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3-D behaviour of photopolymers as holographic recording material

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ABSTRACT

Research dealing with models to predict and understand the behaviour of photopolymers have generated many interesting studies considering a 2-dimensional geometry. These models suppose that the photopolymer layer is homogeneous in depth. Using this approximation good results can be obtained if the thickness of photopolymers is less than 200 μm . However, it is well known that Lambert-Beer's law predicts an exponential decay of the light inside the material. In recent years intensive efforts have been made to develop new holographic memories based on photopolymers. For this application the thickness of the layer is increased, usually to more than 500 μm , and Lambert-Beer's law plays a significant role in the recording step. The attenuation of the index profile inside these materials has been measured, showing that it is an important phenomenon. This attenuation limits the maximum effective optical thickness of the grating and shows that the 2-D models can not be applied in these cases. For this reason in this work a 3-dimensional model is presented to analyze the real behaviour of the photopolymers and study the variations in the index profile in depth. In this work we examine the predictions of the model in the case of a general dependence of the polymerisation rate with respect to the intensity pattern, and the effects of varying the exposure intensity are also compared in 3-D cases. Finally, the limitation of the data storage capacity of the materials due to the Lambert-Beer law is evaluated.

1. INTRODUCTION

Photopolymers have been of interest as holographic recording materials for more than three decades¹⁻³. Their low cost and good optical properties (high diffraction efficiency, acceptable resolution and signal/noise ratio, etc. Ref.4-5) are well known and have been much studied by multinational corporations such as DuPont and Polaroid. In recent years, a new application for this type of material has been developed. The possibility of using thick layers of photopolymers as holographic memories may potentially be the best way forward. The behavior of the material and conservation of the stored information depend on many factors: the chemical composition, thickness, drying time of the material, spatial frequency, recording intensity, etc. Numerous models have been proposed to explain the effects of these factors⁶⁻⁸.

In the 1990's the first models that took into account the importance of the polymerization rate and monomer diffusion processes in the formation of the hologram appeared. In the work of Adhami et al.⁶ equations governing the formation of the hologram were first proposed and solved. Zhao and Moroulis proposed a different model⁸, where the monomer diffusion assumes quite an important role when a sinusoidal exposure is used. In their model one assumes a linear relation between polymerization and incident intensity; in addition a linear relation between the index modulation and polymer concentration is postulated. The differential equations provide a semi-analytical way of explaining why the index modulation is smaller when the separation between strips increases, that is to say, when low spatial frequencies are registered. Subsequently, a lot of new models were presented, taking into account different aspects of the hologram formation process⁹⁻¹¹.

In this paper, the attenuation of the light with depth is included in a first harmonic diffusion model¹¹⁻¹² to obtain the index profile recoded in photopolymers in the case of a general dependence of the polymerisation rate with respect to the intensity pattern, and the effects of varying the exposure intensity are also compared for layers 800 μm thick.

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2. THEORETICAL MODEL

This intensity distribution is assumed constant in y , but attenuated in depth along the z axis. This depth attenuation changes the angular response of the grating stored in the material and determines the optical thickness and ultimately the storage capacity of each material composition [13]. Assuming Beer's law to explain the attenuation of the light intensity with depth, the intensity inside the material must be written as:

$$I(x, z, t) = I_0 [1 + V \cos(K_g x)] e^{-\alpha(t)z} \quad (1)$$

where α is the coefficient of the light attenuation. The initial value of α [$\alpha(t=0) = \alpha_0$] can be obtained if the transmittance and the physical thickness of the layer are known. In our material for layers with a physical thickness of around 800 μm the transmittance is around 0.82%, in others words, the initial values of α_0 for this composition¹⁴ are around 0.006 μm^{-1} . The absorption of the layer changes as a function of the time when the dye is consumed, a first approximation can be written as:

$$\alpha(t) = \alpha_0 e^{-K_\alpha I_0^\beta t} \quad (2)$$

where β is a constant that determines the influence on the intensity as the dye is consumed and depends on many factors (chemical composition of the material, temperature, humidity, etc.), the absorption decay depends on the dye and the intensity used, and K_α defines the rate at which the dye is consumed. The normal evolution of the transmittance of the layer for intensities of 1, 5, 10 and 50 mW/cm^2 and $\beta = 1$ is plotted in Fig. 1, and the constant takes the following value: $K_\alpha = 0.005 \text{ cm}^2 \text{ mW}^{-1} \text{ s}^{-1}$.

Fig. 1.- Transmittance of the layers 800 μm thick as a function of time

We propose the use of the following two equations to describe 3-dimensional polymerization:

$$\frac{\partial [M](x, z, t)}{\partial t} = \frac{\partial}{\partial x} D(t) \frac{\partial [M](x, z, t)}{\partial x} - k_R(t) I^\gamma(x, z, t) [M](x, z, t) + \frac{\partial}{\partial z} D(t) \frac{\partial [M](x, z, t)}{\partial z} \quad (3)$$

$$\frac{\partial [P](x, z, t)}{\partial t} = k_R(t) I^\gamma(x, z, t) [M](x, z, t) \quad (4)$$

where n is the refractive index, $[M]$ is the monomer concentration, $[P]$ is the polymer concentration, and γ indicates the relationship between the intensity and the polymerization rate (k_R).

