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NANOPARTICLE DOPED PHOTOPOLYMER WITH REDUCED SHRINKAGE FOR HOLOGRAPHIC RECORDING

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Abstract: We demonstrate holographic recording with Si-MFI zeolite doped acrylamide based photopolymer film. The influence of silicate nanoparticles on photopolymer shrinkage has been studied and compared with shrinkage in undoped photopolymer layers. The shrinkage of the material (1.03%) is significantly lower for recording in Si-MFI zeolite nanoparticle doped samples than for undoped layers (1.77%).

1. INTRODUCTION

Photopolymers are a good prospect for the manufacture of holographic diffractive elements, as media for holographic data storage, holographic sensors and security holograms [1-3] due to their high sensitivity, high spatial frequency response and the absence of any need of processing [3, 5]. The material investigated in this study is an acrylamide based photopolymer developed at the CIEO [4].

Investigations have been carried out to improve the holographic properties of photopolymer by the addition of nanoparticles [7, 8]. Scattering is the main problem that arises [9]. Porous silicate nanoparticles having an MFI structure and lower refractive index than photopolymer have been used in our experiments. The porous nanoparticles introduce relatively lower scattering and also higher diffraction efficiency and lower shrinkage [10]. The refractive index of the nanoparticle doped photopolymer was previously determined [12].

The expression for the fractional change Δd in grating thickness d is expressed as follow [6].

$$\frac{\Delta d}{d} = \frac{\tan \phi_1}{\tan \phi_0} - 1 \quad (1)$$

where ϕ_0 and ϕ_1 are the initial and final slant angles of the recorded fringes to calculate the shrinkage.

Holographic recording in photopolymer layers is accompanied by light-induced mass transport of photopolymer components. The effect obtained by adding zeolite nanoparticles to acrylamide based photopolymer layers are discussed in [10-12]. We measured angular selectivity for various slant angles in order to determine the thickness and hence the shrinkage as a result of the holographic recording.

In this paper we study the influence of nanoparticles (pure silica MFI-type zeolite (Si-MFI) with a solid concentration 2.5 wt. %) on photopolymer layer shrinkage.

2. EXPERIMENTAL DETAILS

2.1 Sample preparation

The composition of the photopolymer solution is as shown in Table 1.

Table. 1 Photopolymer composition

Acrylamide	0.6g
Methylenebisacrylamide	0.2g
Polyvinyl alcohol (10%stock)	17.5ml
Triethanolamine	2ml
Erythrosine B dye (0.11%wt)	4ml

The photopolymer layer was prepared as described in [4]. Briefly, 0.6g of acrylamide monomer was added to 9 ml stock solution of polyvinyl alcohol (20% wt). Then 2ml of triethanolamine was added. To this solution 0.2 g of *N, N*-Methylenebisacrylamide then 4ml of Erythrosin B dye(0.11 wt stock solution) and finally 6.9ml (2.5 wt.%) of Si-MFI zeolite nanoparticles with a diameter of 30nm (Fig.1) were added to form a nanocomposite. We also added water in the undoped and doped photopolymer solution in order to have the same thickness. Of this solution, 1.4 ml was spread on a 30 mm -120 mm glass plate. The samples were dried for 24 h. Sample thickness after drying was $40 \pm 3 \mu\text{m}$.

2.2 Holographic recording

The setup used to record holographic gratings is shown in Fig.1. The 532-nm beam from an Nd:YVO₄ was split, spatially filtered, expanded to 1cm diameter and collimated. The beam was then split into two using a beam splitter. The two beams were made to overlap at the sample with an angle of 30.85° between them, so the spatial frequency was approximately 1000 lines/mm. The He-Ne probe beam must be corrected for the right incidence angle.

