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ABSTRACT

The development of a panchromatic acrylamide based photopolymer (ABP) for holographic recording application is presented. The scattering of the recording medium was characterised by measuring the Bidirectional Scattering Distribution Function (BSDF). The dynamic range in reflection mode of recording was evaluated by measuring the diffraction efficiencies of the holographic gratings recorded individually at 633nm, 532nm, and 473nm wavelengths at spatial frequencies of 4200 l/mm, 5000 l/mm and 5700 l/mm respectively. Spectral characterisation of the reflection gratings recorded using a combined single RGB beam was carried out and the reconstructed wavelengths were monitored and compared with the recording wavelengths. The recorded and the reconstructed wavelengths were plotted as points on the CIE chromaticity diagram in order to reveal the shifts due to material shrinkage in the corresponding RGB wavelengths. Finally reflection holograms of an object were successfully recorded at all the three

primary wavelengths. The results represent a strong confirmation that this acrylamide based photopolymer can be used as a panchromatic recording material and can be employed in future commercial holographic applications.

1. INTRODUCTION

The first report on the multicolour holographic images obtained by superposition of three holograms each recorded at one of three primary wavelengths appeared in 1964 [1]. Until now both silver halide photographic emulsions (SHPE) [2] and dichromated gelatin (DCG) [3] are the most common materials used for high efficiency full colour reflection hologram recording. However, these materials require wet chemical processing for developing the holograms which is laborious and costly from the point of commercial applications. Self-developing photopolymers [4-6] which do not require development are the ideal choice for real-time recording and reconstruction of holograms.

In the present paper for the first time we report on the sensitising and quantitative and qualitative analyses of an ABP for recording multicolour holographic gratings. Holograms of a real object were also recorded using this material. ABP material was sensitised for recording reflection holographic gratings and holograms at each of three primary wavelengths. Qualitative analysis of the scattering properties of ABP was carried out by measuring the BSDF. Quantitative studies of reflection holographic gratings were carried out by measuring the diffraction efficiency and performing spectral characterisation. For the latter purpose, the chromaticity points of the recorded wavefronts and the wavefronts reconstructed from the hologram were compared using the CIE diagram. The results and the potential issues related to shrinkage in the photopolymer

during holographic recording and its effects on the reconstructed wavelengths are discussed. Reflection holograms were recorded individually at three primary wavelengths on a single photopolymer layer.

2. EXPERIMENTAL

2.1 Recording wavelengths and recording material

The wavelengths used for holographic recording were selected from the CIE chromaticity chart [7] which provides a simple procedure for predicting the range of colours that can be obtained from a mixture of the three primary wavelengths. Figure 1 shows the CIE chromaticity chart. The range of colours that could be reconstructed lies within a triangle whose vertices are the points corresponding to the recording wavelengths. A wide spectral range of colours means that a large area on the CIE diagram could be covered by this triangle. A greater area on the CIE diagram can be covered by a polygon or any other irregular shape formed by using a greater number of spectral wavelengths. However, the use of more spectral wavelengths complicates the holographic recording setup and is constrained by the availability of continuous wave lasers [8].
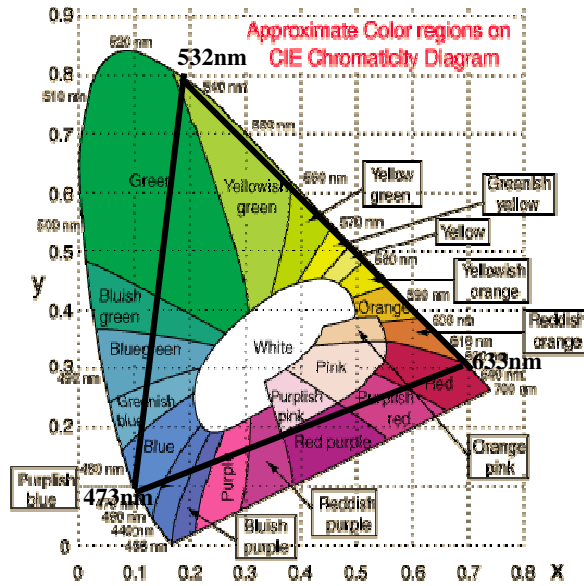
Figure 1 CIE chromaticity diagram [4].

