A wideband sensitive holographic photopolymer

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A novel wideband sensitive dry holographic photopolymer sensitized by rose bengal (RB) and methylene blue (MB) is fabricated, the holographic storage characteristics of which are investigated under different exposure wavelengths. The result shows that the sensitive spectral band exceeds 200 nm in visible light range, the maximum diffraction efficiency under different exposure wavelengths is more than 40% and decreases with the decrease of exposure wavelength, the exposure sensitivity is not change with the exposure wavelength. This photopolymer is appropriate for wavelength multiplexing or multi-wavelength recording in digital holographic storage.

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Because of the potential of providing super high data recording density coupled with high data transfer rates for its intrinsic volume storage and parallel processing characters, digital holographic storage has been extensively studied with the aim of meeting the ever-increasing demand for high-density data storage with fast data recording and retrieval rates in recent ten or more years[1,2]. But there are still some obstacles should be passed before such a system can be got into use, these obstacles include the perfect material that matching all the requirement of digital holographic recording and the recording scheme that coupled with the relative recording media[3].

Tremendous efforts have been made in the development of holographic data storage materials ranging from crystals such as doped $LiNbO_3$ and photorefractive $BaTiO_3$ crystals to photopolymers and azobenzene polyester films in passed ten or more years[4,5]. Because of the characteristics such as high diffraction efficiency, high exposure sensitivity, high resolution, long shelf life, dry real-time processing, and low cost, holographic photopolymer has attracted increasing interest in the study of high-density digital holographic storage[6,7]. It is well known that high-density digital holographic storage is to record many digital holographic pages in the same location in recording medium by various kinds of multiplexing methods: angular multiplexing, wavelength multiplexing, phase multiplexing etc.[8,9], and no matter for what kind of multiplexing method, the recording number N_s of the holograms in the same location in medium is proportional to the thickness of the medium. So, the recording capacity and density of digital holographic storage are proportional to N_s or the thickness of recording medium. But it is difficult to get the thick photopolymer that is more than 500 μm thickness, and on account of the real optical setup of the digital holographic storage, for such photopolymer recording medium, it is too difficult to get ideal information capacity and density by only one kind of multiplexing method with one exposure wavelength[10], we find that the ideal information capacity and density can be given if multi-wavelength is adopted to record the digital holograms. This like but not is the wavelength multiplexing because it is not necessary to the tunable laser source but only a multi-wavelength radiating one. Certainly, the recording photopolymer must be sensitivity to these laser wavelengths. In present work, a novel wideband sensitive holographic photopolymer that sensitized by two kinds of dyes methylene blue (MB) and rose bengal (RB) is fabricated, and the holographic characteristic of it is studied.

The photopolymer is fabricated with the following steps, in dark room, under 40%—60% relative humidity, and 20—25 °C temperature. 1) 2-g polyvinyl alcohol (PVA, Shanghai Reagent Corporation and molecular weight $\approx$ 2500) was put into distilled water and heated to 80 °C, and mixed round it till the PVA dissolved completely to get 10 wt.-% PVA solution. 2) 0.6-g acrylamide (AA, chemical pure, Shanghai Reagent Corporation) and 0.2-g N,N'-methylenebisacrylamide (BAA, chemical pure, Shanghai Reagent Corporation) were dissolved into appropriate mount of distilled water and heated to 30 °C, then put into proper volume of triethanolamine (TEA) aqueous solution and stirred it well with a magnetic stirring device to get a homogeneous solution. 3) Some MB and RB were put into distilled water respectively to get appropriate density dye solutions, mixed the two dye solutions in the proportion of 2 : 1 and stirred it well to get the final dye solution. 4) Put all the monomer solution and 1-mL final dye solution into the PVA solution, a 25-mL photopolymeric solution was got. The basic concentrations of all reagents are: PVA 10%, TEA 0.2 mol/L, AA 0.34 mol/L, BAA 0.052 mol/L, MB 2.4×10^{-4} mol/L, and RB 1.2×10^{-4} mol/L. Appropriate volume of the photopolymeric solution was poured onto a $60\times60\times1$ mm^3 glass substrate and dried for about 24 to 72 hours, then the experimental photopolymer dry film is prepared. The film thickness is about 240 μm.

