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A simple method to fabricate low-scattering holographic polymer-dispersed liquid crystal grating is proposed. The optical efficiencies of the grating are tested. The results show that the scattering losses are almost totally suppressed, at less than 0.25%, and the diffraction efficiency of the grating reaches as high as 99%, indicating a great improvement compared with grating fabricated in the conventional way. In addition, the electro-optical performance maintains good behaviour, and thermo-stability testing shows that the good optical properties of the grating can be maintained in a relatively wide temperature range, satisfying the application requirements of displays and other optical instruments.

Keywords: polymer-dispersed liquid crystal; grating; scattering

1. Introduction

Holographic grating is an important optical element, which has been widely applied in various industries. As a kind of liquid crystal (LC)-based holographic grating, holographic polymer-dispersed liquid crystal (HPDLC) grating is attracting increasing attention from researchers worldwide for its special physical properties compared with conventional holographic gratings, such as electrically tunable optical efficiencies, sub-millisecond response time, smaller size and lighter weight, etc. These properties make HPDLC gratings a bright prospect in the field of displays, optical instrument, laser techniques and sensors [1, 2].

HPDLC grating, which was originally proposed by Sutherland *et al*. almost 20 years ago [3], is formed by exposing a mixture of nematic LCs (NLCs) and photosensitive monomers to an intensity modulated coherent field. The monomers cross-link in the high-intensity zone and form a solid polymer; at the same time, the LC is squeezed out of the polymer and forms LC droplets. The consumption of monomers leads to a periodic chemical potential gradient in the mixture, and subsequently leads to opposing diffusion of monomers and LCs. This diffusion provides further monomers to polymerise, and the monomer concentration is decreased; in addition, more LCs are continually squeezed out of the polymer and coalesce and, as a result, a well-defined structure with alternating polymer-rich and LC-rich lamellas is formed by polymerisation-induced phase separation (PIPS) [1–5].

From its initial proposal to the current day, HPDLC has remained a popular subject of research. Some researchers have focused on the dynamical theories of the PIPS process, aiming to obtain useful microscopic information to design appropriate exposure conditions [6–9]. Others consider that the PIPS process is mainly influenced by the chemical properties of materials, so they aim to improve the phase separations, (i) by using a high-functionality monomer system, such as acrylate, polyurethane or the commercial UV-polymerised NOA systems [10–12]; and (ii) by using additives as the surfactant, such as octanoic acid, stearyl methacrylate, fluorine-substitute acrylate and high dielectric monomer HR-410 [13–16]. In addition, there is much work on devices based on HPDLC gratings, which involve active optical elements, distributed feedback lasers, sensors, displays, muti-information storages, and so on [17–22]. Such research is making a great contribution to the development of HPDLC gratings. However, a serious problem faced by these gratings is the high scattering losses, which not only decrease optical efficiency, but reduce beam quality; thus solutions to this problem are urgently sought.

As Sutherland *et al*. noted, HPDLC scattering mainly arises from two aspects: one is Raleigh scattering, which is proportional to the volume of LCs; the other is differences of LC order in the droplet domains [23]. It has been suggested that decreasing the content of LCs in the HPDLC results in a decrease in the volume of LCs, so that the scattering can be reduced to less than 5% [2], and this reduction is accompanied by an increase of drive voltage. West *et al*. have proposed an intriguing method called ‘shearing’ to make the LC molecules in the domain align in the same direction as the polymer chain after a shear shifting [24], so that the order of LCs is the same and the scattering losses

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are notably decreased. This method is very effective in devices with lower polymer content, such as polymer network liquid crystals (PNLCs) [25]. However, for HPDLC shearing is not feasible, for the following two reasons: the relative high content of solid polymer makes shearing rather difficult; and the shear shift of several or several hundred micrometers will destroy the fine structure of HPDLC grating.

In this paper, a simple method to suppress the scattering losses of HPDLC grating is proposed. The foundations of this method are presented in the next section; testing of the optical efficiencies and performances of the grating is carried out, and the test results are compared with conventional HPDLC grating.