The recording material used in these experiments is an acrylamide based photopolymer (ABP) developed at the Centre for Industrial and Engineering Optics. The photopolymer consists of acrylamide (monomer), N-N methylene bisacrylamide (cross linking co-monomer), polyvinyl alcohol (binder), triethanolamine (electron donor or co-initiator) and primary wavelength sensitive photo initiator dyes. The photopolymer is sensitized using methylene blue (MB), erythrosine B (EB) and acriflavine (ACF) dye to record at red, green and blue wavelengths respectively. The photopolymer composition is given in table 1.

Table 1 Photopolymer composition

10% w/v PVA aqueous solution	Acrylamide	NN'methylene bisacrylamide	Triethanolamine	0.11% w/v of MB dye	0.11% w/v of EB dye	0.11% w/v of ACF dye
17.5 ml	0.8 g	0.25 g	2 ml	3 ml	3 ml	3 ml

The mechanism of hologram recording was explained previously [9-12].

The photosensitive layers were prepared by gravity settling coating technique. 600µl of photopolymer solution was deposited on a 2.5cm x 7cm clear glass slide using a microliter pipette. Samples were left for drying for 6 to 7 hours in a dark room at ambient conditions. The thickness of the layers after drying was 60±5µm when measured using a white light surface profilometer ADE Phase Shift MicroXAM 100. The normalised absorption spectrum of the dry layer was measured using a Perkin-Elmer Lambda 900 UV-VIS-NIR absorption spectrometer and is shown in figure 2. From this spectrum it can be observed that the material has well defined peaks of absorption at 473nm, 532nm and also near 633nm wavelengths. It also shows that no interaction between the different dye molecules takes place as the absorption spectrum of each species is clearly distinguishable.
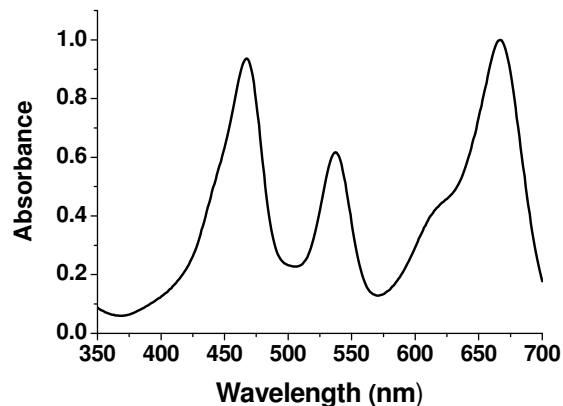

Figure 2 Normalised absorption spectrum of panchromatic acrylamide based photopolymer recording material.

2.2. Experimental set up for BSDF

An important characteristic of the holographic recording material is its low scattering property which can be measured as BSDF. BSDF is the ratio of the intensity of the scattered light $P_{scatter}$ (as function of angle θ) to the incident intensity $P_{incident}$ per unit solid angle $\Omega_{detector}$ as shown in the equation 1 [13, 14] and is measured using the scatterometer shown in figure 3.

$$BSDF(\theta) = \frac{P_{scatter}(\theta)}{P_{incident}\,\Omega_{detector}} \tag{1}$$

