

3 Dimensional analysis of holographic photopolymers based memories

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Abstract: One of the most interesting applications of photopolymers is as holographic recording materials for holographic memories. One of the basic requirements for this application is that the recording material thickness must be 500 μm or thicker. In recent years many 2-dimensional models have been proposed for the analysis of photopolymers. Good agreement between theoretical simulations and experimental results has been obtained for layers thinner than 200 μm . The attenuation of the light inside the material by Beer's law results in an attenuation of the index profile inside the material and in some cases the effective optical thickness of the material is lower than the physical thickness. This is an important and fundamental limitation in achieving high capacity holographic memories using photopolymers and cannot be analyzed using 2-D diffusion models. In this paper a model is proposed to describe the behavior of the photopolymers in 3-D. This model is applied to simulate the formation of profiles in depth for different photopolymer viscosities and different intensity attenuations inside the material.

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1. Introduction

Photopolymers have been of interest as holographic recording materials for more than three decades [1-5]. Their low cost and good optical properties (a high diffraction efficiency, an acceptable resolution and signal/noise ratio, etc. [5-7]) 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 the use of thick layers of photopolymers as holographic memories may potentially be the best way forward.

The behavior of the material and the conservation of the stored information depend on many factors: the chemical composition, the thickness, the drying time of the material, the spatial frequency, the recording intensity, etc. To understand the effects of these factors numerous models have been proposed [8-13].

In the 1990's the first models that included the importance of polymerization rate and monomer diffusion processes in the description of the formation of the hologram appeared. In the work of Adhami *et al.* [14] equations governing the formation of the hologram were first proposed and solved. Zhao and Moroullis proposed a different model [8], where the monomer diffusion assumes a quite important role when a sinusoidal exposure is used. In their model one assumes a linear relation between the polymerization and the incident intensity, in addition a linear relation between the modulation of index and the 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 space frequencies are registered. Following a lot of new models were presented, taking into account different aspects of the formation hologram process [9-13, 15-17]. All of these models assume a sinusoidal exposure along the x axis is:

$$I(x) = I_0 [1 + V \cos(K_g x)] \quad (1)$$

where I_0 is the average intensity ($I_0 = 5 \text{ mW/cm}^2$ in this work), V is the fringe visibility ($V = 1$ in this work), and K_g is the grating wave number.

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 [18] and determines the optical thickness and ultimately storage capacity of each material composition [20-21]. Assuming Beer's law explain the attenuation of the light intensity with depth, Eq. (1) must be rewritten as:

$$I(x, z) = I_0 [1 + V \cos(K_g x)] e^{-\alpha(t)z} \quad (2)$$

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 [22-25] for layers with physical thickness around $800 \text{ }\mu\text{m}$ the transmittances are around 0.82%, in others words, the initials values of α_0 for this composition are around $0.006 \text{ }\mu\text{m}^{-1}$ [24, 25]. The absorption of the layer changes as a function of the time when the dye is consumed, in a first approximation can be written as:

$$\alpha(t) = \alpha_0 e^{-K_\alpha I_0^\beta t} \quad (3)$$

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.), 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 power intensity of 5 mW/cm^2 and $\beta = 1$ is plotted in Fig. 1, and the constant takes the following value: $K_\alpha = 0.005 \text{ cm}^2 \text{ mW}^{-1} \text{ s}^{-1}$.

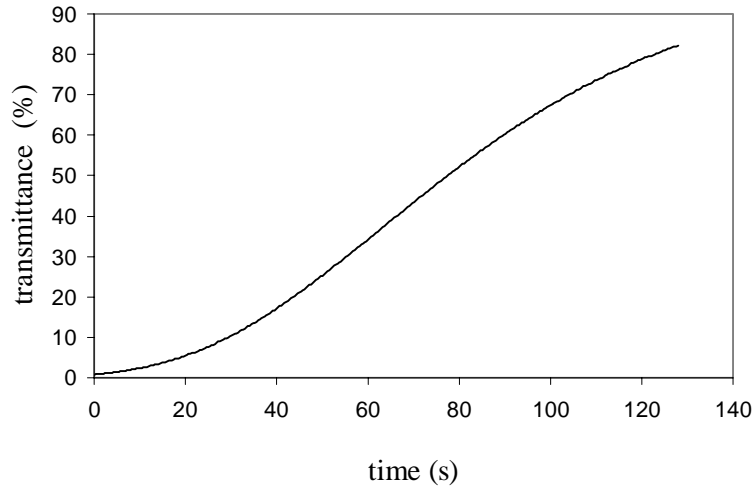


Fig. 1. Transmission of a.800 μm thick layer as function of exposure time.

