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Spatial frequency response of Acrylamide based holographic photopolymer

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Abstract: It has been shown that photopolymers are useful in holographic optical elements [1] and holographic interferometry [2]. However, the list of potential applications increases greatly if reflection holography is possible. Reflection holography requires a material that has the potential to record high spatial frequencies. Here we studied the spatial frequency response of an acrylamide based dye sensitized holographic photopolymer. We have investigated the effect of molecular weight of the binder and concentration of crosslinking monomer on diffusion using real time monitoring of the holographic grating formation. The effect of uniform pre-exposure on diffusion was also investigated. We have also demonstrated that reflection holograms can be recorded in the material.

Key words Spatial frequency, Photopolymer, Holographic grating

1. Introduction

Holography is an image-recording process in which both the phase and the amplitude of the wave field that intercepts the recording medium are recorded [3,4]. Several types of photosensitive recording media are used in holography such as, silver halide photographic emulsion, dichromated gelatins, photoresists, photochromics and photopolymers [5].

Photopolymers have become increasingly important in holography due to their many advantages in recording volume phase holograms. The advantages of photopolymers are high refractive index modulation, self-processing capability, ease of preparation, long shelf life and low cost. Due to the advantage of being self-developing these materials are suitable for many applications such as design of optical elements [1], real time holographic interferometry [2] and optical data storage [6]. The list of the potential applications is increased if reflection holography is possible. To record reflection holograms the material should have high resolution. The spatial frequency response of the acrylamide based dye sensitized photopolymer recording material developed in our research centre was studied by Martin et al., [7] and showed that the material has poor response at higher spatial frequencies. The poor response at high spatial frequency was explained by diffusion of initiating species from bright fringe regions to dark fringe regions, decreasing the amplitude modulation of the recorded grating and causing a decrease in diffraction efficiency. However the rate of diffusion depends on the physical state of the medium through which they move. In the case of this material it depends on the binder and crosslinker. The effect of concentration of crosslinker and the molecular weight of binder on diffusion is presented here.

2. Theory

The standard formulation of this dry photopolymer system [7] contains monomer, electron donor, and photosensitive dye dispersed in a binder matrix. When a photosensitive layer is exposed to an interfering beam of suitable wavelength and intensity, the dye molecules enter into an excited singlet state. From this state, they undergo intersystem crossing, to excited triplet states. The dye molecule in this state will react with an electron donor to produce free radicals. These free radicals react with monomer molecules to start a polymerization reaction. When a photosensitive medium is exposed to interfering beams, monomer molecules in the light exposed areas are polymerized and the unexposed regions are not polymerized. This creates a monomer concentration gradient, which results in the diffusion of monomer molecules from dark fringe regions into the bright fringe regions. This results in a spatial density distribution of refractive index modulation between exposed and unexposed regions of the interference pattern. The diffusion time depends on fringe

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spacing. At low spatial frequencies, where fringe spacing is large the diffusion time is long. As we go to higher spatial frequencies, fringe spacing decreases, the diffusion time becomes shorter.

We have investigated the spatial frequency response of an acrylamide-based photopolymer using a steady state approach. We have recorded diffraction gratings with different spatial frequencies and measured the diffraction efficiencies. We have also investigated the time-resolved measurements of grating growth in real time for a short duration exposure to study the recording dynamics in real time. The experimental set-ups used are shown in the figures 1 and 2

3. Experimental

3.1 Steady state approach

The experimental set-up is shown in the figure 1. A collimated beam of light is incident on a mirror and the photosensitive medium, placed perpendicular to each other at the centre of rotation on a rotational stage. The beam reflected from the mirror onto the photosensitive medium acts as an object beam and a beam directly hitting the photosensitive medium acts as a reference beam. The two beams interfere and result in an interference pattern thus recording a hologram (diffraction grating). The spatial frequency ($1/\Lambda$, Λ= fringe spacing) of the grating depends on the angle (α) between the reference and object beams. The angle between two beams is controlled using a microprocessor-based controller.

$$2\Lambda \sin(\alpha/2) = \lambda \quad (1)$$

Gratings were recorded at different spatial frequencies in a 150μm thick dry photopolymer layers on 5 cm x 5cm glass substrates with 3.5 mW/cm^2 exposure intensity for 4 seconds. A Nd-YAG laser with 532 nm wavelength is used to record the gratings and a He-Ne laser with 633nm wavelength was used to measure the diffraction efficiency. The experiment was repeated for different types of PVA (poly vinyl alcohol) binders, which differ in molecular weight, and for different concentrations of crosslinking monomer (N-N'Methylene bisacrylamide). The results are shown in section 4.

