Characterization of a real-time high-sensitivity photopolymer for holographic display and holographic interferometry

Xiaowei Guo[1], Jianhua Zhu[1]*, Chuanqin Xia[2], Jianfeng Li[1], Li Chen[1]
1. Institute of Information Optics, College of Physics, Sichuan University, Chengdu 610064, China
2. College of Chemistry, Sichuan University, Chengdu 610064, P. R. China

ABSTRACT

In this paper, a red-sensitive photopolymer based on multiple polymerizable monomers is developed for holographic applications. The detailed quantitative experiments on the reagent concentrations, the exposure, the thickness of the photosensitive layer, are conducted to determine an optimum recipe, and bright volume phase holograms with high diffraction efficiency are obtained in our experiments at the exposure level of 4mJ/cm^2. This photopolymer also has higher resolution and self-development capacity, This makes it very convenient and useful for the applications of both real-time and double exposure holographic interferometry, some preliminary experiments of holographic interferometry are demonstrated in this paper for the nondestructive testing and measurement of minor distortion and displacement.

Keywords: holographic recording material, photopolymer, holographic display, holographic interferometry

1. INTRODUCTION

The characteristics of holographic recording material have great effects on the successful applications and the development of holography. Currently available holographic recording materials include silver halide emulsion, dichromated gelatin, photopolymer etc. Silver halide emulsion has high sensitivity, but its diffraction efficiency and signal-to-noise ratio are low, which limits its applications. Dichromated gelatin has high diffraction efficiency and resolution, now has been successfully employed for the fabrication of Lippmann-type display holograms and holographic optical elements. However, dichromated gelatin also has some disadvantages such as low sensitivity, low shelf life, and poor environmental stability which is due to its sensitivity to ambient humidity. Photopolymeric material can avoid above shortcomings, it also has some unique merits, i.e. self-development, high angular selectivity, etc., these technical capacities make it a good candidate for the applications like optical storage, holographic display, holographic interferometry, and so on. [1]

A photopolymer is basically made of one or several monomers, sensitizer and initiator, all on a polymer as film-forming reagent. In recent years, a great deal of attentions have been given to the acrylamide-based polyvinyl alcohol (PVA) photopolymer system, which is initiated by triethanolamine (TEA), because the material has the following advantages: facility of the material preparation, less toxicity, low-cost, high diffraction efficiency and high sensitivity. The major drawback of the system is its low resolution, and it is difficult to obtain high diffraction efficiency at high spatial frequency over 2000 lines/mm, especially when red light are used as the recording beam. [1-5]

In this paper, the preparation of photopolymer film and its imaging mechanism are presented firstly, then the experimental study on the reagent concentrations, the polymerization degree of PVA (film-forming reagent), the exposure, the thickness of the photosensitive layer, are conducted quantitatively to obtain a optimum recipe of photopolymer. Finally, the applications of this photopolymer for holographic interferometry and holographic display are demonstrated in our laboratory for the nondestructive testing and measurement of minor distortion and displacement.

* *zhujh123@hotmail.com; phone 86-28-85255499*

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2. PHOTOPOLYMER PREPARATION AND IMAGING MECHANISM

The photopolymer is basically an acrylamide-based polyvinyl alcohol solution. It consists of acrylamide as primary monomer, methylene bisacrylamide and acrylic acid as promoters and supplementary monomers, methylene blue as sensitizer, triethanolamine as electron donor, all on a aqueous film of polyvinyl alcohol (PVA) as binder. The photosensitive layer is prepared by adding 0.625ml of 1 % w/v methylene blue solution, 1ml of 40% triethanolamine solution, 0.4ml of 2.0% acrylic acid solution, 0.45g of methylene bisacrylamide and 2.4g acrylamide to 100ml of 5.0% polyvinyl alcohol solution. We prepared four batches of photopolymer plates with different thickness by pouring 10ml、18ml、27ml、36ml of mixed solution over 24cm×8cm glass plate, respectively. Then the plate is dried in the dark under normal laboratory conditions ($T \cong 20-25^{\circ}C, RH \cong 50-70\%$). The thickness of prepared photopolymer film approximately corresponds to 25μm, 45μm, 70μm, 100μm.