In this paper the attenuation of the light with depth is included in a first harmonic diffusion model [13], to obtain an estimation of the parameters involved in hologram formation in photopolymers with physical thickness around 0.8 mm. Using this model the distributions of the residual monomer, the polymer and the index modulation inside the layer can be simulated. In our research group the Polyvinylalcohol/Acrylamide (PVA/AA) based materials are used [21-23]. In this work values of the polymerization and diffusivity determining using are the values obtained by the 2-dimensional model for layers of 800 μm thick. This type of photopolymer is frequently chosen to be the main compound in holographic memories [24-25]. Their low price, easy preparation and the fact that complicated developing processes are not necessary makes them attractive for use on a large scale in read only WORM (write once read many) type memories.

2. Theoretical model

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

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

$$\frac{\partial[P](x, z, t)}{\partial t} = k_R(t) I^\gamma(x, z, t) [M](x, z, t) \quad (5)$$

where n is the refractive index, $[M]$ is the monomer concentration, $[P]$ is the polymer concentration, γ indicates the relationship between the intensity and the polymerization rate (k_R). In this paper, we assume $\gamma=1$, linear intensity response, to simplify our calculations.

We assume that the polymerization rate decreases due to the Trommsdorff effect, is given by:

$$k_R(t) = k_R \exp(-\varphi I_0 t) \quad (6)$$

where φ is the attenuation coefficient of the polymerization rate.

We defined the diffusion time constant, τ_D , as:

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

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 eqn. 3 and 4 can be solved in a similar manner to that used for the 2-D case [13].

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 will be divided into G different sub-films each of thickness d_g (Fig. 2); the total thickness of the photopolymer, d , can be obtained as the sum of the thickness of the different sub-films.

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

where G is the number of sub-films used.

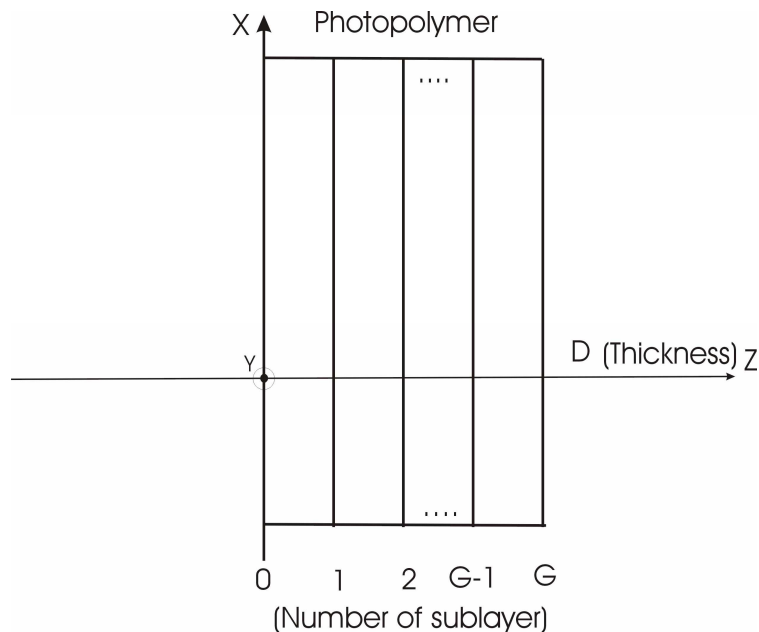


Fig. 2. Holographic grating structure

The exposing intensity in each sub-grating can be calculated using eqn. (2) and the initial monomer concentration of each is M_0 , 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 are 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 gratings will be a first harmonic diffusion model, because the presence of the higher harmonics can often be neglected in photopolymers [26].

The influence of the refraction indexes of the each photopolymer component and the polymer and monomer harmonics on the index modulation can be expressed as [13]:

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

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 refraction index of the layer before the exposure.

In the material used in this work the different refractive indexes take the follow values:

$$n_m = 1.486$$

$$n_b = 1.474$$

$$n_{dark} = 1.478$$

This values were obtained using the Lorentz-Lorenz equation and the method used is described in reference [27]. The calculations are based on refractometer measurements using water solutions.

As in others models referenced in this paper [10,11,13] the relation between polymerization and diffusion is represented by the dimensionless parameter R :

$$R = \frac{D K_g^2}{k_R I_0} \quad (11)$$

For materials where the diffusion rate is faster than the polymerization rate, $R > 1$ and high values of index modulation can be recorded in this case, higher fidelity recordings. For materials where the polymerization is faster than monomer diffusion $R < 1$ and low values of index modulation are recorded, more distortion occurs.

3. Results and discussion

The diffusivity of the monomer molecules (D) is a critical parameter [28]. In this section different kind of material systems are analyzed. In the first place to thoroughly explore the different behaviours, values of D ranging from $2 \times 10^{-13} \text{ cm}^2/\text{s}$ (highly viscous systems) to

$5 \times 10^{-9} \text{ cm}^2/\text{s}$ (liquid systems) were studied [28]. In the second place different light attenuations throughout the depth of the material are simulated. These changes in the material absorption is due to two different factors: variations of the dye concentration or different kind of dye used in the photopolymer composition.

