Fabrication of high-efficiency Fresnel-type lenses by pinhole diffraction imaging of sol-gel hybrid materials

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Sol-gel hybrid materials containing a large quantity of photoactive molecules exhibited large changes in both refractive index and volume on UV exposure. The materials were used for fabrication of Fresnel-type lenses using a simple method: pinhole diffraction imaging. With this technique, problems associated with the contact method could be overcome and Fresnel-type lenses with good focusing performance could be fabricated easily. Importantly, a high diffraction efficiency approaching 85% could be obtained. © *2004 American Institute of Physics*.
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A Fresnel lens (FL) with focusing and diffraction ability is an important component for beam dispersion and conversion for use in optical interconnection and optical signal processing in compact and complicated optical data storage systems and optical integrated circuits.[1–4] In particular, the diffraction efficiency of FLs has received much attention as a crucial factor for exact and efficient beam alignment.[1,5–9] FLs are generally classified into two types depending on their response to electrical signals, electrically switchable FL and static FL, both of which are typically fabricated by lithographic or etching techniques.[1–9] With these techniques, many studies on the fabrication of FLs with higher diffraction efficiency and focusing effects have been performed.[4–9] However, these methods are rather complex and need several steps, which in turn cause limitations to obtaining higher diffraction efficiency and control of the focusing properties. For this reason, many studies have concentrated on simplifying the fabrication process of FL.[10] In order to achieve higher performance in optical elements, materials must have high photosensitivity, meaning a large refractive index change, and undergo a volume change upon light irradiation, because the efficiency depends directly on the photosensitivity of the materials used. In recent years, sol-gel hybrid (SGH) materials and photosensitive polymers have been used as potential materials for fabricating highly efficient diffractive optical elements.[7,10,11] However, lithographic and etching techniques need to be adapted to obtain sufficient change of refractive index and volume in those materials, making the fabrication process complex. In addition, in the case of photosensitive polymers, higher thermal stability is required in their application for use in optical elements.

Recently, photosensitive SGH materials doped with large amounts of photoactive molecules were found to exhibit larger refractive index and volume change on UV exposure without any etching step, a process called photoinduced self-developing.[12–14] The photosensitivity of these materials could be increased by the simple addition of photoactive molecules. In addition, these materials have the potential to be used in simple single-step photopatterning due to the simultaneous changes in both refractive index and volume. Thus, these photosensitive SGH materials are good candidates for highly efficient diffractive optical elements. However, the photopatterning of an optical element with a complicated shape, such as a FL, is generally based on the contact method that uses patterned photomasks; these contact methods require exact alignment between films and mask. Additionally, the photosensitive SGH materials require several treatments, such as predrying and severe contact before photopatterning, which reduce the film quality. Eventually, in the case of photosensitive materials, the fabrication of diffractive optical structures using contact methods has some limitations in obtaining higher efficiencies.

In this study, we thus sought to resolve problems involving the contact method and fabricate a high-efficiency Fresnel-type lens (FTL) on photosensitive SGH films by introducing a noncontact method named pinhole diffraction (PHD) imaging. In particular, the effect of the density of the photoactive molecule on photosensitivity has been investigated, with a focus on increasing the photosensitivity. Finally, we manufactured a well shaped FTL with high diffraction efficiency and good focusing properties by using PHD imaging without any etching treatment.

In our experiments, transparent photosensitive SGH films were prepared using methacryloxypropyltrimethoxysilane (Aldrich), perfluoroalkylsilane (Toshiba), and zirconium n-propoxide (Aldrich) chelated with methacrylic acid (Aldrich) as precursors, as described in previous reports for fabricating FTL.[12–14] All precursors were hydrolyzed with 0.01 N HCl. After 20 h stirring for full SG reaction, any residual products such as alcohols were removed at 50 °C with an evaporator. Benzyldimethylketal (BDK, Aldrich) as a photoinitiator, and methylmethacrylic acid (MMA, Aldrich) as a photoactive monomer, were added into the hybrid solution prior to the coating. Of particular note, the problem of the large content of solid state BDK could be solved by adding the same amount of liquid state MMA and increasing the sensitivity of SGH materials. After stirring the solution for 1 h at room temperature, a homogeneous photosensitive SGH solution was obtained. This solution was spin-coated on a cleaned glass substrate with 3000 rpm spinning speed. The coated films were exposed by using a He-Cd laser (wavelength 325 nm) and then consolidated by baking at 150 °C for 5 h.

