

# PROCEEDINGS OF SPIE



SPIE—The International Society for Optical Engineering

## *Holographic Materials IV*

**T. John Trout**

*Chair/Editor*

**27 January 1998**

**San Jose, California**

*Sponsored by*

SPIE—The International Society for Optical Engineering

IS&T—The Society for Imaging Science and Technology

*Published by*

SPIE—The International Society for Optical Engineering



**Volume 3294**

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Please use the following format to cite material from this book:

Author(s), "Title of paper," in *Holographic Materials IV*, T. John Trout, Editor, Proceedings of SPIE Vol. 3294, page numbers (1998).

ISSN 0277-786X  
ISBN 0-8194-2734-9

Published by  
**SPIE—The International Society for Optical Engineering**  
P.O. Box 10, Bellingham, Washington 98227-0010 USA  
Telephone 360/676-3290 (Pacific Time) • Fax 360/647-1445

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Printed in the United States of America.

# Optimal composition of a holographic recording material

Salvador Blaya<sup>2</sup>, Luis Carretero<sup>1</sup>, Antonio Fimia<sup>2</sup>, Ricardo Mallavia<sup>2</sup>, María T. Garzón<sup>2</sup>, Roque F. Madrigal<sup>2</sup>, Immaculada Pascual<sup>1</sup>, Augusto Beléndez<sup>3</sup>, Roberto Sastre<sup>4</sup>, Francisco Amat-Guerri<sup>4</sup>.

1. Laboratorio de Óptica. Departamento Interuniversitario de Óptica. Universidad de Alicante, Apdo. 99. Alicante 03080 Spain, e-mail: luis@aclis.labopti.ua.es
2. Departamento de Ciencias Experimentales y Tecnología. Universidad Miguel Hernández. Paseo Melchor Botella s/n, Apdo. 03206, Elche (Alicante) Spain.
3. Departamento de Física, Ingeniería de Sistemas y Teoría de la señal. Universidad de Alicante. Apdo. 99. Alicante 03080 Spain.
4. Instituto de Química Orgánica e Instituto de Ciencia y Tecnología de Polímeros, C.S.I.C, Juan de la Cierva 3, E- 28006 Madrid Spain

## ABSTRACT

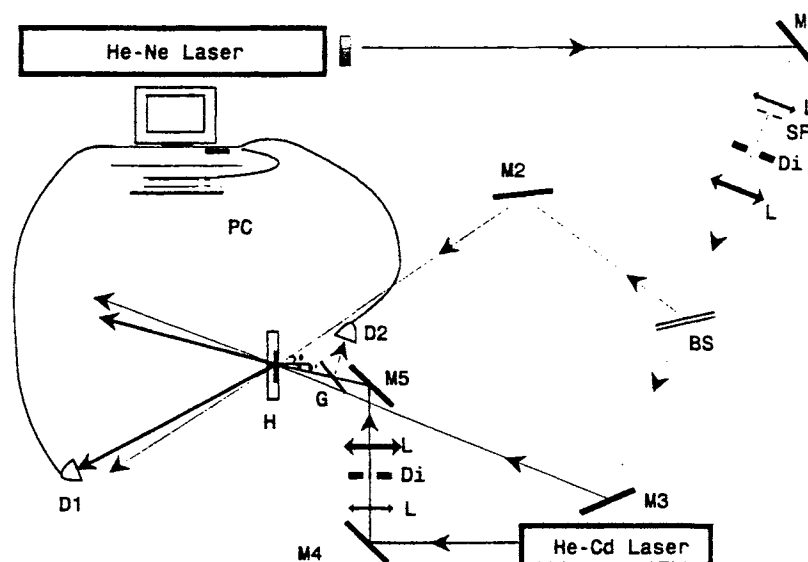
We study the effect of the addition of a crosslinking agent (*N,N'*-metylenbisacrylamide) in a photopolymerizable matrix for real time holography. The optimization of the concentration of this component has been realized attending to the holographic parameters like energetic sensitivity and diffraction efficiency. Diffraction efficiencies near to 80 % have been obtained with energetic exposures of 12 mJ/cm<sup>2</sup>.

