The changes of holographic characteristics of photopolymer induced by temperature

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The changes of holographic characteristics of photopolymer induced by temperature are studied experimentally. The results show that the exposure sensitivity increases with the increase of temperature. The maximum diffraction efficiency and the final maximum diffraction efficiency increase with the increase of temperature when the temperature is lower than T_g (glass transition temperature), while they decrease with the increase of temperature when the temperature is higher than T_g. The effect of the change of temperature on the saturation refractive index modulation is very weak.

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Volume holographic recording of information holds promise for the next generation digital mass storage systems with high capacity density and fast transfer rate[1]. For holographic storage to become a practical system, a stable recording medium with nearly ideal properties is required. The desirable properties include high diffraction efficiency, rapid in-place development, no wet processing, no grating shrinkage, long shelf life, temperature and moisture stability, high exposure sensitivity and good resolution, etc[2]. It has long been recognized that lithium niobate, the traditional medium of study, is an improper choice for a practical commercial system because of its inadequate refractive index modulation ($\Delta n \sim 10^{-4}$) and low exposure sensitivity (10^{-3} cm^2/mJ)[3]. Photopolymers are attractive alternatives to lithium niobate because they are low cost, can be processed and designed to have large index modulation ($\Delta n \sim 10^{-3}$–10^{-2}) and high exposure sensitivity (10^{-2} – 10^{-1} cm^2/mJ) easily[4,5].

We know that the diffusion process of monomers plays an important role in the formation of the holographic grating in photopolymers, and the diffusion process of monomers has a critical relationship with temperature. Thus, the change of temperature may have large effects on the holographic characteristics of photopolymers. Dhar *et al.* have studied the effects of temperature on the dimension stability and Bragg shifts of the photopolymers[6]. In present paper, we focalize on the changes of holographic characteristics of photopolymers with temperature, such as maximum diffraction efficiency, exposure sensitivity, and refractive index modulation.

Our photopolymer consists of monomers acrylamide (AA) and N, N'-methylenebisacrylamide (BAA), photosensitizer erythrosine B (ERB), photoinitiator triethanolamine (TEA), and the binder polyvinylalcohol (PVA). All these components were of the best available grade and were used without further purification, and the samples were prepared in dark room under the condition (20 – 25 °C temperature and 40 – 60 % relative humidity) as:

1. Basic PVA ($MW \approx 1750$) was dissolved in distilled water and heated to 80 °C to yield a PVA aqueous solution having 10 wt.% of PVA.

2. Crystals of ERB were dissolved in water to obtain the desired dye concentration.

3. A certain volume of TEA was dissolved in water.

4. Added the TEA and ERB solutions into the PVA solution. Finally monomers AA and BAA were added. Mixing the solution well, we got the photopolymeric solution and the total volume was approximately 25 ml.

5. Appropriate volume of the photopolymeric solution was poured on a 6.5 × 6.5 cm^2 glass plate and natural dried 36 − 48 hr, and a photopolymer dry film sample was obtained. The photopolymer dry film is about 230 μm thick, the absorption peak wavelength of samples is at 528 nm.

The experimental setup is shown in Fig. 1. Laser 1 (Ar$^+$, 514 nm) is the recording laser, and for avoiding the effect of absorption, laser 2 (LD, 650 nm) is used as the reading laser. M_1, M_2, and M_3 are mirrors, BS is beam splitter, SH$_1$ and SH$_2$ are shutters, D is dynamometer, BD is a beam deflector whose angle can be controlled with controller CT, and H is the holographic photopolymer sample. L_1 and L_2 are lenses with focal length f, consisting of a $4f$ system, BD is placed in the input plane, and H is placed in output plane.
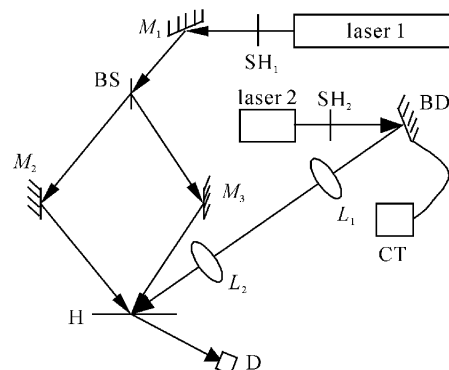

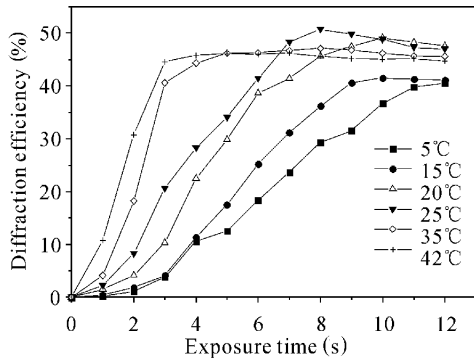
Fig. 1. Experimental setup.

