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TEMPERATURE DEPENDENCE ON THE GRATING FORMATION IN PHOTOREFRACTIVE POLYMERIC COMPOSITE

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We investigated the dependence of the grating formation on temperature in photorefractive polymeric composite, especially the index contrast of the grating. For photorefractive polymeric materials, temperature is one of the most important factors together with the external electric field, because it is closely related with photogeneration efficiency, carrier mobility, electro-optic coefficient tensor, and so on. The diffraction efficiency of the photorefractive polymeric composite decreased with increasing the temperature, and it could be explained with the magnitude of space-charge field and the electro-optic behavior at various temperatures.

Keywords: low glass temperature polymer; photorefractive effect; temperature dependence

INTRODUCTION

Photorefractive effect is a well-known phenomenon in which the refractive index of material is spatially modulated due to the light-induced redistribution of charges [1]. The redistribution of charges inside the material induces space-charge field, which subsequently modulates the refractive index of materials via an electro-optic effect [1–3]. The sample temperature is one of the most substantial factors determining the photorefractive behavior of a given material. Since the temperature significantly affects photo-charge generation efficiency, carrier mobility, electro-optic effect and so on, both build-up speed and index contrast of the photorefractive grating inevitably depend on the temperature.

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For electro-optic behavior in the photorefractive polymeric material with low T_g , the index grating is mainly induced through the chromophore reorientation along applied electric field. The order parameter of material containing reoriented chromophores under an electric field is well defined by the oriented gas model [4]. According to the model, the order parameter should be decreased with increasing temperature at a given electric field. For space-charge field in the polymeric materials, the magnitude is determined by several factors related with traps, such as trap density and trap depth [5].

In this work, the diffraction efficiency of photorefractive polymeric composite was measured at different temperatures, and it was analyzed with the temperature dependence of space-charge field and electro-optic behavior. To measure the magnitude of space-charge field, we used the characterization method presented recently in our previous paper [6]. Also, the diffraction efficiency was calculated on the basis of the measured value of space-charge field and electro-optic behavior and it was compared with that obtained by degenerated four-wave mixing.

EXPERIMENTAL

Sample Preparation

In this work, low T_g photorefractive material was prepared by doping the optically anisotropic chromophore, 2-{3-[(E)-2-(dibutylamino)-1-ethenyl]-5,5-dimethyl-2-cyclohexenyliden} malononitrile (DB-IP-DC), into photoconducting polymer matrix, poly[methyl-3-(9-carbazolyl) propylsiloxane] (PSX-Cz) sensitized by 2,4,7-trinitro-9-fluorenone (TNF) as shown in Figure 1. PSX-Cz and DB-IP-DC chromophore were synthesized by the previously described methods [7]. TNF (Kanto Chem. Co. Inc.) was used after purification. The composition of polymeric composite was PSX-Cz: DB-IP-DC: TNF = 69:30:1 by wt%. The glass transition temperature (T_g) of the composite determined by differential scanning calorimetry (Perkin Elmer DSC7) was 27°C. The device was prepared by sandwiching the softened composite between two ITO coated glass plates [7]. The thickness of active layer was 100 μm .

Measurement

We prepared a sample holder, which allows the temperature of sample to be adjusted from 20 to 50°C with error range of $\pm 0.2^\circ\text{C}$. There is a hole with the diameter of 1.5 cm at the center of the heating plate of the sample holder, and laser beams illuminating on the sample pass through it. The temperature of the sample is monitored by thermometer (Fluke 50S) whose probe is contacted on the glass plate of the sample.

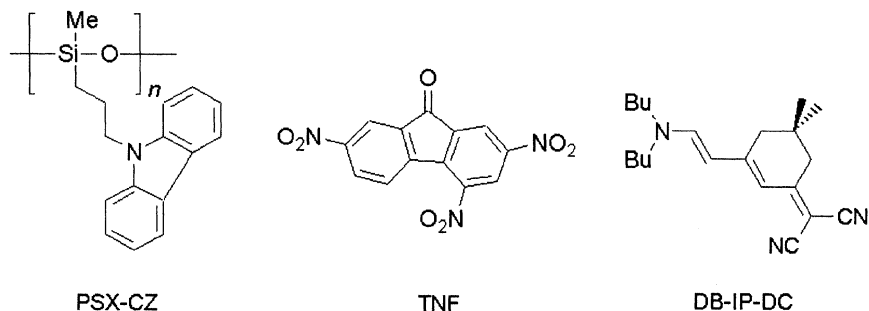


FIGURE 1 Chemical structure of components of photorefractive composite: poly [methyl-3-(9-carbazolyl) propylsiloxane] (PSX-CZ), 2,4,7-trinitro-9-fluorenone (TNF), and 2-[3-[(*E*)-2-(dibutylamino)-1-ethenyl]-5,5-dimethyl-2-cyclohexenylidene] malononitrile (DB-IP-DC) chromophore.

