

Exposure schedule for multiplexing holograms in photopolymer

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ABSTRACT

We present dynamics of the grating formation process in phenanthrenequinone- (PQ-) doped poly(methyl methacrylate) (PMMA) photo-polymer. The exposure schedule for multiplexing holograms to achieve equal-strength storage in a single location is described. The analysis method is valid for any saturated media, thus the results can be applied to other photopolymer materials with similar kinetics.

Keywords: Photopolymer material, PMMA, Holographic data storage, multiplexing schedule, dynamic range

1. INTRODUCTION

Photorefractive and photopolymer materials have recently been drawing intense attention in the area of holographic storage because of their unique properties of building up photo-induced refractive index phase grating in real time during holographic exposure [1-8]. Grating formation in these media does not require wet chemical processes and it is usually called as "self-developing". When these materials are used as recording medium in holographic memory, thousands of data pages can be recorded on one location by using a suitable multiplexing scheme. As the number of holograms superimposed on one location is increased, a technique for multiple storage to achieve equal-strength gratings becomes important. In photorefractive materials, two exposure methods have been practiced: scheduled exposure and incremental exposure [9-11]. The main idea for these two methods is compensating the erasing effect of the previously recorded holograms. Unlike photorefractive materials, photopolymer materials possess a nature of self-fixing and saturation effect during holographic exposure. Each hologram is automatically fixed after recording so that the previously exposed holograms are not partially erased by subsequent exposure. Instead, subsequent exposures in polymer media cause the material to saturate such that the growth rate of the grating strength decreases with the exposure order. As a result, recording with a constant exposure schedule will result in holograms with non-uniform diffraction efficiencies [12]. To overcome this saturation problem, a special exposure schedule should be carefully designed. In this paper, we present our studies on the recording dynamics of our photopolymers. Based on the measurement of dynamics, we propose two exposure schedules for achieving equal-strength gratings of multiple storage. By using our exposure schedule, three hundred and fifty-five plane wave holograms with uniform diffraction efficiency have been stored at a single location in a $2 \times 2 \times 0.75 \text{ cm}^3$ cube photo-polymer using scheduled exposure.

2. MATERIAL PREPARATION

In our experiments, polymer blocks were made by two stages at different temperatures. In the first stage, samples were prepared by dissolving initiator, AIBN (~0.5%) and PQ molecules (up to 0.7%) in solvent MMA. The solution was purified before use to remove un-dissolving particles such that scattering centers are reduced. It was then poured into square glass flasks and put in a pressure chamber at room temperature for about 120 hours until the solution turned into homogeneously viscous. In this stage, because of the slow polymerization at low temperature, nitrogen resulted from the thermo-decomposition of AIBN and heat released from the chain propagation of the MMA monomers could leave the flask completely so that the bubbles did not exist in the solid blocks. In the second stage, the temperature of the chamber was

elevated to 45 °C (for 24 hours) to promote the fast thermo-decomposition of AIBN. Chain propagation reaction of the polymerization were accelerated and completed. The samples became solid blocks. The glass flask was used to allow the fabricate the samples in a variety of dimensions, ranging from 1 x 1 to 2.5 x 2.5 cm² with thickness of 1-25 mm. In following sections, all polymer samples used are doped with 0.6% PQ molecules. The transmission spectra for 1.2-mm- and 2.4-mm-thick samples are shown in Figure 1. The absorptions for both the samples reach maximum of $\geq 97\%$ below 454 nm. For the wavelength of 514.5 nm, which was used in all experiments described in this paper, the absorption of 1.2-mm-thick sample is 30.2% and that for the 2.4-mm-thick sample is 44%.

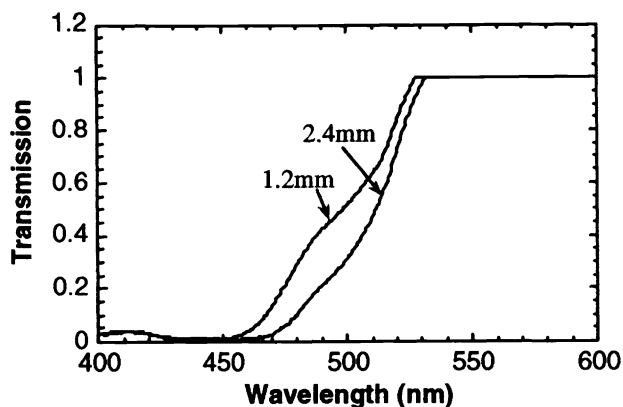


Figure 1. Transmission spectra for PQ-PMMA samples with varying thicknesses.

