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One-step fabrication of surface-relief diffusers by stress-induced undulations on elastomer

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ABSTRACT

In this study, we demonstrated that microscale surface undulations induced by acid treatment could serve as the surface relief on diffusers coated with a layer of PDMS polymer. Since the orientation of undulations was found to be always disordered, these undulations would scatter light uniformly. The periodicity of the undulations could be adjusted by the control of duration of the dipping of the elastomer into H_2SO_4/HNO_3 solutions and by the volume ratio of H_2SO_4/HNO_3 the solution, resulting in the modulation of diffusing ability of diffusers. The optical properties, transmittance, and light diffusivity, were characterised. This proposed approach offers potential for mass production of surface-relief diffusers. In addition, the proposed method allows the creation of undulations on arbitrary substrates.

1. Introduction

In recent years, diffusers, the function of which is to spread the incident light from sources over a wide angular range with the aim of preventing the light source from being seen directly by viewers and to keep the brightness uniform, have been used as light-homogenizing elements in light-emitting diode (LED) display panels, liquid crystal display (LCD) backlights and other lighting systems, as well as in beam-homogenizing devices. In general, diffusers can be classified into two groups: one is the volume type [1]; the other is the surface-relief type. Volume-type diffusers depend mainly on transparent micro-beads or fillers located inside the plates to scatter light. However, it is difficult to distribute the fillers uniformly inside the plates, which significantly affects the optical performance of volume-type diffusers. On the other hand, surface-relief diffusers rely on microstructures, such as rough surfaces or microlens arrays, to scatter light. It is easy to generate microstructures located uniformly on the surface of plates. Recently, various methods have been developed to generate microstructures on surface of plates, such as holographic recording method [2], silver halide sensitized gelatin (SHSG) method [3], electrospray method [4], replica molding [5], hot embossing [6] and photofabrication [7]. However, most methods involve complex fabrication processes and require expensive instruments.

Stress-induced undulations on the surface of an elastomer are often observed when the elastomer is exposed to oxygen plasma or coated by metal films. Compared with the other techniques [2–7], stress-induced undulations on elastic materials [8–11] have the potential to generate large-scale and low-cost microstructures. However, metal films deposited on an elastomer [8,9] are not suitable to serve as diffusers, because the metal films may be damaged easily under physical deformation. In our previous work, a simple novel way to develop self-organized microscale undulations on a large area by chemical oxidization methods was demonstrated [12].

In this study, we tried to develop microscale surface undulations on different substrates such as planar and curved substrates to serve as diffusers. The optical performance of the diffusers was then examined. Since the orientation of the undulations was always disordered, these undulations were able to scatter light uniformly. The periodicity of undulations could be adjusted by the control of duration of the dipping of the substrate into H_2SO_4/HNO_3 solutions, resulting in the modulation of the diffusing ability of the diffusers.

2. Experiment

2.1. The preparation of materials

The liquid PDMS polymer was mixed with PDMS silicone elastomer (Sylgard 184) and curing agent in the weight ratio of

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10:1. Glass or plastic plates were selected and the PDMS films with 100 μm thickness were deposited onto them through a spin-coating process or, for curved substrates, by dipping the substrate into the PDMS liquid. These PDMS films were subsequently cured in an oven at 70 °C for 20 min.

2.2. *The development of microscale undulations*

A strong acid that mixed sulfuric acid solutions (H_2SO_4, content 95%) and nitric acid solutions (HNO_3, content 66–71%) with the appropriate volume ratio was prepared. The solidified PDMS films were immersed into the H_2SO_4/HNO_3 solutions for durations in the range of a few seconds to several minutes. Finally, the acid-modified PDMS films were dipped into clean water to remove the residual acid and then dried by an air gun or in an oven. Microscale surface undulations could be observed on the surface of PDMS.