Fig. 2. Diffraction efficiency vs exposure time at different temperatures.

The ratio of reference intensity to object intensity is 1 : 1, total recording power is 8 mW, and reading power is 20 mW. Because the shrinkage or inflation of the sample caused by the recording will result in the Bragg shift of Bragg angle[6], the incidence angle of reading beam must be tuned a little bit to compensate this shift, this can be implemented by BD and $4f$ system. The experiments are completed in the dark room with different constant temperatures, the humidity of the room is controlled at 45% RH.

At first, we measured the diffraction efficiency versus recording time at different temperatures: 5, 15, 20, 25, 35, and 42 °C, as shown in Fig. 2. The diffraction efficiency is defined as the ratio of the power of diffraction beam to the power of reading beam.

From Fig. 2, we got the exposure sensitivity versus the temperature curve, as shown in Fig. 3. The exposure sensitivity is defined as

$$S = 1/E_0, \tag{1}$$

where E_0 is the exposure energy when the diffraction efficiency approaches maximum value.

It can be seen that with the increase of temperature the exposure sensitivity increases monotonously. This is easy to be understood. It is well known that the exposure sensitivity relies on the polymerization rate, and the polymerization rate relies on the diffusion rate of the monomers inside the sample. The diffusion rate increases with the increase of temperature, so the polymerization rate increases with the increase of temperature, and thus the exposure sensitivity also increases with the increase of temperature. On the other hand, the increase in value of exposure sensitivity at the temperature under 25 °C is smaller than that at the temperature above 25 °C. The reason is that 25 °C is close to the glass transition temperature (T_g) 26.3 °C of our photopolymer sample, the sample is solid when the temperature is lower than 25 °C, the diffusion of the monomer is difficult, and the increase in value of exposure sensitivity is small; but the sample becomes soft when the temperature is higher than T_g, this is in favor of the diffusion of the monomers, so the increase of exposure sensitivity is large.

From Fig. 2, we also can get the variation rule of maximum diffraction efficiency (the maximum diffraction efficiency during exposure) with temperature, as shown

in Fig. 4. We can see that the maximum diffraction efficiency increases at first with the increase of temperature, and when the temperature reaches 25 °C, the maximum diffraction efficiency gets its maximum value. And then, when the temperature is higher than 25 °C, the maximum diffraction efficiency will decrease with the increase of temperature. This is because when the temperature is lower than T_g, the grating intensity increases with the increase of diffusion rate, so it increases with the increase of the temperature. So the maximum diffraction efficiency increases with the increase of temperature. When the temperature is higher than T_g, the soft sample is easy to cause distortion, this goes against the maintenance of the grating intensity, so the maximum diffraction efficiency has a little bit decrease when the temperature is higher than T_g.

Secondly, it is good to understand the effect of temperature on the diffusion of monomers and the forming of the holographic grating by monitoring the diffraction efficiency change with the time after exposure for a long time (150 s) at different temperatures. The result is shown in Fig. 5.

Evidently, because the monomers have not polymerized completely during the exposure, the residual monomers will continue to diffuse from dark area to bright area and to polymerize, so the grating intensity will continue to increase with the increase of time. With the diffusion and polymerization, the concentration of the monomer is smaller and smaller, so the increase of grating intensity and diffraction efficiency is slower and slower until the monomers are polymerized completely, then the grating intensity and diffraction efficiency get saturation. At different temperature point, the diffusion rate is
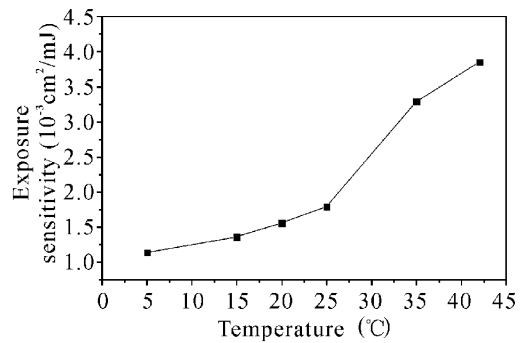

