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HOLOGRAPHIC RECORDING IN CORONA CHARGED ACRYLAMIDE-BASED MFI-ZEOLITE PHOTOPOLYMER

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The influence of corona charging on holographic recording in acrylamide-based photopolymer nanocomposite containing MFI zeolite nanoparticles has been studied. The holographic recording was carried out in two different geometries—transmission grating recording and total internal reflection grating recording. During the recording process, the layers were charged in a corona field. It was observed that independently of the corona polarity, in the case of transmission geometry of recording, the corona charging led to a decrease in the diffraction efficiency (DE) of the grating. In the case of the total internal reflection grating, the DE increased in the corona field presence.

Keywords: Holographic recording; corona charging; zeolite nanoparticles; photopolymer nanocomposites.

1. Introduction

The influence of corona charging on holographic recording in different media, both organic and inorganic films, has been investigated recently.¹ It has been observed that the induced polarizability, caused by electric charging, influences the holographic characteristics of the recording materials. Our recent investigations² show that the addition of an electric corona charge seems to be a promising method for increasing the diffraction efficiency, or the sensitivity of the different types of azopolymer recording media. Similar results have been observed by Nastas *et al.*³ and by Reinke *et al.*⁴ in their investigations of the influence of electric charge on the diffraction efficiency of holograms, recorded in thin chalcogenide films. The aim of the present paper is to investigate the influence of corona charging on holographic recording in acrylamide-based photopolymer nanocomposite containing MFI zeolite nanoparticles. The diffraction efficiency behaviour of transmission and reflection gratings was studied in real time.

Two types of polymer samples—undoped and doped with MFI nanoparticles, were studied. The investigated doped systems contain a soft photopolymer matrix with embedded porous nanoparticles. The standard photopolymer solution developed in the Centre for Industrial and Engineering Optics, Dublin Institute of Technology⁵ was used as a matrix. It consists of 9ml stock solution of Polyvinyl alcohol (20 w/w%), 2ml triethanolamine, 0.6 g acrylamide, 0.2 g *N,N*-methylene bisacrylamide and 4ml Erythrosin B dye of 1.1mM dye stock solution. Si-MFI zeolite nanoparticles of 30 nm size were used. The aqueous suspension of nanoparticles was sonicated for 20min prior to mixing with the photopolymer solution in order to ensure disintegration of any possible agglomerates. The aqueous suspension of the nanoparticles was mixed with the liquid photopolymer syrup, resulting in a concentration of 5.0 wt.% of nanoparticles in the final solution.

In order to obtain equal thicknesses of the undoped layers and layers doped with MFI zeolite nanoparticle, distilled water was added to the undoped solution to obtain the same total volume of each of the two stock solutions of 45ml. An amount of 0.7ml of each well-mixed solution was gravity settled on a levelled 2.5 cm×5 cm ITO glass substrate. The thickness of both the doped and undoped layers after drying for 24 h in darkness under normal laboratory conditions [$t_0 = (21-23)^\circ\text{C}$ and $\text{RH} = (40-60)\%$] was about 70 μm .

3. Holographic Recording

The recording of the holographic gratings in the layers was carried out simultaneously with their charging in a corona field. The electric field was created by a 5 kV DC high voltage power supply. The holographic recording was performed using two different geometries — transmission grating recording and total internal reflection grating recording with an original optical set-up (BG Patent No. 1391/31.11.2010).

The main advantage of the used device lies in the possibility of holographic recording with corona discharge over a large area of photosensitive material.

The experimental set-up is shown in Fig. 1.



Fig. 1. Experimental arrangement for simultaneous holographic recording and corona charging

4. Holographic Transmission Grating Recording

As illustrated in Fig. 2, a symmetrical two-beam recording set-up was used for holographic grating recording in the Bragg regime with angle $2\theta = 32.23^\circ$ between the recording light beams (3a and 3b), and the corresponding spatial frequency was 1050mm^{-1} . The total intensity of the laser beams was $10\text{mW}/\text{cm}^2$ and the recording wavelength was 532 nm .