2.3. *The generation mechanism of microscale undulations*

Oxidization layers, the product of chemical reaction that occurred when the PDMS polymer was dipped into the H_2SO_4/HNO_3 solutions, were capped on the surface of PDMS after acid treatment and subsequently undulations were spontaneously developed due to the large difference in volumetric contraction rates in the bilayer system. Young's moduli of the oxidization layer and of the underlying elastical materials were significantly mismatched, resulting in the generation of a compressive stress. In order to release the compress stress, the surface would form undulations [11].

3. Results and discussion

3.1. *The development of undulations on planar substrates*

Fig. 1(a) shows the images of the microscale surface undulations on planar substrates, photographed by the use of inverted microscope. Fig. 1(b) shows the 3D AFM topography of microscale undulations. Based on these images, it could be found that the orientation of surface undulations was random and that the profile of the undulations was sinusoidal. Hence, these undulations could be considered as bar-like microlenses located randomly on the surface of substrates, which promote the diffusion of light.

As the generation of bar-like structures was due to compressive stresses induced by the large difference in volumetric contraction rates of the two layers, the thickness of the oxidization layer was directly related with the surface morphology. Hence, by varying the dipping duration and the volume ratio of H_2SO_4/HNO_3 it is possible to change the thickness of the oxidization layer and to vary the surface morphology of the PDMS layer. Fig. 2 shows the dipping time as a function of the undulations' periodicity obtained for dipping into different volume ratios of H_2SO_4/HNO_3. The experimental results show that the undulations' periodicity ranged from 3 to 130 μm, with the periodicity increasing in response to an increase of the dipping duration and to an increase in the volume ratio of H_2SO_4/HNO_3 solutions. The distribution of undulations' periodicity was narrow in the initial stages but was broader in later stages. The further increase of dipping duration would result in a nonuniform distribution of undulations' periodicity because undulations would merge together [11]. In addition, the periodicity increases with the increase of the volume ratio of H_2SO_4/HNO_3 solutions from 1:1 to 4:1. On the other hand, the surface undulations were hardly developed when the volume ratio of H_2SO_4/HNO_3 solution was below 1:1. It indicates that the oxidization layer thickness increased with the proportion of H_2SO_4 in the solution when the proportion of H_2SO_4 exceeded a certain amount.

