who worked on acrylate copolymers [3] and by KUZYK and DIRK on various dyes [4]. Azo dyes are of particular interest because they are readily prepared with a wide range of donor and acceptor groups and are systems

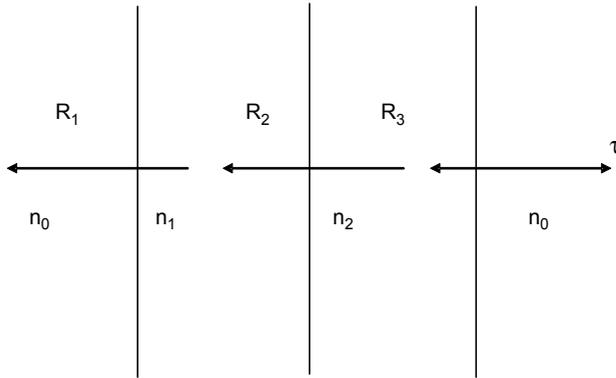


Fig. 1. Power τ transmitted perpendicularly through materials with different refractive indices n_0 , n_1 and n_2 .

that provide the excellent conjugation necessary for high molecular susceptibilities. In addition to guest–host polymers, side-chain copolymer, main-chain and cross-linked polymers could be employed [5–7].

2. Theory

2.1. Power reflectance and transmittance

Assuming that the input power in Fig. 1 is perpendicular to the boundary, the output power, the power reflected at normal incidence, can be written in the form of a Fresnel reflection by a simple derivation using complex amplitudes with respect to the associated interfaces as follows:

$$R_1 = \left(\frac{n_0 - n_1}{n_0 + n_1} \right)^2 \quad (1a)$$

$$R_2 = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2 \quad (1b)$$

$$R_3 = \left(\frac{n_2 - n_0}{n_2 + n_0} \right)^2 \quad (1c)$$

The effective transmitted power τ is given by:

$$\tau = 1 - (R_1 + R_2 + R_3) - A_1 - A_2 \quad (2)$$

where A_1 and A_2 are the propagation losses in layers n_1 and n_2 , respectively. Both A_1 and A_2 have the exponential thickness and depend on the input power at the entrance of each layer. For convenience, the above equation may be written in the form of

decibels. In our case, layer n_1 is the electro-optic effect material layer, and R_1 and R_2 will be affected if the value of n_1 is disturbed by the electric field.

2.2. Electro-optic effect

The influence of the electro-optic effect refers to changes in the refractive index of a material induced by the application of an external electric field, which modulates the optical properties. The applied field is not the electric field of any light wave, but it is a separate external field [8]. Usually, alterations in the refractive index are slight. The influence of electro-optic effect is categorized according to the first- and second-order effects. The applied electric fields are known to align the dye molecules. When this happens, the effect is not a change in the refractive index, which certainly is also happening, but is caused by a change in the optical extinction due to the imaginary part of the refractive index, or the absorption coefficient. The refractive index n is a function of the applied electric field E , *i.e.*, $n = n(E)$, such that it can be expanded as a Taylor series in E . The new refractive index $n_1(E)$ would be represented by the following equation

$$n_1(E) = n_1 + a_1E + \frac{1}{2}a_2E^2 + \dots \quad (3)$$

where the coefficients of the expansion are $n_1 = n_1(0)$, $a_1 = dn/dE$ at $E = 0$, and $a_2 = d^2n/dE^2$ at $E = 0$ [9]. For reasons that will become apparent, a_1 and a_2 are called the linear electro-optic effect and the second-order electro-optic effect coefficients, respectively. The change in n due to the first E term is called the Pockels effect. The change in n due to the second E^2 term is called the Kerr effect, and the coefficient a_2 is generally written as λK , where K is the Kerr coefficient. In the case of the Pockels effect, the precise effect of the applied electric field depends on the crystal structure and symmetry of the material under consideration. The main contribution to birefringence of the electro-optic effect crystal is due to the Kerr effect, as represented by the equation [10]:

$$\Delta n_p \ll \Delta n_k \quad (4)$$

The Kerr electro-optic effect is defined as follows: under the influence of an electric field, an optical anisotropy with an induced optical axis appears. The axis is directed along the applied electric field. When light radiation passes through a sample located in the electric field, a phase difference φ appears between the orthogonally polarized light components [11]:

$$\varphi = \frac{2\pi}{\lambda} \Delta n l = 2\pi K I E^2 \quad (5)$$

where Δn is the induced birefringence, l is the length of the optical path in the zone of the field application, λ is the light wavelength, E is the electric field strength, I is the light intensity and K is the Kerr coefficient of the material. Assuming that

the electric field is uniform, the field strength E can be expressed via the applied voltage V_0 and the distance between the electrodes d . Expression (1) then takes the form:

$$\varphi = \frac{2\pi BIV_0^2}{d^2} \quad (6)$$

The Pockels effect is expressed in the following equation:

$$\Delta n = a_2 E \quad (7)$$

This equation is an oversimplification because, in reality, the effect of an applied field along a particular crystal direction on the refractive index for light with a given propagation direction and polarization must be considered [12]. For reasons that will become apparent, it is convenient to write Eq. (3) in terms of two new coefficients, $P = -2a_1/n_3$ and $K = -a_2/n_3$, known as the electro-optic coefficients, such that:

$$n_1(E) = n_1 - \frac{1}{2} P n_1^3 E - \frac{1}{2} K n_1^3 E^2 + \dots \quad (8)$$