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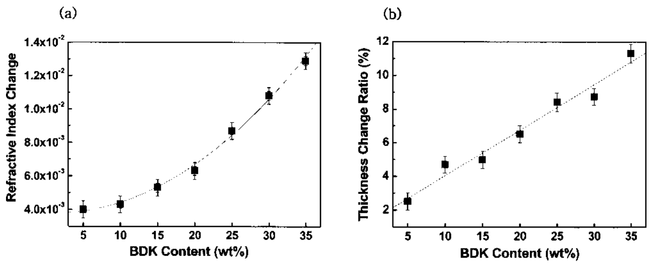

FIG. 1. Refractive index changes (a) and thickness changes (b) in UV exposed SGH film as a function of BDK content.

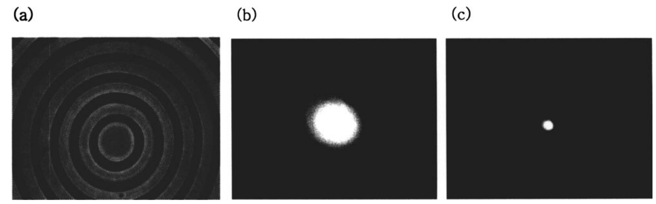

FIG. 3. (a) Optical micrograph of a patterned FTL having a focal length of 1 mm. (b) CCD image of collimated 632.8 nm light at focal plane (without patterned FTL). (c) CCD image of collimated 632.8 nm light at focal plane (with patterned FTL).

In order to achieve better performance from FTL, such as good focusing properties and higher diffraction efficiency, materials should have, above all, high photosensitivity. Thus, before fabricating and checking the performance of FTL, the effect of photosensitivity in the materials was characterized by increasing the photoactive molecule content on UV exposure. The changes in the refractive index and thickness of the film before and after irradiation with a He-Cd laser were measured by the prism coupler method using the 632.8 nm wavelength of a He-Ne laser. The optical energy density of the laser was 330 mJ/cm^2, the same as the UV dose used for fabricating the FTL. Figure 1 shows the changes of refractive index (a) and thickness (b) as a function of the photoactive molecule content. The changes in refractive index and thickness increased in proportion to the content of BDK and MMA. In order to achieve higher photosensitivity, the addition of large amounts of photoactive molecules was required. The limitation imposed by mixing the large amounts of solid state BDK with the SGH matrix could be solved reasonably well by adding liquid state MMA in the same quantity as the photoactive molecule. As shown in Fig. 1, the largest change of refractive index and thickness occurs in SGH materials having a BDK content of 35 wt % of SGH matrix, these being 1.3×10^{-2} and 11.3%, respectively, upon a UV dose of 330 mJ/cm^2, while the sensitivity ($S=\Delta n/E$) exhibited the high value of 3.9×10^{-2} cm^2/J. BDK and MMA in the exposed areas were fixed within the SGH matrix by various photoinduced reactions, followed by heating and drying. On the other hand, BDK and MMA in the unexposed area were volatile and easily removed by baking. After baking, the different characteristics of photoactive molecules between the UV exposed area and unexposed area make large changes in the refractive index and thickness, and these differences increased according to the increases in BDK and MMA content. These highly photosensitive materials are thus very good candidates for fabricating a high-efficiency FTL.

Fabrication of the FTL using the highly photosensitive SGH materials was attempted. Figure 2(a) illustrates the schematic configuration used to create PHD patterns, while Fig. 2(b) shows the PHD image. The PHD beam has circular diffraction patterns of different sized apertures, usually called the zone plates (ZP).[15] The shape of the PHD fringes exhibits almost same form as the FL and these fringes could be applied to make a FTL on SGH films without a patterned mask. A FTL is made by the spatially controlled irradiation of the PHD beam with periodic changing of the irradiation intensity. The most important factors in the experimental system are the PH size and the position of lens 2, which can control the respective aperture sizes of PHD patterns shown in Fig. 2(b). In our PHD method, the setup conditions, such as PH size and lens position, are easily changed, while the innermost and outer areas of PHD patterns, which are directly dependent on the focal length of the FTL, can be controlled. Therefore, the focal length of the FTL is easily controllable through this PHD method.

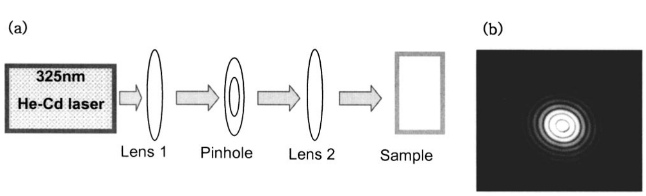

FIG. 2. (a) Schematic configuration for PHD patterns. (b) CCD image of PHD patterns.