The basic imaging mechanism for the radical polymerization can be described as the following processes [6]. The first step is the initiation process, which involves the production of free radicals, then these radicals bind to the monomers to create the chain-initiating species. After initiation, these species propagate by combining with other monomer molecules to form a large polymer chain. Finally, when the radical of the growing polymer bonds with a free radical a dead polymer is formed.

In our materials, PVA is not used only as a binder, it also acts as an active reagent. The introduction of coexistent monomers, on the one hand, can speed up the process of polymerization reaction of monomers, and on the other hand, it can also form a more stable cross-linking structure with PVA polymer to increase the refractive index modulation and diffractive efficiency of hologram. The possible multiple polymerization mechanism in the photosensitive layer can be expressed as follows:

$$D \xrightarrow{h\nu} D^{*} \xrightarrow{HED} ED^{*} \xrightarrow{R,M} R' + M' \xrightarrow{\begin{cases} A \\ 2A \\ \end{cases}} \begin{cases} A\text{-}R_n & \text{(engraftation)} \\ A\text{-}M\text{-}A & \text{(crosslinking)} \\ R\text{-}M\text{-}R & \text{(copolymerization)} \end{cases}$$

Where D denotes dye, D^{*} excited dye, HED electron donor, ED^{*} the radical of electron donor, R the monomers, M the cross-linking monomer, R' the radical of monomer, M' the radical of cross-linking monomer, and A is PVA.

When the photosensitive layer is irradiated by He-Ne laser with a wavelength of 633 nm, photosensitive dye D is excited to high energy state D^{*}, then it transfers the exciting energy to electron donor HED to produce free radicals ED^{*}. At the second step, free radicals bind to monomers R and M to produce the radicals R', M' and further chain-initiating species. These species propagate by combining with other monomer molecules to form a large polymer chain R_n, and copolymerization takes place between R and M. Meantime, PVA may engraft into R_n and build crosslinking structure with M, these supplementary structures also contribute to the increase in refractive index modulation. After the exposure process is finished, infrared light or ultraviolet light is employed to irradiate the holograms entirely to carry out the fixation process. This additional step will decompose the residual dye or optical initiator and finish the polymerization process of the remaining monomers, then the gratings with refractive index modulation is obtained.

3. EXPERIMENTAL OPTIMIZATION OF RECIPE

3.1 The concentration of acrylic acid

Acrylic acid exhibits two functional aspects in our photosensitive recipe. On the one hand it acts as a monomer for polymerization process, On the other hand it can maintain the system's humidity at a certain extent due to its high viscosity, which will make the radicals diffuse easily to improve the speed of photochemical reaction and sensitivity.

However the introduction of acrylic acid may somewhat reduce the temporal stability of photosensitive layer, especially in high ambient humidity where acrylic acid absorbs more water and leads to the swelling of PVA and the decrease in diffraction efficiency of hologram. As a result, the concentration of acrylic acid must be adjusted to a proper value. Fig.1 shows the relation between the concentration of acrylic acid and the diffraction efficiency of hologram, The chemical composition of the material is AA 5.0%, BAA 2.4%, TEA 2.0%, PVA 5.0%, where the thickness is 45μm and the exposure intensity used is 0.68mw/cm^2. We can see that the optimum concentration is 2%. Fig.2 shows the comparative results with and without acrylic acid, we can find that the introduction of acrylic acid can increase the diffraction efficiency and sensitivity obviously.