The BSDF was obtained at two different wavelengths - 473nm and 633nm individually so as to cover a wide range of the visible region. This BSDF was obtained by measuring the scattered light intensity as a function of angle using laser wavelengths (at 473nm and 633nm) by illuminating the sample with a converging beam which was spatially filtered. The intensity of the incident light on the sample placed at the goniometer axis was 0.8 µW and 0.65 µW for blue and red wavelengths respectively. The spot radius of the laser beam on the sample was 1 cm. The detector (Newport 1830C Optical Power Meter) was placed on the moving arm to measure the angular dependence of the intensity. The detector signal and goniometer arm were controlled by a LabView programme. The distance from sample to the detector was 24.5 cm and an aperture of 1mm diameter was placed in front of detector. This complete scatterometer setup was enclosed in a black box to minimize stray light reaching the detector.
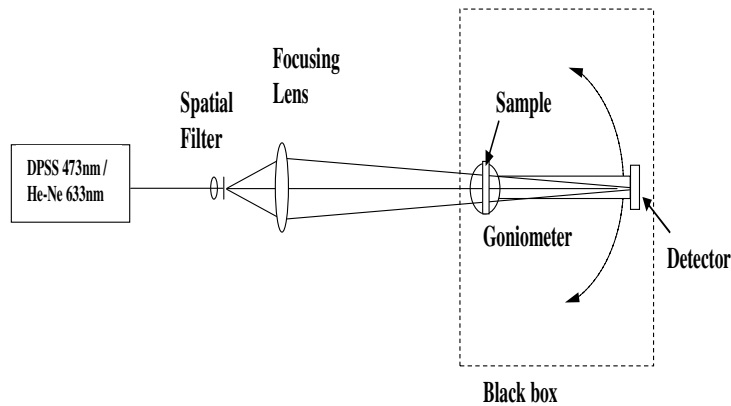
Figure 3 Schematic diagram of scatterometer to measure BSDF.

2.3. Experimental set up for recording reflection gratings

Reflection gratings were recorded in the photopolymer layers sensitized to three wavelengths red, green and blue using a symmetrical geometry shown in Figure 4. It consists of a He-Ne laser of wavelength 633 nm and two diode pumped solid state (DPSS) lasers (532nm and 473 nm). These three laser beams were combined using a mirror and two dichroic mirrors for green and blue wavelengths. The combined laser beams were spatially filtered using a single spatial filter and then collimated. The collimated beam was split into two beams using a beam splitter. The two beams were then diverted onto the photopolymer layer using two adjustable mirrors to record the holographic gratings.
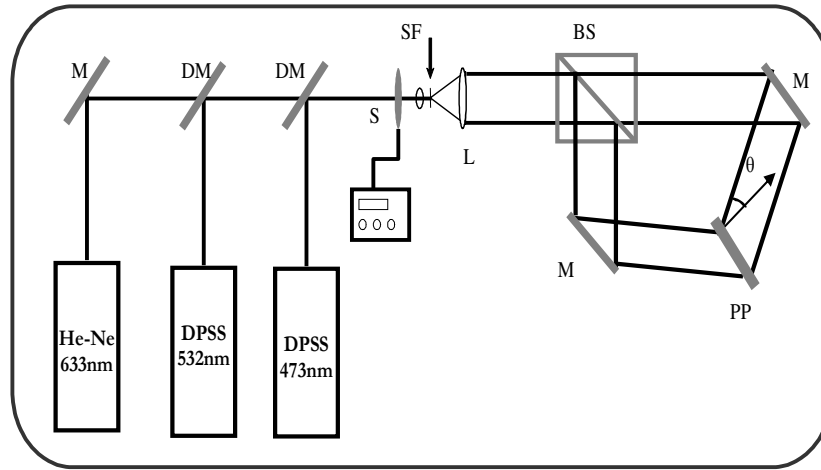
Figure 4 M-mirror, DM-dichroic mirror, SP-spatial filter, S-shutter, L-collimating lens, BS-beam splitter, PP-photopolymer layer.