To design a strategy for recording multiple holograms in this material, we first briefly discuss the recording mechanism [13-17]. Under light illumination, PQ molecules become radicals and bond with a single monomer, MMA. Our chemical analysis indicated that typically the residual monomer MMA in our samples is about 10 %. When light distribution is not uniform, the primary chemical bond reaction occurs in the bright region. Because of the minimal PQ diffusion coefficient at low temperature, free MMA molecules then diffuse from dark into bright regions due to spatial variation of the density level. Consequently, a difference in the refractive index of the polymer matrix in dark region and that of PQ:MMA compounds in bright region is created, i.e. a phase grating is recorded. Under this situation, the matrix of host polymer would not be influenced by light distribution and therefore the shrinkage effect can be reduced to minimal through the recording procedure. Non-shrink effect makes this photopolymer material an attractive candidate for volume holographic memory. Performing experiments with light-illumination on a yellow mixture liquid of MMA monomers and PQ molecules provides the evidence of this process. We observed that the liquid samples did not turn into solid, but becomes colorless. This implies that under illumination PQ molecules did not cause photo-induced polymerization mechanism. In addition, we also analyzed the chemical component of the exposed polymer samples by gas chromatograph/mass spectrometer. The result is shown in Figure 2. It can be seen that the peaks corresponding to the single pair of PQ:MMA and ionized PQ:MMA are much stronger than that of large molecules. Therefore, we believe that in our samples, bonding of PQ molecules with residual MMA monomers is the main photo-induced reaction. In a previous paper [18], authors have presented that thermal treatment of an hologram recorded in the PMMA polymer can increase the diffraction efficiency of grating. We believe that this increasing effect may result from thermal-degradation of residual MMA monomers.

3. DYNAMICS OF THE POLYMER MATERIAL

The recording dynamic of materials is an important criterion to characterize a material's ability of the multiple storage in a single recording area. To examine the recording dynamics, we have performed experiments with peristrophically multiplexed hologram storage by using optical setup in Figure 3 [19]. A pair of expended ordinarily polarized 514.5-nm Argon-laser beams was symmetrically incident into the sample with an intersection angle of 32 degrees outside the sample. The peristrophical multiplexing was achieved by rotating the polymer sample mounted on a rotation stage with 1-degree rotation between holograms. Shutters were used to control the recording and the read out of holograms. Three hundred and

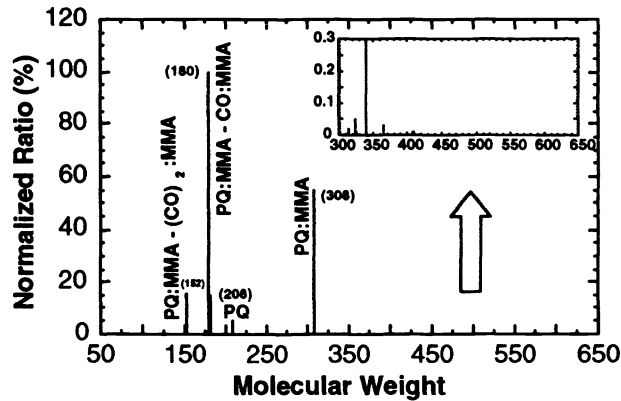


Figure 2. Molecule weight analysis spectra of the exposed polymer sample

fifty-five plane wave holograms were recorded with equal exposure energy ($\sim 8\text{mJ/cm}^2$ per exposure) at a single location of the samples with various thicknesses. For each sample, the diffraction efficiency of each hologram was measured, and the square roots of the measured diffraction efficiencies were summed up to plot a running curve of the cumulative grating strength (defined as $C = \sum_{i=1}^N \sqrt{\eta_i}$, N is the total number of exposed holograms) with respect to the exposure energy. The resulting curves for polymer blocks of different thickness are given in Figure 4. It can be seen that the final value of the cumulative grating strength is a function of the sample thickness. The curves in Figure 4 are related to the recording dynamics for different samples. In order to model the temporal behavior of the recording dynamics of our polymer, the running curves in Figure 4 were fitted as an exponential growth formula (solid lines in Figure 4) of the following form:

$$C = C_{\text{sat}}(1 - \exp(-E/E_\tau)) \quad (1)$$

where C_{sat} represents the saturation value of the grating strength and E_τ is the characteristic exposure energy constant. Notice that the value of C_{sat} is related to the dynamic range of our polymer sample. If an exposure schedule can be designed to record equal strength holograms that utilize all of the available dynamic range of the polymer material, then the final diffraction efficiency of each hologram will be ($\eta_{\text{final}} = (C_{\text{sat}}/N)^2$). The $M/\#$ is a system metric for a holographic memory systems, depending on experimental conditions such as material thickness, recording geometry, and physical parameters. According to the definition of the $M/\#$ ($\eta_{\text{final}} = (M/\#/N)^2$) for the photorefractive material [20], then the dynamic range of the cumulative grating strength is related to the $M/\#$ of the polymer material. Therefore, we can call this dynamic range as the $M/\#$ for photo-polymer.