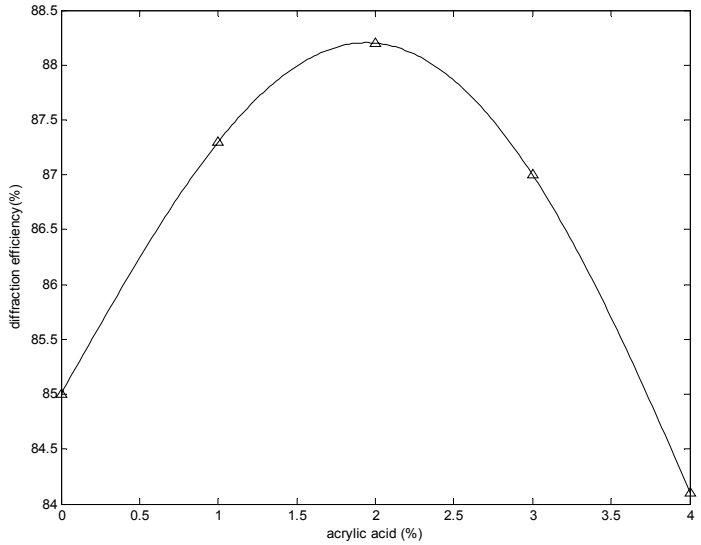

Fig.1 The diffraction efficiency of hologram as a function of the concentration of acrylic acid

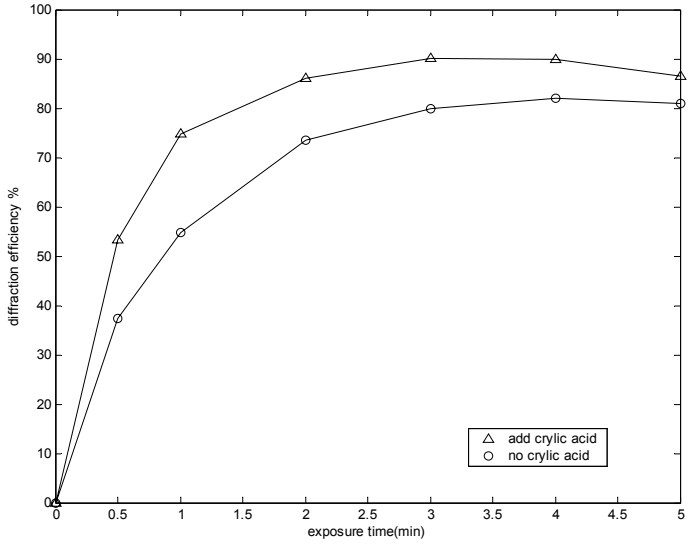

Fig.2 the comparative results with and without acrylic acid

3.2 The polymerization degree of PVA

Two types of PVA polymers with different polymerization degree are used for the comparative experiments. Their degrees of polymerization are 342 and 1750, corresponding molecular weights are 15000 and 72000, and degree of hydrolysation is 86~89 mol%. We found that the PVA polymer with low polymerization degree had better solubility and uniformity. From Fig.3 it can be seen that the photosensitive layer using low polymerization degree PVA can achieve diffraction efficiency higher than 70%, and its photo-sensitivity is also higher, the experimental result agrees with that presented by Salvador Blaya [7]. Moreover, we also found that the monomers in PVA with low polymerization degree will crystallize less during drying. It seems that PVA with low polymerization degree can reduce the deposition of the monomer on the surface of the film, and consequently improve the homogeneity of photosensitive film.

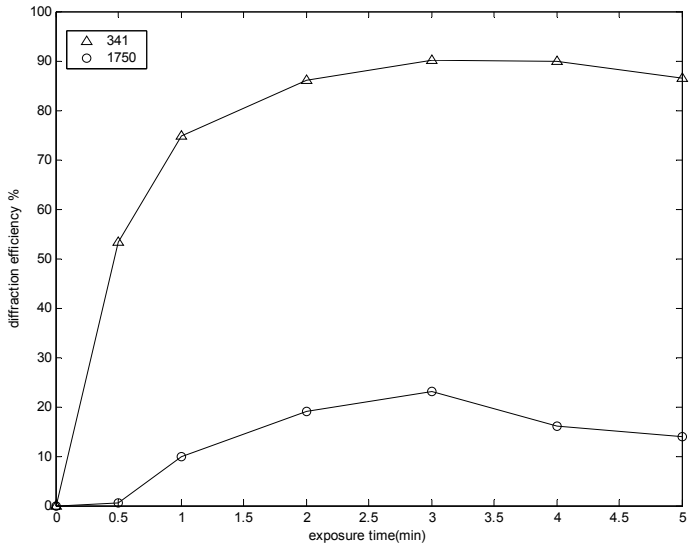

Fig.3 Diffraction efficiency dependence of exposure time at different PVA polymerization degree