Diffraction efficiency of photorefractive material was determined by the conventional degenerated four wave mixing method using He-Ne laser [7]. The intensity of s-polarized writing beams was 30 mW/cm². The incident angles of two writing beams were 30° and 60° from the sample normal. The intensity of p-polarized reading beam was 0.06 mW/cm².

We measured the magnitude of the space-charge field using the characterization method that had been reported in our previous paper. The basic scheme of the method can be summarized as follows. The chromophore group that is previously aligned along external electric field is reoriented by newly-formed space-charge field. The variation of the birefringence is induced by the reorientation and it is closely associated with the space-charge field. Using numerical analysis based on the oriented gas model and the index ellipsoid method, we can determine the magnitude of the space-charge field from the variation. Details on this method were well described in Ref. 6.

To characterize the electro-optic behavior of a given chromophore, the standard two crossed polarizers setup was used. The polarization axes of analyzer and polarizer are set to 45 and -45° with respect to incident plane, respectively, and the sample was tilted by 30° from the propagation direction of the probe beam.

RESULTS AND DISCUSSION

Figure 2 shows the diffraction efficiency of the sample as a function of sample temperature. The diffraction efficiency is considerably decreased with increasing the temperature. It may be attributed to the decreases of space-charge field and/or electro-optic coefficient with increasing

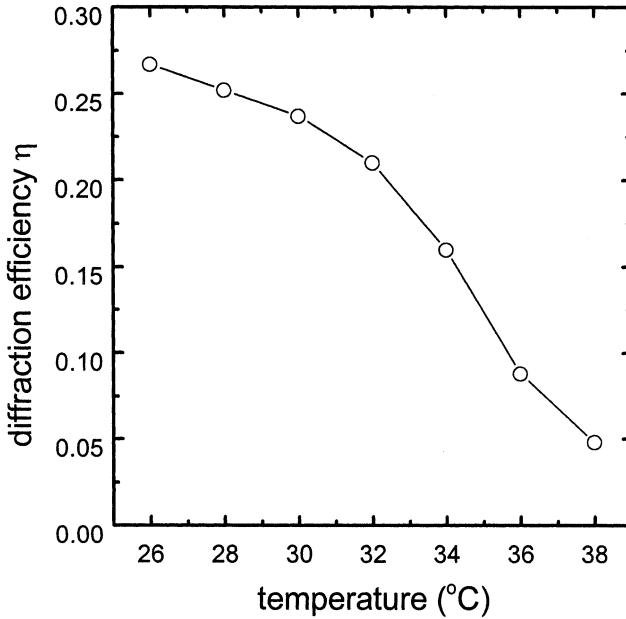


FIGURE 2 Variation of the diffraction efficiency of photorefractive polymeric composite with sample temperature.

temperature. But we cannot be certain which one is more dominant to reduce the diffraction efficiency because the index contrast of the photorefractive grating is given by $\Delta n \propto \gamma_{eff} E$, where γ_{eff} is an effective electro-optic coefficient and E is the magnitude of the space-charge field [2].

We measured the variations of the birefringence and the space-charge field with temperature as shown in Figure 3. In the measurement of birefringence and space-charge field, applied field was 30 V/ μm . The magnitude of the space-charge field decreased linearly by 36% with increasing the temperature from 24 to 36°C. The decrease of space-charge field must be closely related with the variations of trapping, detrapping, and recombination rates [5]. In order to improve the dependence of space-charge field on temperature, experimental and theoretical works on traps should be needed. The other important factor, effective electro-optic coefficient, was observed at various temperatures using a pair of crossed polarizers setup. From the obtained transmittance, we can quantitatively determine the variation of birefringence of the sample with temperature under a given electric field. It also decreases by 17% with increasing the temperature by 12°C. Consequently the reduction of the diffraction efficiency by increasing the temperature is due to both space-charge field and electro-optic effect.