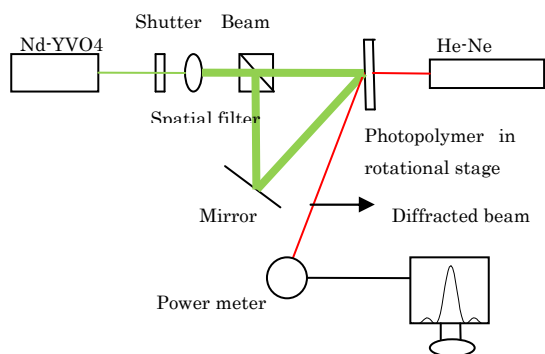


Fig. 1 Optical set-up for recording transmission holographic phase gratings

The photosensitive layer was mounted on a high-precision computer-controlled rotational stage (Newport M-URM100ACC). Diffraction efficiency was monitored using a He-Ne laser which was diffracted by the recorded grating but not absorbed by the photopolymer. Gratings were recorded using intensity of 5 mW/cm^2 and exposure time of 16s.

3. RESULTS AND DISCUSSION

The dynamic light scattering curve of zeolite nanoparticles prior to mix with the photopolymer is shown in Figure.2. The inset shows the TEM image of crystalline particles and from the curve we are able to determine the particle diameter (M). In this case the Si-MFI nanoparticles have a diameter of 30nm.

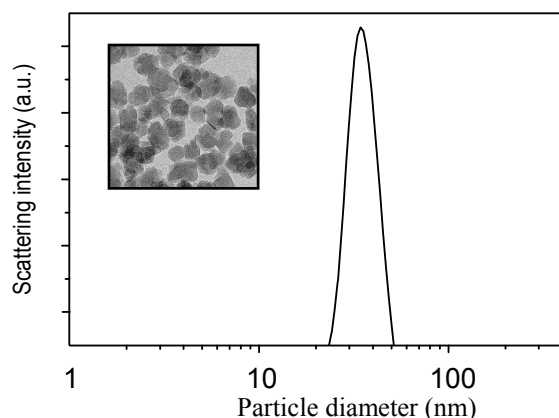


Figure 2. Dynamic light scattering curve of Si-MFI zeolite nanocrystals in water suspension prior mixing with photopolymer; the inset shows a TEM picture of the crystalline particles ($M=30 \text{ nm}$).

Fig 3(a) shows the diffraction efficiency growth curve of $+5^\circ$ slanted gratings for undoped and doped photopolymer having the same total exposure. It is clear that the diffraction efficiency is higher for the doped samples. The fitted angular selectivity curves for slant angle $+5^\circ$ are shown in

Fig 3(b), as plots of normalized diffraction efficiency versus angle inside the photopolymer layer, where the position of the Bragg peak of the unslanted grating in undoped photopolymer is $+12.13^\circ$ and for 2.5% MFI doped it is $+12.73^\circ$ inside the photopolymer layer for 1000 lines/mm.

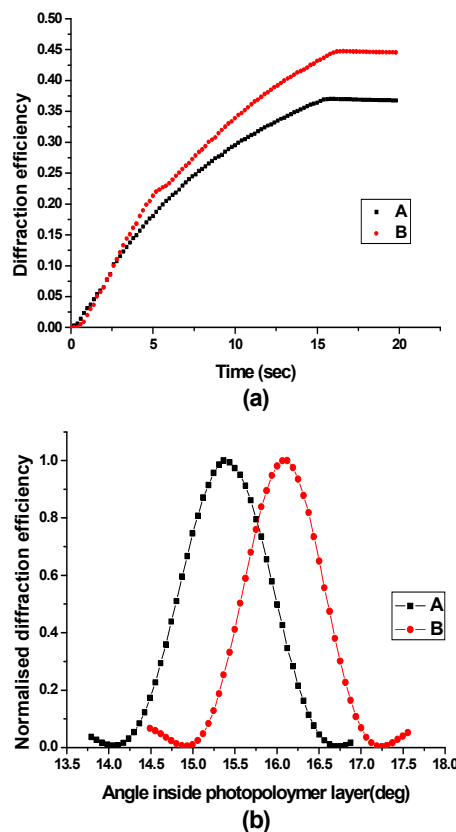


Fig. 3 Diffraction efficiency growth (a) and angular selectivity curves (b) for gratings recorded in A- Undoped; B- Si-MFI doped photopolymer, 16sec, the corresponding peak positions in (b) are B- 16.123° , A- 15.360°

From Fig. 3(b) the shift of the Bragg peak is determined and it is 0.54° in the case of undoped photopolymer and 0.37° in 2.5 wt. % MFI doped layers. The difference in the change of position of the Bragg curves arises from the change in refractive index as the nanoparticles doped layers have lower refractive index than standard undoped photopolymer layers. We also measured the shift of the Bragg peak at slant angles -10° , -5° and $+10^\circ$. An approximately linear relationship between the shift in the Bragg peak and slant angle is observed. Fig.4 shows the shift in Bragg peak versus slant angles for undoped (A) and Si-MFI doped (B) layers. We can see a smaller shift of the Bragg selectivity curve for nanoparticle doped layers.

