

# Improved maximum uniformity and capacity of multiple holograms recorded in absorbent photopolymers

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**Abstract:** In order to use photopolymers in the recording of holographic memories, high physical thickness is required. This generates many problems associated with the attenuation of light in the recording due to Beer's law. One of the more significant disadvantages is the fact that there are differences between the physical thickness of the material and the optical thickness of the holograms recorded. The optical thickness characterizes the angular selectivity of the holograms and determines the separation between two consecutive holograms in angular peristrophic multiplexing. In this work we propose a new method to record many holograms multiplexed with similar diffraction efficiency values taking into account the different effective optical thickness of each hologram.

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**OCIS codes:** (090.0090) Holography; (090.2900) Holographic recording materials; (090.7330) Volume holographic gratings

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## 1. Introduction

Recently, great advances have been made in the use of photopolymers as data storage media [1-3]. Photopolymer materials exhibit good optical properties (low losses, low scatter, high refractive index modulations and high density information storage) and are relatively inexpensive. Due to these reasons, some companies have presented the first holographic disks based on photopolymers [4-5]. One of the requirements to obtain competitive holographic memories is that the material must have a thickness of 500  $\mu\text{m}$  or more [6]. This gives rise to differences between the physical thickness of the material and the effective optical thickness of the holograms recorded. The effective optical thickness determines the Bragg selectivity and the separation between two consecutive holograms in some multiplexing techniques [3]. Some of these aspects have been studied in hologram formation in these thick layers [7-8]. On the other hand, schedules have been proposed based on experimental and theoretical studies. For example J.T. Sheridan and co-workers proposed an interesting schedule based on a two dimensional non-local diffusion model to obtain holograms with the same diffraction efficiency using these materials [3]. In these studies the authors assume that the diffraction efficiency only depends on the refractive index modulation of the grating because the thickness of the gratings is considered constant; in other words the physical and effective optical thickness have the same value. This assumption is valid for thin layers, but for thick photopolymer layers with high light absorption we need new methods to study the multiplexing schedule.

In our recent experimental studies [9] we observed some problems when multiplexing many holograms using iterative algorithms [10]. These problems are due to the variation in the effective optical thickness of the gratings recorded during the multiplexing process. In Fig. 1 the experimental effective optical thickness of 60 gratings as a function of the hologram number is shown for peristrophic multiplexing. We fitted the angular response of each grating to obtain the effective optical thickness. The classical algorithm useful for non-overlapped gratings is the one proposed by Kogelnik [11]. The first 10 gratings have very low effective optical thickness and it is difficult to read each grating separately and obtain the hologram thickness. The average hologram diffraction efficiency is 2%; afterwards, the transmission of the layer at the end of the multiplexing process is around 50%. In Fig. 1 we can see two important effects. Firstly we can see the low effective optical thickness of the first 10 holograms stored. Based on this result, more angular separation between these holograms is required and the capacity of the material decreases. The second interesting effect is observed in the analysis of the last 10 holograms. We can see a maximum effective optical thickness of around 470  $\mu\text{m}$ , and then the thickness of the following holograms is lower. This can be

explained if all the monomer is consumed on the illuminated polymer side, and the following gratings are recorded deeper in the material.

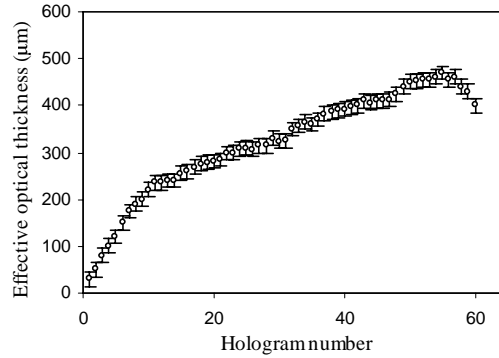


Fig. 1. Experimental effective optical thickness as a function of hologram number to record 60 holograms with diffraction efficiency around 2%.