3.1 Different material viscosities

The standard values of the parameters that governs the polymerization in PVA/AA thick layers are around: $k_R \sim 0.016 \text{ cm}^2 \text{ mW}^{-1} \text{ s}^{-1}$; $\tau_D \sim 10 \text{ s}$ for 1125 lines/mm ($D \sim 10^{-11} \text{ cm}^2/\text{s}$); $\varphi \sim 0.006 \text{ s}^{-1}$; and $\alpha_0 \sim 0.006 \mu\text{m}^{-1}$ [29]. These values indicate that $R \sim 1.25$. Using these parameters the distribution of refraction index inside the material for different time exposure have been simulated. The results are plotted in Fig. 3 and the maximum refraction index inside the material is 1.479 at $d = 50 \mu\text{m}$ and $t = 100 \text{ s}$. In this Fig. the attenuated refraction index profile can be seen. It is interesting to observe that no grating is recorded in the last $200 \mu\text{m}$ for exposure times shorter than 40 s. This aspect is very important in the fabrication of holographic memories, because the effective optical recording thickness is thinner than the physical thickness of the material layer and the resulting storage capacity is lower.

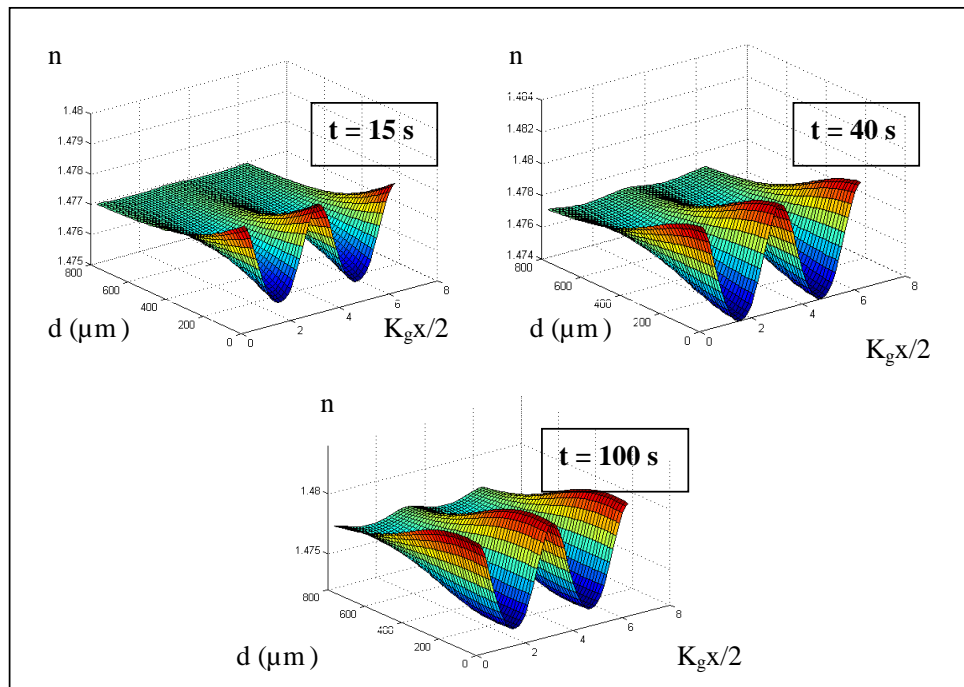


Fig. 3. Refractive index distribution within the photopolymer for three different recording times (15 s, 40 s and 100 s) for standard parameters ($D \sim 10^{-11} \text{ cm}^2/\text{s}$ and $R \sim 1.25$).

Other interesting aspect to analyze for this type of material is the growth of the refraction index modulation versus time and thickness. This aspect is represented in Fig. 4(a) where the maximum index modulation recorded is $n_f = 4 \times 10^{-3}$. The distribution of the residual monomer inside the hologram is plotted in Fig. 4(b). As can be seen in this Fig., the values of residual free monomer in the first $400 \mu\text{m}$ are lower than 10%, but in the last $400 \mu\text{m}$ the values of free monomer are much higher. This factor determines the conservation of the stored information when the photopolymer film is exposed at ambient light [21] and the dynamic range of the material [23].