3.3 The exposure

The relation between diffraction efficiency and exposure is given in Fig.4. The thickness of photosensitive layer is about 45 μm, and spatial frequency of grating is approximately 1000 lines/mm. From Fig.4 we can see that at the first period of exposure, local photo-polymerization is linearly proportional to the exposure energy, the diffraction efficiency increases sharply with exposure. When the exposure energy reaches to 4 mJ/cm^2, the modulation of refraction index saturates and the diffraction efficiency will not increase with the exposure. Furthermore, the diffraction efficiency decreases for some degree, this may be caused by the cross-talk of recorded holographic gratings during consequent exposure. Some of the noise sources are (1) surface deformation of the recording material, (2) random scatter caused by the granularity of the recording material, and (3) nonlinear recording of the signal wave. When the exposure increases, the cross-talk increases much quickly and results in more output signal being scattered, and the diffraction efficiency decreases. [8] High diffraction efficiency over 90% can be obtained at the exposure level of 4 mj/cm^2, which shows the high photo-sensitivity of this photopolymer material.

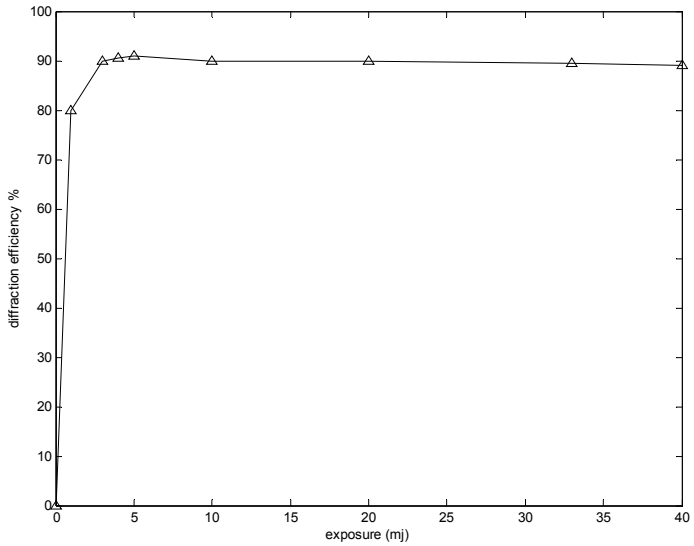

Fig.4 Diffraction efficiency as a function of exposure of photopolymer hologram

3.4 The thickness and spatial frequency response

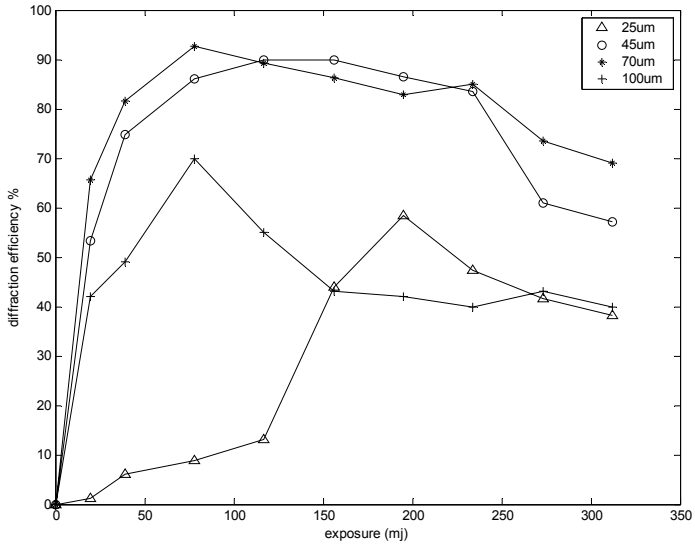

Fig. 5 Diffraction efficiency as a function of exposure at the spatial frequency of 2000 lines/mm