Terms higher than the third order can safely be neglected. The above equation can be simply written as follows:

$$n_1(E) \approx n_1 + PE + KE^2 + \dots \quad (9)$$

The electro-optic coefficients P and K normally depend on the direction of the applied electric field and the polarization of the light. Figure 2 shows the polarization vs. electric field (P - E) hysteresis loop for a typical ferroelectric crystal [13]. Typical values of the Pockels coefficient lie in the range of 10^{-12} to 10^{-10} m/V (1–100 pm/V). Changes in the refractive index induced by the electric field are typically small. Due

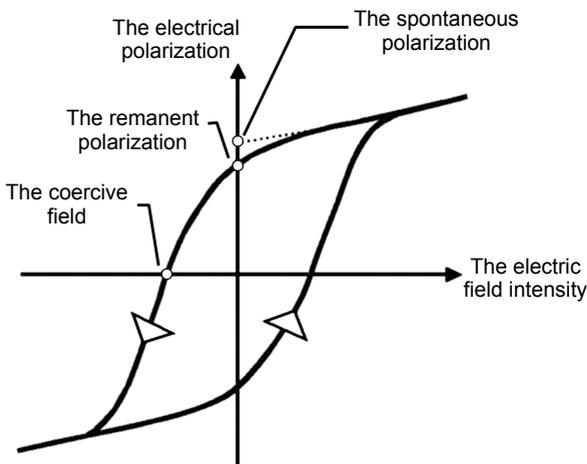


Fig. 2. The polarization vs. electric field (P - E) hysteresis loop for a typical ferroelectric crystal.

to the change in n_1 caused by the presence of E , a change in R_1 and subsequently R_2 will result, assuming A_1 and A_2 experience a negligible change. Therefore, the value of the transmittance power τ will be varied. The electric field varies with the voltage and width of the waveguide by the relation $E = V/d$. For example, if the applied voltage is 100 V and d is 10 μm , then the electric field is 100 kV/cm.

3. Experimental details

Two different kinds of polymeric materials were used: the guest–host polymer and side-chain copolymer. For the first group, the guest chromophore was DO-3. The host polymer was polysulfone, and acrylic copolymer was the side-chain group. They consisted of 1 g/20 mL of polymer. The guest–host polymer was approximately 1.0% w/w of polymer. Undoped polymer was also prepared. The solutions were filtered through a 0.4- μm pore-size nylon filter. Thin films were spin coated onto an aluminium-sputtered optical glass slide with approximately 70–90 μm in width of a narrow waveguide 2 cm in length across the slide. Aluminium electrodes were connected to a maximum voltage supply of 2 kV. The supply voltage was varied, and the percentage of transmission was evaluated at room temperature using a spectrophotometer. The transmission was monitored to observe the relaxation time of the switching.

4. Result and discussion

Figure 3 shows the transmittance versus the wavelength of the acrylic copolymer with 50 V of applied voltage with E parallel to the light beam. We found that due to the long relaxation time of the polymer molecules and with the application of 50 V of voltage, the reduction in τ is proportional to time. The application of 50 V of voltage affected the change in the optical transmission by nearly 10%. The reduction in the transmittance when exposed to the field is primarily due to the increase in R_1 or

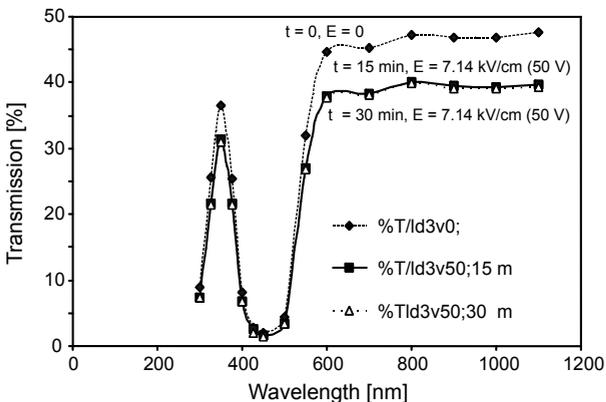


Fig. 3. Transmittance in copolymer acrylate under the application of 50 V voltage showing the saturation level after 15 minutes.

R_2 in Eq. (1). It is clear that this change is originated by an incremental change in n_1 . The observation of the decrease is significant in the range of 600 to 1100 nm. With a more detailed inspection, a slight red shift was found from 485 to 475 nm.