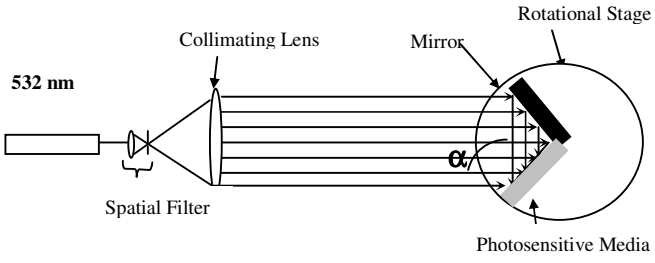

Figure 1 Experimental set-up for steady state measurement

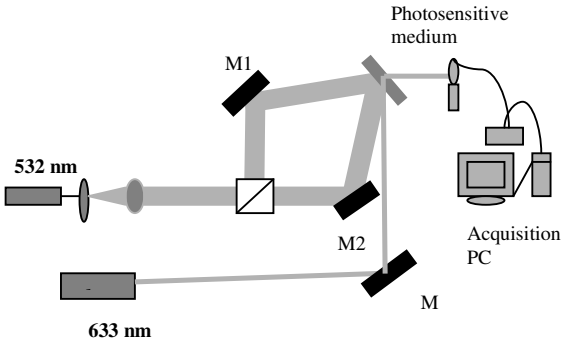

Figure 2. Experimental set-up for real-time monitorin

3.2 Real-time monitoring

The experimental set-up for real-time monitoring is shown in figure2. A spatially filtered, collimated beam (λ= 532 nm) of light is split into two beams with a beam splitter. The two beams are made to incident onto two adjustable mirrors and are overlapped in the plane of photosensitive medium and the plate normal bisecting the inter beam angle. This is to ensure that the grating is unslanted. The angle between the two beams gives the spatial frequency of the grating using the Bragg condition (equation 1). A He-Ne laser (λ= 633 nm) incident at Bragg angle was used to read the diffracted beam. The diffracted beam was recorded and sent to a data acquisition system interfaced to computer.

4. Results and Discussion

4.1 Steady state measurement

4.1.1 Influence of Binder on spatial frequency response

From figure 3 we observe that as the molecular weight of binder increases the diffraction efficiency also increases. It is also clear that for the lowest molecular weigh binder (13K-23K) the fall-off in diffraction efficiency that normally occurs at higher spatial frequencies occurs much earlier. In fact the optimum spatial frequency for high diffraction efficiency in this binder appear to be around 650 lines/mm, in comparison with the optima for the higher molecular weight binders

which are closer to 1000 lines/mm. Each of the data points is the average of two separate experiments, and the error bars indicate the deviation.

This can be explained by the fact that diffusion processes are faster in lower molecular weight binders. If the diffusion of free radicals into the dark fringe areas is responsible for the poor response at high spatial frequency then we would expect to observe such a worsening of the effect in lower molecular weight binders. Higher molecular weight binders would therefore be expected to perform better, but with the samples available to us in this study no improvement could be made on the standard (20K-200K) binder. Higher molecular weights were found to be too difficult to dissolve. The other method of changing the physical state of the photopolymer layer and thus the diffusion rate is to change crosslinker concentration.

4.1.2 Influence of Crosslinker concentration on spatial frequency response

In figure 4, we observe that, as the concentration of crosslinker increases the diffraction efficiency also increases. The presence of crosslinker in the photopolymer system would have been expected to result in a more complex formation of crosslinked chains which could slow down the diffusion process and improve the material's response at high spatial frequencies. However, the increase in diffraction efficiency is observed at all spatial frequencies.

This overall improvement is possibly due to the improved fidelity of the recorded profile (because of lower diffusion) at all spatial frequencies or altered polymerization rates. But, as stated in section 3, all the gratings were recorded with 4 seconds exposure. To study a process like diffusion, which is very fast and to measure the grating growth dynamics in real-time, the exposure time should be in same the order of magnitude of the diffusion time. Such a study was carried out and is explained in section 4.2.