Spatial frequencies were selected to ensure high spatial frequency recordings, and were calculated using the Bragg diffraction equation (equation 2).

$$2n\Lambda \sin \theta_B = \lambda \qquad (2)$$

Where λ - wavelength of the recording light, $n=1.51$ (average refractive index of the recording material), Λ is the fringe spacing and θ_B *is the* inter beam angle. This inter beam angle inside the photopolymer layer can be determined using snell's law. In our experiment, the incident angle 40° was chosen to illuminate the photopolymer layer outside and this angle corresponds to half of the recording inter beam angle which is $\theta_B=64.8°$ inside the photopolymer layer. This value corresponds to spatial frequencies 4200 l/mm, 5000 l/mm and 5700 l/mm for the wavelengths of 633nm, 532nm, and 473nm respectively.

The photopolymer layers were exposed for different exposure times ranging from 20s to 120 sec with an increment of 20 seconds for each new recording. The diffraction efficiencies of each reflection grating were then measured by exposing the grating to one of the recording beams. The intensity in the diffracted order was measured using a photo detector. The experiments were repeated for the green (532nm) and blue (473nm) wavelengths. The recording intensities for the red, green and blue wavelengths were 1.8mW/cm^2, 3mW/cm^2 and 4mW/cm^2 respectively. The percentage diffraction efficiency, η, was calculated using equation 3.

$$\eta = \frac{I_{1st}}{I_{incident}} \times 100 \qquad (3)$$

Where I_{1st} is 1st order intensity and $I_{incident}$ is incident intensity. The results are shown in figure 6. The Fresnel loss was calculated to be $\approx$15% for the incident angle of 40° at all three primary wavelengths.

2.4. Experimental set up for spectral characterisation of multicolour reflection grating

We have characterised the multicolour reflection holographic gratings recorded in our panchromatic photopolymer by exposing the material for 80 sec to the combined RGB beam with an intensity of 1.8mW/cm^2, 3mW/cm^2 and 4mW/cm^2 at 633nm, 532nm, and 473nm wavelengths respectively. The spectral response curve is shown in figure 8. The spectral characteristics of the recorded reflection gratings were studied by probing them with a halogen white light of 5 mm diameter (Avantes, AvaLight-HAL [15]) coupled into a 400µm optical fibre and collimated with a collimating lens. The reconstructed light from the grating was coupled into a second optical fibre connected to an Avantes AvaSpec-2048 spectrum analyser. The set up is shown in figure 5.

The angle of incidence of the reconstructed beam is approximately the same as the recording beam.
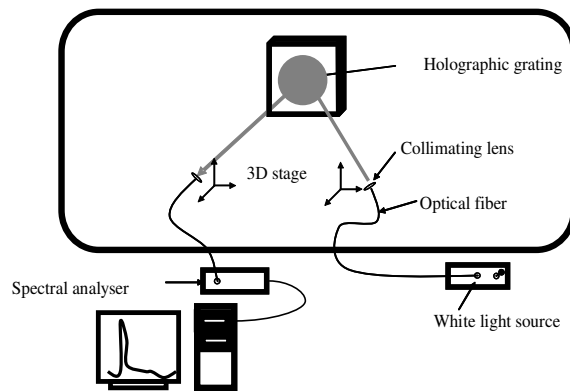

Figure 5 Experimental setup used for spectral characterisation of the recorded reflection holograms/gratings.

3. RESULTS

3.1 Measurement of BSDF

The BSDF of the air, glass and uniformly polymerised layer at 473nm are shown in figure 6. From the graph it can be observed that the scattering of the uniformly polymerised layer is close to that of the glass and air signals which indicates that the material is characterised by very low scattering. The inset of figure 6 shows the BSDF data of the uniformly polymerised layer measured at 633nm and 473nm. As expected the scatter is greater at the shorter wavelength of 473nm than the higher wavelength at 633nm. From these experiments it can be concluded that the total scattering of the layers with thickness of 60µm is very low and negligible in the present composition of ABP. This is because the photopolymer material is grain less and soft unlike