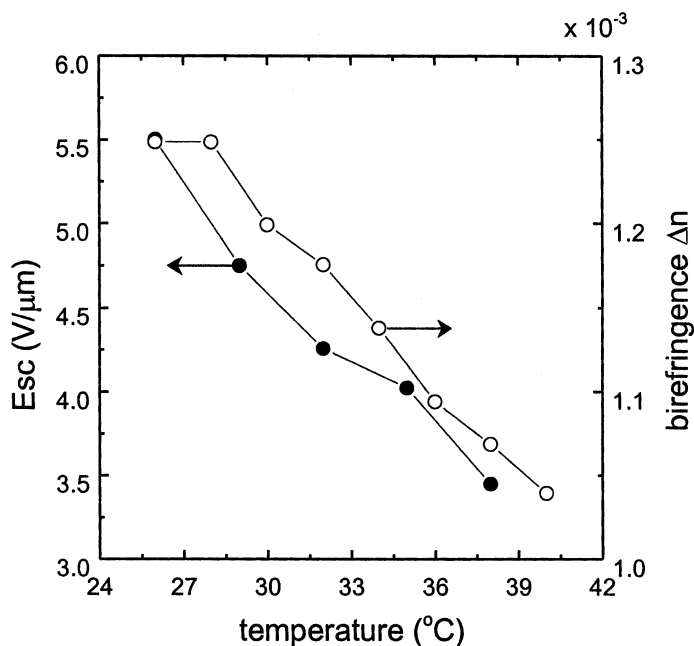


FIGURE 3 Magnitude of the space-charge field formed in the polymeric sample (left-axis) and birefringence of sample under electric field (right axis) at different temperatures. In these measurements, applied field was 30 V/μm.

Figure 4 shows the comparison between the diffraction efficiency and the calculated one on the basis of the oriented gas model and the index ellipsoid method. This calculation was accomplished in the similar way to the Figure 5 in Ref. 6. When the photorefractive grating was recorded in the sample, the locally modulating refractive index can be calculated from electro-optic behavior of the sample and the magnitude of space-charge field. The formal was well-defined from the experimental data and the calculation based on the oriented gas model and the latter was obtained experimentally. Then the diffraction efficiency can be determined from the spatial distribution of refraction index via the coupled mode theory [2]. According to the results in Ref. 6, the index contrast calculated from the determined space-charge field is ca. 40% larger than one experimentally obtained by degenerated four wave mixing measurement. For the reason, we normalized the measured and the calculated diffraction efficiencies to compare them in Figure 4. As can be seen Figure 4, they shows good agreement and it becomes clear that the decreases of the space-charge field and the effective electro-optic coefficient cause the reduction of the diffraction efficiency, when the temperature increases.

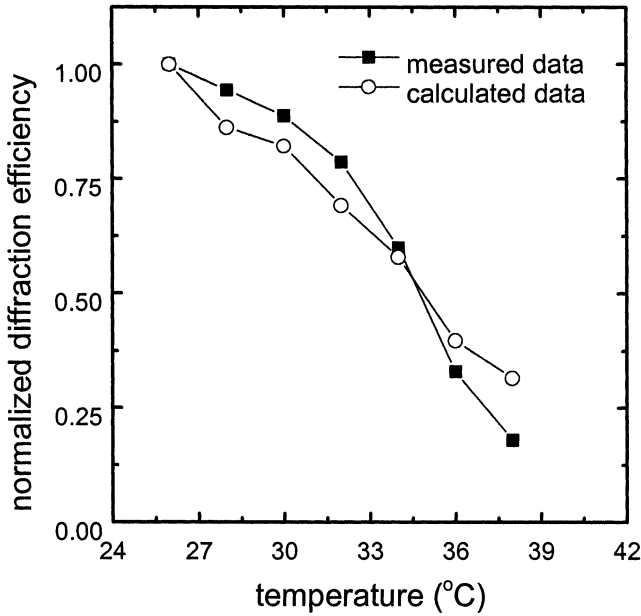


FIGURE 4 Comparison of calculated and measured diffraction efficiencies (○: calculated data, and ■: measured data).

CONCLUSION

In this work, we investigate the dependence of the photorefractive grating on temperature in a organic photorefractive material. The diffraction efficiency decreases with increasing the temperature, and as the reason, the space-charge field decreases by ca. 36% and the birefringence determined by the effective electro-optic coefficient decreases by ca. 17%. We calculated the variations of the diffraction efficiency estimated from these reductions on the based of the oriented gas model and the index ellipsoid method and we compared it with the measured one. We can explain the reduction of the diffraction efficiency with the space-charge field and the electro-optic effect.

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