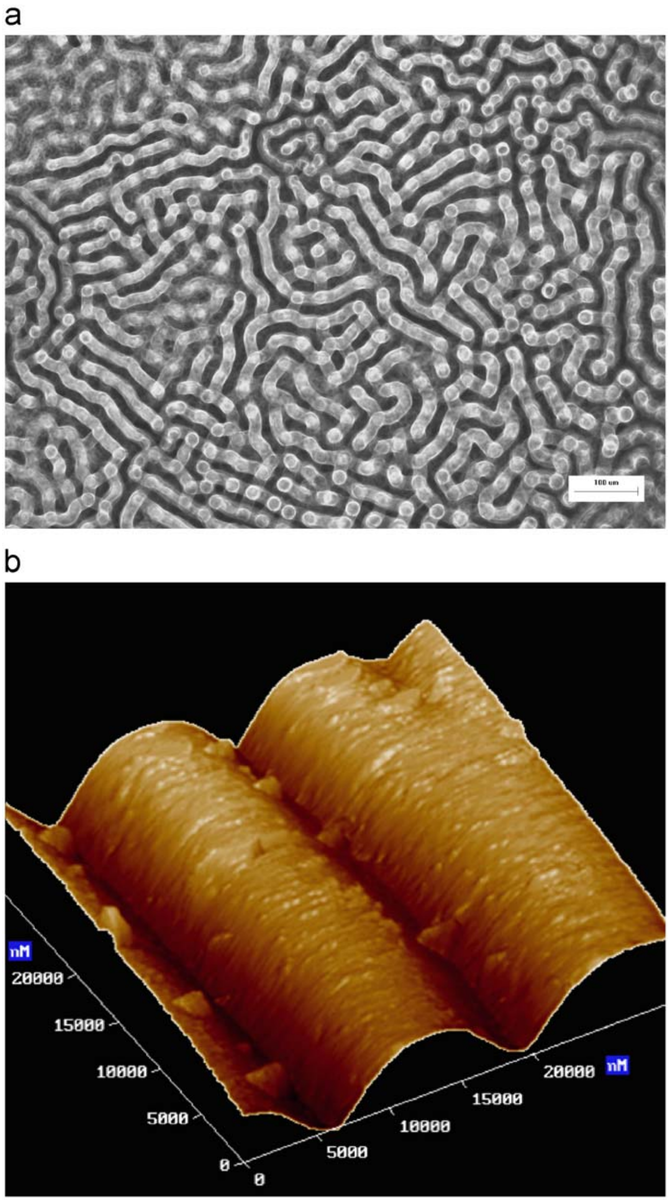

Fig. 1. (a) The optical image of microscaled surface undulations on planar substrates and (b) the AFM image show the 3D topography of microscaled undulations.

3.2. *The optical performance of undulations on planar substrates*

The diffusing pattern of the diffusers was evaluated by a imple optical setup. A schematic diagram of the setup is shown in Fig. 3(a). A collimated He–Ne laser beam with 632.8 nm wavelength illuminated the diffusers and the diffused beam was projected on the screen. The optical pattern projected on the screen was recorded by digital camera and the brightness was measured by the light meter. By this apparatus, three kinds of optical patterns could be obtained and be roughly classified as shown in Fig. 2. The first was an optical pattern consisting of a series of concentric circles in region A (shown in Fig. 3(b)), the second was an optical circular pattern with 0th order beam in region B (shown in Fig. 3(c)) and the other was a circular pattern without a 0th order beam in region C (shown in Fig. 3(d)). The appearance of concentric circles was because the undulations had almost a single periodicity when undulations were developed for small dipping durations. The distribution of the undulations' periodicity was found to be restricted within 0.5 μm. When the dipped duration increased to reach region B, the distribution of undulations' periodicity gradually broadened, with the diffuser producing concentric circles. In this case, the distribution of the undulations' periodicity was limited from 3 to 8 μm. Although the 0th order beam of the optical pattern could be observed, the boundary between each ring in the concentric circles was indistinct. When the dipping duration increased to reach region C, the distribution of the undulations' periodicity was broadened further, resulting in a uniform circular pattern. The distribution of the undulations' periodicity ranged between 10 and 19 μm. It suggested that the bar-like microlenses located on the PDMS surface were not of uniform size, causing the generation of different focal lengths. This allows the scattered laser beam to spread uniformly and produce a large diffusing angle.

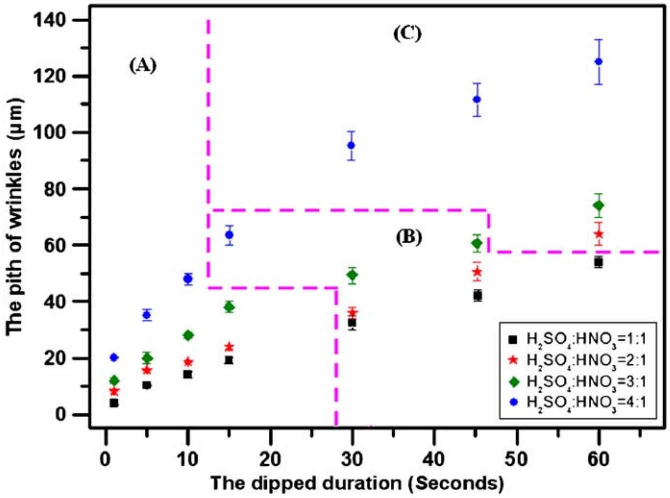

Fig. 2. The dipped time as a function of the undulations' periodicity under different volume ratios of H_2SO_4/HNO_3, wherein three domains were defined by optical pattern, including a series of concentric circles (region A), a circular pattern with 0th order beam (region B) and without 0th order beam (region C).

