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PVA/acrylamide photopolymers as holographic recording material at short wavelengths

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ABSTRACT

Photopolymers can be considered holographic recording materials because of their many attractive features. Among these, we could mention their ability to self-develop, the fact dry processing can be used with them, their good stability and thick emulsion layers, and their high sensitivity, diffraction efficiency and resolution. Photopolymers have been developed for conventional imaging systems, real time interferometry and holographic optical elements. These materials have applications in other optical devices such as high-density storage and optical data processing. This paper is a review of the study of photopolymers employed as holographic recording materials used in applications such as holographic memories and the obtaining of holographic optical elements.

1. INTRODUCTION

It is well known that holography enables an image to be reconstructed in three dimensions, as opposed to photography which reproduces an image in only two dimensions. However, this characteristic is not the most technically useful in other fields of science and technology. Thus, for example, the characteristic on which the development of basic elements in sectors such as communications and computers is

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based is the ability to divert in free space, at any desired angle, a beam of incident light onto a point of the plate on which the hologram is recorded. Such holograms are known as holographic optical elements [1]. Another characteristic of holography that enables the development of areas such as information storage [2] is the capacity to store many object waves in a single hologram when volume holographic elements are used.

In the manufacture of holographic optical elements as well as in the storage of information, the key factor is the utilization of an appropriate recording material. There is a great demand for holographic recording materials capable of giving impetus to each of the various applications, such as optical memories [3-5], optical processing [6] or holographic interferometry [7]. It is therefore necessary to make further advances in the development of holographic recording materials since the success of modern optical devices depends on the behavior and versatility of the optical material employed.

The demand for better recording materials requires that they have characteristics such as high resolution, high energetic sensitivity, great spectral sensitivity, simple processing and erasion of information.

This present study is centered on an analysis of the behavior of phototopolymers employed as holographic recording material to be used in the two applications mentioned above. In the first place, all aspects related to obtaining and depositing the photopolymeric material, preparation of the final solution, depositing, uniformity of the dry film and its characteristics such as thickness and transmittance are considered. In the second place, the behavior of the photopolymer must be analyzed from the holographic point of view, considering the diffraction efficiency achieved, sensitivity and signal to noise ratio or scattering of the stored holograms.

2. MATERIAL AND METHODS

The interest in photopolymers as holographic recording materials arose more than three decades ago and has increased enormously in recent years. Photopolymeric materials have holographic properties such as high reliability, great and controllable film thickness, dry processing, reasonably long shelf life, and, very importantly in the case of large consumer production, a low price. Because of these properties, holographic photopolymers are now regarded as key materials for viable implementations of holographic-based storage systems where a high optical storage capacity, fast access time, high data transfer rate storage and intense storage are needed.

Since the late 1960s, a variety of photopolymer materials have been used to produce holograms [8]. The earliest reported photopolymer materials for holographic recording were liquid formulations of acrylamide materials. Close *et al.* in 1969 [9] were the first to record volume holograms in a photopolymer using acrylamide and metal acrylates as monomers and a ruby laser tuned at 694 nm. Jenney [10] also studied this photopolymer and observed a modulation of both refractive index and the surface.

In the early 1970s a photopolymer composition in dry-film form was reported that was a forerunner to the material now supplied by Du Pont. This material was studied by Colburn *et al.* [11] and Booth [12].

Continued research led to the introduction in the 1980s of new improved photopolymer compositions analyzed in the work of Ingwall *et al.* [13], and Smothers *et al.* [14].

Finally in the last decade, a great variety of photopolymers were used as can see in the work of Sutherland *et al.* [15] who demonstrated that by using liquid crystals in an acrylate the diffraction efficiency reaches 100% for holograms registered at 488 and 514 nm. Shannon *et al.* [16] showed that the orientation of the molecules in liquid crystals can be controlled by polymerization, which allows the orientation to be used in information storage. Zhang *et al.* [17] obtained thin gratings in liquid crystals by polymerization and surpassed the theoretical

value of 34% for the diffraction efficiency. Piazzolla *et al.* [18] proposed a theoretical model on holographic gratings formed in commercial photopolymers.

If we make a review of all the photopolymers that appear in the literature [19, 20] we can see that, depending on the needs or requirements of the different applications, the principal characteristics of a recording material to be taken into account are response in real time, processing of the material, energetic and spectral sensitivity, diffraction efficiency, resolution, linearity, storage capacity, rapid access, reversibility, etc. The degree of importance or priority of these characteristics is determined by the application for which the recording material is to be used.

On the other hand the way in which a photopolymer behaves when irradiated with an appropriate light source gives rise to different reactions, such as photopolymerization, photocrosslinkage, crystals formed as liquid polymers, photorefraction under electrooptical effects, etc. The monomers and polymers on which most research has been done are acrylates, acrylamides, polymethylmethacrylates, polyvinylalcohol, polyacrylic acid, polyvinylcarbazole, polystyrene, and polyesters. There is also a great variety of photosensitive substrates, photosensitive photochromic materials, and metal ion doped polymers. Furthermore, different commercial formulations exist such as those of DuPont and Polaroid.

If we consider recording materials from a holographic perspective, two large groups can be differentiated according to the type of hologram generated. If we produce an amplitude hologram, a variation in the absorption coefficient takes place so a low diffraction efficiency is reached. If on the contrary we produce a phase hologram, a variation in the phase take place, either by variation of the refractive index, by variation of the thickness or both, so a much higher diffraction efficiency is reached.

