Enhancement of the recording stability of a photorefractive polymer composite by the introduction of a trapping layer

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Enhancement of the recording stability of a photorefractive polymer composite by the introduction of a trapping layer

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A photorefractive polymer composite containing a trapping layer which exhibits a significant enhancement in its photorefractivity is reported. The photorefractive polymer composite containing a trapping layer showed a grating life time as high as 600 s, while the grating life time was only 7 s in the photorefractive polymer composite without a trapping layer. The grating growth rate was found to be unaffected by the introduction of a trapping layer. The diffraction efficiency and gain coefficient also increased by the introduction of a trapping layer. The enhancement in the photorefractive properties is due to the increase in the space-charge field developed in the photorefractive polymer composite containing a trapping layer. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1606885]

Since the first report of the organic polymer-based photorefractive (PR) materials in 1991,[1] PR polymers have been investigated intensively for their potential applications in optical information memory and processing.[2–5] Recently, PR polymer composites gained a great amount of attention for its advantage of large PR effect, structural flexibility, and good processability. However, one major concern which has received little attention for the application of PR polymer composites is the problem associated with the relatively poor recording stability due to temporal or thermal relaxation of the space-charge field which originates from the liberation and recombination of trapped charge carriers. Some progress on improving the recording stability of PR polymer composites was made recently by simply using additional trapping materials,[6–8] but unfortunately this has been accompanied by a tradeoff in performance, that is, a longer grating growth time. This is considered to be inevitable because additional trap sites introduced in the PR polymer composites by a simple blending act as barriers toward charge transport in the materials. Thus, the more efficient way of trapping is required for the maintenance of the space-charge field (E_{sc}) for long periods of time without affecting the grating growth time.

In the present work, we introduce additional trap sites as a layer at the surface of the PR polymer composite layer and discuss the PR properties of the PR polymer composite. Unlike the simple blend of trapping materials with the photorefractive polymer composite, traps introduced as a new layer in this work are expected to not influence the growth rate because charge transport process through the bulk of the photorefractive polymer composite layer is completely independent on the trapping process in the added trapping layer.

Two different samples were prepared to verify the effect of the trapping layer on the recording stability and the growth rate of the PR polymer composite. Case I is the conventional PR polymer composite which does not contain a trapping layer and case II is a PR polymer composite consisting of a trapping layer and a PR polymer layer. The composite is based on poly(vinyl carbazole) (PVK) doped with a nonlinear optical chromophore Disperse Red1 (DR1), the liquid plasticizer, ethyl carbazole (ECZ), and C_{60} as the photosensitizer. For the PR polymer composite preparation, the PVK: DR1: ECZ: C_{60} was dissolved in the ratio 75:5:20:0.2 wt % in chlorobenzene. The mixture was dripped onto indium tin oxide (ITO) coated glass and left to dry. For case II, we used poly(vinyl phenol) as the trapping layer. It was formed by spin coating the polymer solution on the ITO coated glasses. The sample configuration of case II is described in Fig. 1.

Two-beam coupling (TBC) experiments were performed by interfering two *s*-polarized beams with equal intensity of 2.6 mW/cm^2 from a He–Ne laser operating at 633 nm. The external beam incidence angles were 15° and 45° with respect to the sample normal. In the degenerate four-wave mixing (DFWM) experiments, two *s*-polarized beams with equal intensity write a grating which is probed by a weak Bragg matched beam counterpropagating to one of the writing beams. All samples were electrically poled at 40 V/μm before DFWM and TBC experiments in order to align the nonlinear optical chromophore, DR1, in the PR polymer composites. Both DFWM and TBC experiments were carried out under external electric field of 40 V/μm.

The diffraction efficiency, η, defined as the intensity of the diffracted beam divided by the intensity of the probe beam in the DFWM experiment is shown as a function of writing time in Fig. 2. For both cases, the diffraction efficiency increases with time and saturates at steady-state diffraction efficiency at about 200 s. As shown in Fig. 2, the growth rate is found to be almost similar for both cases from
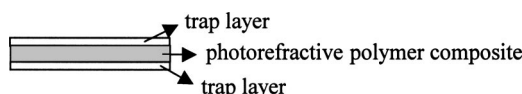

FIG. 1. Sample configuration of photorefractive polymer composite containing trap layer.