Figure 4 shows the change in the transmittance difference of the copolymer with respect to the transmittance value at zero field for three values of the electric field. A plot in Fig. 5 represents the actual transmittance at the respective fields. The transmittance decreases at a rate of 0.0336% for every 1-kV/cm increase in electric field. The transmittance versus n_1 , as given by Eq. (1), is plotted in Fig. 6 for the copolymer with a zero-field index of 1.55. The plot is drawn to show that the refractive index increases with an increase in the field. Moreover, as found from the variation in time of the applied voltage, there is a decrease in the transmitted light with time for the acrylic copolymer, while the reverse was found for the guest–host polymer. However, both polymers showed an effect with time; the sample displayed time

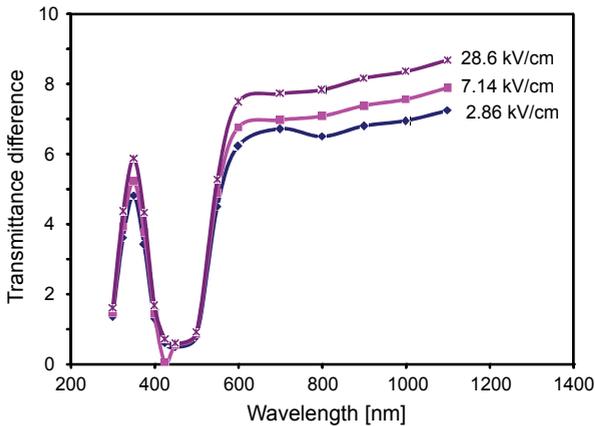


Fig. 4. Transmission difference (compared to transmission at $E = 0$) in acrylate copolymer under the application of various levels of electric field.

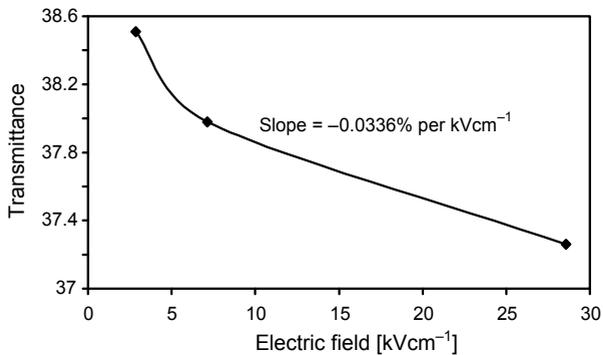


Fig. 5. Transmission percentage decrease with electric field in acrylate copolymer at room temperature.

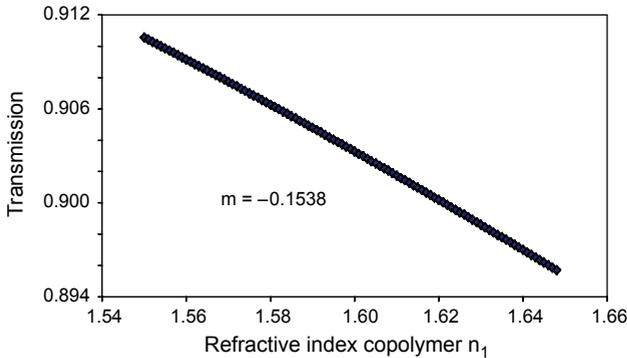


Fig. 6. Plot of transmission vs. n_1 from Eq. (1), showing that the increase in R_1 and R_2 reduces the total transmission. Here, A_1 and A_2 are assumed to be fixed at zero.

dependence in relation to its relaxation time. From a microscopic point of view, this result is due to the induced dipole moment in the polymer. There are several models that describe the chromophore mobility. The Debye relaxation time model is expressed by the relationship:

$$\varepsilon(t) = \varepsilon_0 e^{-t/\tau} \quad (10)$$

where τ is the characteristic decay constant. It was found that the decay of the curve in Fig. 3 in relation to time illustrates this phenomenon due to the dissimilar microenvironment of dye. Hence, the mobility must include the characteristic time and number of molecules of the dye species. HAMPSCH *et al.* [14] used a biexponential model to fit either a fast or slow decay time upon removal of the poling field. Slow relaxation may be associated with the polymer matrix mobility, and fast relaxation may be due to the dye. The refractive index n for the acrylate copolymer, measured using an ellipsometer, was 1.7–1.8, and the polysulfone guest–host polymer-doped DO-3 was 0.83–1.88 at 632.8 nm. Because transmission is related to reflectance by $T = (1 - R)$ and to n by Eqs. (1a)–(1c), the transmission of the acrylate copolymer in air will be approximately 83.3% and 91.83% for a small difference in the refractive index of only 0.1. For transmission greater than 88%, n normally would be 1.60 or below, but undoped polysulfone exhibited 88% transmittance at near-IR wavelengths with n of 1.685.

5. Conclusions

A doped acrylate copolymer with DR-1 showed a decrease in transmission intensity with applied field, while the guest–host polymer system depicted otherwise. The dye hyperpolarizability and composition may affect the transmission intensity, which

could be related to the refractive index. The induced dipole of the dye showed chromophore mobility to poling and perhaps with higher temperature, poling of the polymer and the electro-optic coefficient could be evaluated.

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