Therefore we present in this paper a model to optimize the scheduling, taking into account the different optical thickness of the gratings stored in the thick layers. We simulated the exposure time to record more than 200 gratings for different chemical compositions and characteristics of the dried layer. Nevertheless, for our simulations we use values of parameters characteristic of the thick layers based on PVA/acrylamide developed by our research group (such as the absorption, inhibition process, etc.). We studied the differences between the effective optical thicknesses of each grating and the influence of different parameters on the optimal times to obtain gratings with the same diffraction efficiencies.

## 2. Theoretical model

In previous studies we analyzed the importance of making some assumptions in order to present a simplified model which describes the photopolymer behavior. The first important approximation is that we only consider two harmonics in the concentration of monomer and polymer. We have demonstrated in previous papers that in gratings recorded in the thick layers of our material the higher harmonics are more than 20 times lower than the two main harmonics, which occurs because the monomer diffusion is  $10^{-10} \text{ cm}^2 \text{ s}^{-1}$  or higher [12]. Another important assumption is that we are working with the local case, we disregard the non-local polymerization introduced by Sheridan and co-workers in the polymerization process [13]. Then the equation that governs the gratings' formation can be written as:

$$\frac{\partial[M](x, z, t)}{\partial t} = \frac{\partial}{\partial x} D(t) \frac{\partial[M](x, z, t)}{\partial x} - k_R(t) I^\gamma(x, z, t) [M](x, z, t) + \frac{\partial}{\partial z} D(t) \frac{\partial[M](x, z, t)}{\partial z} \quad (1)$$

$$\frac{\partial[P](x, z, t)}{\partial t} = k_R(t) I^\gamma(x, z, t) [M](x, z, t) \quad (2)$$

Where  $[M]$  is the monomer concentration,  $[P]$  is the polymer concentration,  $D$  is the monomer diffusivity and  $\gamma$  indicates the relationship between intensity and polymerization rate ( $k_R$ ). Assuming Beer's law, which explains the attenuation of the light intensity with depth, the incident light inside the material can be written as:

$$I(x, z) = I_0 [1 + \cos(K_g x)] e^{-\alpha(t)z} \quad (3)$$

Where  $\alpha$  is the coefficient of the light attenuation. The initial value of  $\alpha$  [ $\alpha(t=0)=\alpha_0$ ] can be obtained if the transmittance and the physical thickness of the layer before the recording process are known and depends on the dye concentration, the drying process and the recording wavelength.  $I_0$  takes the value of  $5 \text{ mW cm}^{-2}$  in this work.

We assume that the polymerization rate increases very quickly due to the Trommsdorff effect [14] and then decreases because it is limited by the viscosity of the material when the polymer is generated. It is given by:

$$k_R(t) = k_R \exp(-\varphi_k [P]) \quad (4)$$

Where  $\varphi_k$  is the attenuation coefficient of the polymerization (we take  $\varphi_k=5$  in this work). Therefore the monomer diffusion inside the material decreases when the polymer is generated as follows:

$$D(t) = D_0 \exp(-\varphi_D [P]) \quad (5)$$

Where  $\varphi_D$  is the attenuation coefficient of the monomer diffusivity (we take  $\varphi_D=5$  in this work, slow diffusivity attenuation).

One important novelty introduced in the study of thick layers is the cut-off intensity presented by some materials for very low intensities. In PVA/AA materials used in our research laboratory the lower cut-off intensity is around  $50 \text{ } \mu\text{W/cm}^2$  [15]. The importance of the inhibition period for the first hologram, due to the existence of oxygen and impurities inside the layer, should be borne in mind and we took into account this effect for each sub-layer. In our material we measured the energy density necessary to start the grating formation, which is around  $6 \text{ mJ/cm}^2$ .

Another important approximation made in our model is that the monomer diffusion along the Z axis can be disregarded. The validity of this approximation is shown in reference [12] where a 3-dimensional model is solved by the finite differences method. 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,  $d$ , is the sum of the thickness of the different sub-films.

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

where  $G$  is the number of sub-films used.