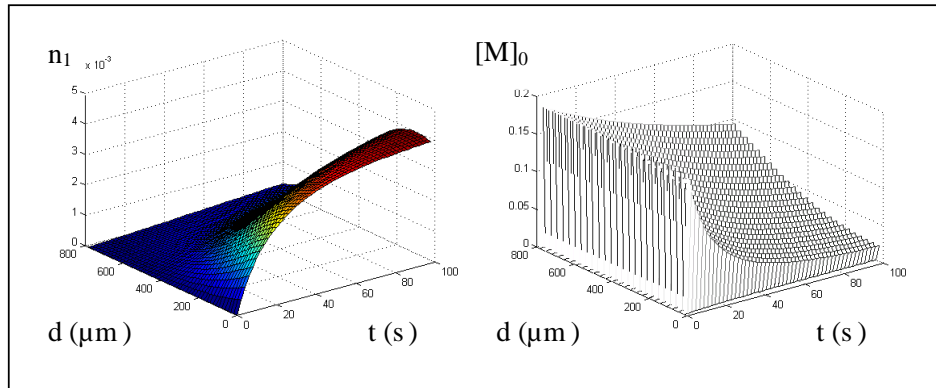


Fig. 4. Refractive index modulation distribution within the photopolymer versus thickness and time for standard parameters ($D \sim 10^{-11} \text{ cm}^2/\text{s}$ and $R \sim 1.25$).

It is interesting to analyze the behaviour of materials with high diffusion time constants and low diffusivity, $\tau_D \sim 500 \text{ s}$ ($D \sim 2 \times 10^{-13} \text{ cm}^2/\text{s}$, $R \sim 0.02$). In these types of materials the higher harmonics of polymer and monomer concentrations cannot be neglected [12], in other words the profile recorded in the photopolymer is not sinusoidal. Nevertheless this model provides interesting qualitative information on the distribution of the refractive index in depth. In Fig. 5 the distribution of the refractive index is presented for three different exposure times (15, 40 and 100 s). The maximum of the refractive index is smaller ($n = 1.4775$ for $d = 300 \mu\text{m}$ and $t = 100 \text{ s}$) due to the large diffusion time constant. One interesting characteristic of this materials is that the maximum refractive index is located within the material (at a depth around $150 \mu\text{m}$ for $t = 40 \text{ s}$ and around $300 \mu\text{m}$ for $t = 100 \text{ s}$). The reason for this effect is the decrease in the parameter R with depth when the light is attenuated within the material (see Eq. (10)). Another particular characteristic of this type of material is that the distribution of index profile in depth is more uniform than in the cases with greater diffusivity.

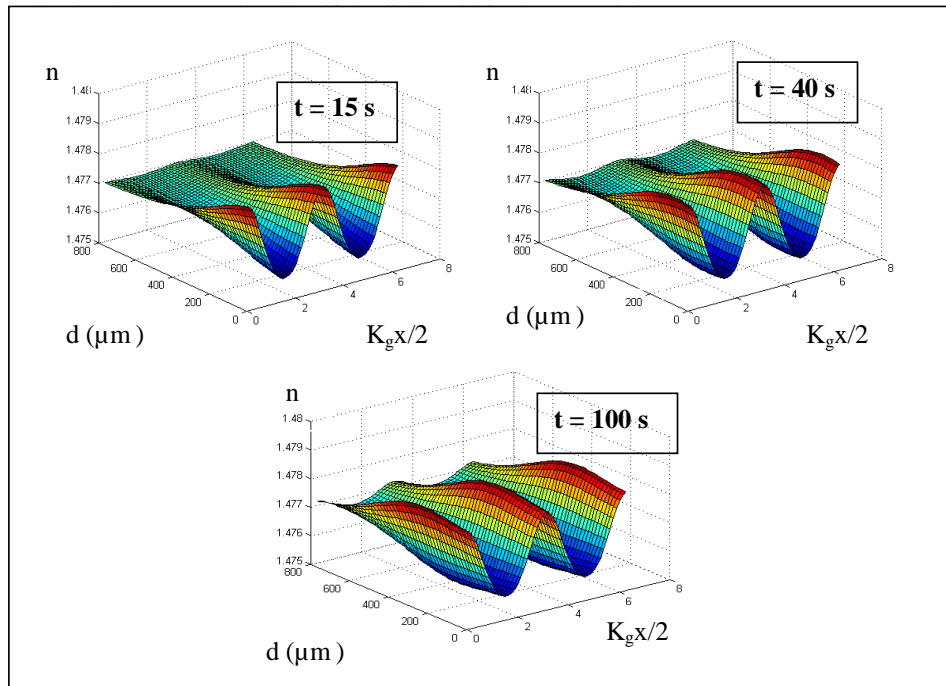


Fig. 5. Refractive index distribution within the photopolymer for three different recording times (15 s, 40 s and 100 s) for usual parameters of viscous photopolymers: $D \sim 2 \times 10^{-13} \text{ cm}^2/\text{s}$ and $R \sim 0.02$.