2. Materials, foundations and testing

The materials used to fabricate HPDLC grating comprised commercial NLCs (TEB30A, $n_o = 1.5222$, $n_e = 1.6925$ and $\Delta n = 0.1703$, provided by Slichem, Shijiachuang, China) and acrylate-based monomers, mixed together with the weight ratio of 3:7. In order to ensure high cross-linking of the polymer and obtain suitable photoreactive kinetic parameters, two kinds of monomers should be mixed to form a mixture with the average functionality of 3.5; one is penta-functional dipentaerythritol hydroxyl pentaacrylate (DPHPA, Aldrich), and the other is di-functional phthalic diglycol diacrylate (PDDA, Eastern Acrylic Chem. Tech. Co., Ltd. Beijing); these are mixed with the molar ratio of 1:1. The mixture of LCs and monomers was stirred for about 12 h at a temperature higher than the clearing point of LCs. To make the monomers polymerise at the wavelength of 532 nm, a small amount of photoinitiator Rose Bengal and coinitiator n-phenylglycine were added to the mixture. In addition, considering the better electrical tunability of HPDLC grating, about 8 wt% perfluorodecyl acrylate was added as the surfactant. The refractive index of monomers after polymerisation was tested as 1.523 by Abbe refractometer, which is approximately equal to the ordinary refractive index of LCs. The syrup was injected into a 12 μm-thick cell and irradiated with coherent beams for several minutes.

To reduce the scattering losses of HPDLC grating, some fundamental theories are first considered. As shown by the equations of Raleigh scattering [26],

$$I_s = \frac{8\pi^4 N r^6}{d^2 \lambda^4}\left(\frac{m^2-1}{m^2+2}\right)^2 (1+\cos^2\phi) I_0 \qquad (1)$$

where N is the number of LC droplets in the grating, r is the radius of the droplets; λ is the wavelength of the testing beam and φ represents the angle of the beam incident on the droplets; d is the distance between the grating and the detector. The parameter m is defined as the refractive index ratio between any two droplets, and it can be expressed as [27]:

$$m = \sqrt{1 + \frac{\Delta S}{\dfrac{6n_i^2}{n_e^2 - n_o^2} + S}} \qquad (2)$$

in which, S and ΔS, respectively, represent the average order parameter of the LCs and the differences of order parameter between the droplets; n_i is the average refractive index; as usually defined, n_e and n_o are the extraordinary and ordinary refractive index, respectively.

Combining Equations (1) and (2), it can be found that the scattering could be eliminated if ΔS equals zero. Thus, a necessary and important issue is to make the LC molecules in the HPDLC grating align uniformly. So, in this work we used conventional rubbing to align the LC in the grating. The rubbing direction was selected as the same as that of grating grooves (shown as the black arrow in the inset of Figure 1). Although some tiny LC droplets enclosed by the polymer wall are very hard to align by rubbing, the scattering of these droplets was negligible because of their very small volume.

The set-up for the testing of optical efficiencies and electro-optical performance is shown in Figure 1. A He-Ne laser was used as the light source. A quart-wave plate and polariser were used to obtain a polarised incident beam. The sample was placed on a holder with Bragg angle to the incident beam. The intensities of the diffractive beams were detected by CCDs connected to computers for data collection. A signal generator was used to apply a 50 Hz sine wave to the sample for electro-optical testing. The polarisation direction of the incident beam can be adjusted by rotating the polariser, and two typical polarisation directions were defined as S (polarisation is same as the director of LCs) and P (polarisation is vertical with the director of LCs), as shown in the inset of Figure 1. At the beginning of testing, an empty cell was placed on the holder and rotated to Bragg angle. We then tested the intensity of the transmitted beam, labelled as I_T; thus reflections and absorptions caused by the glass could be subtracted. The zeroth and first-order intensities of the gratings were tested by two detectors and defined as I_0 and I_1. It was confirmed that the absorbance of HPDLC materials near 632 nm is so small that it can be ignored. Therefore, the scattering losses (η_s) and the diffraction efficiency (η_1) of HPDLC grating can be calculated as follows, and our testing is based on the two equations.