According to this holographic point of view, we can classify photopolymers in two large groups. On the one hand, those that enable a high diffraction efficiency to be reached or, in other words, generate phase holograms, such as photopolymerizable systems, acrylamides, acrylates, methylmethacrylates (MMA) or those which are photocrosslinkable systems, polyvinylcarbazole (PVK), dichromated polyvinylalcohol (DCPVA), polyacrylic acid (PAA), and on the other hand, photopolymers that reach low diffraction efficiency, in other words generate amplitude holograms, such as doped polymers (photochromic compounds, bacteriorhodopsin). Within the first group we shall focus on photopolymerizable systems.

Photopolymerizable systems are used to generate phase holograms with a high diffraction efficiency produced by a change in the refractive index as already mentioned. Its composition consists, basically, of a polymeric film that acts as a binder, a photoinitiator system made up of a sensitizer or dye and a radical generator, and one or more monomers.

Photopolymerizable systems can be divided into two large groups: liquid compositions and dry films. The latter can in turn be subdivided into acrylates, methylmethacrylate and acrylamides depending on the type of monomer used. In our study we used photopolymerizable systems deposited on dry films and based on acrylamides. Table 1 shows the characteristics of photopolymerizable systems deposited on dry films similar to those obtained in our laboratory. In the photopolymerizable systems usually used as holographic recording material in the form of a dry film, the photosensitizer and the radical generator form the photoinitiator. These two functions may be accomplished by the same element or by different elements. The absorption of light by the sensitizer in the irradiated zones of this film gives rise to the formation of reactive species, generally free radicals, that begin a

Table 1. Photopolymerizable recording materials: Holographic properties

MATERIAL	Thickness (μm)	Wavelength (nm)	Sensitivity (mJ/cm²)	Resolution (lines/mm)	Diffraction efficiency (%)
DuPont HRF-700 X (acrylates)	10-200	476-532	10-40	3000-5000	99
DuPont OMNIDEX (acrylic monomers)	40	476-532	50-100	3000-5000	99
Polaroid DMP-128 (vinylic monomers)	1-30	442-647	5-30	5000	80-95
Martin *et al.* [21] (acrylamide-bisacrylamide)	150	514	100-300	2000-5000	90
Weiss *et al.* [22] (acrylamide-bisacrylamide)	5-100	514	12-35	1000	90

chain reaction of the monomer or monomers forming the corresponding polymer, whose refractive index is different from that of the original monomer which remains undisturbed in the zone not exposed to radiation.

Figure 1 shows the different steps in a normal photopolymerization process, where I is the photoinitiator, M is the monomer, $R^{\bullet}$ is the free radical, $Pi^{\bullet}$ is the polymer radical, k_i, k_p, k_{tp} are the constants of the initiation, polymerization and termination reaction respectively, and hv is the energy of the incident photon.

$$I \xrightarrow{h\nu} 2R^{\bullet}$$

$$R^{\bullet} + M \xrightarrow{k_i} P_1^{\bullet}$$

$$P_n^{\bullet} + M \xrightarrow{k_p} P_{n+1}^{\bullet}$$

$$P_n^{\bullet} + R^{\bullet} \xrightarrow{k_{tp}} P_nR$$

Figure 1. Photopolymerization process.

3. EXPERIMENTAL SETUP

The recording material used is composed of a matrix of polyvinylalcohol (PVA), acrylamide (AA) as monomer, triethanolamine (TEA) as radical generator and yellowish eosin (EA) as sensitizer. The photopolymerizable solution was prepared by adding 1.5 ml of 8 g/l yellowish eosin together with 8 ml of acrylamide and triethanolamide to 50 ml of PVA. The resulting solution was deposited on a 20 x 40 cm^2 glass plate using an automatic depositor and adjusting the thickness of the film. The plate was dried for a period of 24 h in the dark and under normal laboratory conditions (T = 21-23°C, RH = 40-60%). Once dried it was cut into plates measuring 6.5 x 6.5 cm^2 to be used in our experimental setup. The concentration of each of the components can be seen in Table 2.

Table 2

Acrylamide (AA)	4.4×10^{-1} M
Triethanolamine (TEA)	2.0×10^{-1} M
Yellowish eosin (EA)	2.5×10^{-1} M
Polyvinylalcohol (PVA)	10%
Initial thickness	500 μm
Acrylamide (AA)	4.4×10^{-1} M
Triethanolamine (TEA)	2.0×10^{-1} M
Yellowish eosin (EA)	2.5×10^{-1} M
Polyvinylalcohol (PVA)	15%
Initial thickness	1000 μm

The composition of the photosensitive solution used is the same except for the concentration of polyvinylalcohol (PVA), which varies depending on the initial thickness, as shown in Table 1. One of the problems arising is the uniformity of thickness of the plate. Once the film is deposited, it is necessary to control the thickness of the film obtained, since this parameter is a fundamental factor affecting the diffraction efficiency of the material. According to Kogelnik's theory [23], the thickness determines the maximum diffraction efficiency achieved with a recording material, and the value of the product of index modulation in the maximum intensity zone and thickness $[n_1(t)d]$determines the evolution of the diffraction efficiency. The variation in optical thickness (Δnd) of this photopolymerizable material with the same chemical composition produces changes in the shape of the diffraction efficiency versus time curve and in the position of maximum diffraction efficiency. In addition, the thickness influences the sensitivity of the material, which increases as the thickness is increased. For this reason the thickness of the plate was measured with an ultrasound meter (Universal Defelsko Mod 100-B2). The thickness was also determined measuring the transmittance from the material with a spectrophotometer (Hitachi U-2000) at the working wavelength of 514 nm. Making use of Lambert-Beer's law [24]:

$$T = 10^{-\varepsilon d[Dye]}$$

the thickness d is related to the transmittance T. Experimentally it is observed that with greater thicknesses the transmittance is less, while low thicknesses give the highest values of transmittance, for a constant concentration of the components of the photopolymer film. Therefore, a correlation exists between the transmittance obtained and the measured thickness.