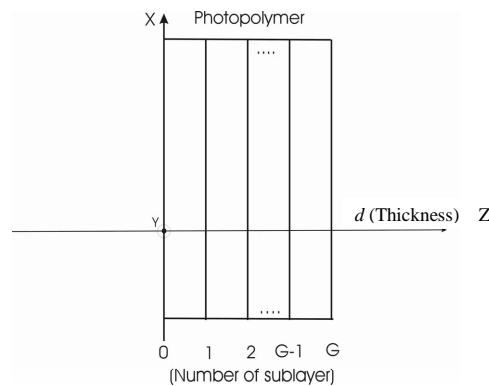


Fig. 2. Holographic grating structure

The exposure intensity in each sub-grating can be calculated using eq. (3) 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 [8].

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

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 the higher harmonics can often be disregarded in photopolymers [12, 16].

In reference [12] the time between two consecutive gratings necessary to obtain a uniform distribution of monomer in the material again is studied. This time is around 10 seconds; in the schedule presented in this work, we assume that the monomer concentration is homogeneous before recording a new grating.

In order to record many holograms with the same diffraction efficiency ( $DE$ ), it is important to analyze the dependence of the diffraction efficiency on the monomer and polymer concentrations. Classically this dependence can be written as follows [11]:

$$n_1 d_{\text{eff}} = \frac{\lambda_0 \cos \theta}{\pi} \sin^{-1} \sqrt{\frac{DE}{ATT}} \quad (6)$$

We assume that the explicit equation of Kogelnik's theory can be used to obtain the relation between the  $DE$  and  $n_1$  (average refractive index modulation in the layer). Nevertheless, there are two important facts to take into account in the parameters  $d_{\text{eff}}$  and  $n_1$ . Firstly, it is important to understand that the thickness used is the effective optical thickness of the gratings ( $d_{\text{eff}}$ ) [7]. Secondly, the refractive index modulation is attenuated in depth, so an average value should be used [17]. On the other hand, the refractive index modulation depends on the distribution of the monomer and polymer, but if at the end of the schedule we consume all the monomer or the residual monomer is homogeneously distributed. We can write:

$$n_1 = \frac{(n_{\text{dark}}^2 + 2)^2}{6n_{\text{dark}}} \left[ \left( \frac{n_p^2 - 1}{n_p^2 + 2} - \frac{n_b^2 - 1}{n_b^2 + 2} \right) [P]_1 \right] \quad (7)$$

where  $n_{\text{dark}}$  is the refractive index of the layer before exposure ( $n_{\text{dark}} = 1.478$ ),  $n_b$  is the binder refractive index ( $n_b = 1.474$ )  $n_p$  is the polymer refractive index and depends on the final length of the polymer chains ( $n_p = 1.512$  when BMA is used as crosslinker), and  $[P]_1$  is the first harmonic of the polymer concentration.

These values were obtained using the Lorentz-Lorenz equation. The calculations are based on refractometer measurements using water solutions.

Therefore if we want to multiplex many holograms with the same diffraction efficiency we need the product  $n_1 d_{\text{eff}}$  to be constant. This condition has been introduced in our model when  $DE$  is fixed. On the other hand, the effective optical thickness  $d_{\text{eff}}$  determines the width of the central lobe in the gratings recorded and it is the parameter that governs the separation needed between two consecutive holograms so that they may be read separately. This was a problem in the first experimental results obtained by us in previous studies [9] where the first holograms have a very small effective thickness and are overlapped. An accurate prediction

of the effective optical thickness and performance over time necessary to obtain equal diffraction efficiencies in each grating are the main goals of this work.

### 3. Results and discussions

The importance of the separation between two consecutive holographic gratings or data pages for them to be reproduced separately is fundamental in order to maximize the capacity of the information stored in photopolymers. In the first approximation of the problem of multiplexing many holograms in the same volume of light absorbent material, it is easy to understand that the effective optical thickness of the holograms will be greater initially and the limit of the effective optical thickness is the physical thickness of the layer. This first part of the work consists of the simulation of this type of materials and discussion of the importance of the bleaching process and absorption coefficient variation. Secondly, we present the analysis of some materials where the effective optical thickness never achieves values close to the physical thickness of the layer. This occurs in materials where the polymerization velocity is faster than the dye consumption rate.