Furthermore for this type of low diffusivity material another characteristic studied is the behaviour of the index modulation versus time (Fig. 6(a)) and the high values of the residual monomer (Fig. 6(b)). In Fig. 4(a) we can see the maximum value of the index modulation, $n_I = 1.5 \times 10^{-3}$, is achieved around $300 \mu\text{m}$ inside the material when $t = 100 \text{ s}$. On the other hand the curves that present n_I as a function of exposure time, for low values of R , are similar to those obtained in [13]. In this case, for viscous systems, the unpolymerized monomer is higher than the first case, this is an important drawback if one wishes to preserve the stored pattern (data/information), but is an important advantage if one wishes to record many holograms at a single location in the material using multiplexing methods [30].

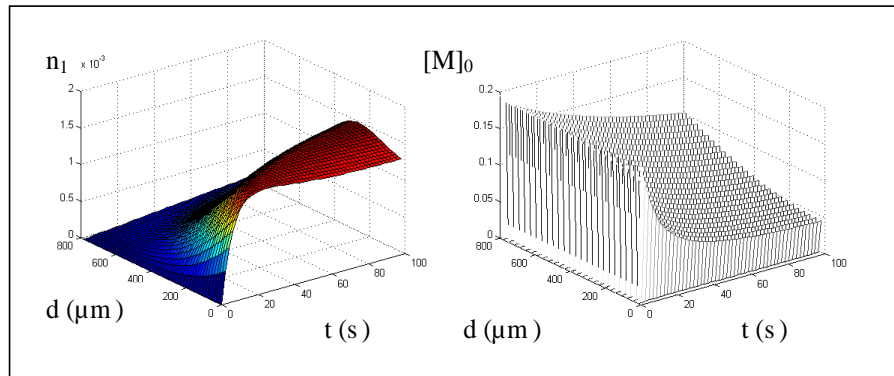


Fig. 6. Refractive index distribution within the photopolymer versus thickness and time for usual parameters of viscous photopolymers: $D \sim 2 \times 10^{-13} \text{ cm}^2/\text{s}$ and $R \sim 0.02$.

The last type of materials analysed are materials with high diffusivity ($D \sim 5 \times 10^{-9} \text{ cm}^2/\text{s}$ and $R \sim 60$). These types of materials are found to have low values of higher harmonics of index modulation [12] and thus to give high fidelity recording. The index profile recorded within the material is represented in Fig. 7 for three different exposure times. As can be seen, for this type of material the differences between the maximum and minimum values of refraction index are larger, due to a higher index modulation stored. The maximum value of refractive index achieved is ($n = 1.48$ at $d = 1 \text{ }\mu\text{m}$ and $t = 100 \text{ s}$). On the other hand the gradient of index modulations in depth is higher too, in other words, the refractive index is less uniform inside the material. As the diffusivity is high, the maximum refractive index recorded in such a material is recorded very close to the surface (see Fig. 7 and see Fig. 8(a)).

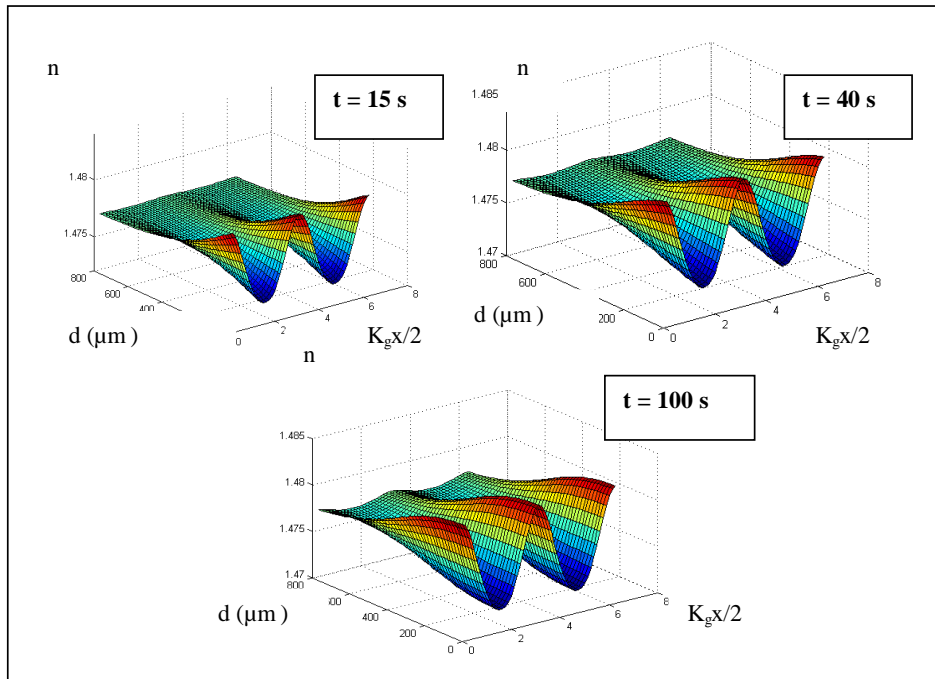


Fig. 7. Refractive index distribution within the photopolymer for three different recording times (15 s, 40 s and 100 s) for usual parameters of liquid systems: $D \sim 5 \times 10^{-9} \text{ cm}^2/\text{s}$ and $R \sim 60$.