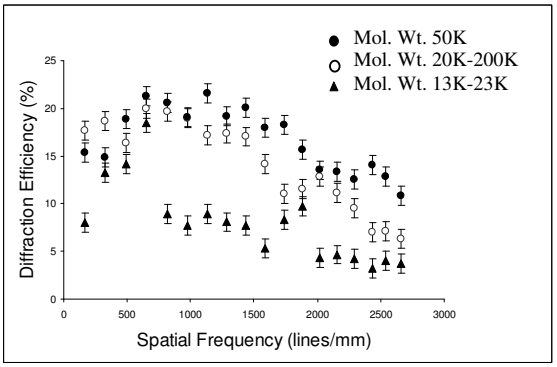

Figure 3 Influence of binder on spatial frequency response

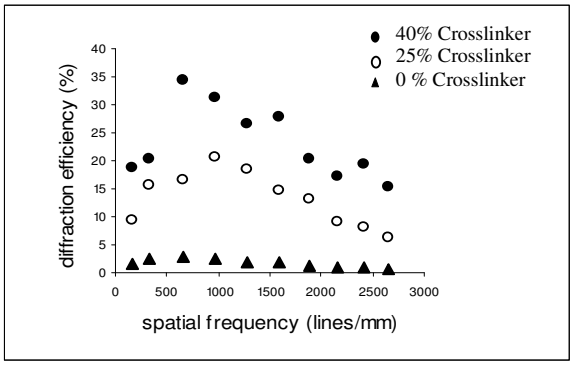

Figure 4 Influence of crosslinker concentration on spatial frequency

4.2 Real-time monitoring

The experimental set-up used to study real-time monitoring of diffraction efficiency growth of the grating is shown in figure 2. The diffraction grating was recorded for a duration of 0.2 seconds with an exposure energy of 6.9mW/cm$^{2.}$ These conditions record 1-2% diffraction efficiency grating. The reason for the short duration of exposure is to observe the processes that occur immediately after the initial exposure is switched off. An example of measured transmission grating kinetics is shown in figure 5.

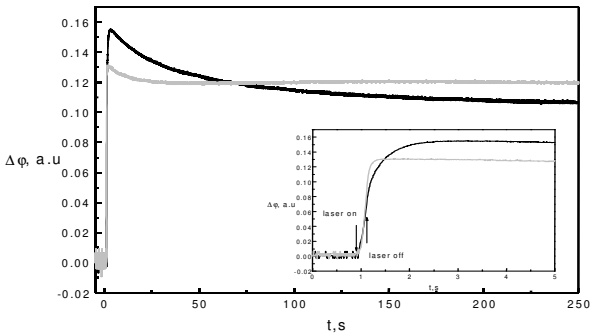

Figure 5. Recording dynamic in layers containing one monomer (gray line) and two monomers (black line)

Binder	Diffusion constant	
	Post process1	Post process2
13,000-23,000 MW	7.80x10^{-8} cm^2/*s*	9.84x10^{-10} cm^2/s
50,000 MW	7.29x10^{-8} cm^2/s	8.50x10^{-10} cm^2/s
20,000-200,000 MW	2.70x10^{-8} cm^2/s	4.44x10^{-10} cm^2/s

Table 1. Diffusion constants measured for different molecular weight binders

In figure 5, after switching off the recording beams (shown with arrows) an initial increase was followed by a slow decrease in the refractive index modulation. In the layers containing only one monomer (acrylamide) both of the post processes are faster than in layers with two monomers (acrylamide and methylene bisacrylamide). The first process which has a positive contribution to the refractive index modulation, was ascribed as monomer diffusion from dark fringe regions to the bright fringe regions and the second process which has a negative contribution to the final refractive index modulation could be the diffusion of free radicals or short chain polymer molecules from the bright fringe regions to dark fringe regions. The second process could be responsible for the poor high spatial frequency response. The experiments were repeated with an exposure time of 0.2 seconds 20 mW/cm^2 for different molecular weight binder compositions. Data from the growth curves were fitted using Microcal origin software. The time constants for the two post processes were extracted and calculated the diffusion constants for each binder composition (shown in Table 1).

5. Reflection holography

Reflection holograms were recorded at 3950-lines/mm spatial frequency for 30s exposure time with an exposure energy of 24 mW/cm^2 in different molecular weight binders and the diffraction efficiencies achieved are shown in the table 2.

Binder	Reflection efficiency
13,000-23,000 MW	3.2 %
50,000 MW	4.7 %
20,000-200,000 MW	10.7 %

Table 2. Reflection grating efficiencies

6. Conclusions

The influence of the binder molecular weight on the spatial frequency response was studied. From the results it is concluded that the range of molecular weight of the binder used in our experiments has no positive influence at higher spatial frequencies. The influence of the concentration of the crosslinking monomer on the spatial frequency response was also studied and found to improve efficiency. The results of real-time monitoring experiments showed that there are two diffusion processes. The first diffusion process which is faster and has positive contribution to refractive index modulation was ascribed as monomer diffusion and the second process with slow decrease, which has negative contribution to refractive index modulation could be diffusion of free radicals or short chain polymer molecules from the bright fringe regions to dark fringe regions. This second diffusion process could be responsible for the poor high spatial frequency response of the investigated system. The diffusion constants for the two processes were calculated. Reflection holograms were recorded with different binder compositions, the highest having a reflection efficiency of 10%.

7. Acknowledgements

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