In Fig. 1, the solid curve is the absorption spectrum of the photopolymer sample, and the two dashed curves are the absorption spectra of two dyes MB and RB respectively. We can see that the absorption spectrum of the film is almost the superposition of the two dyes MB and RB except a little bit shift of the absorption peaks. It indicates that there is no new matter created during the fabricating of the photopolymer sample. The figure shows that the sensitive band of the photopolymer is widened from about 100 nm for the sample that sensitized by only single dye to more than 200 nm for sample

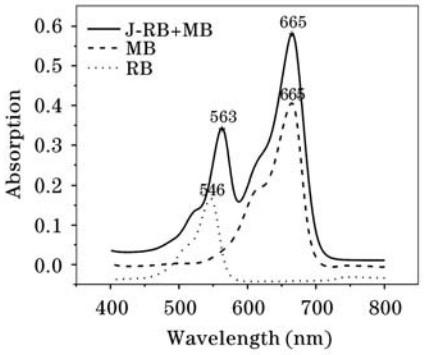

Fig. 1. Absorption spectra of the aqueous solutions of RB ($\lambda_{max} = 546$ nm), MB ($\lambda_{max} = 665$ nm), and the photopolymer film J-RB+MB ($\lambda_{max} = 665$ nm).

that sensitized by two dyes. The absorption spectrum of the photopolymer covers the range from 450 to 700 nm in the visible light wavelength band, and it is especially appropriate to expose with the convenient laser wave such as He-Ne laser (633 nm), Ar^+ laser (514.5, 496.5, 488 nm), etc.. We know that if the digital holographic storage disk is processing with these 4 or more wavelengths, its capacity will be more than 1 Tb[10].

Figure 2 is the optical experimental setup, a 90° setup is adopted to record unslanted grating in the photopolymer sample. M_1, M_2, M_3, and M_4 are mirrors, L_1 and L_2 lenses, L_3 and L_4 are Fourier lenses with the focus length f, SH_1, SH_2, and SH_3 are shutters, PM is the power meter, O is the object to be recorded, H is the photopolymer sample, and CCD is the charge couple device.

When we measure the exposure characteristic curves (diffraction efficiency versus exposure energy or exposure time), remove all the lenses and O, the beam from M_4 is object beam, and the beam from M_3 is reference beam; CCD is replaced by PM, shut off SH_3 and read the first level diffraction power on PM every one second. The diffraction efficiency is defined as the ratio of the first level effective diffractive power to the power of reading reference beam. When we measure the transmittance curves (transmittance versus incident time), shut off SH_3, only the beam from M_3 passes through the sample, read a transmissive power on PM every one second. The transmittance is defined as the ratio of the transmissive power to the power of incident beam.

Figure 3 shows the exposure characteristic curves with four exposure wavelengths: 632.8, 514.5, 496.5, and 488 nm, the total exposure intensity is about 21.2 mW/cm^2.

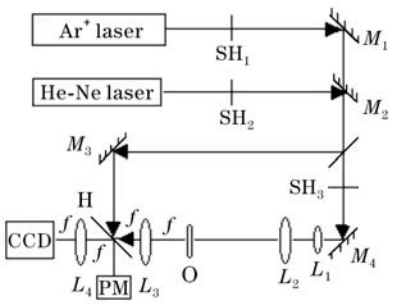

Fig. 2. Optical experimental setup.

From the figure, we can see that no matter which exposure wavelength is adopt, the diffraction efficiency increases with the increase of the exposure time at first, then it reach its maximum value, all the maximum diffraction efficiencies are more than 40% with four exposure wavelengths, and at last the diffraction efficiency saturates at a little bit low level. The reason is that with the increase of the exposure time, the polymerized monomers increase, then the refractive index modulation increases, so the diffraction efficiency increases with the increase of the exposure time[6]. But when the exposure time gets farther more, the scattering light in the photopolymer induced by the optical fault of the film gets large enough to result in the polymerization of the residual monomers in the dark region, this decreases the refractive index modulation in the sample, so the diffraction efficiency has a little bit decrease, and at last when all the monomers in the sample are polymerized, the diffraction efficiency gets its saturation value.

Because of the different absorptions of the different exposure wavelengths (in Fig. 1), the maximum diffraction efficiency decreases with the decrease of exposure wavelength at the same exposure time, as shown in Fig. 3. We can see that from Fig. 1 for four different exposure wavelengths, the absorption intensity decreases with the decrease of the exposure wavelength, so for same exposure time, there are less monomers polymerized with shorter exposure wavelength, its refractive index modulation is smaller than that with the longer exposure wavelength, and the maximum diffraction efficiency is smaller than that with the longer exposure wavelength.

We know that the exposure sensitivity is an important parameter of the holographic medium, its definition is the reciprocal of the exposure energy on unit area when the diffraction efficiency reaches or closes to its maximum value. Under this definition, we can see from Fig. 3 that the exposure time of four different exposure wavelengths are almost the same (6—7 s), so the exposure sensitivity is the same for different exposure wavelengths, it seems in contradiction to the general understanding. In general, because of the less absorption with shorter exposure wavelength, the more monomers are residual in the dark area in the interference field, with the increase of the exposure time, these monomers diffuse to the bright area from dark area, more power is need to be absorbed to make them polymerization, so the exposure sensitivity should be smaller than that of the longer wavelength exposure. The reason of this special phenomenon may be also the scattering.