Fig.5 and Fig.6 show the influence of thickness on diffraction efficiency at spatial frequency of 2000 lines/mm and 3000 lines/mm respectively, From these experimental results we know that the thickness affects not only diffraction efficiency but also photosensitivity. In Fig.5, with the increase of thickness from 25μm to 70μm, the sensitivity of photopolymer film and the diffraction efficiency of hologram increase obviously. We think that the increase in thickness leads to the enhancement of volume effect and the achieved modulation degree of refraction index. But when thickness increases to 45 μm the diffraction efficiency increases slowly, and up to 100 μm the diffraction efficiency decreases rapidly which may be attributed to the serious absorb, scatter and reflection in films with the increase in thickness. As a result, the plates of 45 μm and 70 μm are and proper and favorable in our experiments. In Fig.6 we only use the plates of 45μm and 70μm for the experimental comparisons. It is observed that at spatial frequency of 3000 lines/mm, the

diffraction efficiencies up to 50% are reached for both 70μm and 45μm, which shows this photopolymer material is suitable for the applications such as display holography, especially for the recording of reflection hologram.

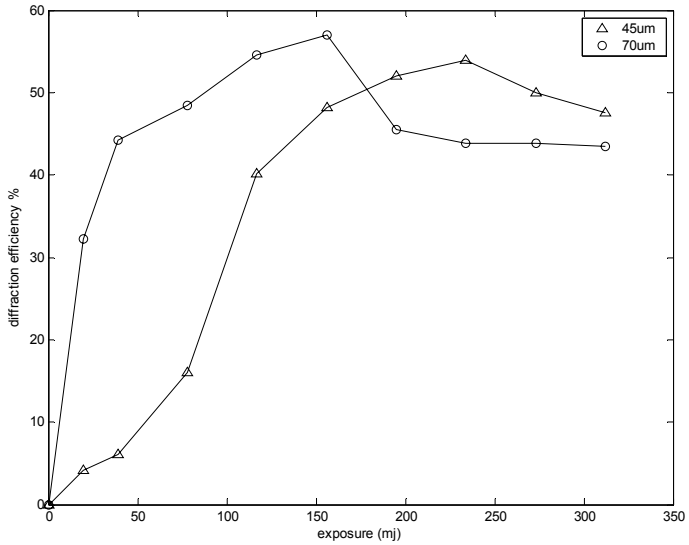

Fig.6 Diffraction efficiency as a function of exposure at the spatial frequency of 3000 lines/mm

If comparing Fig.4, Fig.5 and Fig.6 carefully, we can also draw a conclusion that the photosensitivity falls with the increase in spatial frequency, the research on the factors affecting the resolution and their mechanism is under way.

4. HOLOGRAPHIC APPLICATIONS

As most of the other photopolymer-type materials, our photopolymer has good self-development capacity, which makes it very convenient and useful for the holographic applications. No requirement for post-processing or repositioning after exposure is an attractive aspect for holographic interferometry or holographic storage. Some preliminary experiments of both real-time and double-exposure holographic interferometry are demonstrated in this paper for the nondestructive testing and measurement of minor distortion and displacement.

4.1 Real-time holographic interferometry

Conventional real-time holographic interferometry usually employ silver halide emulsion or dichromated gelatin as recording material, the original object and reference waves interfere with each other to make a holographic latent image at first. After the photographic plate has been developed and fixed, it is carefully arranged to its original position. The exact superposition of object and image wave requires the deviation of replacement to be no more than a fraction of a fringe width, this is very difficult to perform. As a result in actual work, the hologram is processed 'in situ' using a specially designed plate shelf and container in which the recording plate is immersed and developed. Then the reference beam and transformed object beam synchronously illuminate the hologram, the reconstructed original object wave and the actual object wave interfere to obtain an interference pattern which gives the characteristics of the deformation and displacement. The advantage of this technique is that any small movement or deformation of object caused in real-time process by thermal, electrical, mechanical effects can be observed. Therefore it is called as real-time holographic interferometry or live-fringe holographic interferometry.