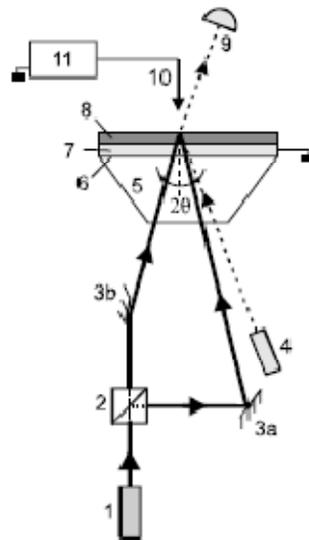


Fig. 2. Holographic set-up for recording of transmission gratings: 1 — recording laser, 532 nm; 2 — beam splitter; 3a, b — mirrors; 4 — monitoring He-Ne laser, 632.8 nm; 5 — prism; 6 — refractive index matching liquid; 7 — ITO electrode; 8 — recording medium; 9 — power meter; 10 — needle; 11 — high voltage power supply.

A low-intensity He-Ne laser (4) was employed as a read-out beam to monitor the buildup dynamics of the grating during the holographic recording.

The corona discharge was performed with a needle (10) at a distance of 1 cm from the sample, using a voltage of +5 kV or -5 kV (11). The corona experiments were performed using glass coated with ITO (indium tin oxide) (7) as a substrate for the polymer layers. ITO coated glass (7) was used as the grounded electrode for corona charging.

The holographic recordings in the undoped photopolymer films and in films doped with 5 wt.% MFI zeolite nanoparticles, were carried out simultaneously with their charging in positive and negative corona fields. Six samples from each of the groups were investigated.

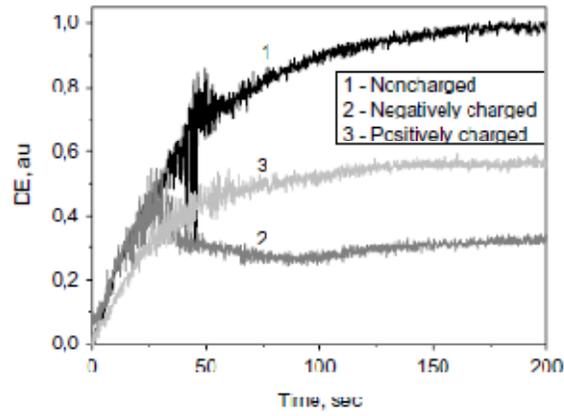
The evolution of the diffracted light intensity and its dependence on the corona charging are shown in Fig. 3. The results presented in Fig. 3 reveal that the presence of the corona field during the recording decreases the diffraction efficiency (DE) in both doped and undoped layers independently of the polarity of the corona field. The decrease of the DE in negative corona charging is more than a factor of three in both the undoped and MFI doped samples. The positive corona charging decreases DE by up to 60% in both the undoped and MFI doped samples.

5. Holographic Recording of Gratings by Total Internal Reflection (TIR)

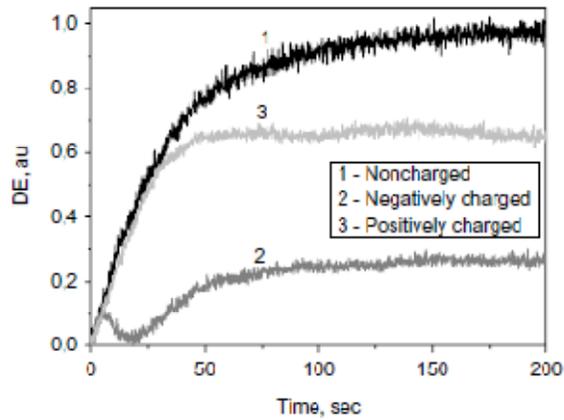
Three high spatial frequency slanted gratings are simultaneously recorded in the Stetson regime,¹ using the holographic setup presented in Fig. 4. The reference beam (3b) is totally reflected from the photopolymer — air interface at the 45° incidence angle. The object beam (3a) is incident normally to the photopolymer layer surface. Two reflection gratings were simultaneously recorded with spatial frequencies of 1440mm⁻¹ and 3470mm⁻¹, respectively. The first recording was realized by the interference between reference wave (beam 3b) and object wave (beam 3a). The second recording was realized by the interference between TIR reference wave and object wave (beam 3a). The evanescent wave slanted grating with 2323mm⁻¹ spatial frequency was recorded by the interference between beam 3a and surface propagated evanescent wave, created by the total internal reflection of the beam 3b. Two unslanted gratings were also recorded by the interference between beams 3a and 3b and its Fresnel reflections. These gratings, however, are with negligible contribution to the total diffraction efficiency due to their low fringe visibility. The total laser beams' intensity was 10mW/cm². A low-intensity He-Ne laser (10) was employed as a read-out beam to monitor the buildup dynamics of the gratings during the holographic recording.