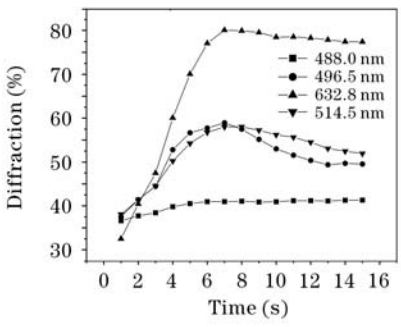

Fig. 3. Diffraction efficiency versus exposure time under four different exposure wavelengths.

It is easy to be understood from Figs. 1 and 4. Figure 4 is the transmittance versus exposure time curves with four different wavelengths, we can see that the shorter the exposure wavelength, the less the transmittance and absorption. Where does the power of the incident light go? The most possible explanation may be also the scattering: the shorter the exposure wavelength, the more the scattering power, this is just consistent with the classical scattering theory. When the exposure wavelength is longer (632.8 nm), there is less scattering in the sample, almost all the monomers in dark area diffuse to bright area to be polymerized, so the refractive index modulation is larger, and the maximum diffraction efficiency is larger (80%). But when the exposure wavelength is shorter (514, 496.5, 488 nm), there is much light scattering into the dark area, it makes the polymerization of some monomers in the dark area in the interference field, and only the un-polymerized monomers in the dark area diffuse to bright area to be polymerized. When the exposure time is long enough, all the monomers both in the bright area and dark area in the interference field are polymerized. So there is no necessary more power to make the monomers polymerized, the maximum refractive index modulation is smaller, the maximum diffraction efficiency is smaller (< 80%), and the exposure sensitivity is not smaller than that of the longer exposure wavelength just like that described above.

From Fig. 4 we can see that for 632.8-nm exposure wavelength the transmittance increases with the increase of the exposure time, and it reaches a saturation value at final, this is because that there is almost no scattering during the exposure, with the increase of the exposure time the more and more dye molecules are bleached and the transmittance gets saturation till all the dye molecules are bleached. But for 514.5-, 496.5-, and 488-nm exposure wavelength, the transmittance increases with the increase of the exposure time at the first several seconds, then it decreases with the increase of the exposure time, this is just the result of the scattering described above. At first, with the bleaching of dye molecules, transmittance increases with the exposure time, the scattering is a secondary problem, but with the increase of the exposure time the scattering is getting more and more important, more monomers in the dark area are polymerized, these random distributing polymers act the new scattering centers and enhance the scattering, so the transmittance is getting small and small.

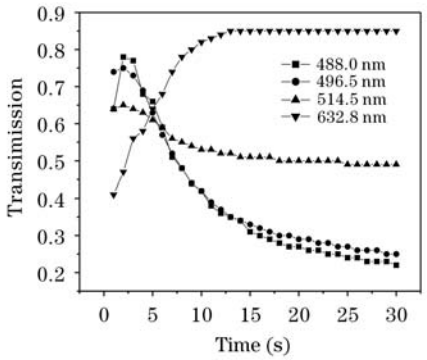

Fig. 4. Transmittance versus exposure time.

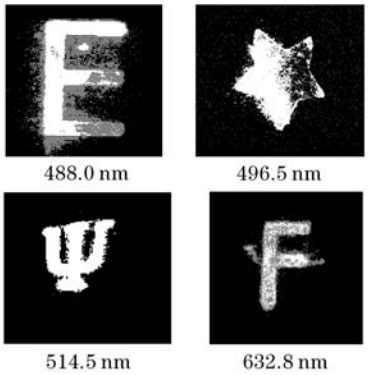

Fig. 5. Reconstruction images at different wavelengths.

Figure 5 shows the reconstructed holograms recorded with the four wavelengths in the same location in sample. The imperfect of photo pentacle is the result of fault of light spot irradiating on the object. There is almost no crosstalk in four reconstructed holograms, indicating that this wideband sensitive photopolymer is appropriate for multi-wavelength holographic storage.

In conclusion, the sensitive spectra of the photopolymer sample sensitized by two dyes MB and RB are widened from about 100 to 200 nm because of the co-sensitization of two kinds of dyes. The maximum diffraction efficiency of every exposure wavelength is more than 40%, and decreases with the decrease of exposure wavelength because of the scattering of the shorter exposure wavelength, but the exposure sensitivity does not change with the exposure wavelength for the same reason. The analysis indicated that the scattering acts an important role in the process of holographic recording, and it is not good for the increasing of the exposure sensitivity and maximum diffraction efficiency or information capacity. The high quality reconstructed holograms recorded with different exposure wavelengths at the same location on the sample showed that the sample has the potential for multi-wavelength holographic storage.

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