We assume that the decrease in polymerization rate due to the Trommsdorff effect after the process is initiated is given by:

$$k_R(t) = k_R \exp(-\phi I_0^\gamma t) \quad (5)$$

where ϕ is the attenuation coefficient of the polymerization rate.

$$D(t) = D_0 \exp(-\phi I_0^\gamma t) \quad (6)$$

We defined the diffusion time constant, τ_D , as:

$$\tau_D = \frac{1}{D_0 K_g^2} \quad (7)$$

If the thickness of the material is around 1mm and the period of the grating recorded is lower than 1 μm , then we can assume that:

$$\frac{\partial[M](x,z,t)}{\partial x} \gg \frac{\partial[M](x,z,t)}{\partial z} \quad (8)$$

Using this approximation the monomer diffusion along the z axis can be neglected, and equations. 3 and 4 can be solved in a similar manner to that used for the 2-D case.

Our simple approach to solve these equations in order to obtain qualitative information about the behaviour of the material in depth is as follows: The photopolymer film is divided into G different sub-films each of thickness d_g ; the total thickness of the photopolymer, d , is obtained from the sum of the thicknesses of the different sub-films.

$$d = \sum_{g=1}^G d_g \quad (9)$$

where G is the number of sub-films used

The exposure intensity in each sub-grating can be calculated using eq. (2) and the initial monomer concentration of each is , the same as that of the global grating. Knowing the initial conditions for each layer, the 2-dimensional model can be used to examine the 3-D problem.

In order to consider a particular photopolymer film useful for holographic memory applications, its thickness must be greater than 500 μm . In our research group the thickness of typical photopolymers layers is between 500 μm and 1000 μm . For this reason the minimum value of $G = 100$. In this work the value taken for the thickness is 800 μm .

In this paper the model used to solve the diffusion equations in each grating will be a first harmonic diffusion model, because the presence of higher harmonics can often be neglected in photopolymers [26].

The influence of the refractive index of each photopolymer component and the polymer and monomer harmonics on the index modulation can be expressed as¹³:

$$n_1 = \frac{(n_{dark}^2 + 2)^2}{6n_{dark}} \left[- \left(\frac{n_m^2 - 1}{n_m^2 + 2} - \frac{n_b^2 - 1}{n_b^2 + 2} \right) [M]_1 + \left(\frac{n_p^2 - 1}{n_p^2 + 2} - \frac{n_b^2 - 1}{n_b^2 + 2} \right) [P]_1 \right] \quad (10)$$

where: n_p , is the polymer refractive index, n_m , is the monomer refractive index, n_b , is the binder refractive index, n_{dark} , is the refractive index of the layer before exposure. In the material used in this work the different refractive indexes take the follow values: $n_m = 1.486$; $n_b = 1.474$; $n_{dark} = 1.478$. These values were obtained using the Lorentz-Lorenz equation [15], based on refractometer measurements using water solutions.

3. RESULTS AND DISCUSSION

In this section the influence of the recording intensity on the profile and index modulation recorded in the photopolymer using a 3-D analysis is evaluated. To do this, the results obtained for different values of γ and I_0 are simulated. The values used in these simulations are shown in table 1.

Table 1. Values of the parameters used in the simulations.

I_0	γ	γ	γ	γ
1 mW/cm ²	0.3	0.5	0.7	1
10 mW/cm ²	0.3	0.5	0.7	1
50 mW/cm ²	0.3	0.5	0.7	1

In the first case (Figure 2) the profiles stored in the grating after 20 seconds for different values of γ (1, 0.7, 0.5 y 0.3) are shown. In this figure it can be seen for materials with high values of γ , greater attenuations in depth of refractive index are obtained, but the index modulation of the material near the surface is also greater. Therefore, the effective optical thickness of the holographic gratings stored in these materials will be less.

In the next case, we consider higher values of the mean recording intensity. Figure 3 shows the profile stored in the material after 20 s when $I_0 = 10\text{mW}/\text{cm}^2$. In this case, in addition to obtaining a smaller attenuation of the index with thickness for the lower values in the range, the attenuation is seen to be less when lower values of recording intensity are considered. This result is to be expected if we bear in mind that these materials are an exception to the rule of reciprocity. Therefore, if we want to record uniform diffraction gratings, it is advisable to use a high intensity in order to obtain higher values of effective optical thickness.

CONCLUSIONS

When the results of the three-dimensional analysis of thick layers are evaluated for different values of γ and I_0 , two proposals immediately arise: to use the mechanism of polymerization with low values of γ to obtain more uniform holograms (higher values of effective optical thickness and large values of data storage capacity) and to use high values of the recording intensity with short exposures in multiplexing methods to obtain more uniform gratings. Nevertheless if the values of γ are close to 1 the material presents better energetic sensitivity and higher values of refractive index modulation.

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Fig.3. Refractive index profile when $I_0 = 10 \text{ mW/cm}^2$ and for different values of $\gamma = 1, 0.7, 0.5$ and 0.3 .

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