To study the behavior of the photopolymer as a holographic recording material we obtained diffraction gratings using a holographic setup in order to determine the diffraction efficiency of the material, sensitivity and scattering.

The experimental device used to store diffraction gratings is shown in Figure 2. The holographic gratings were created with an Argon laser at a wavelength of 514 nm. The laser beam was split into two secondary beams, referred to as the object beam and the reference beam, with an intensity ratio of 1:1. The diameters of these beams were increased to 1 cm with an expander while spatial filtering was ensured. The object and reference beams were recombined at the sample at an angle of 16.8° to the normal with an appropriate set of mirrors, and the spatial frequency obtained was 1125 lines/mm. The diffracted intensity was monitored in real time with a He-Ne laser positioned at Bragg's angle tuned to 633 nm where the material does not absorb, with an incident intensity of 8 mW/cm^2. The diffracted intensities were detected with two detectors connected to a computer and measured every 10 seconds. The sensitivity of the setup was ± 0.05° which assured we were working in Bragg's angle. It should be borne in mind that the angle sensitivity curve $\Delta\theta$ = 1°, therefore positioning of the plate must be done very accurately.

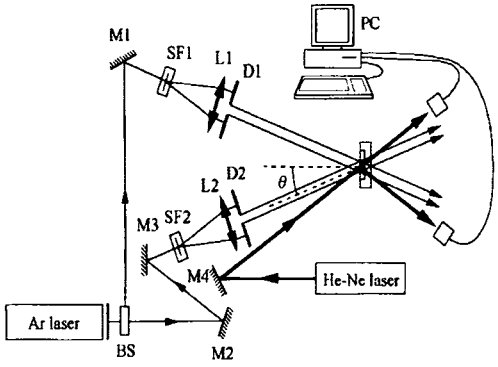

Figure 2. Experimental Setup: BS, beam-splitter, M_i, mirror, SF_i, spatial filter, L_i, lens, D_i, diaphragm, PC, personal computer.

4. RESULTS AND DISCUSSION

To analyze the behavior of the photopolymer employed as the holographic recording material we represented diffraction and transmission efficiency versus exposure in order to establish the influence of the thickness and the intensity.

In Figure 3 we can analyze the behavior of the photopolymer for two different thicknesses, observing the diffraction efficiency reached. When photopolymer films of 70 ± 5 μm are used, a diffraction efficiency of 75% and energetic sensitivity of 300 mJ/cm^2 are obtained. The working intensity is 10 mW/cm^2.

The energetic sensitivity is defined as the minimal energy necessary to reach maximum diffraction efficiency. When the thickness of the film is greater (110 ± 5 μm), it is possible to achieve a diffraction efficiency of about 80% and greater energetic sensitivities of around 200 mJ/cm^2. When the material is illuminated with a wavelength that corresponds to the absorption band of yellowish eosin, a photoreduction reaction takes place producing radicals that initiate the polymerization reaction. One of the parameters that influences the polymerization reaction is the incident intensity, since depending on this the whitening process of the dye is more or less rapid [25]. In order to study the influence of the intensity on the behaviour of the photopolymer, different intensities (I = 1, 10, 15 mW/cm^2) were used with a film thickness of 70 ± 5 μm.

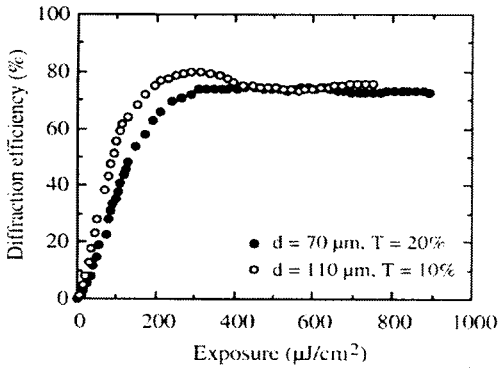

Figure 3. Diffraction efficiency versus exposure for different thicknesses.

Figure 4 shows diffraction efficiency as compared to exposure for intensities of 1, 10 and 15 mW/cm^2. In this case the energetic sensitivities are different and increase as the intensity is reduced. For the three values of intensity, the maximum diffraction efficiency reached is 70%. The slope of each of these curves is related to the polymerization rate, which increases as the slope of the curve increases.

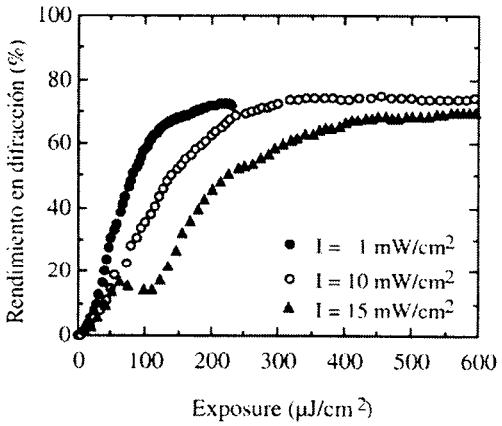

Figure 4. Diffraction efficiency versus exposure for intensities of 1, 10, 15 mW/cm^2.