## 1.INTRODUCTION

The photopolymerization -based processes<sup>1</sup> have been used in a lot of fields like the fabrication of integrated circuits, microlithography, optical data storage and holography. The mechanism of recording holograms in these materials implies photopolymerization processes in the illuminated zones that it gives variations of refraction index or volume. The advantage of these systems is that it can be treated as pure volume holograms with high diffraction efficiency and it can be monitored at real-time. A typical photopolymeric recording material system is composed by a polymeric matrix that it supports monomers and the photoinitiator system. In our case the material is composed by polyvinylalcohol as binder monomers as acrylamide (AA) and *N,N'*-metylenbisacrylamide (BMA), an electron-donor triethanolamine (TEA) and methylene blue (BM) as dye. Acrylamide have been used in holography widely.<sup>2-4</sup> An improvement was achieved when *N,N'*-metylenbisacrylamide was added to speed up the polymerization reaction.<sup>5-7</sup> In this case index modulation is produced when a molecular space lattice of acrylamide chains is formed, and BMA builds crosslinks to form a copolymer which is transparent. In this work we present the characteristics and the optimal concentration of this compound that gives a holographic recording material with sensitivities of 12 mJ/cm<sup>2</sup> and diffraction efficiencies of 80 % with a spatial frequency of 1000 lines/mm.

## 2. EXPERIMENTAL

Real-time measurements of the transmission hologram recorded were carried out on the system described in Figure 1. The holographic gratings are created with a He-Ne laser tuned at a wavelength of 633 nm. The power intensity carried by each beam was  $2 \text{ mW/cm}^2$  and the relation of intensities between them was 1:1. The object and the reference were recombined at the sample at an angle of  $20^\circ$  from the normal with an appropriate set of mirrors and the spatial frequency obtained is 1,000 lines/mm. The diffracted intensity was monitored with a He-Cd laser positioned at the Bragg's angle tuned at 441 nm where the material does not absorb. In order to obtain the intensity of this beam (incident) at each time, a photodiode detector (D2 in Figure 1) positioned in the reflection direction of a glass situated in the trajectory of this beam. The incident intensity was the result of the product of the intensity measured and the previously calibrated one with a second photodiode detector (D1) in the transmission beam. The diffracted beam was detected with a second photodiode detector both detectors being connected to a personal computer and the diffraction efficiency was calculated as the ratio of the diffracted and incident intensities.



**Figure 1** A schematic representation of the setup for the recording of holographic growth with time curves. BS: beam splitter. SF: spatial filter. D: detector. L: Lens. Di: diaphragm. M: mirror. G: glass. H: holographic plate. PC: personal computer.

All the components of the material were of the highest grade available and used without any further purification provided by SIGMA. The photosensitive aqueous solution was prepared when over 50 ml of polyvinylalcohol (provided by Riedel-de-Häen and  $M_w \approx 25000$ ) of 10 % by weight, 2.5 ml of methylene blue of 6.25 mM and 8 ml of a solution the necessary

concentration of bisacrylamide. 2.5 M of acrylamide and 1.5 M of triethanolamine were added. The film was prepared by coating the photosensitive solution over a 20 x 40 cm<sup>2</sup> glass (BK7) with a TLC coater provided by CAMAG allowing it to dry 20 h under normal conditions (65 % R.H. and 20 °C). The resulting thickness of the film was measured with an apparatus P.I.G. 455 supplied by Neurtek. In Table 1 the concentration of the optimal composition is summarized. This composition gives 80 % of diffraction efficiency with energetic exposures of 12 mJ/cm<sup>2</sup>.

**OPTIMAL CONCENTRATION THE PHOTSENSITIVE MATERIAL**

TRIETHANOLAMINE (TEA)	0.2 M
METHYLENE BLUE	2.1x10 <sup>-4</sup> M
ACRYLAMIDE	0.34 M
PVA	5 %
BISACRYLAMIDE	0.053 M

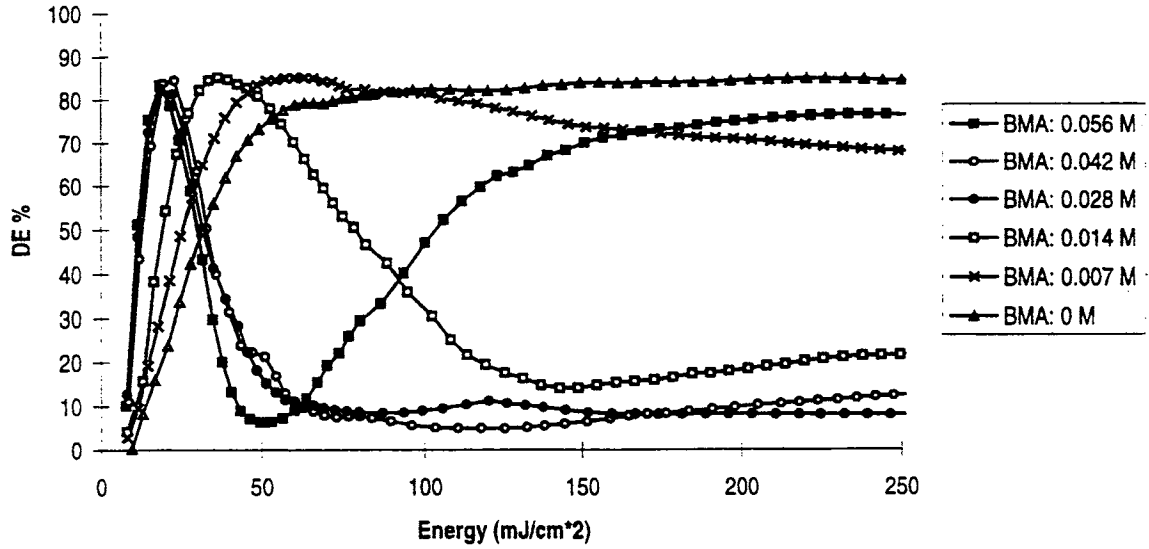
Table 1. Composition of the photopolymer coating solution.