#### 3.1 High velocities of the bleaching process

The initial value of the light attenuation coefficient in depth of each photopolymeric material can be directly determined measuring the initial transmittance of the layer for the recording wavelength. In this section we use the measured valued  $\alpha_0=0.008 \mu\text{m}^{-1}$  for 800  $\mu\text{m}$  thick layers recorded with a wavelength of 514 nm, with yellowish eosin as the dye [18]. The variation in the attenuation of the light inside the material has a critical importance in the evolution of the effective optical thickness of the multiplexed holograms. It may be studied using only one incident beam and fitting the variation in the layer transmittance as a function of the time (for each incident intensity). This evolution has different velocities for different drying process times, drying temperatures, chemical compositions, etc. Therefore, very similar materials can present different behaviors.

The absorption of the layer changes as a function of the time when the dye is consumed; in a first approximation it can be written as:

$$\alpha(t) = \alpha_0 e^{-K_\alpha I_0^\beta t} \quad (8)$$

where  $\beta$  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  $K_\alpha$  defines the rate at which the dye is consumed. The normal evolution of the transmittance of the layer for a power intensity of 5 mW/cm<sup>2</sup> and  $\beta = 1$  is plotted in Fig. 3, for two different values of  $K_\alpha$  0.001 cm<sup>2</sup>mW<sup>-1</sup>s<sup>-1</sup> and 0.0005 cm<sup>2</sup>mW<sup>-1</sup>s<sup>-1</sup>

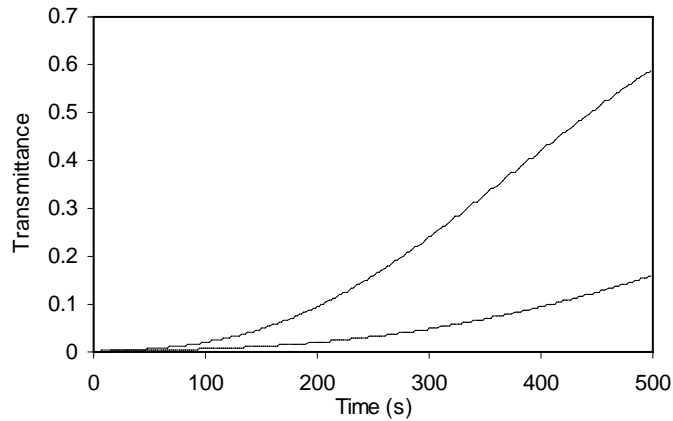


Fig. 3. Transmittance of the layer as a function of the exposure time for  $K_a=0.001\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (continuous line) and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (discontinuous line).

Bleaching velocity simulations of the 250 gratings recorded with a diffraction efficiency of 0.4% for these two cases were carried out. In this simulation, the total sum of the diffraction efficiency at each Bragg's angle is around 100%. This type of photopolymer is similar to the material analyzed in references [19-20], but with lower light absorption. For both light absorption variations we can observe that there is a similar behavior as regards the time required to record the first 100 holograms (Fig. 4). After that, we can see an exponential increase in the time needed to achieve the diffraction efficiency of 0.4%, because the materials are near saturation. This effect can be seen when the photopolymer is near saturation, as can be seen in the experimental papers [9-10]. The common parameters used in the two simulations are  $k_R=0.019 \text{ cm mW}^{-1/2}\text{s}^{-1}$ ,  $D_0=10^{-10}\text{s cm}^{-2}$  and  $\gamma=0.5$ .

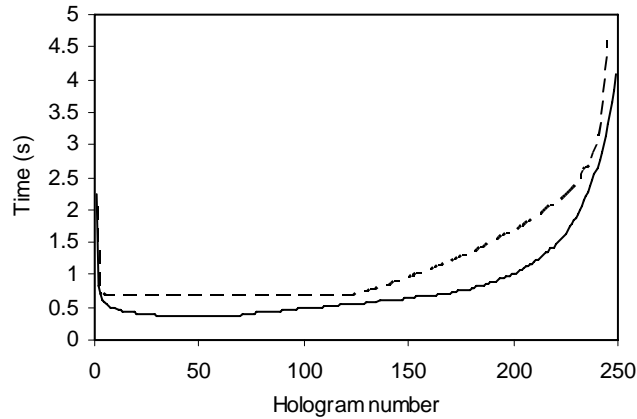


Fig. 4. Time scheme needed to record 250 holographic gratings with 0.4% diffraction efficiency for two different velocities of bleaching process,  $K_a=0.001\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (continuous line) and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (discontinuous line).