As we can see this recording material behaves in a way that does not comply with the law of reciprocity, as in the case of other holographic recording material such as dichromated gelatin [26].

Holographic behavior for high thicknesses

Holographic memories are one of the alternatives to current information storage systems. When holographic memories are used, it is necessary to store many holograms in a given volume. Because of this it is important to know how a photopolymer behaves when working with high thicknesses,

and what characteristics it must have for its application in holographic memories [27].

To obtain photopolymer films of great thickness, a solution made up of 2.5 ml of a solution of 8 g/l of yellowish eosin and 15.8 ml of a solution of acrylamide (whose concentration varies) and triethanolamine is used. Finally, 99.2 ml of 15% PVA is added to this solution. The resulting solution is deposited with an automatic depositor giving an initial thickness of 1000 μm. Once the plates are dried (36 hours in normal conditions: T = 22 ± 2°C and a relative humidity of the 50 ± 10%) holographic diffraction gratings are stored in them using the experimental setup and working with an incident intensity of 10 mW/cm^2.

The principal problem arising when working with great thicknesses is the appearance of noise gratings produced by an increase in the thickness and concentration of PVA.

The study is carried out using two different thicknesses, 110 and 150 μm, varying in both cases the concentration of acrylamide from 0.296 M to 0.667 M so that the holographic behavior of the material is determined in terms of the diffraction efficiency achieved, sensitivity and scattering.

Figure 5 shows diffraction efficiency, transmission efficiency and the sum of both versus exposure for a thickness of 110 μm. Figure 6 shows diffraction efficiency, transmission efficiency and the sum of both versus exposure for a thickness of 150 μm. If we compare the curves corresponding to the same thickness (d = 110 μm) for different concentrations (Figure 5), it can be seen that the maximum diffraction efficiency reached is 80% for all the concentrations. However, the sensitivity is different for each concentration and increases as the concentration of acrylamide is increased, except in the case of a concentration of 0.593 M (Figure 5d) when a decrease in sensitivity is produced. It can be said that the form of the curve varies with concentration. For the first two concentrations (Figure 5a and b) the curve reaches a maximum and remains constant, for greater concentrations (Figure 5c) once the maximum diffraction efficiency is reached the curve drops and if we continue to increase the concentration (Figure 5d), the curve presents two peaks.

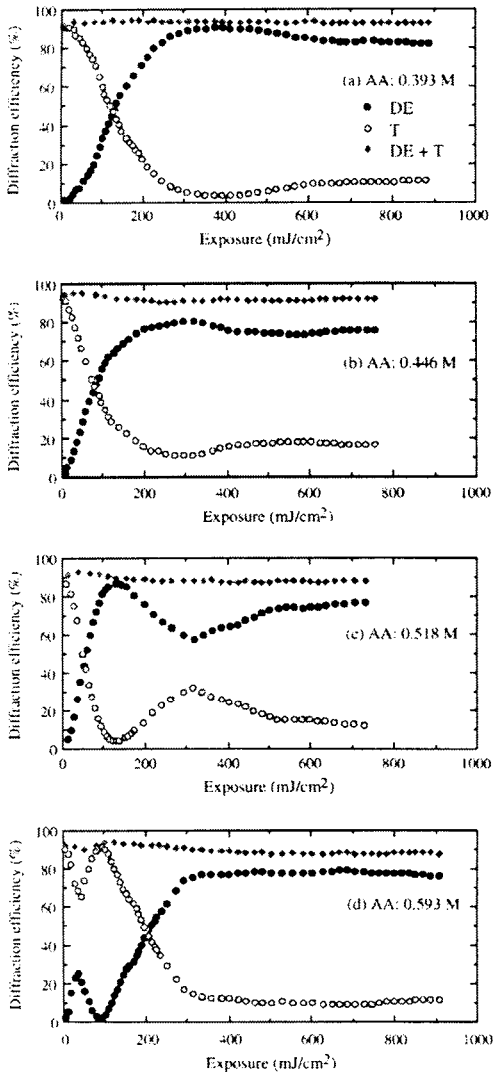

Figure 5. Diffraction efficiency (DE) and transmittance (T) versus exposure for a photopolymer film of thickness 110 μm and different concentrations of acrylamide (AA).

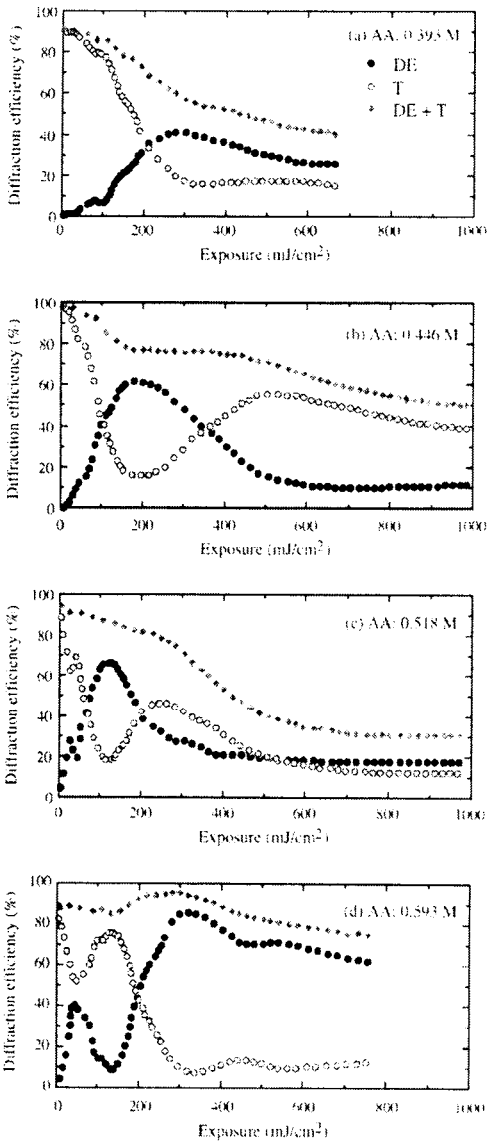

Figure 6. Diffraction efficiency (DE) and transmittance (T) versus exposure for a photopolymer film of thickness 150 μm and different concentrations of acrylamide (AA).