### 3. RESULTS AND DISCUSSION

When concentration of the monomeric species is raised the rate of polymerization increase. In a previous work<sup>9</sup> we showed that in a material where the unique monomeric compound is acrylamide, the increase of the concentration produces high sensitivities. This effect is shown when we use two monomers. In Figure 2 we present the diffraction efficiency as a function of exposure for different concentrations of the crosslinking agent (bisacrylamide). The drawback of this composition is that the concentration of both monomers can not be increased indefinitely due to their solubility in the PVA matrix.

The effect of the concentration is clearly explained if we use the Kogelnik's theory<sup>10</sup>. We have used the expression 1 like index modulation as a function of time, where  $n_{o1}$  is the final modulation index obtained and  $\alpha$  is a parameter related with the rate of copolymerization and intensity.

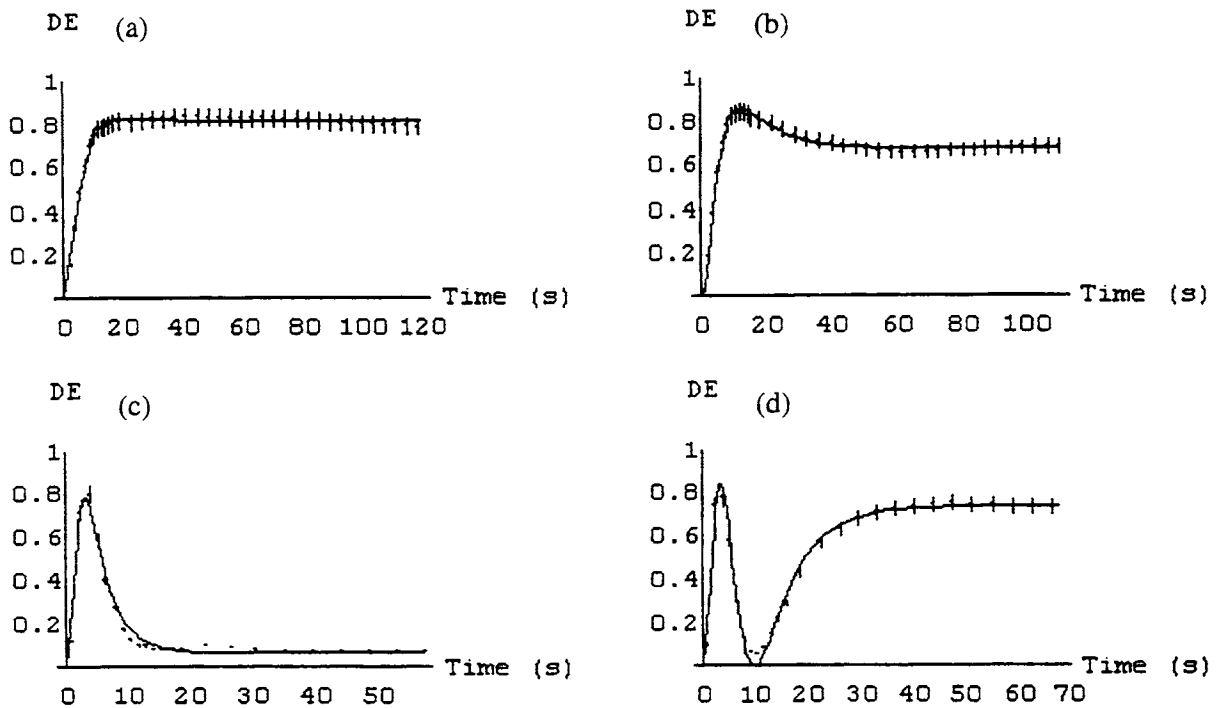
$$n_1 = n_{o1} (1 - e^{-\alpha}) \quad (1)$$



**Figure 2:** Effect of the variation of *N,N*-methylenbisacrylamide over the diffraction efficiency recorded. The composition of the material is: AA: 0.335 M; TEA: 0.199 M; BM:  $2.6 \times 10^{-4}$  M; PVA: 10%. The intensity was  $4 \text{ mW/cm}^2$  and the thickness  $75 \text{ }\mu\text{m}$ .