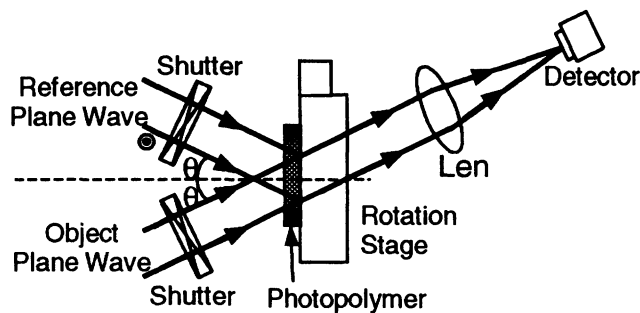


Figure 3. Optical set-up for measuring recording dynamics

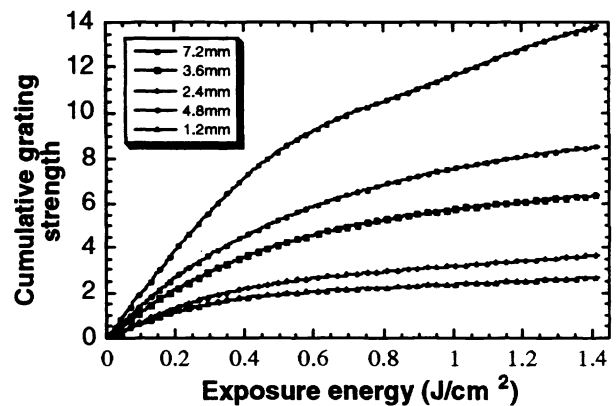


Figure 4. Cumulative grating strength as a function of exposure energy for varying thicknesses.

We have calculated the $M/\#$ of our polymers with respect to the sample thickness by using the saturation values of the curves in Figure 4. As expected, the $M/\#$ increases linearly with the sample thickness. We are attracted to the dependence of exposure energy constant with respect to the sample thickness, which is shown in Figure 5. It can be seen that the required energy constant increases as the thickness increases. This phenomenon may result from the material absorption. Because of the absorption, the beam are depleted through the sample such that the interference fringes become weaker near the back end of the sample.

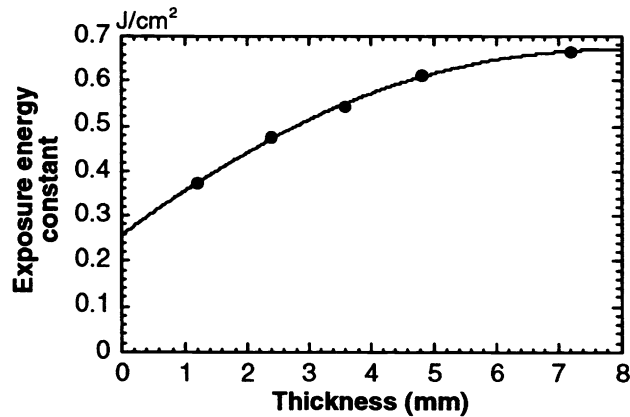


Figure 5. Exposure energy constant as a function of sample thickness.

4. EXPOSURE SCHEDULE

We have plotted the diffraction efficiency of three hundred and fifty-five holograms from a peristrophically (1-degree rotation between holograms) multiplexed storage in a 4.8-mm thick sample, which is shown in Figure 6, it is seen that recording with equal exposure energy results in non-uniform diffraction efficiency. For the beginning exposures, the diffraction efficiency of each hologram remains a constant and then decreases with the exposure order as the recording material begins to be exhausted. It represents that the material exhibits a quasi-linear exposure response until the material saturates. To alleviate this non-uniformity, a special recording schedule to compensate for the effect of the material saturation should be used.