The corona discharge was performed with a needle (11) at a distance of 1 cm from the sample, using a voltage of +5 kV or -5 kV (12). The corona experiments were performed using ITO (indium tin oxide) glass (7) as a substrate for the polymer layers. ITO glass (7) was used as the grounded electrode for corona charging.

The holographic recordings in undoped films and in films doped with 5 wt.% MFI zeolite nanoparticles were carried out simultaneously with their charging in positive and negative corona fields. Six samples from each of the groups were investigated. The evolution of the diffracted light intensity and its dependence on the corona charging are shown in Fig. 5. The results presented in Fig. 5 reveal that in this geometry of recording, the influence of the corona field is different for doped and undoped layers. In undoped layers, the corona charging decreases the DE and the positive charging leads to the stronger decrease. In doped layers, the presence of the corona field increases the DE in both cases of polarity — positive and negative polarity. The influence of negative corona charging is much stronger than the positive one.



(a)



(b)

Fig. 3. Diffraction efficiency of transmission holographic gratings as a function of exposure time for: (a) undoped and (b) MFI zeolite doped acrylamide-based photopolymers.

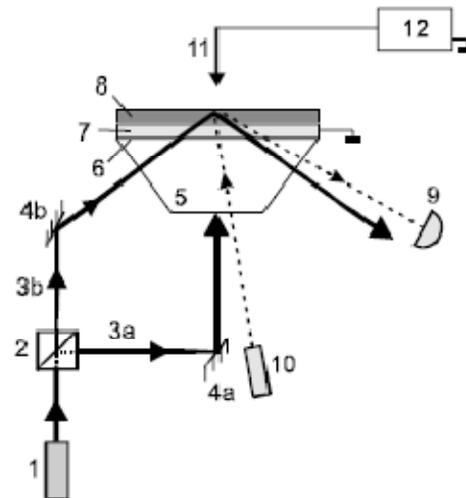
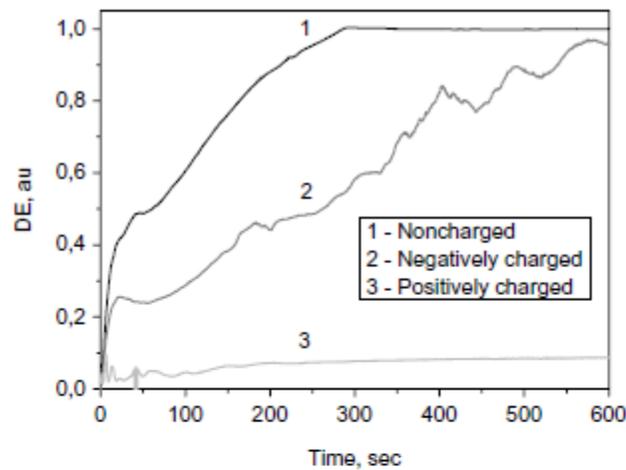
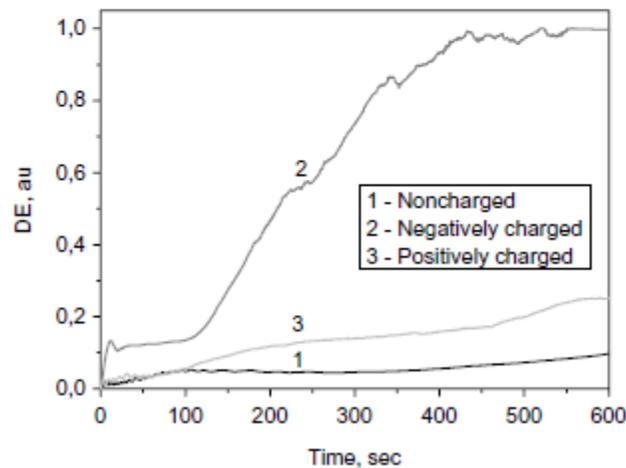