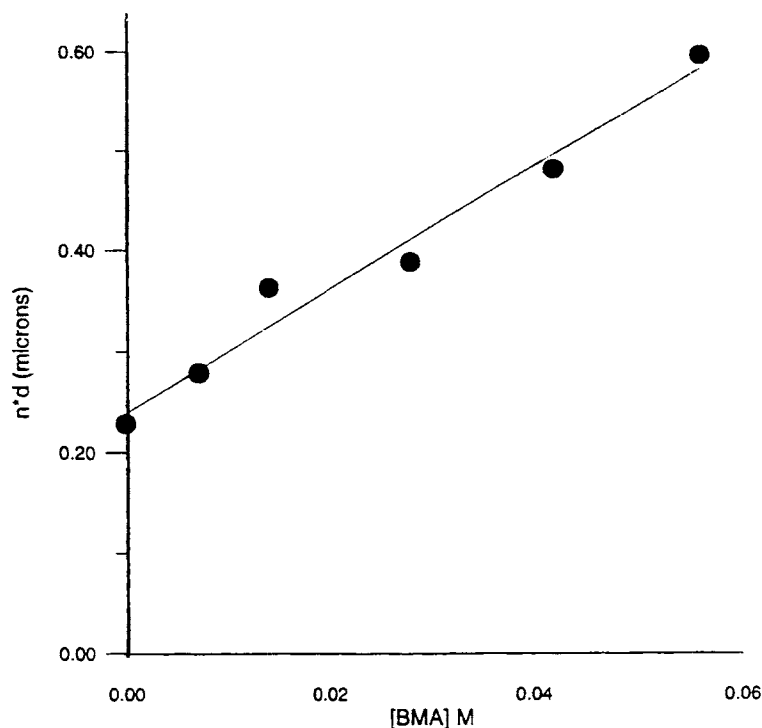
Equation 2 is the final expression used to fit the temporal variation of diffraction efficiency, where  $\Lambda$  is the factor related with the scattering losses and absorption and  $d$  is the thickness. The wavelength  $\lambda$  is  $441 \text{ nm}$  and  $\theta_B$  is the corresponding Bragg angle at this wavelength. Fitted curves are showed in figure 3, where a good agreement between the experience and theory is produced.

$$\eta(t) = \Lambda \sin^2 \left( \frac{n_{o1} (1 - e^{-\alpha}) \pi d}{\lambda \cos \theta_B} \right) \quad (2)$$



**Figure 3:** Experimental and fitted curves of temporal variation of diffraction efficiency for materials with different concentrations of bisacrylamide. (a) without BMA; (b) 0.007 M; (c) 0.028 M; (d) 0.053 M.

The results of the nonlinear fit are presented in figure 4. The product of the final modulation index and thickness (“optical path” of the formed grating) shows a linear relationship between the “optical path” of the grating and the concentration of bisacrylamide exists. When the concentration of BMA is raised the quantity of doubled bounds in the system increase, however the most important point could be the formation of layers instead of lineal polymeric chains that it is produced in a single monomeric polymerization process. However  $\alpha$  does not change with the increase of BMA.



**Figure 4:** Change of Optical path (nxd) versus concentration of bisacrylamide.

#### 4. CONCLUSIONS

In previous papers we study the optimal composition of the system composed uniquely by acrylamide as monomer, in this case the energetic sensitivity is of  $45 \text{ mJ/cm}^2$ . When bisacrylamide is introduced a great improvement in the sensitivity is produced as we show in figure 2. The effect of the concentration of the crosslinking agent has been studied. Optimal concentration that gives diffraction efficiencies of 80 % with an energetic exposures of  $12 \text{ mJ/cm}^2$  can be obtained when the hologram is recorded with an intensity of  $4 \text{ mW/cm}^2$  and a resolution of 1000 lines/mm. By using a theoretical model we have showed that the increase in sensitivity is due to the increase of the final modulation index

#### 5. ACKNOWLEDGMENT

The authors gratefully acknowledge Research Project PTR-0115 (CICYT) support for part of this work and to the commission interministerial de Ciencia y Tecnología (C.I.C.Y.T.) of Spain for financial support project MAT96-1767-E.



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