Generally, good diffusers must have a low percentage of the 0th order beam and an arbitrary diffusing angle. In this study, the better fabrication parameters should be located in region C in Fig. 2. Hence, we selected three diffusers in region C to examine their optical performance. The diffuser α was made using an H_2SO_4/HNO_3 solution with the ratio of 3:1 with the dipping duration being during 60 s. The undulations' periodicity ranged from 68 to 78 μm. Diffusers β and γ were made by using an

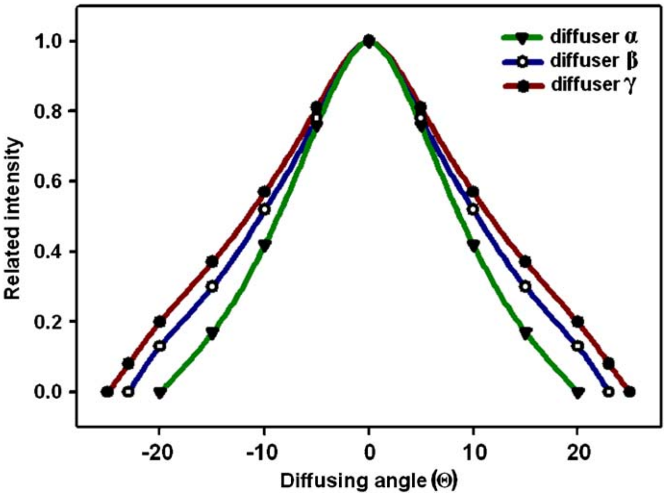

Fig. 4. The related brightness as a function of diffusing angles.

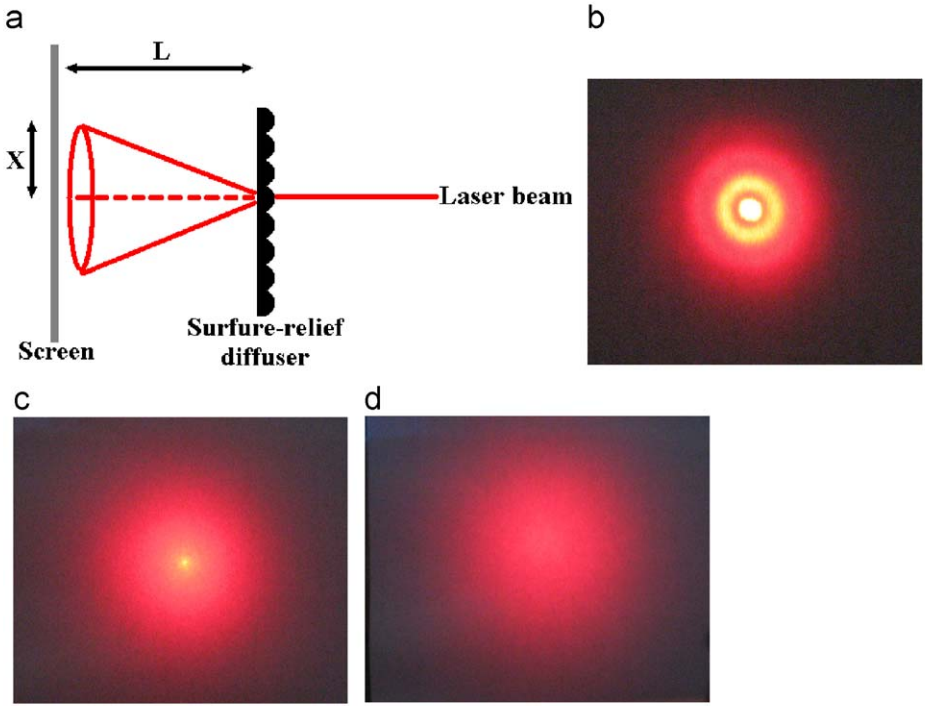

Fig. 3. Diffusing spot measurement: (a) schematic showing the experimental setup. The images of diffusing optical pattern, (b) an optical pattern consisted of a series of concentric circles, (c) an optical circular pattern with 0th order beam and (d) an optical circular pattern without 0th order beam.