But it is not easy to accomplish these goals by using conventional recording materials [9], which greatly limits the practical applications of holographic interferometry. One of the most attractive characteristics of our photopolymer

system is its dry or self-development capacity, it avoids the complicated wet chemical processing of the above recording materials. After the holographic recording process, a dry optical or thermal development by overall ultraviolet or infrared radiation is followed to fix the holographic image. This makes it very convenient and useful for the applications of both real-time and double exposure holographic interferometry as the repositioning problem of reconstructed wavefront is eliminated completely. A demonstration of the suitability of this photopolymer to holographic interferometry was carried out as follows.

A white flat disk is fixed on a shelf that can move freely up and down. The two expanded beams from He-Ne laser are splitted by a beam splitter, one illuminates the flat disk while the other is incident directly upon the recording plate as reference beam, and the light scattered from the disk onto the recording plate is the object beam. The dimension of recording plate is 6cm×8cm and thickness is 70μm. At the end of the exposure, the intensity of the illuminating light was reduced, both to allow readout without risk of further recording, and to increase the visible fringe contrast. [9]

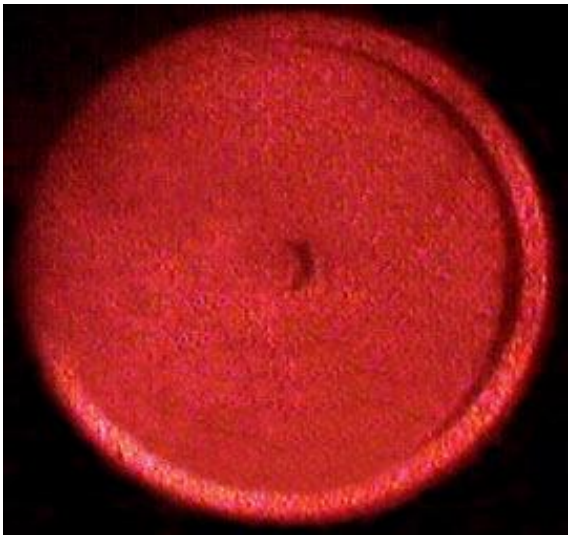

Fig.7 the reconstruction image of the white flat disk

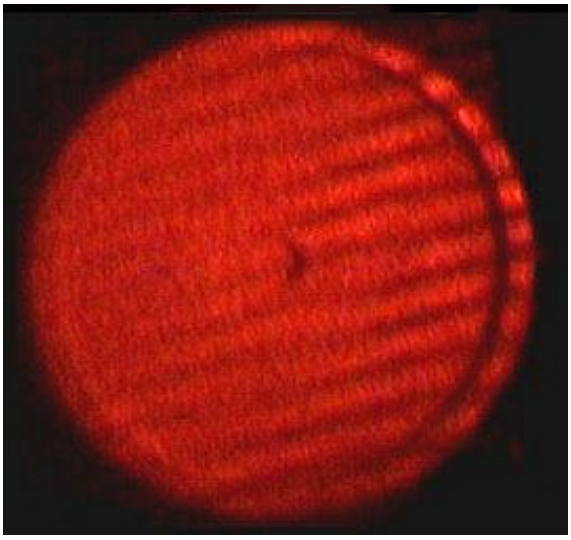

Fig.8 The wide interference fringes produced by a minor displacement

Fig.9 The narrow interference fringes produced by a larger displacement

The reconstructed hologram can be viewed directly by blocking the object with a piece of black card. Figure 7 shows a reconstruction image of the disk, at this moment there is no interference fringes on the disk surface as there is no movement or distortion occurred. Figure 8 shows wide interference fringes produced by a minor displacement, larger motion causes narrow interference fringes superimposed on the surface of disk, the photograph is shown in Figure 9. So by counting the number of interference fringes, we can roughly judge the moving distance of disk.

4.2 Double-exposure holographic interferometry

Double-exposure holographic interferometry is similar to real-time holographic interferometry. The difference between them is that two holograms are recorded on the same holographic plate at different time in Double-exposure experiment instead of recording a hologram. The first one records the original object wave while the second one records transformed object wave. When the reference wave illuminates the holographic plate, the two reconstructed object waves interfere with each other because of the differences in their amplitudes or phases. The three-dimensional image covered by the interference fringe pattern can be viewed, and the fringe characterizes the movement or distortion of object wave field.