silver halide photographic emulsions. From the above results we believe that this photopolymer material is suitable reflection holographic recording.
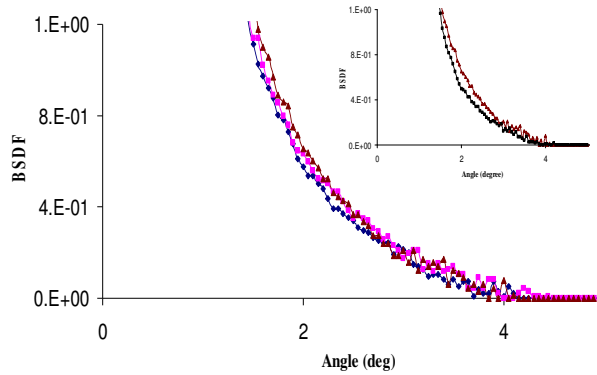

Figure 6 BSDF data for the air (blue circle), glass (pink square) and uniformly polymerised layer (brown triangle) measured at 473nm. The inset picture shows the BSDF of the uniformly polymerised photopolymer layer measured at 633nm (black square) and 473nm (brown triangle).

3.2 Measurement of Diffraction efficiency of Reflection gratings

The total recording intensity of the two interfering beams was 1.8mW/cm^2 at 633nm. From figure 7 it can be observed that, as exposure time increases, diffraction efficiency increases. The recording time for maximum diffraction efficiency for the three wavelengths is between 80 and 100s. However, the maximum diffraction efficiencies are different for the three wavelengths. The maximum diffraction efficiency for the gratings recorded at 633nm, 532nm and 473nm wavelengths are 11.5%, 6%, and 1.6% respectively. This is at least partly due to the fact that the spatial frequency increases as the wavelength decreases. Another possible reason could be that the quantum yield of generation of free radicals is different for the three dyes which is an

important parameter which drives the polymerization process and results in a refractive index modulation between the bright and dark regions.
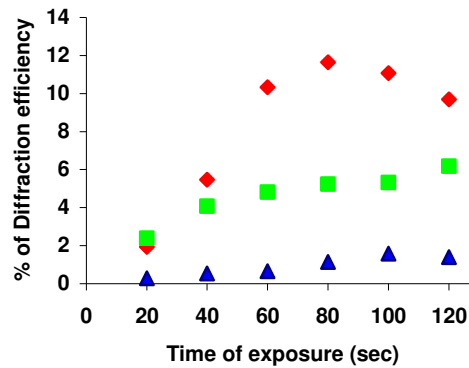

Figure 7 Diffraction efficiency (%) of reflection gratings as a function of time of exposure at red, green and blue wavelengths.

3.3 Spectral Characterisation of Multicolour reflection grating

One of the disadvantages of most holographic recording materials is that they often undergo dimensional changes either during recording or during any post exposure treatment. These dimensional changes would some times lead to a decrease or increase in the diffraction efficiency, deviation from the Bragg angle, shift in the reconstruction wavelength etc. These can cause serious problems in display holography and holographic data storage. Figure 8 shows the spectral characterisation of the ABP layer done using the setup shown in figure 5 (experimental section 2.3). The recording and reconstruction wavelengths were compared and are shown in table 2
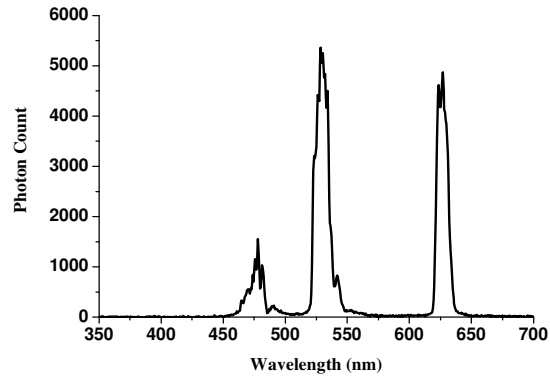
Figure 8 Spectral characteristics of the reconstructed wavefront from the reflection holographic grating recorded using 473nm, 532nm and 633nm.

Table 2 shows the recording wavelengths and the corresponding reconstruction wavelengths.