For a thickness of 150 μm (Figure 6), it can be seen that when we increase the concentration the maximum diffraction efficiency increases from 40% to 80%. As in the case of a thickness of 110 μm, the sensitivity increased with monomer concentration. The behavior of the curves for all concentrations is the same, once the curve reaches the maximum, it drops. As occurred with a thickness of 110 μm, when the concentration of acrylamide is 0.593 M (Figure 6d) the curve presents two peaks.

A possible explanation for these two peaks of the curve is the existence of two different termination mechanisms [28]. As we have already mentioned, polymerization is initiated by the absorption of light by the dye which generates radicals that react with the monomer and initiate the polymerization reaction. There are two possible mechanisms by which the reaction is terminated: the reaction comes to an end because two polymer molecules join together, this is called bimolecular termination or because the radicals unite with polymer molecules resulting in so-called monomolecular termination (see Figure 7, where I is the photoinitiator, M is the monomer, $R^{\bullet}$ is the free radical, $Pi^{\bullet}$ is the polymer radical, k_i, k_p, k_t, k_{tp} are the constants of the initiation, i, polymerization, p, and termination, t and tp, reactions respectively, and hν is the energy of the incident photon.

$$I \xrightarrow{h\nu} 2R^{\bullet}$$

$$R^{\bullet} + M \xrightarrow{k_i} P_1^{\bullet}$$

$$P_n^{\bullet} + M \xrightarrow{k_p} P_{n+1}^{\bullet}$$

$$P_n^{\bullet} + P_m^{\bullet} \xrightarrow{k_t} P_{n+m}(P_n + P_m)$$

$$P_n^{\bullet} + R^{\bullet} \xrightarrow{k_{tp}} P_nR$$

Figure 7. Photopolymerization mono – bimolecular process.

When the monomer concentration increases both types of terminations may take

place. For large concentrations of monomer, the rate of polymerization increases and small polymer molecules are formed. These molecules join together giving rise to bimolecular termination. As a result of polymer formation, the viscosity of the medium increases and the mobility of the monomer molecules is reduced and so the rate of polymerization decreases. When the monomer molecules reach the zones occupied by the radicals, polymerization is initiated again and is terminated by polymer molecules uniting with radicals. In Figures 5d and 6d the slope of the curve corresponding to the diffraction efficiency indicates the rate at which this process takes place. The slope corresponding to the first maximum indicates the rate of polymerization during the first part of the reaction in which bimolecular termination takes place. This type of termination reduces the mobility of the monomers and so the curve drops until the monomers reach the radicals once more and polymerization is reinitiated and then monomolecular termination takes place.

The bimolecular mechanism can be identified by the intensity used. It is possible to eliminate the first maximum from these curves when the concentration of acrylamide is high by varying the incident intensity. As can be observed in Figure 8 when the intensity used is lower, the first maximum disappears. If the intensity is increased polymerization takes place quickly, that is to say the polymerization rate increases, therefore it is possible for the two types of termination to take place simultaneously. When the intensity is low the polymerization is slower and only one type of termination takes place, with one of the two peaks of the diffraction efficiency curve being eliminated. If the intensity is sufficiently low, a period of inhibition may appear at the bottom of the curve.

Finally it is possible to analyze the influence of the acrylamide concentration on the scattering for different thicknesses. A measure of scattering at a given point is given by the difference between the sum of the diffracted intensity and transmitted intensity at the given point and the initial intensity. Figure 9 shows scattering as a function of acrylamide

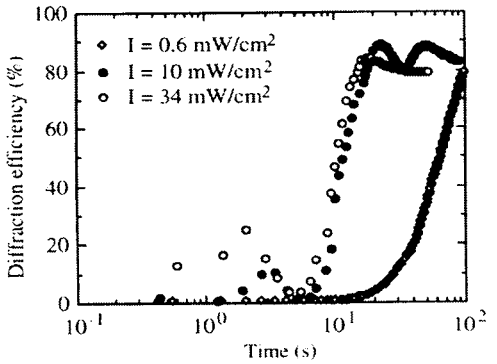

Figure 8. Diffraction efficiency versus exposure time for different intensities and high concentrations of acrylamide.