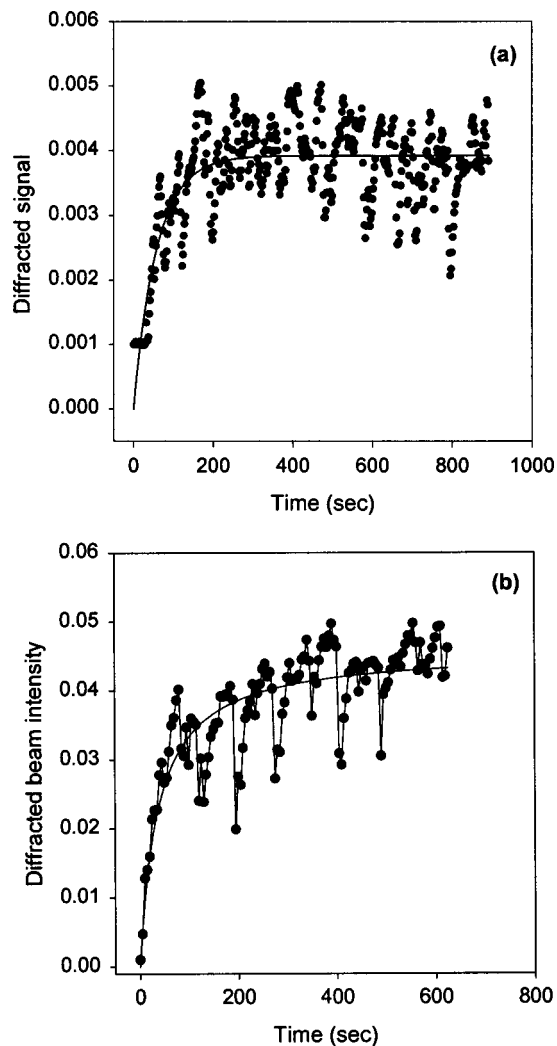[a)]Author to whom correspondence should be addressed; electronic mail: pjk@mail.kaist.ac.kr

FIG. 2. Grating growth characteristics of photorefractive polymer composites for (a) Cases I and (b) II.

FIG. 3. Erasing behaviors of photorefractive polymer composites for (a) Cases I and (b) II.

the initial slopes of the two curves. For both cases, it takes about 30 s and 100 s to reach 50% and 80% of the steady-state diffraction efficiency, respectively. It should be noticed that the absolute values of the growth time are relatively large compared to other reported values. It may be attributed to the fact that gratings were written with relatively low light intensities and low external electric fields (40 V/μm). In Fig. 2, a chaotic oscillation of the diffraction signal was observed for both cases I and II. This oscillation seems to be caused by the competition of the two types of gratings that have different characteristic growth times.[9] In our case, it is thought to originate from the competition between the gratings formed by photoisomerization of DR1 and the PR grating. Maximum diffraction efficiencies obtained for cases I and II were 0.36% and 3.56%, respectively.