The last results are simply the reproduction of the well-known experimental results [9-10]; the important part of this work is the prediction of the effective optical thickness to minimize the separation between the holograms and to maximize the material storage capacity. For the

two materials simulated in this section, the effective optical thickness is the same as the physical thickness of the layer after 67 holograms are recorded in the first case and 124 in the second case (Fig. 5). This effect can be observed because the dye consumption is faster than the hologram formation, and can be demonstrated fitting the angular responses of each hologram.

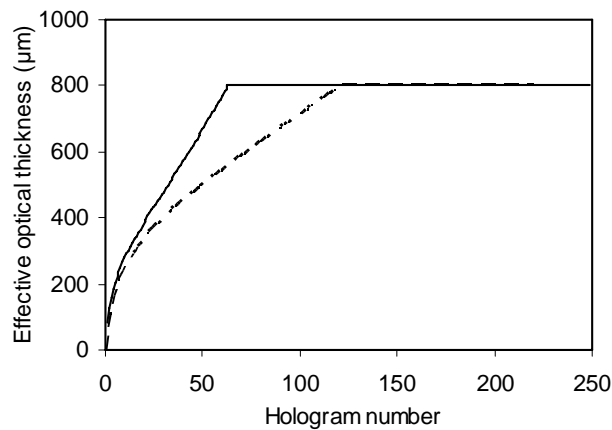


Fig. 5. Effective optical thickness for 250 holographic gratings recorded with 0.4% diffraction efficiency for two different velocities of bleaching process,  $K_a=0.001\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (continuous line) and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  (discontinuous line).

### 3.2 High velocities of grating formation

Let us analyze in this section the importance of the velocity of the grating formation in the time scheme and in the effective optical thickness of the multiplexed gratings. The polymerization rate and the monomer diffusion inside the material depend on many factors, and the same compositions of material can present important differences if the drying conditions of the layer are different. On the other hand, when the photopolymer composition presents an excessive dye concentration or elevated photopolymerization rates (this case occurs when crosslinker monomers are used in the material composition [20]), two interesting effects may be seen. The time required to record holograms with 0.4% diffraction efficiency is lower (see Fig. 6, where  $k_R=0.026\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ ,  $D_0=3\times 10^{-10}\text{scm}^{-2}$  and  $\gamma=1$ ) as could be expected. Nevertheless, the most interesting effect can be seen in Fig. 7, where the effective optical thickness of the gratings recorded never achieves the value of the physical thickness of the grating. This effect can be seen in previous experimental papers [9] where the maximum effective optical thickness achieved after the recording of 60 gratings with an average diffraction efficiency of 2.4 % was 550  $\mu\text{m}$ .



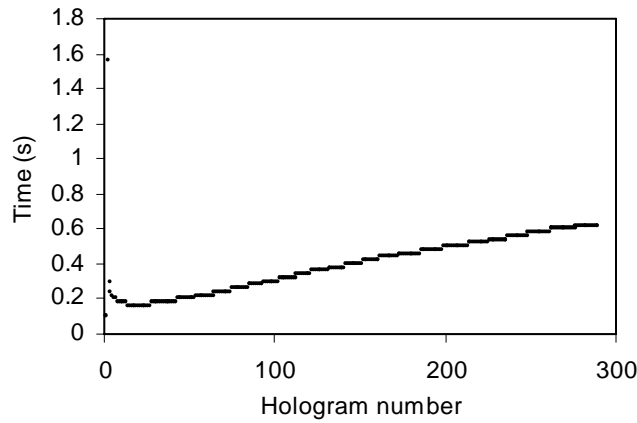


Fig. 6. Time scheme needed to record 300 gratings with 0.4% diffraction efficiency for high values of polymerization rate:  $k_R=0.026\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ .

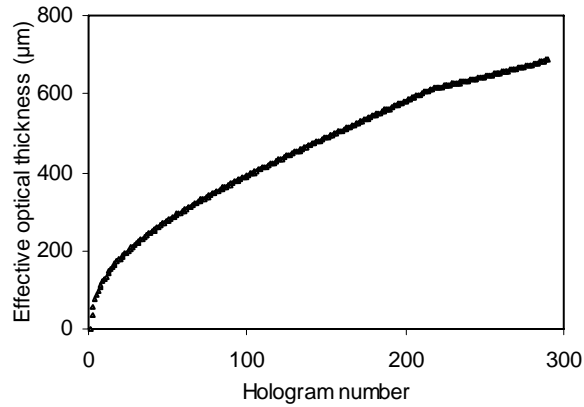


Fig. 7. Effective optical thickness for 300 gratings recorded with 0.4% diffraction efficiency for high values of polymerization rate:  $k_R=0.026\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$  and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ .

It is possible to make a deeper analysis of the last effect observed. In order to better appreciate the limit of the effective optical thickness, we simulated 600 grating with 0.4% diffraction efficiency (Fig. 8). But now the value of the initial light absorption considered is higher  $\alpha_0=0.012\text{ }\mu\text{m}^{-1}$ . This value can be obtained when a diode-pumped frequency-doubled Nd:YAG [9] laser at 532 nm is employed to store the multiplexed gratings in one layer as analyzed in Fig. 1. As we can see in Fig. 8, at the end of the multiplexing process the layer is near saturation and the time needed to multiplex one grating with 0.4% diffraction efficiency grows exponentially.

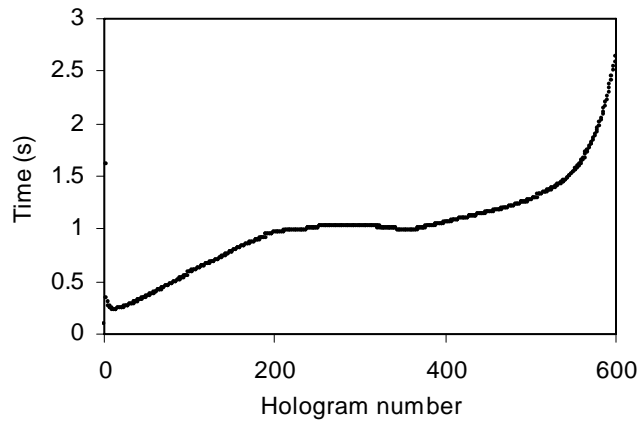


Fig. 8. Time scheme needed to record 600 holographic gratings with 0.4% of diffraction efficiency for high values of polymerization rate:  $k_R=0.026\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ ,  $\alpha_0=0.012\text{ }\mu\text{m}^{-1}$  and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$

An important fact can be seen in Fig. 9, the evolution of effective optical thickness as a function of the multiplexed holograms presents a maximum around 600  $\mu\text{m}$  after 350 holograms are recorded and then the effective optical thickness decays. This effect has been observed before in PVAAA based polymer and is due to the consumption of 90% of the monomer in the first 190  $\mu\text{m}$  of the layer.

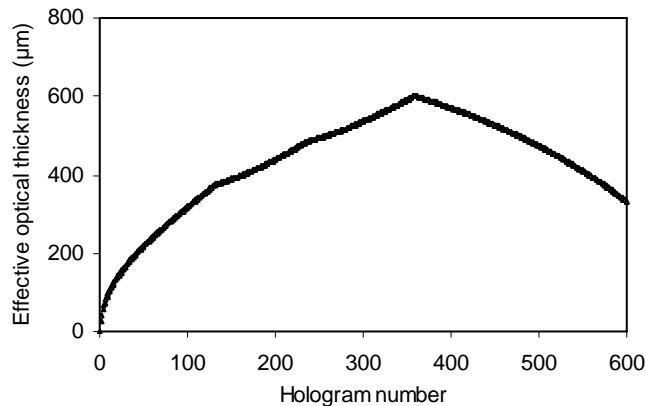


Fig. 9. Effective optical thickness for 600 holographic gratings recorded with 0.4% diffraction efficiency for high values of polymerization rate:  $k_R=0.026\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ ,  $\alpha_0=0.012\text{ }\mu\text{m}^{-1}$  and  $K_a=0.0005\text{cm}^2\text{mW}^{-1}\text{s}^{-1}$ .