As in the same case of 2-D models the values of index modulation are higher for high values of R (large values of diffusivity). This characteristic is shown in Fig. 8(a) where higher values of the monomer gradient in axis z can be seen. In Fig. 8(b) the residual monomer within the material as function of time is shown. In this case low values of residual monomer are presented. As can be seen in the first 200 μm almost all monomer has been consumed after 40s.

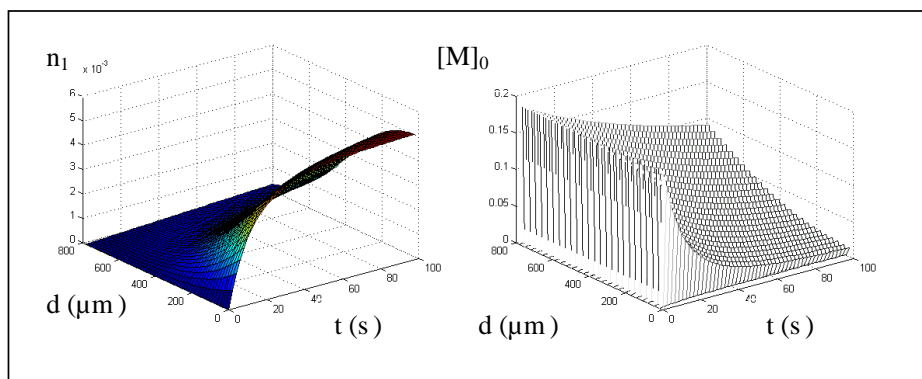


Fig. 8. Refractive index distribution within the photopolymer as function of thickness and time for usual parameters of liquid systems: $D \sim 5 \times 10^{-9} \text{ cm}^2/\text{s}$ and $R \sim 60$.

3.2 Different light attenuation inside the material

In order to optimize holographic materials for WORM memories three factors have to be considered, the stored capacity of the holograms, the dynamic range and the energetic sensitivity. The dye concentration plays the most important role in the final values of these factors. If the dye concentration is increased it produces an increase of the light attenuation (α), the polymerization rate constant (k_R), and the noise [31]. Now greater values of α ($0.01 \mu\text{m}^{-1}$) and k_R ($0.025 \text{ cm}^2 \text{ mW}^{-1} \text{ s}^{-1}$) will be considered. The new distribution of the refraction index in depth is plotted in Fig. 9 for three different exposure times (15, 40 and 100 s). As can be seen in this Fig. no diffraction grating is recorded in last 400 μm of material. It is important to note that the low effective thickness reduces dramatically the storage capacity of material.

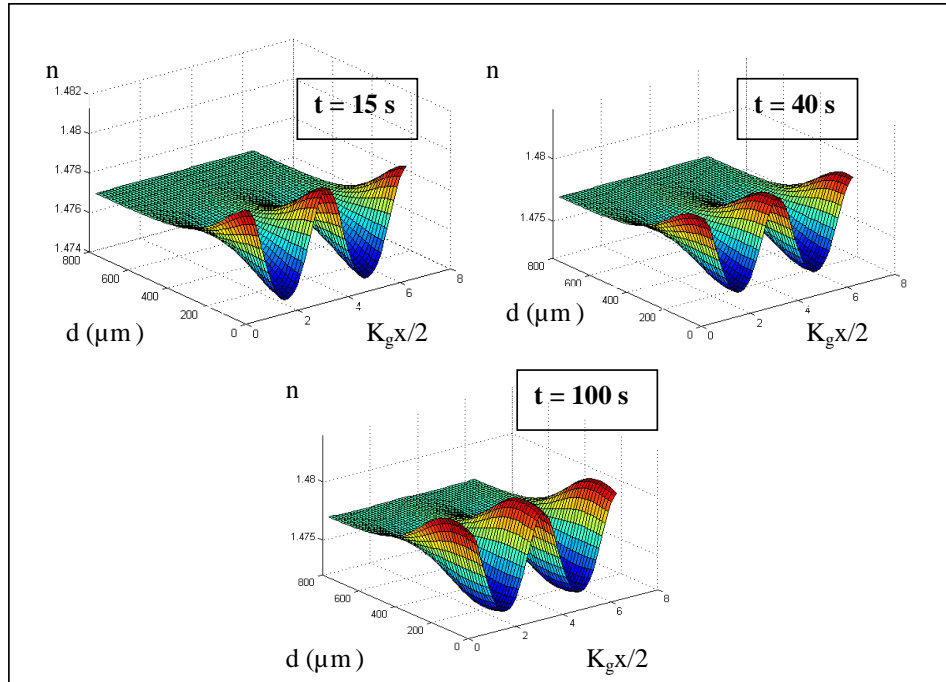


Fig. 9. Refractive index distribution within the photopolymer for three different recording times (15 s, 40 s and 100 s) and for a high dye concentration ($\alpha = 0.01 \mu\text{m}^{-1}$).

