Optical Materials 52 (2016) 212–218

Contents lists available at ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

High efficiency panchromatic photopolymer recording material for holographic data storage systems

Vayalamkuzhi Pramitha [a], Bhargab Das [b], Joby Joseph [c], Rani Joseph [d], Krishnapillai Sreekumar [e], Cheranellore Sudha Kartha [a,*]

[a] Department of Physics, Cochin University of Science and Technology, Cochin 682022, India
[b] Advanced Materials and Sensors Division, CSIR-Central Scientific Instruments Organization, Chandigarh 160030, India
[c] Department of Physics, Indian Institute of Technology Delhi, New Delhi 110016, India
[d] Department of Polymer Science & Rubber Technology, Cochin University of Science and Technology, Cochin 682022, India
[e] Department of Applied Chemistry, Cochin University of Science and Technology, Cochin 682022, India

ARTICLE INFO

Article history:
Received 26 November 2015
Received in revised form 24 December 2015
Accepted 27 December 2015
Available online 4 January 2016

Keywords:
Holographic gratings
Diffraction efficiency
Photopolymer recording media
Holographic data storage

ABSTRACT

Studies carried out to gauge the potential of a metal-ion doped panchromatic photopolymer recording material for application in real-time holographic data storage is presented. The photopolymer films are spin coated on glass plates to ensure better surface uniformity. Volume holographic transmission gratings with peak diffraction efficiency of 80% could be stored in the photopolymer films of 100 μm thickness. An efficiency of 70% is achievable even for gratings recorded with exposure energy as low as 10 mJ/cm^2. A checkerboard pattern data page recorded in the photopolymer film using a defocused 4-*f* recording geometry could be reconstructed with good image quality. The experimental results illustrate the competency of the developed photopolymer for holographic data storage applications.

1. Introduction

Volume holography is considered as a promising candidate for future storage technologies because of its high storage capacity and fast data-write and read rates [1–3]. The development of suitable recording materials has been one of the primary challenges in the advancement of holographic data storage (HDS) systems. HDS can fulfil its promise as a universal storage technology only when highly efficient, high capacity, highly stable and cost-effective recording media will become available. Even though a wide variety of recording media have been developed as holographic data storage media, most of them have some drawback that prevent their use in practical holographic systems. Compared to other recording media, photopolymers are potential candidates for write-once-read-many times (WORM) data storage applications due to characteristics such as good light sensitivity, real-time image development, large dynamic range, good image stability and relatively low cost [4–17]. Photopolymers based on olygoether(meth)-acrylates are promising on account of their advantages such as better optical transparency and acousto-optical properties [18]. The self-processing dye sensitized acrylamide-based photopolymer systems have attracted great deal of attention because of their high diffraction efficiency (DE) and low cost. Incorporation of different additives into these films was attempted for improving their storage lives, of which, incorporation of metal ions was reported to be the best, since metal ions formed ionic crosslinks with the poly (vinyl alcohol) (PVA) matrix, thereby inhibiting the diffusion of unreacted monomers and stabilizing the recorded grating [19–24]. Metal ions such as chromium (Cr^{6+}), ferric (Fe^{3+}) and cupric (Cu^{2+}) have been doped into polymer matrices such as poly(vinyl alcohol) and poly(acrylic acid) to realize various holographic recordings.

We had earlier introduced silver doped photopolymer recording material as a promising candidate for holographic applications [25]. Gratings recorded in these films could be stored for years with good diffraction efficiency. The panchromaticity of the silver doped photopolymer and its suitability to record multiple gratings in a 100 μm thick film was also successfully demonstrated [26,27]. Even though the dye sensitized acrylamide-based photopolymers have been widely studied for data storage applications, the metal-ion doped acrylamide-based systems remain less explored. Moreover, most of the reported works have evaluated the capability of a thick photopolymer film (500 μm or approximately 1 mm) for holographic storage [11–15]. Through this paper, we report the potential of our 100 μm thick panchromatic silver-doped

* Corresponding author.
E-mail address: csk@cusat.ac.in (C.S. Kartha).

http://dx.doi.org/10.1016/j.optmat.2015.12.042
0925-3467/

photopolymer material for actual data storage applications. Silver-doped photopolymer films prepared by gravity settling method exhibited a diffraction efficiency of 75% for exposure energy of 80 mJ/cm^2 [25,26]. Surface analysis of these films revealed non-uniformity in thickness especially near the edges. Since uniform thickness over the entire film surface is essential for practical holographic applications, films were prepared by spin coating, as it is an excellent technique for preparing uniform films over a large area. Transmission holographic gratings were recorded in the photopolymer films with varied concentrations to optimize the composition. Efforts were made to study the stability of the recording material as well as that of the stored gratings. The effect of sealing the film on the shelf life of the material and the storage life of the holographic gratings was studied. The photopolymer films with optimized composition, shelf-life, storage-life and stability were then tested for holographic data storage applications. Even though a 4-*f* architecture with the recording medium placed at the Fourier plane of an objective lens is one of the most commonly used geometries for recording data pages [28,29], a defocused 4-*f* HDS system was employed in the present work to record the image of a checkerboard pattern data page in the photopolymer film. This is because, for an amplitude modulated data page, a high-intensity DC peak (or DC spot) occurs in the Fourier plane. Defocusing the recording material away from the Fourier plane distribute light more evenly over the aperture of the hologram leading to high fidelity recording. The paper is divided into two main sections. The first part discusses the spectral and energetic sensitivity of the films, optimization of the film composition, stability of recorded gratings, aging process and effect of sealing the film. This is followed by the recording of holographic data page in the film.