In order to better understand Fig. 9, let us analyze the monomer distribution simulation after 300 holograms and 600 holograms are recorded. In Fig. 10 we can see the gradient in depth of the monomer and diffusion concentration. After 300 holograms all the monomer in the first 160  $\mu\text{m}$  has been consumed. At this moment, the effective optical thickness of the layers recorded is around 540  $\mu\text{m}$ . Lastly, it is important to note that after 600 gratings are recorded only in the last 400  $\mu\text{m}$  of the layer is there some free monomer.

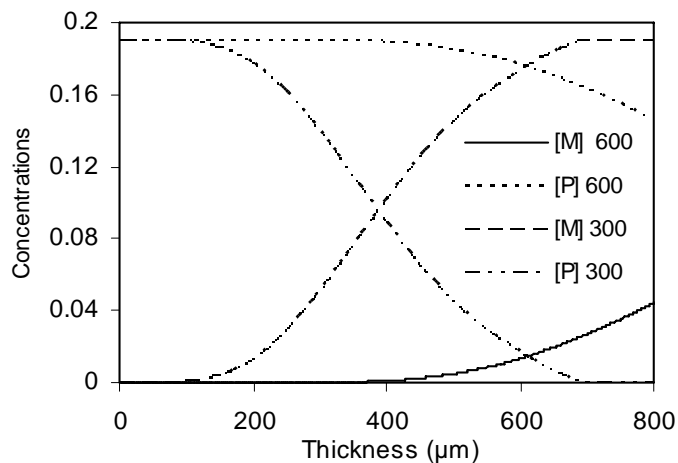


Fig. 10. Average monomer and polymer concentrations as a function of the depth after 300 and 600 holographic gratings are recorded with 0.4% diffraction efficiency.

Finally, let us explain better the effect analyzed and predicted in this work using the diagram presented in Fig. 11. Initially, for a small hologram number, the hologram's effective optical thickness (the length of the zone where the hologram exists) is very low due to the short penetration depth of the light inside the material and only the monomer that is near the incident recording wave is consumed. When the hologram number increases, the monomer in this zone has been consumed and the following holograms are recorded deeper in the layer but the hologram's effective optical thickness decreases.

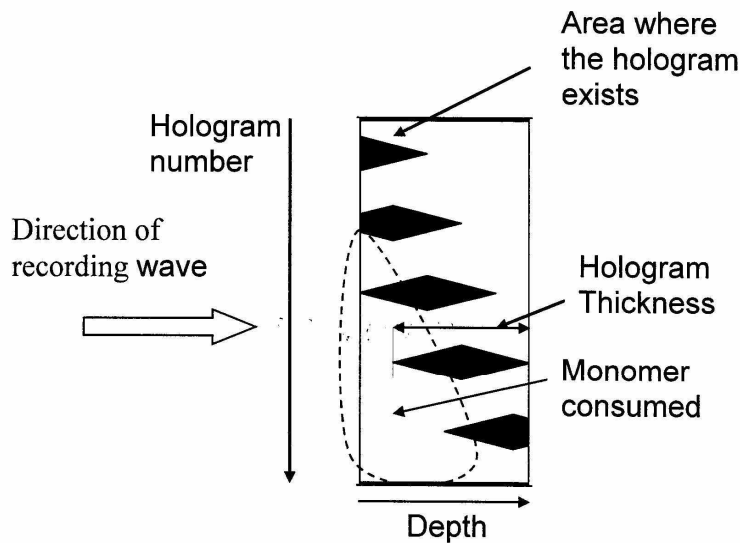


Fig. 11. Diagram of the hologram formation as a function of hologram number.

#### **4. Conclusion**

The work has a great importance for the design of the multiplexing scheme in holographic memories based on photopolymers. The method presented can predict the times required to obtain many holograms multiplexed with equal diffraction efficiency and maximum storage capacity. Different types of materials have been studied in order to analyze the different dependence of the effective optical thickness on the number of holograms multiplexed. The analysis of the evolution of the effective optical thickness as a function of the exposure time can be used to characterize the hologram formation and to calculate some parameters such as the polymerization rate of the hologram.

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