H_2SO_4/HNO_3 solution with the ratio of 4:1, with dipping durations of 30 and 60 s, respectively. The undulations' periodicity of diffuser β ranged between 88 and 101 μm and the undulations' periodicity of diffuser γ ranged from 115 to 134 μm. Fig. 4 shows the related brightness as a function of diffusing angles, wherein the related brightness is defined as the measured flux divided by the maximum flux. Therefore, the diffusing angles were calculated from the equation

$$\theta = 2\tan^{-1}(x/L)$$

where x is the distance from the center of the screen to the edge of an optical pattern and L represents the spacing between the diffuser and screen. The diffusing angle of diffusers α, β and γ were found to be 40°, 46° and 49°, respectively. The percentage of the light in the 0th order beam, defined as the flux in the center of the diffusing beam divided by the overall flux of a laser beam, was also calculated for each diffuser, and was found to be 2.16%, 1.38% and 1.03%, respectively. Based on this result, diffusers β and γ had a better performance than diffuser α. It implies that the diffusers had a broad distribution of undulation periodicity and a larger size of undulations, resulting in good optical performance. In addition, the optical performance of diffusers β and γ had no obvious differences, indicating that there is a limit to the diffusing angle that could be obtained.

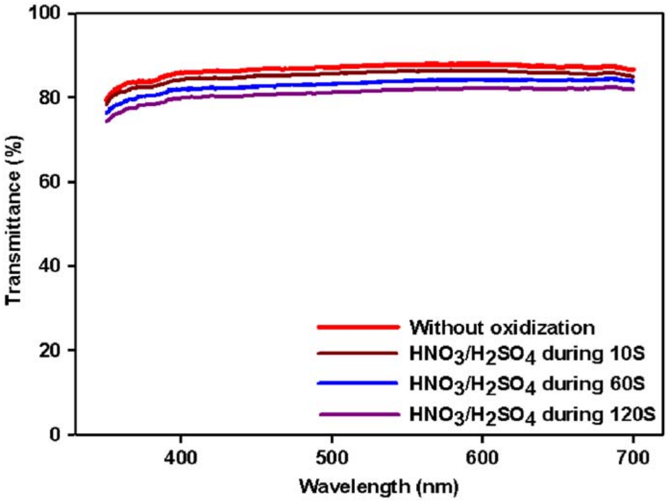

Fig. 5. A transmittance curve of oxidized PDMS plates dipped into H_2SO_4/HNO_3 solutions during four durations ($t = 0$, 10, 60 and 120 s) as a function of incident wavelength ranged between 350 and 700 nm.

The transmittance also plays a critical role in the evaluation of diffusers. Hence, the influence of the transmittance of oxidized PDMS polymer in the visible spectrum should be taken into account. Fig. 5 shows a transmittance curve of oxidized PDMS plates dipped into H_2SO_4/HNO_3 solutions (the volume ratio of 3) for four durations ($t = 0$, 10, 60 and 120 s, respectively) as a function of incident wavelength ranging between 350 and 700 nm, and the result was characterized by the use of a UV–vis spectrophotometer. Experimental results indicate that the transmittance decreased with increasing thickness of the oxidization layer. The transmittance of diffusers α, β and γ was given by 82%, 79% and 74%, respectively. The thickness of the oxidization layers influenced the transmittance, which is a disadvantage for this proposed method. However, the plates with microscale undulations could be serve as replication templates to transfer surface morphologies onto high-transmittance materials, which may be an approach to overcome this disadvantage in the further.