To compare the dark decay behavior of cases I and II, the two writing beams were blocked and the external electric field was also removed after reaching the steady-state diffraction efficiency. Figure 3 shows a difference in the decay behavior for the two cases. A decrease in the diffraction efficiency with time was drastic for case I, while much slower decay was observed for case II. For case I, the diffraction efficiency decayed completely within several seconds after blocking the two writing beams and removing the external
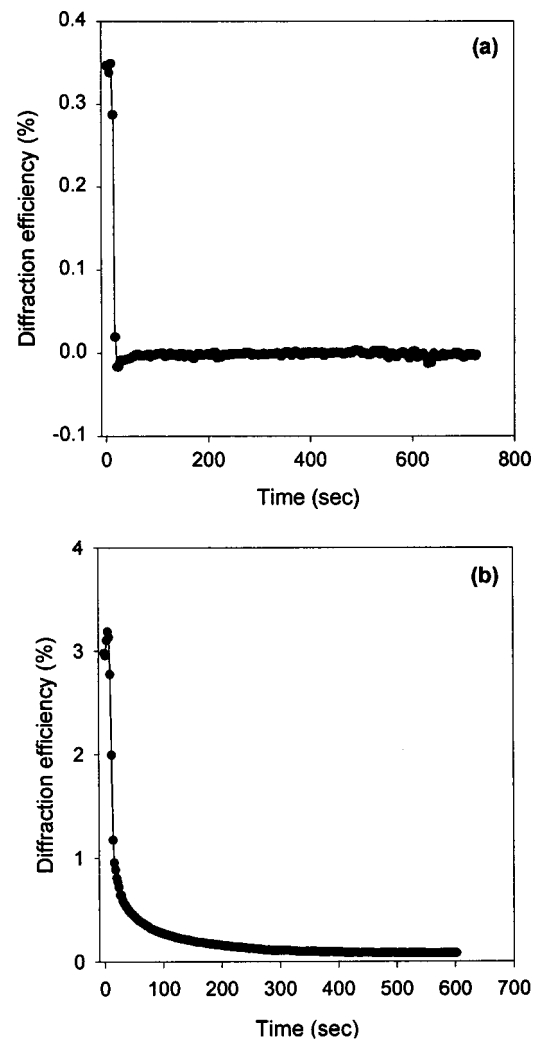
electric field, while about 20% of diffraction efficiency was maintained until 100 s and almost completely decayed after about 600 s for case II. This concludes that the recording stability of the PR polymer composite is greatly enhanced by the introduction of the trapping layer.

Proof of the PR nature of these gratings is usually provided by the observation of asymmetric two-beam coupling.[10] From the TBC experiment, gain coefficients (Γ) of 4.85 and 10.6 cm^{-1} were obtained for cases I and II, respectively. Several optical properties of the two cases including gain coefficients and maximum diffraction efficiencies are summarized in Table I. It should be noticed that the transmittance of case II is not significantly reduced by the additional trapping layer. It means that there is almost no light loss at the interface between the trapping layer and the PR polymer composite layer due to good interfacial adhesion between the two layers.

TABLE I. Optical properties of the polymer composites.

	d (μm)[a]	t (%)[b]	Γ (cm^{-1})	η (%)
Case I	167	85	4.85	0.36
Case II	130	86	10.6	3.56

[a]Sample thickness.
[b]Transmittance.

Appl. Phys. Lett., Vol. 83, No. 15, 13 October 2003

Kim, Lee, and Park   3047

TABLE II. Physical properties of polymer composites.

	$\phi(°)$	E_q (V/μm)	N_T (cm^{-3})	E_{sc} (V/μm)
Case I	1.21	16	1.42×10^{16}	182
Case II	0.65	31	2.65×10^{16}	453

From the earlier results, it is evident that the trapping layer has a significant influence on the PR properties of the polymer composite. Higher gain coefficient and diffraction efficiency, and enhanced recording stability were obtained for case II. This enhancement in the PR properties should result from the higher effective trap density for case II, which should lead to a higher space-charge field buildup. To confirm our presumption, we evaluate several important physical parameters including the effective trap density (N_T), the trap-limited space-charge field (E_q), and the total space charge-field (E_{sc}) that contain information on the physical processes occurring in the PR polymer composite. The evaluated physical parameters using the coupled-wave theory[11] and the standard Kukhtarev model[12,13] are listed in Table II. As shown in Table II, the space-charge (E_{sc}) for cases I and II are 182 and 453 V/μm, respectively. The higher value of E_{sc} for case II can be attributed to the increase in the effective trap density (N_T).

In conclusion, by introducing a trapping layer, we could prepare a type of photorefractive polymer composite which could enhance recording stability, gain coefficient, and diffraction efficiency without sacrificing the grating growth rate. This approach of introducing trap sites as a new layer is expected to be a promising method for producing photorefractive polymer composites with high recording stability as well as fast growth rate.

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