Recording wavelength (nm)	Reconstruction wavelength (nm)
633	626.3
532	529.2
473	478.17

From the results of the spectral characterisation studies it is evident that the panchromatic acrylamide based photopolymer recording material is suitable for recording reflection gratings at all the three primary wavelengths. The spectral response curve (figure 8) shows that reconstructed wavelengths of the reflection gratings recorded at 633nm, 532nm and 473nm are at 626nm, 529nm and 478nm respectively. The colour triangles plotted on the CIE chromaticity diagram by joining the chromaticity points for these recorded and reconstructed wavelengths are shown in figure 9. It is observed from these preliminary results that the reconstructed wavelengths were shifted towards shorter wavelengths due to the shrinkage (approx. 1.11%) for

red and green wavelengths. Surprisingly we observed a shift towards longer wavelength at blue wavelength (approx 1.1 %) which is speculated to be due to the swelling.
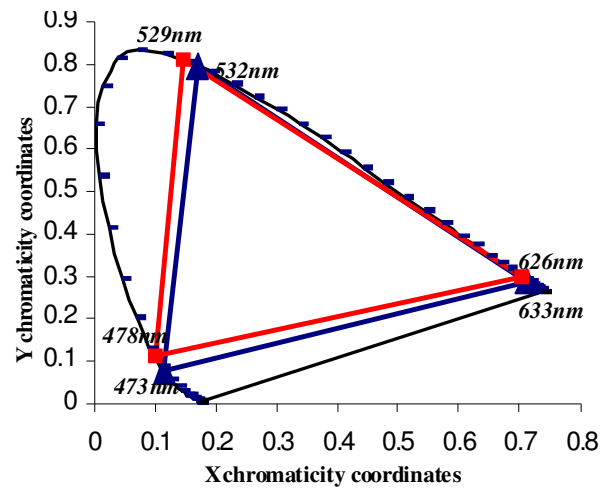

Figure 9 Comparison of the chromaticity points for the recorded and reconstructed wavelengths of the multicolour holographic grating.

To demonstrate the use of this panchromatic photopolymer for display holography application, reflection holograms of a 10 euro cent coin were recorded at 633nm, 473nm, and 532nm wavelengths at three different spots(for each wavelength) on the same 60µm thick photopolymer layer using Denisyuk single beam geometry.

Photographs of the reconstructed images are shown in figure 10.

Figure 10 Photograph of white light holographic reconstruction from a hologram recorded at the three primary wavelengths. The object used was a 10 euro cent coin.

CONCLUSION

An acrylamide based photopolymer holographic recording material was sensitized at red, green and blue primary wavelengths by using three photosensitive dyes. BSDF of the photo polymer recording material was measured and the scattering of the material was found to be very low and so can be efficiently used for holographic recording applications. Reflection gratings that were recorded at 4200 l/mm, 5000 l/mm and 5700 l/mm showed maximum diffraction efficiencies of 11.5%, 6%, and 1.6% at 633nm, 532nm, and 473nm respectively. Spectral properties of the multicolour holographic gratings recorded in the same location in the photopolymer layer with the combined RGB beam were characterised using a spectrum analyser. The recorded and reconstructed wavelengths were compared in the CIE chromaticity diagram. Results show small spectral shifts in the reconstruction wavelengths which can be attributed to the swelling/shrinking of the photopolymer. Individual primary wavelength reflection holograms were successfully recorded and reconstructed in a single panchromatic photopolymer layer. Although the work, to produce full colour hologram combining all the three primary beams in one single light source is at the embryonic stage, dedicated research is still being carried out to improve the diffraction

efficiency of the holograms by optimising the composition of the recording material (dye concentrations, monomers etc) and exposure conditions and also doping the photopolymer with specific nanoparticles to minimize if not completely suppress the dimensional changes. Hence the results presented in this paper provide substantial evidence that using the ABP for panchromatic holographic recording display applications is feasible.

Acknowledgements

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