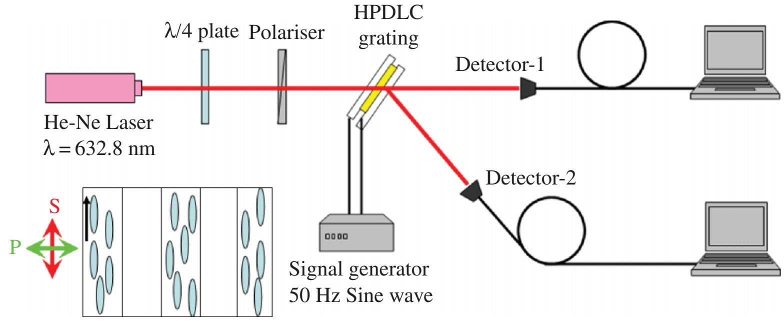

Figure 1. Testing set-up for HPDLC grating. The inset in the lower left corner is a simple scheme of the grating.

$$\eta_s = \frac{I_T - I_0 - I_1}{I_T} \times 100\% \tag{3}$$

$$\eta_1 = \frac{I_1}{I_T} \times 100\% \tag{4}$$

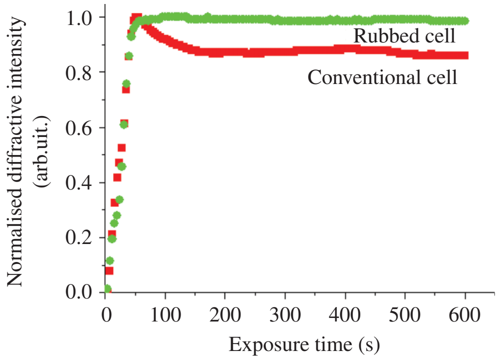

Figure 2. Normalised diffractive intensity vs. exposure time curves of rubbed and conventional cells.

3. Results and discussions

Several LC cells without and with rubbing processes were used to fabricate the conventional and low-scattering losses HPDLC gratings, respectively. The changes of first-order diffractive intensity with exposure time for the gratings fabricated by conventional cell and rubbed cell were detected by CCDs and compared with each other. As shown in Figure 2, in the case of the conventional cell, the intensity reached a maximum when the cell was exposed for about 60 s, and followed an evident decrease, finally reaching the stable state at 150 s. Bunning *et al*. ascribe the decreasing diffraction intensity to the scattering losses of HPDLC grating [1]. As a comparison, the grating fabricated by rubbed cells presented a different change of intensity, which reached the maximum at the almost the same time as the conventional one, and then the intensity reached saturation with no evident decrease. Such results indicate two important points: first, surface rubbing does not affect the formation of the grating; second, the scattering losses can be effectively suppressed through rubbing.

To further investigate the optical efficiencies of HPDLC grating based on rubbing, and compare the results with those of conventional grating, the first-order diffraction efficiency and scattering losses of our samples were tested according to the set-up shown in Figure 1 and calculated by Equations (3) and (4). These results are listed in Table 1; the upper three rows are the results of three conventional samples, and the lower three are those of rubbed samples. The beam intensity through the empty cell I_T was almost constant. However, the first-order diffraction efficiency and the scattering losses show large differences. As compared in the last line of Table 1, the average diffraction efficiency of the rubbed sample reaches 99.0%, which is very close to the theoretical value, 100%, and we can see that the scattering losses are only 0.24%; in contrast, the diffraction efficiency and scattering losses of the conventional sample are 75.0% and 20.7%, respectively. We consider that the significant rise of the diffraction efficiency for the rubbed sample is mainly caused by the effective decreasing of scattering losses after the rubbing process. Therefore, the results in Table 1 indicate that the main factor in the decrease of the diffraction efficiency was the scattering, and this reflects the necessity to solve the scattering problem.

The electro-optical performance of the rubbed HPDLC grating was then tested. A sine signal with a frequency of 50 Hz was applied to the sample and the first-order diffraction efficiency corresponding to the voltage was recorded by CCD. To examine the alignment of LCs in the grating, the efficiencies for S-polarisation and P-polarisation incidences were tested and compared. A good electro-optical performance for S-polarisation incidence was obtained, as shown in Figure 3. The diffraction efficiency of S-polarisation decreased with the increase of applied voltage. The

Table 1. Optical efficiencies of HPDLC gratings (unit of intensity: mW/cm^2).