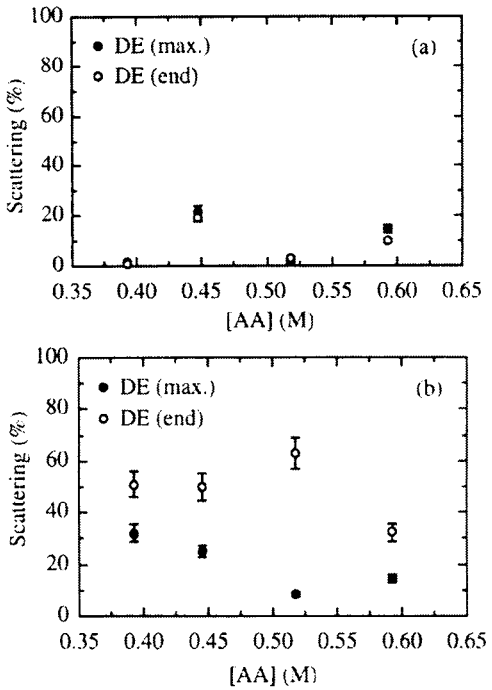

Figure 9. Scattering versus acrylamide concentration [AA] for two different thicknesses: (a) 110 μm and (b) 150 μm.

concentration measured at the points corresponding to the maximum and final diffraction efficiency for the two thicknesses considered, 110 and 150 μm.

For a thickness of 110 μm we can see that the concentration of acrylamide does not affect the scattering and that in all cases the value of this parameter is the same for maximum and final diffraction efficiency, its value being between 2 and 20%.

For a thickness of 150 μm however, scattering is seen to depend on the concentration of acrylamide, so that scattering is reduced at the maximum diffraction efficiency points for high concentrations of acrylamide (0.593 M gives rise to 15% scattering). However, at the final diffraction efficiency points, greater scattering is observed than at the maximum diffraction efficiency points, with a value of 60% being reached for a concentration of 0.518 M, and for the highest concentration of acrylamide studied, scattering is reduced to 30%.

We can conclude that it is possible to reduce scattering for high diffraction efficiencies using high concentrations of acrylamide and great thicknesses. This results in the material being considered suitable for use in holographic memories due to the form of molecular growth during polymerization and to the type of chemical termination reaction.

Obtaining holographic optical elements

One of the holographic applications most studied is that of obtaining holographic optical elements (HOEs) as was mentioned in the introduction. In this study we obtained low and high frequency HOEs in a photopolymer employing a copy method [29].

The copy method consists in copying a previously generated master by placing this master in direct contact with the photopolymer, with the master and the photosensitive layer of the copy placed in contact. A collimated beam from the source incident on the master and the copy is used to expose the photopolymer. When we use partially coherent light, this type of light enables us to work with more economical sources and devices, and provides stable conditions that are not as strict as those used in conventional holographic devices. Additionally, the low coherence of the source gives a better signal to noise ratio.

In the copy of low frequency HOEs a mercury lamp was used, that is to say a source of partially coherent light, without interferential filter, in order to take advantage of all the emission lines of the lamp to which our photopolymer is sensitive. The master was designed by digital techniques and the design saved in a PostScript file, which was printed by a Linotronic 630 printer on photographic film. The fidelity of the implementation of the digital design depends on the resolution in the positioning of the printed spot of the printer and on the diameter of the spot printed by the graphic. The combination of these two parameters limits the maximum spatial frequency that can be implemented on the master to 30 lines/mm. In this study low frequency symmetrical gratings were copied between 4 and 32 lines/mm and the diffraction efficiency reached was around 27% (Figure 10), with a sensitivity of 85 mJ/cm^2.

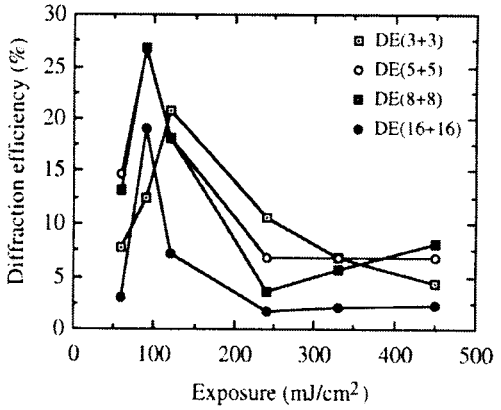

Figure 10. Diffraction efficiency versus exposure for low frequency gratings.

In the copy of high frequency HOEs, an Argon laser tuned at 514 nm was used. The master was obtained in silver halide sensitized gelatin (SHSG) with a wavelength of 633 nm of a 15 mW He-Ne laser. The SHSG is based on the combination of the whitening corresponding to the photographic emulsions and the processing of dichromated gelatin. In this way, the final product is a high-performance material with high diffraction efficiency, low noise level, stability to light and similar transparency to dichromated gelatin, but with the energetic and spectral sensitivity of a photographic emulsion. The master had a frequency of 1000 lines/mm.

On the other hand, if we want to assure maximum modulation in the copy process, it is important for the ratio of the intensity of the object beam to that of the reference beam, into which the original beam is split, to be approximately 1:1; in other words, the diffraction and transmission efficiency is of the same order, therefore we chose a master with a diffraction and transmission efficiency of 50% at a wavelength of 514 nm. As the original to be copied had a high frequency, it was necessary to locate it in Bragg's angle.

Copies of gratings of 1000 lines/mm were obtained with a beam ratio of 1:1, and diffraction efficiencies of 40% (Figure 11) were reached with an energetic sensitivity of 300mJ/cm^2.

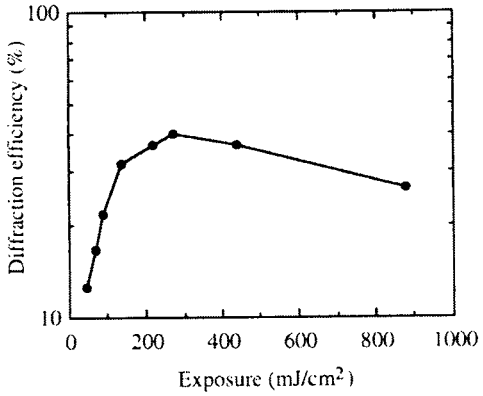

Figure 11. Diffraction efficiency versus exposure for high frequency gratings.