2. Materials and methods

The photopolymer film consists of poly(vinyl alcohol) (PVA) (binder or the host matrix), methylene blue (MB) (photosensitizing dye), triethanolamine (TEA) (electron donor), acrylamide (AA) (monomer) and silver nitrate ($AgNO_3$) (crosslinker). 15% PVA solution was prepared by dissolving 15 g of PVA (molecular weight 1, 25,000) in 100 ml distilled water. Acrylamide crystals were dissolved in PVA solution and the resulting solution was sensitized with methylene blue and triethanolamine. The mixture was then stirred well to get a homogenous solution. To this solution, silver nitrate solution was added and stirred again. A fixed volume of photopolymer solution (6 ml) was deposited on the glass substrate (6 cm × 6 cm × 2 mm) kept over the vacuum chuck of the spin coater) (SPIN 150, SPS Europe). Films were prepared for different spin speeds for 2 s. After the spin process, the films were taken carefully from the chuck and kept on a levelled glass tray for drying. After the films were dried, thickness was measured using stylus profiler (Dektak 6 m). Films with 100 μm thickness were used for further studies. Films were prepared by varying the concentration of the constituents. Keeping PVA as 15% w/v, acrylamide concentration was varied between 0.47 and 0.9 M, methylene blue was varied between 0.014 and 0.06 mM, TEA was varied between 0.05 and 1 M and $AgNO_3$ concentration was varied between 0.05 and 0.09 mM. The surface morphology of the photopolymer film before and after recording of gratings was observed by an atomic force microscope (AFM) (JEOL, JSPM 5200). The fabricated films were found to be highly uniform. Spectral and energetic sensitivity of the films were determined by optical absorption and real-time transmittance studies. UV–Visible-Near-IR spectrophotometer (JASCO V-570) was used for recording the optical absorption spectra of the films as shown in Fig. 1 wherein the panchromaticity of the photopolymer film is clearly depicted [26].

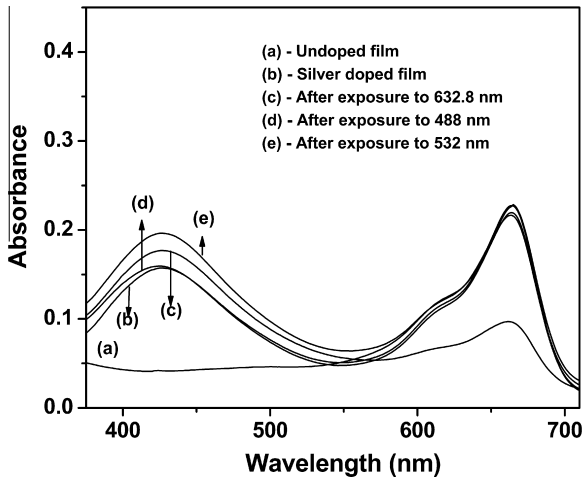

Fig. 1. Absorption Spectra of the photopolymer film; (a) undoped film; (b) silver doped film; (c) silver doped film exposed to 632.8 nm; (d) silver doped film exposed to 488 nm; and (e) silver doped film exposed to 532 nm.

The transmittance characteristics of the photopolymer film upon laser exposure was monitored by illuminating the film to laser beam of irradiance 1 mW/cm^2 for four minutes and monitoring the transmitted power at regular intervals using an optical power meter. Laser outputs from He–Ne laser (632.8 nm, Melles Griot, CW), Ar^+ laser (488 nm, Melles Griot, CW) and frequency-doubled Nd:YAG Laser (532 nm, Compass 215M-20, Coherent, CW) were used for the measurements. The relative transmittance T/T_0 was also determined, where T is the real-time transmittance of silver doped films and T_0 is the transmittance of the PVA/AA films without dye and silver nitrate. It was observed that the transmittance increases even for low exposure energies, indicating the good energetic sensitivity of the films. This is because, even at low exposure, the dye molecules (MB) were getting excited and got converted to the leucoform. The variation of relative transmittance with laser exposure was reported earlier [26]. The transmittance vs. exposure energy plot showed that irradiation using longer wavelength (632.8 nm) resulted in an increase in transmittance with exposure time, and reached a final saturation value when all the dye molecules are bleached. However, on irradiation with 488 nm and 532 nm lasers, the relative transmittance was found to be decreasing for higher exposure energies. This can be attributed to scattering in the film, since the shorter the exposure wavelength, the more is the scattering power, consistent with the classical scattering theory [26,30]. Diffraction efficiency analysis was then carried out by recording transmission gratings in the film by interferometric technique as discussed elsewhere [25]. The recorded grating was illuminated by laser beam and the diffracted beam power was measured by an optical power meter positioned at Bragg's angle.

3. Results and discussions

In order to determine the concentration of each constituent required for achieving maximum DE, transmission gratings were recorded in the films using a 632.8 nm He–Ne laser. The interbeam angle was 40°, total recording power was 4 mW/cm^2 and intensity ratio of the recording beams was 1. The exposure energy was varied between 5 mJ/cm^2 and 150 mJ/cm^2. Gratings were reconstructed using the same He–Ne laser and diffraction efficiency values were calculated. The effect of concentration variation of each constituent on the diffraction efficiency of the recorded grating was explained in detail in our previous papers and hence was