Samples	No.	I_T	I_0	I_1	η_1 (%)	η_s (%)	$\bar{\eta}_1/\bar{\eta}_s$ (%)
Conventional	**1**	1.540	0.100	1.121	72.8	20.7	75.0/20.7
	2	1.511	0.050	1.163	77.0	19.7	
	3	1.561	0.050	1.172	75.1	21.7	
With rubbing	**1**	1.550	0.012	1.535	99.0	0.19	99.0/0.24
	2	1.561	0.016	1.541	98.7	0.26	
	3	1.553	0.008	1.541	99.2	0.26	

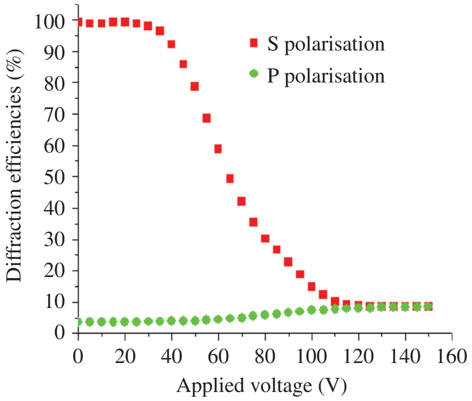

Figure 3. Electro-optical performance of rubbed HPDLC grating.

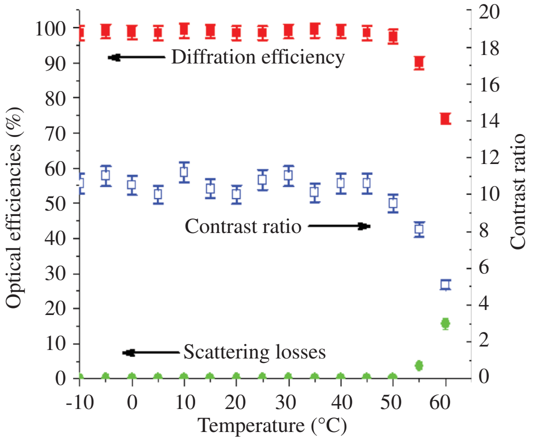

Figure 4. Thermo-stability testing of rubbed HPDLC grating.

threshold voltage and the saturated voltage were 35 V and 120 V, respectively, and the contrast ratio, defined as the ratio of maximum and minimum of diffraction efficiency, was about 11, which satisfies the applications of the grating. For P-polarisation, there was only a small increase of diffraction efficiency with the increase in voltage, and the contrast ratio was tested as less than 2; these results show a good alignment of LCs after the rubbing process, and are in good agreement with the results obtained in Figure 1.

In addition, the thermo-stability of the rubbed grating was evaluated from three aspects: diffraction efficiency, scattering losses and contrast ratio. The sample was placed on a precisely controlled hot-stage (Linkam LT120S, UK), the temperature was changed from −10–60°C in steps of 5°C, and the optical efficiencies and contrast ratio at every temperature were tested. As shown in Figure 4, the diffraction efficiency did not change with temperature, until heated above 50°C. The scattering results showed the same tendency. From these results we believe that there is no notable change of LC alignment when the temperature is below 50°C. The electro-optical performance was evaluated by testing the contrast ratio. As Figure 4 shows, the contrast ratio was around 10–11 at temperatures below 50°C. Some fluctuations may be related to the instability of the testing source. At temperatures higher than 50°C, an evident decrease was found. This tendency is in accordance with that seen in the optical efficiency results. Thus, we believe that the rubbed HPDLC grating can work in a very wide temperature range. Such a wide range can be attributed to two points: the NLCs used in the experiment are wide temperature range materials (nematic: 0–61°C); and the polymer network distribution in LC-rich zone plays an important role in stabilising LC alignment, therefore the grating works normally even below 0°C, which is beyond the liquid crystal phase range for our NLCs. To further widen the range, we can use a NLC with a wider temperature range or enhance the cross-link density of the polymer network.

4. Conclusions

A low scattering losses HPDLC grating was fabricated by rubbing the substrates of the LC cell. The rubbing process led to a uniform alignment of LCs in the grating, and decreased the differences of order parameter between the LC domains, which are the main reasons for the low scattering. The optical efficiencies of the grating were tested, and the results show that the scattering losses were effectively suppressed and decreased to less than 0.25%, which is two orders of magnitude lower than conventional HPDLC grating; the diffraction efficiency of the rubbed grating was as high as 99%, almost 35% higher than that of the conventional one, and the threshold voltage was low, only about 35 V. In addition, thermo-stability testing showed that the rubbed grating maintained good optical efficiencies and electro-optical performance across a relatively wide temperature range, satisfying the application requirements of displays and other optical instruments.

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