Storage of diffuse objects

Another application of photopolymers considered is the storage of diffuse objects.

Diffuse objects were stored, analyzing the influence of the beam ratio and intensity on the optical quality of the transmission hologram images reconstructed in real time. The signal to noise ratio and the diffraction efficiency were used as a measure of the optical quality. The experimental setup used can be seen in Figure 12.

Both recording and reconstruction of the hologram was done with a 5 W Argon laser tuned at 514 nm. The diffuse object used was a test target USAF in front of which was placed a diffuser. Only the zone corresponding to 1.6 lines/mm was stored. The virtual image situated in the object plane was focalized by the L_2 and captured by a CCD camera. The holographic images were captured in real time with a frequency between 5 and 10 s depending on the incident intensity.

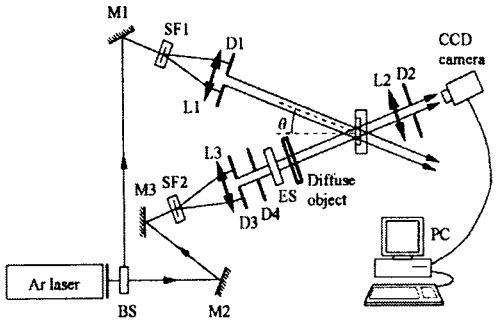

Figure 12. Experimental setup for the storage of diffuse objects: BS, beam splitter, MI, mirror, SFI, spatial filter, LI, lens, DI, diaphragm, ES, electronic shutter, PC, personal computer.

To evaluate the optical quality of the images captured by the CCD, we used the

signal to noise ratio (SNR) and the diffraction efficiency defined by the following expressions:

$$SNR = \frac{I_{max}}{I_{min}}$$

where I_{max} and I_{min} are given by:

$$I_{max} = \bar{I}_1 \quad \text{with} \quad I_1 = \sum_{j=1}^{128}\left(\sum_{p=1}^{5}\sum_{i=N_{2p}}^{N_{2p+1}} I_{ij}\right)$$

and

$$I_{min} = \bar{I}_2 \quad \text{with} \quad I_2 = \sum_{j=1}^{128}\left(\sum_{p=0}^{5}\sum_{i=N_{2p+1}}^{N_{2p+2}} I_{ij}\right)$$

so that:

$$\sum_{p=0}^{5}(N_{2p+1} + N_{2p+2}) = 128$$

where I_{ij} is the intensity of the pixel in the position ij and N_i marks the beginning and the end of each fringe.

Since the signal to noise ratio of the object image depends on both the total incident intensity and the beam ratio, all signal to noise measurements were normalized with respect to the signal to noise ratio of the object.

The diffraction efficiency, DE, is calculated as:

$$DE(\%) = \frac{I_d}{I_i} \times 100$$

where:

$$I_d = \sum_{i=1}^{128}\sum_{j=1}^{128} I_{ij}$$

$$I_i = K I_0 = \sum_{i=1}^{128}\sum_{j=1}^{128} I_{0_{ij}}$$

where I_d is the diffracted intensity captured by the CCD, I_0 the intensity of the object beam, I_i the incident intensity and K the beam ratio.

Figure 13 shows the normalized signal to noise ratio SNR/SNR_0 as a function of time (Figure 13a) and diffraction efficiency (Figure 13b). As can be seen the behavior is different for low (0.6 and 1.2 mW/cm^2) and high (2.4 and 4.8 mW/cm^2) incident intensity [30]. For low intensities the signal to noise ratio reaches a maximum value and then remains stable. When the intensity is increased it reaches a maximum value and then, after certain periods of time, decreases. In other words, the noise takes longer to be stored. This dependency on

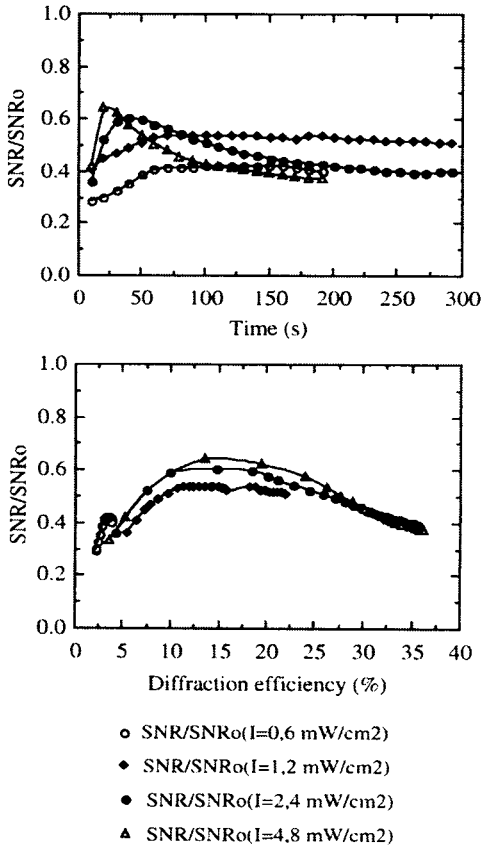

Figure 13. Normalized signal to noise ratio as a function of: (a) time, and (b) diffraction efficiency, for a beam ratio of 5.

the intensity is a result of the polymerization rate being greater for high intensities, therefore the hologram is formed more quickly (steep slope of the curve considered). At the same time as the diffraction grating is stored, the intermodulation noise resulting from interference between the object waves from different points of the object is also stored, giving rise to low frequency gratings. The presence of this intermodulation noise leads to the drop in the curve.