Here we use transparent light bulb to perform the double-exposure holographic interferometry experiment. At first the bulb was powered off, the temperature field and air density around it was uniform, then the first hologram is recorded. After a while the bulb was powered on, ambient temperature rise up sharply due to the heat from the bulb, which changes air density and air refraction index, the second hologram with phase deformation is recorded. The reconstructed double-exposure holographic image of bulb temperature field is given in figure 10. From figure 10, it can be seen that the interference fringes are sparse away from the filament which shows that the temperature change is smaller, while the dense interference fringes near the filament shows the sharp change of temperature field. The exact phase change can be calculated by some reported techniques. [10-11]

Fig.10 Double-exposure hologram of an bulb showing the change of temperature field

4.3 Holographic display

Our photopolymer material has very high sensitivity, which can shorten the recording time and reduce the effect of vibration, so high quality holograms can be produced easily. A higher diffraction efficiency over 55% can be obtained even when the spatial frequency of hologram reaches to 3000 lines/mm. This makes it suitable for the recording of various holograms, some transmission and reflection holograms are recorded successfully in our laboratory. But the brightness of reflection hologram is not high and needs further improvement.

5. CONCLUSION

By recipe optimization in the concentration of Acrylic acid, the polymerization degree of PVA, the thickness of the photosensitive layer and exposure, a photopolymer system of acrylamide-based polyvinyl alcohol is successfully developed for holographic applications of display and interferometry. Bright volume phase holograms with high diffraction efficiency over 90% were obtained in our experiments at the exposure level of 4mJ/cm^2. The experimental demonstrations on holographic interferometry and holographic display show the potential application prospects of our photopolymer material.

REFERENCES

1. S. Martin, C. A. Feely, V. Toal, "holographic recording characteristics of an acrylamide-basec photopolymer", *Appl. Opt.*, *36*, pp.5757-5769, 1997
2. J. A. Jenney, "Holographic recording with photopolymers," *J. Opt. Soc. Am., 60*, pp.1155-1161, 1970
3. R. Mallavia, A. Fimia, C. Garcia, "two dyes for holographic recording material panchromatic ion pair from Rose Bengal and Methylene Blue", *J. Mod. Opt.*, *48,* pp.941-945, 2001
4. C. Carre, D. Lougnot, et al, "Photopolymerizable material for holographic recording in the 450-550nm domain: Characterization and Applications (Ⅱ)", *J. Opt. (paris)*, *23*, pp.73-79, 1992
5. Calixto, "dry polymer for holographic recording", *Appl. Opt.*, *26*, pp.3904-3910, 1987

6. C. Neipp, S. Gallego, M. Ortuno, A. Marquez, et al, “Characterization of a PVA/acrylamide photopolymer.Influence of a cross-linking monomer in the final characteristics of the hologram”, *Opt. Comm.*, *224*, pp.27-34, 2003
7. S. Blaya, L. Carretero, R. Mallavia, A. Fimia, et al, “optimization of an acrylamide-based dry film used for holographic recording” , *Appl. Opt.*, *37*, pp.7604-7609, 1998
8. Huawen Yao, Mingju Huang, Zhongyu Chen, Lisong Hou, Fuxi Gan, “Optimization of two- monomer-based photopolymer used for holographic recording”, *Materials Letters*, *56*, pp.3–8, 2002
9. S. Martin, C. Feely, J. T. Sheridan, V. Toal , “Applications of a self developing photopolymer material: Holographic interferometry and high efficiency diffractive optical elements”, *Proc. SPIE*, *3264*, pp.60-70, 1998
10. K. Creath, “Phase measurement interferometry techniques”, In *Progress in Optics*, *XXVI*, edited by E. Wolf, pp.349-393, 1988
11. M. Takeda, H.Ina. S. Kobayashi, “Fouried transform method of fringe pattern analysis for computer based topography and interferometry”, *J. Opt. Sco. Am.*, *72*, pp.156, 1982