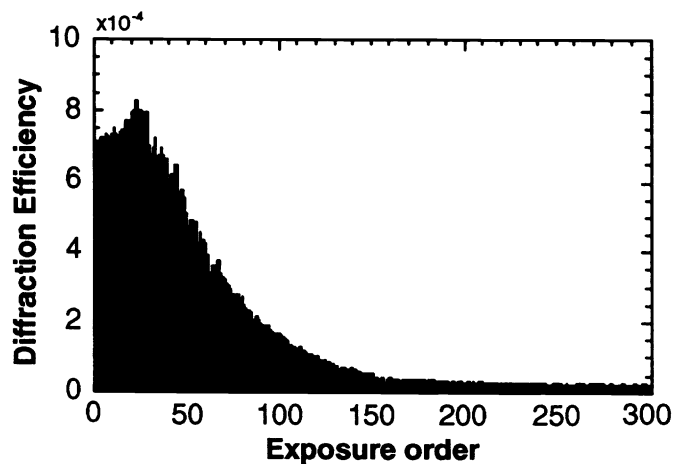


Figure 6. Diffraction efficiency of 355 plane wave holograms recorded with identical exposure energy of 8 mJ/cm².

4.1 Scheduled exposure method

For a given sample, allocating the dynamic range of the polymer material equally can equalize the grating strength for each hologram. The corresponding exposure energy for each equal hologram can be assigned by mapping the grating strength for each hologram into the corresponding point on the horizontal axis in Figure 4 by using the relation of the transfer curve. Analytically, by differentiating Eq.(1), the growth rate of the cumulative grating as a function of the exposure energy can be obtained. Therefore, assuming that each exposure energy is very small (compare with the dynamic range), the desired exposure schedule can be written as

$$t_n = R \frac{E_\tau}{I} \exp\left(\frac{1}{E_\tau} \sum_{i=1}^{n-1} E_i\right) \quad (2)$$

where t_n is the exposure time of the n -th hologram, R is the ratio of the desired grating strength with respect to the saturation grating strength for each thickness, E_τ is the exposure energy constant from Eq. (1), E_i is the exposure energy for recording the i -th hologram, and I is the intensity of the incident light. It can be seen that in Eq. (2), the recording time for each hologram is pre-determined by a special exposure schedule. Therefore, we call this method as ‘‘Scheduled Exposure Method’’ (SEM).

For a 4.8-mm thick sample, an exposure schedule for 350 holograms was calculated by using Eq.(2). If the incident intensity is 4-mW/cm², then figure 7 shows the exposure time for each hologram. Note that the exposure time for each hologram is no longer a constant and it increases as the sample becomes saturated. The diffraction efficiencies of the 350 holograms that were recorded with this special schedule are shown in Figure 8. It can be seen that the diffraction efficiencies are much more uniform than that in Figure 6.

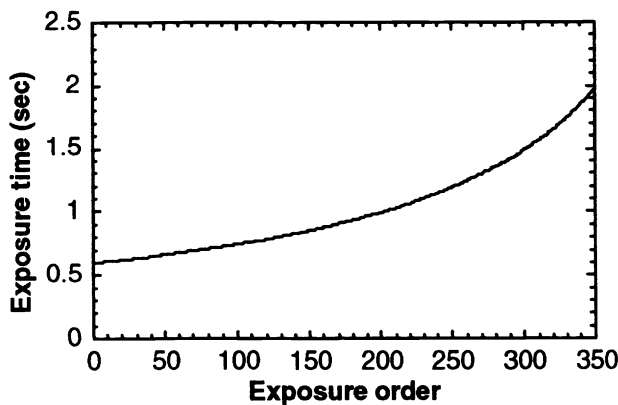


Figure 7. Scheduled exposure for 350 holograms

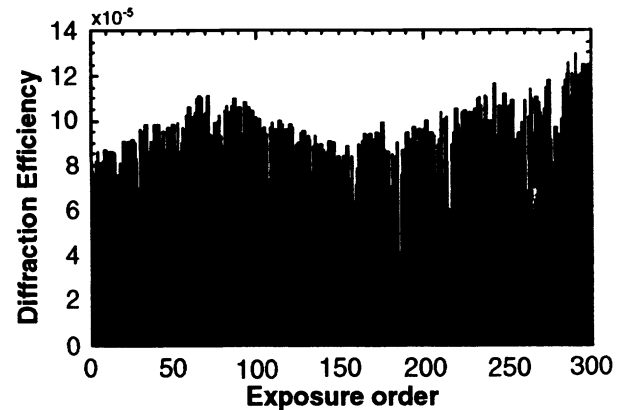


Figure 8. Diffraction efficiency as a function of exposure order with scheduled exposure.