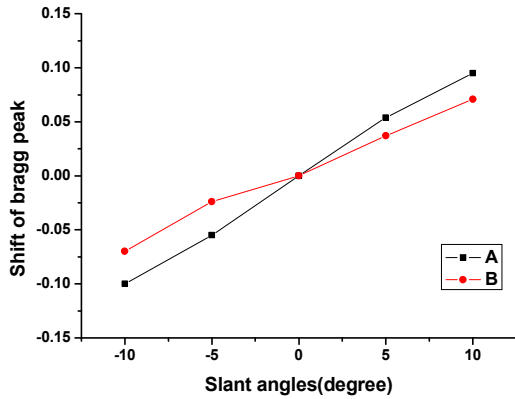


Fig. 4 Bragg peak shift versus slant angle for spatial frequency 1000 lines/mm for undoped (A) and 2.5% doped (B) layers

Using Eq. (1) one can fit the linear dependence of $\tan(\phi_1)$ on $\tan(\phi_0)$. The slope gives the percentage shrinkage of the material (Fig.5). The shrinkages evaluated from the fitted curves are 1.77%, 1.03% for undoped (a) and doped (b) photopolymer respectively.

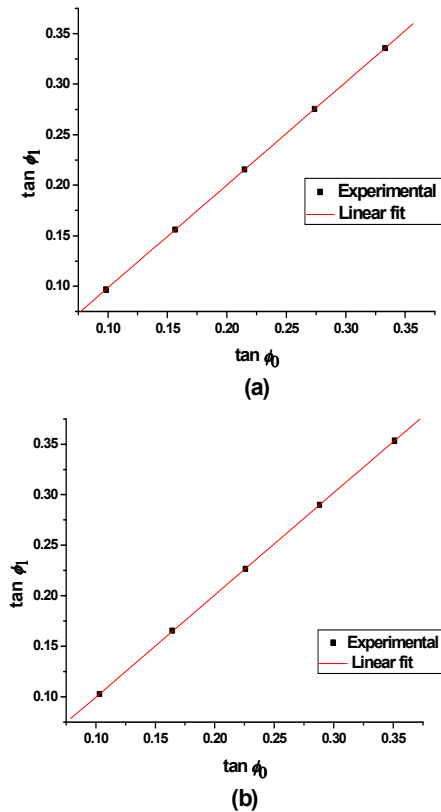


Fig.5 Dependence of the final slant angle after recording on the initial slant angle undoped (a) and 2.5 wt. % doped (b) layers.

The increase in the diffraction efficiency in

doped photopolymer with zeolite nanoparticle as a result of the counter diffusion process inside the layer during holographic recording is observed. The counter diffusion process occurs when monomers diffuse in to bright regions as a result of change in concentration gradient and the nanoparticles migrate from bright to the dark region during the holographic recording. We can attribute the increase in diffraction efficiency to the increased refractive index modulation caused by nanoparticles redistribution

4. CONCLUSIONS

Transmission diffraction gratings of spatial frequency 1000 ln/mm were recorded in an acrylamide based photopolymer film having $40 \pm 3 \mu\text{m}$ thickness with and without nanoparticles at constant exposure. The Bragg curves and Kogelnik's coupled wave theory were used to fit the angular selectivity curves of the gratings. Further the grating thickness and final slant angles from the Bragg curve are used to calculate the shrinkage. Higher shrinkage is noted for recording with undoped photopolymer.

The next step is the modification of the photopolymer material in order to decrease further the shrinkage due to photopolymerisation. We are studying the effect of different nanoparticles incorporated with different concentrations in the material. The results will be presented elsewhere.

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