Fig. 4. Holographic set-up for recording of gratings by total internal reflection: 1 — recording laser, 532nm; 2 — beam splitter; 3a — normally incident beam; 3b — beam undergoing total internal reflection; 4a, b — mirrors; 5 — prism; 6 — refractive index matching liquid; 7 — ITO electrode; 8 — recording medium; 9 — power meter; 10 — monitoring He-Ne laser, 632.8 nm; 11 — needle; 12 — high voltage power supply.



(a)



(b)

Fig. 5. Diffraction efficiency of gratings recorded by TIR as a function of the exposure time for: (a) undoped and (b) MFI doped acrylamide-based photopolymers.

6. Discussion

The influence of the applied electrical field at the time of holographic recording is different for the two arrangements we used — TIR and transmission grating recording. This is most probably due to differences in the processes that take place. In the first case of TIR grating recording, three slanted diffraction gratings are simultaneously recorded. One of them has a lower spatial frequency (1440mm^{-1}), and the recording is performed by the normally incident wave and the wave that falls is incident at 45° . The second grating which has more than twice higher spatial frequency (3470mm^{-1}) is obtained as a result of the interference of a normally incident wave and a totally internal reflected wave at the sample-air boundary. Both gratings are recorded in the sample volume. Near the surface of the photopolymer, an evanescent wave grating with 2323mm^{-1} is also recorded. In the case of transmission grating recording, using the geometry shown in Fig. 2, only one unslanted diffraction grating is recorded with a spatial frequency of 1050mm^{-1} . During corona charging, a charge of the same polarity is deposited onto the surface. Positively or negatively charged oxygen-containing ions deposited on the sample surface, during the charging process, generate a constant electric field in the sample volume. Obviously, the electric field that is created in the case of a transmission grating recording worsens the recording in the acrylamide-based photopolymers in contrast to the chalcogenide glasses and azopolymers for which an increase of the DE has been earlier observed by Nastas *et al.*,³ Reinke *et al.*,⁴ Couture *et al.*⁷ and Natansohn *et al.*⁸

The addition of Si-MFI nanoparticles does not have any influence on the transmission unslanted grating recordings, but significant changes are observed in the TIR grating recordings. There are two effects that need to be considered here. First, in this geometry of recording, two slanted gratings in the photopolymer volume are recorded and their dynamics will be affected by any dimensional change occurring in the sample during the recording process. It is known that the presence of Si-MFI nanoparticles suppresses dimensional changes during photopolymerisation.⁹ Thus one could expect that any additional dimensional change caused by corona charging of the layer will also be partially or entirely suppressed by the presence of the zeolite dopants. The fact that there is no dimensional change was supported by AFM scans of the surfaces of the recorded layers, showing no surface relief modulation. The formation of a surface relief grating enhanced by the simultaneous presence of zeolite nanoparticles and corona charges can therefore be excluded as a factor causing the changes in corona field. Secondly, in the case of the second recording geometry using total internal reflection, the created surface propagating evanescent wave makes an essential contribution to what is happening. The grating is recorded in a very thin (about one wavelength) layer near the photopolymer surface. The charged particles created by the corona discharge are also situated in a thin surface layer and most probably the observed improvement of the DE is influenced mainly by the properties of the grating recorded by the evanescent reference wave. The corona charges could interact with the zeolite nanoparticles, which are negatively charged and thus increase the DE by enhancing the modulation of the refractive index. When the corona is negative, DE grows faster than in a positive corona.

7. Conclusion

Corona charging has different influence on holographic recording in acrylamidebased MFI zeolite doped photopolymer nanocomposite. The holographic recording was performed by using two different geometries — transmission grating recording and recording by using total internal reflection. In the case of unslanted transmission grating recording, the corona charging decreased the diffraction efficiency independently of the polarity of the corona charge. In the second case, however, where the reference laser beam was totally internally reflected from the sample-air interface, an increase in the diffraction efficiency was observed in MFI nanoparticle doped photopolymer for corona charging of both polarities.

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