Once the representations of the index modulation and residual monomer as a function of thickness and exposure time are represented (Fig. 10), it is possible to observe that the index modulation in the zones near the photopolymer surface is higher and the free monomer near the surface is very low. Nevertheless this type of gratings present high gradients of index modulation and residual monomer in depth, in other words, they are clearly highly nonuniform.

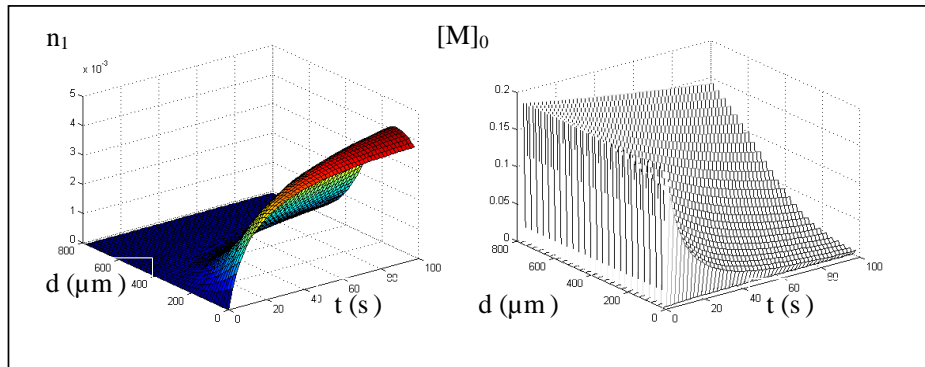


Fig. 10. Refractive index distribution within the photopolymer as function of thickness and time for high dye concentration ($\alpha = 0.01 \mu\text{m}^{-1}$).

Opposite in behaviour to the previous case are material with low dye concentration. The behaviour of this type of photopolymer can be modeling reducing the values of α (in this case $0.003 \mu\text{m}^{-1}$) and k_R (in this case $0.008 \text{ cm}^2 \text{ mW}^{-1} \text{ s}^{-1}$). In Fig. 11 the grating profile for three different exposure times are represented. The figures obtained show that in this case all the physical thickness of the photopolymer layer contains stored gratings. Nevertheless attenuation of the grating profile in depth can still be observed. This attenuation is the origin of smoothing in the off-Bragg response curves modelled by Uchida [32] for the first order and has also been reported in the higher orders by Neipp et al. [18], who demonstrated that an attenuated refractive index profile is stored in volume gratings recorded in PVA/Acrylamide photopolymer materials.

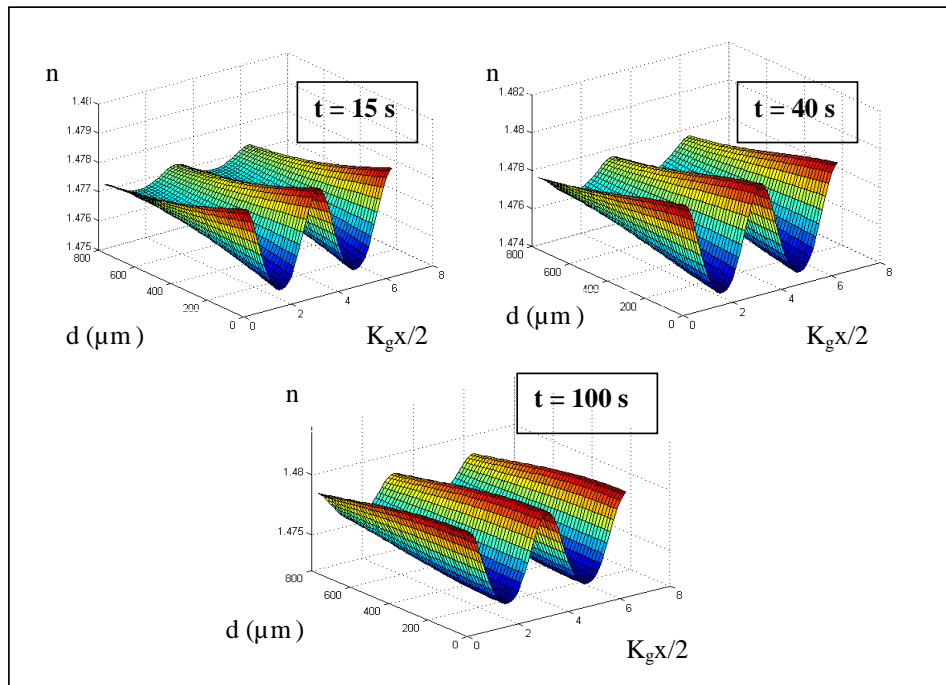


Fig. 11. Refractive index distribution within the photopolymer for three different recording times (15 s, 40 s and 100 s) and $\alpha = 0.003 \mu\text{m}^{-1}$.