3.3. The optical performance of undulations on LEDs

LEDs have been often used as light sources in LCD systems. However, it was difficult to form plane light sources from high-intensity LEDs, causing the generation of hot spots in displays [13]. If diffusers can be fabricated on the plastic curved casing of LEDs, it may be possible to produce a more uniform illumination. However, it is difficult to fabricate microstructures on curved substrates using standard fabrication technologies [2–7], especially on devices with

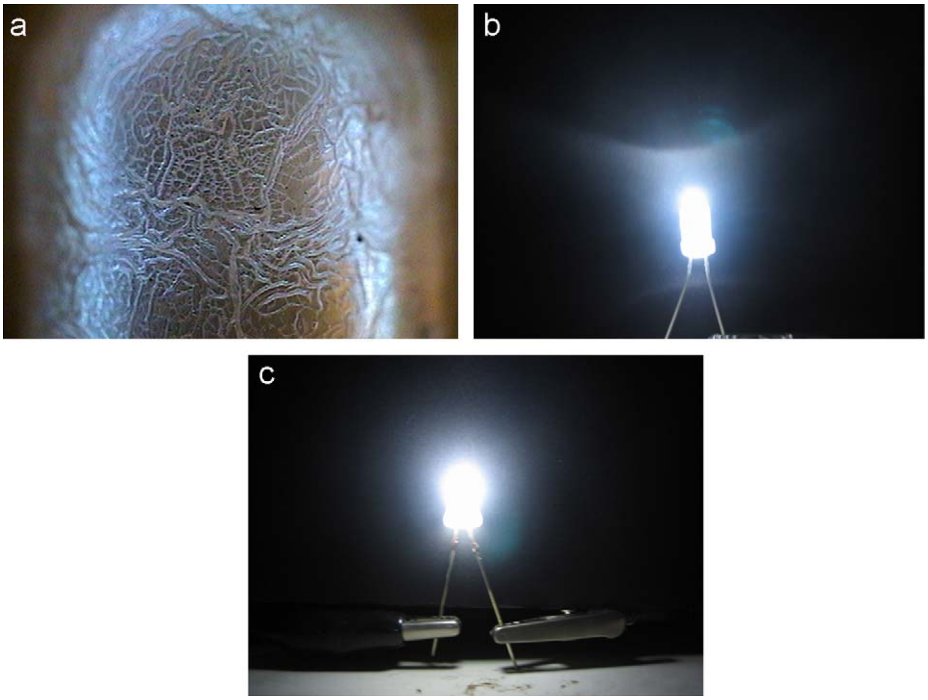

Fig. 6. The optical images of (a) disordered microscaled undulations on plastic curved casing of LEDs and the radiation pattern of a LED, (b) without diffusers and (c) with diffusers.

small volume. In this work, a commercial LED with a diameter of 5 mm was used. The LED was coated with a layer of PDMS polymer by dipping the LED into a PDMS solution and subsequently solidifying it. After acid treatment (H_2SO_4/HNO_3 with the ratio of 3:1) for a period of 60 s, undulations were developed on the curved casing of the LED. Fig. 6(a) shows the optical image of the disordered undulations with various periodicities on the plastic curved casing of the LED. The radiation pattern of the LED was then measured. Figs. 6(b) and (c) show the images of the radiation pattern of the LED without diffuser and with diffuser, respectively. Light emitted from the top of the LED without diffusers was concentrated in certain angles in Fig. 6(b). On the other hand, light emitted from the LED with diffusers spread in all directions in Fig. 6(c). Experimental results demonstrated that the diffusers fabricated on the LEDs improved the uniformity of the illumination.

4. Conclusion

Surface-relief diffusers could be quickly fabricated by developing undulations on PDMS polymer. The undulations generated spontaneously when oxidization layers were capped on PDMS films after acid treatment, due to the large difference in volumetric contraction rates on a bilayer system. The thickness of an oxidization layer was directly related with the surface morphology, and the thickness of oxidization layers could be changed by the modulation of the duration of acid treatment and the volume ratio of H_2SO_4/HNO_3. Experimental results showed that the distribution range of undulations' periodicity became broad, resulting in the better optical performance of diffusers. In addition, undulations could be developed on arbitrary substrate, such as planar or curved substrates. It was helpful to promote small-volume lighting devices to diffuse uniform light in photoelectric systems. Hence, the proposed approach offers a great potential for mass production of surface-relief diffusers.

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