4.2 Incremental exposure method

Another method for equalizing the grating strength of the holograms is to record all holograms successively using identical exposure energy, which is much smaller than the response exposure energy, with a special order of recording sequence until the steady-state is reached. The recording sequence is schematically shown in Figure 9. First, all the desired images are successively recorded in the polymer with an identical exposure energy and with a recording sequence from 1 to N (N is the number of the stored holograms). To compensate for the saturation effect during recording, in the second cycle, the recording sequence is reversed (i.e. from N to 1). It is reversed again (i.e. from 1 to N) for the third cycle, and so on. This recording procedure is repeated until the steady state of the polymer is reached. By using the recording dynamics (Eq. (1)) and a recursive derivation, the cumulative grating strength of j -th hologram at the end of the q -th recording cycle can be

obtained as the following relation,

$$C_j = \frac{C_{sat}\Delta E}{E_\tau} \left[\exp\left(-\frac{\Delta E}{E_\tau}(j-1)\right) + \exp\left(-\frac{\Delta E}{E_\tau}(2N-j)\right) \right] \left[1 + \exp\left(-\frac{\Delta E}{E_\tau}2N\right) + \exp\left(-\frac{\Delta E}{E_\tau}4N\right) + \exp\left(-\frac{\Delta E}{E_\tau}6N\right) + \dots \right]$$

$$\approx \frac{C_{sat}\Delta E}{E_\tau} \left[2 - \frac{\Delta E}{E_\tau}(2N-1) \right] \frac{1 - \exp\left(-\frac{\Delta E}{E_\tau}2qN\right)}{1 - \exp\left(-\frac{\Delta E}{E_\tau}2N\right)}, \quad \text{if } (2N-1)\Delta E \ll E_\tau \quad (3)$$

where N is the total number of the holograms, ΔE is exposure energy for each exposure, q is the number of the recording cycles. It can be seen that in relation (3), under the assumption of small exposure energy, the cumulative exposure energy for each hologram is a constant. Therefore, we can achieve the equal strength recording of holograms. It can be seen that this method is similar to the incremental recording for photorefractive materials in Ref. [11]. Therefore, we also call it as "Incremental Exposure Method" (IEM).

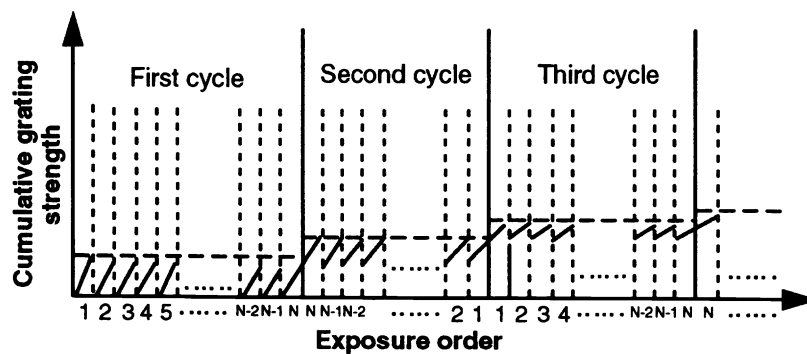


Figure 9. Schematic diagram of the recording consequence for incremental exposure method

5. CONCLUSIONS

We have presented a method for synthesizing large high-optical-quality polymer volumes, which are made by PQ-doped PMMA host matrix polymer. Up to 2.5 cm³ volume polymer block has been made. We have measured the recording dynamics of these polymer samples. Based on these studies, the criterion for evaluating the recording dynamic range of the polymer has been defined and the $M/\#$ of the polymer has been defined. This definition is generally available for any saturated material with similar recording dynamics. Using the recording dynamics of our polymer samples, two special recording schedules have been proposed to uniform hologram for multiple storage. In SEM recording procedure, the holograms are recorded one after each other by using the pre-determined recording time of each hologram. The recording procedure is completed in one cycle. This procedure has the advantage of easy addressing for the reference beams. Because of the saturation effect of the polymer, the recording time for a new hologram is longer than that of previously recorded hologram. Then, the recording times for very last holograms are very long. In this situation, the polymer is close to saturation and the noise gratings will build up, and the fanning effects of the incident beam become maybe strong such that the fidelity of the stored image is degraded. In IEM recording procedure, holograms are recorded with identical exposure energy with a designed sequence of recursive exposures. Because of the requirement of many repeated recording cycles, the time needed to display the image on input spatial light modulator may take a long time such that the recording procedure is slow down. In addition, it is not easy to achieve exact reference beam condition (such as angle in angle-multiplexing) for all the cycles. Nevertheless, because the energy for each exposure is much smaller than the exposure energy constant, the image fidelity will be better than that of the SEM recording procedure.

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