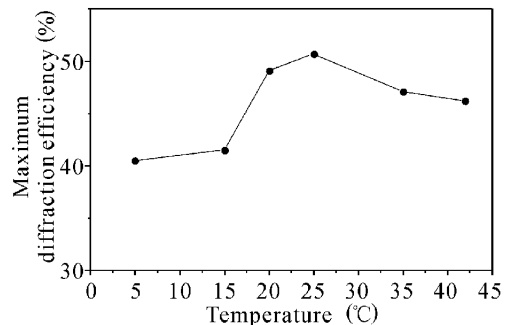Fig. 3. Exposure sensitivity vs temperature.

Fig. 4. Maximum diffraction efficiency vs temperature.
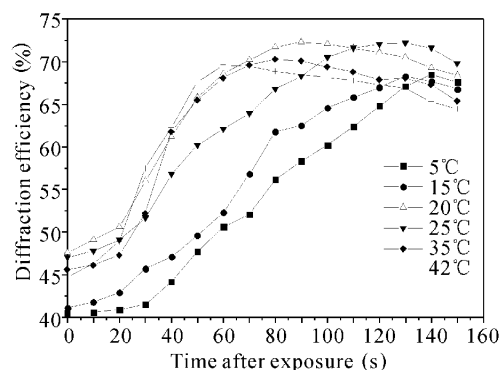
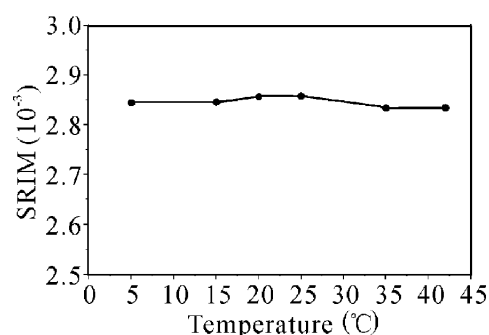Fig. 5. Diffraction efficiency vs the time after exposure.

Fig. 6. Saturation refraction index modulation vs temperature.

different, so the increase rate of the diffraction efficiency is different. The lower the temperature, the slower the increase rate. This is just the result of experiment, as shown in Fig. 5.

Just at the situation during exposure (recording), the diffraction efficiency gets its maximum value at 25 °C, and when the temperature is lower than 25 °C, the final maximum diffraction efficiency (the maximum diffraction efficiency after the exposure, i.e. the maximum in every curve in Fig. 5) increases with temperature, while it decreases with the temperature when the temperature is higher than 25 °C. The reason of this phenomenon is just like the reason discussed above.

With the coupled wave theory[7], we know the saturation refraction index modulation (SRIM) can be calculated as

$$\Delta n = \frac{\lambda \sin^{-1}(\eta \cos\theta_r \cos\theta_s)^{1/2}}{\pi d}, \qquad (2)$$

where d is the sample thickness, λ is the recording laser wavelength, θ_r and θ_s are the incidence angles of reference beam and signal beam to the normal of the sample surface, respectively, and η is the final maximum diffraction efficiency. Using the experimental results shown in Fig. 5 and the parameters used in our experiment, we calculated the curve of SRIM versus temperature. The result is displayed in Fig. 6. We can see from it that the change of the SRIM is very small (about $10^{-6} - 10^{-7}$) when the temperature changes. This indicates that the temperature has a very little effect on the SRIM. The reason is that the monomers will polymerize completely if the time keeps up long enough no matter at higher temperature or lower temperature, so SRIM will reach its saturation value that is not changing with the temperature finally.

We have studied the effect of the temperature on the holographic characteristics experimentally. The results show that the exposure sensitivity increases with the increase of temperature. The maximum diffraction efficiency during exposure and the final maximum diffraction efficiency after exposure increase with the increase of temperature when the temperature is lower than T_g, while they decrease with the increase of temperature when the temperature is higher than T_g. The change of the temperature has a weak effect on the saturation refractive index modulation.

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