With respect to the diffraction efficiency (Figure 13b), the behavior of the material is very similar except for the intensity of 0.6 mW/cm^2, the only one at which the noise of the CCD is stored. Nevertheless each curve indicates the possible diffraction efficiency for a given SNR/SNR_0 or, in other words, the diffraction efficiency range of the stored images. For an intensity of 4.8 mW/cm^2 a normalized signal to noise ratio of 0.65 is reached with a diffraction efficiency of 15%.

We also studied the influence of the beam ratio on the quality of the holographic image. The beam ratios analyzed were K = 3, 5, 10, 20, for a constant intensity, I = 1.2 mW/cm^2 as can be seen in Figure 14. This Figure shows the normalized signal to noise ratio as a function of time (Figure 14a) and diffraction efficiency (Figure 14b). When the beam ratio is increased the maximum signal to noise ratio obtained increases. The best results were obtained for K = 20 with a signal to noise ratio of 0.94 giving a holographic image with a signal to noise ratio very similar to that of the object image captured. Figure 14b shows the normalized signal to noise ratio as a function of diffraction efficiency for different beam ratios. This graph shows the diffraction efficiencies that may be reached when the image of the diffuse object is stored and at the same time the signal to noise ratio. As can be seen the greatest diffraction efficiency is about 20% and the SNR/SNR_0 at this point is 0.5, due to the fact that at the same time as we store the image of the diffuse object we also store the intermodulation noise. For $SNR/SNR_0 = 0.94$ and K = 20 a diffraction

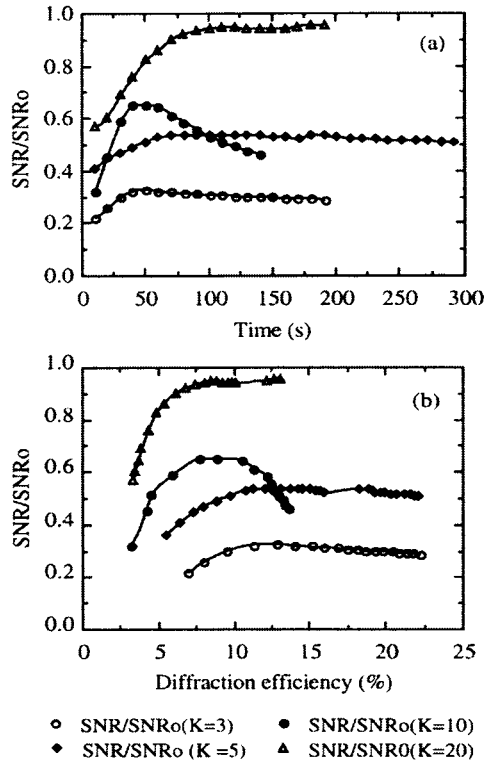

Figure 14. Normalized signal to noise ratio as a function of: (to) time, and (b) diffraction efficiency, for an intensity of 1.2 mW/cm^2.

efficiency of 13% is reached.

It should be pointed out that the diffraction efficiency reached when storing diffuse objects is less than that obtained when storing diffraction gratings, as has been seen when a comparison was made with the curves obtained in the first paragraphs of this study. This decrease in diffraction efficiency is due to the fact that when the images were captured by the CCD, it was necessary to cut off the object beam in order to store the image of this object reconstructed by the hologram. By eliminating one of the beams interfering at the plate, the plate was illuminated only by the remaining beam, that is a uniform beam since the interferential pattern at the plate disappeared. Upon irradiating the photo-

polymer plate with a uniform beam, the maximum modulation of refractive index is reduced, thereby reducing the value of the maximum diffraction efficiency.

On the other hand the diffraction efficiency measurements obtained in the previous paragraphs of this study were made using a wavelength of 633 nm, while in this paragraph corresponding to the storage of diffuse objects, diffraction efficiency measurements were made with a wavelength 514 nm at which the behavior of the photopolymer is different, since at this wavelength absorption takes place by the material employed.

4. CONCLUSIONS

A photopolymeric system has been optimized analyzing both the influence of the thickness and the intensity used, with greater sensitivities being found for greater thicknesses and lower intensities. The diffraction efficiency reached in all cases was of the order of 80%.

We obtained and studied thick photopolymer films (about 150 μm) in which the acrylamide concentration was varied. Differences were found in the diffraction efficiency versus exposure curves. These differences were seen at high concentrations when two different slopes were obtained, implying two different polymerization rates and so two different terminations of the photopolymerization process. These two different slopes are due to the appearance of two peaks in diffraction efficiency that can be identified by the intensity used. At the same time a decrease in scattering was observed for high efficiencies using high concentrations of acrylamide and high thicknesses, and this makes it possible to use the photopolymer in information storage applications such as holographic memories.

High and low frequency diffraction gratings were obtained by the copy method, with efficiencies of about 27% for low frequency gratings and 40% for those of high frequency.

Finally, diffuse objects were stored in this photopolymer and the recording material was studied at different intensities and beam ratios. The best result for SNR/SNR_0 of 0.94 was found for a beam ratio of 20 and an incident intensity of 1.2 mW/cm^2. In this case the diffraction efficiency was 13%.

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