The values of index modulation and residual monomer as a function of thickness and exposure time are represented in Fig. 12(a) and Fig. 12(b) respectively. The values obtained using the diffusion model show lower values of index modulation (low energetic sensitivity) due to the reduction of the polymerization rate constant. Also, it is important to mention that the values of residual monomer are again very high again, this is an important problem if we wish accurately preserve the information stored in photopolymer film.

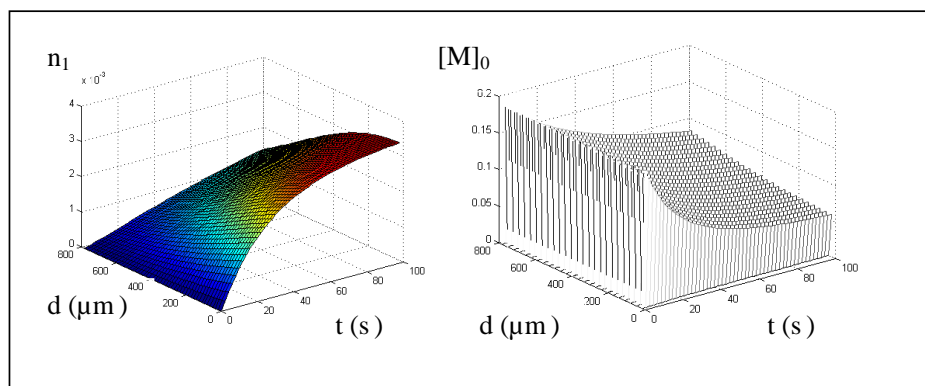


Fig. 12. Refractive index distribution within the photopolymer as function of thickness and time with $\alpha = 0.003 \mu\text{m}^{-1}$.

Taking in account of our results for three values of light attenuation analyzed, the main result to be considered in the fabrication of holographic memories is the attenuation of light inside the material. This phenomenon occurs due to Beer's law and is the origin of the decay of the refractive index profile within the depth and it will limit the storage capacity [20]. On the other hand, one effect of the attenuation is to smooth the diffraction efficiency curve. This effect is well-known, Uchida for instance [30], explained it using analytical solutions of the diffraction efficiency of the first order obtained from a two coupled wave method, and Neipp *et al.* explained it using numerical solutions using an algorithm based on Rigorous Coupled Wave Theory [18]. Nevertheless the application of these ideas to the experimental manufacture of holographic memories presents some problems due to the low index modulations stored when the dye concentration is low [24].

4. Conclusions

A simple flexible method has been proposed for modelling the 3-D formation of holographic gratings in thick photopolymers. This material is available to be used as holographic memory, but due to Beer's law the capacity of this material can be reduced. Three different diffusivities of the material have been studied. The materials with high viscosity present higher uniformity in the refractive index profile in depth but the index modulation is lower and the values of residual monomer is high. One characteristic of this type of material is the presence of maxima of refractive index located far from the input exposing of the photopolymer layer. The materials with high diffusivity produce high values of index modulation near the material surface, but the index modulation decreases quickly in depth. All the monomer in the zones near the surface is consumed.

The importance of the dye concentration in the material has been evaluated for three different values of the light attenuation inside the material. As a result, the limitation of optical thickness and storage capacity in materials with high values of dye concentration has been demonstrated. Using the 2-dimensional diffusion model applied in each sub-layer the necessity to obtain low values of the light attenuation inside the material in order to obtain higher values of the optical thickness have been demonstrated. In other words, with these simulations we have demonstrated the attenuated in the refractive index profile stored in the volume gratings recorded in PVA/AA photopolymer materials. On the other hand using high values of monomer and dye we can obtain materials with high values of dynamic range and good energetic sensitivities. In addition, experimental results show that when the values of dye are really low the values of effective optical thickness are larger but the values of diffraction efficiencies are lower than 10 %, taking into account this aspect [20,24].

Two proposals immediately arises from our analysis: Use of the unused depth as a reservoir of monomer, using monomer diffusion from the back to the front to replenish that used during exposures, or record from both sides, thus doubling the layer capacity.

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