Figure 3(a) shows an optical micrograph of a FTL patterned by PHD on photosensitive SGH films. The FTL exhibited the higher odd zones and lower even zones by the negative-type sensitivity depending on the spatially controlled exposure of PHD, which was related to the photoreactions between the SGH matrix and the photoactive molecules, including BDK and MMA within the exposed areas. These series of photoinduced reactions are related to various reactions, such as photopolymerization due to photodecomposition of BDK; photolocking, such as the reaction between BDK radicals and the attachment of BDK radicals to the matrix; and photomigration due to the concentration gradient of photoactive molecules between unexposed and exposed areas.[12–14,16]

For the FL different from the general diffraction gratings with regular diffraction angles and grating pitches, the diffraction angles and the periods between adjacent rings on the ZP are normally multiple and gradient, and lead to a lens with circular diffraction beams and various focal points. The primary point in respective focal points has the strongest and maximum intensity, and the diffracted beams of the FL eventually converge to near the primary focal spot by the diffraction effect. In this letter, the FTL fabricated by PHD method exhibited almost same diffraction and focusing property as a FL. As shown in Fig. 3(a), the first ring diameter of our FTL is around 40 μm and a focal length of our FTL experimentally measured by using a CCD camera is around 1 mm for the He-Ne laser ($\lambda=632.8$ nm). Figures 3(b) and 3(c) show the focusing property of a FTL from a CCD camera. The CCD camera connected to a computer was set at a distance of around 0.1 cm from the FTL. In the absence of the FTL, the He-Ne laser beam did not show any change of beam size, as shown in Fig. 3(b). In the presence of the FTL, however, the diffracted laser beams exhibited the focusing effect and were intensified, as shown in Fig. 3(c). A FTL with the short focal length of ~1 mm made by PHD would be potentially useful for application in compact optical systems.

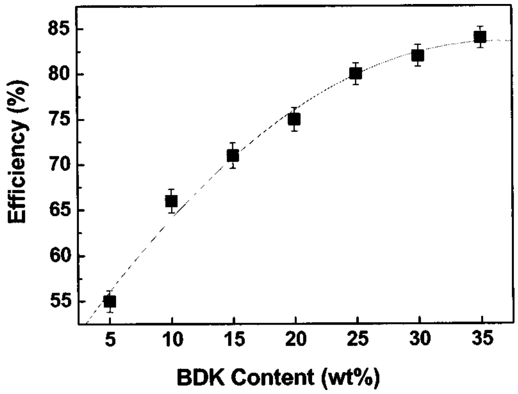

FIG. 4. Diffraction efficiency of FTL as a function of BDK content.

In order to evaluate the quality of our FTL, the diffraction efficiency was checked in addition to the focusing effect. The diffraction efficiency for a FL is defined as $\eta=(P*-P_o)/P_t$, where $P*$ is the transmission laser power at a focal point, P_o is the transmission laser power with no focusing, and P_t is the laser power passing through the sample.[8] For all material compositions, the diffraction efficiencies measured using the He-Ne laser ($\lambda=632.8$ nm) were highly dependent on UV dose and had the highest value at a dose around 330 mJ/cm^2, this being chosen for fabricating the lenses. Figure 4 shows the diffraction efficiency of FTL as a function of photoactive molecule content. The diffraction efficiency increased in proportion to the content of BDK and MMA. The enhancement of diffraction efficiency relied heavily on the change of photosensitivity in SGH films with the increase of photoactive molecule content. As shown in Fig. 4, diffraction efficiency was only around 50% at low concentrations of BDK (5 wt % of SGH matrix). However, the efficiency could be enhanced with an increase in the photoactive molecule content, and a very high efficiency approaching 85% was obtained for a BDK content of 35 wt % of the SGH matrix. This value is almost the highest value among the diffraction efficiencies of FTL reported to date. Our PHD technique using photosensitive SGH materials results in the large changes of refractive index and volume between odd and even zones on FTL, with no damage to the film surface, and the best surface uniformity, leading to a high-efficiency FTL. Moreover, the FTL fabricated using photosensitive SGH materials exhibited not the rectangular and binary-type index and volume changes, but the gradient index and volume changes between odd and even zones, and our FTL using SG system has the very high and stable transmittance for visible and IR wavelengths. The transmittance of SG film is greater than 90% for visible and IR wavelengths, which led to the high efficiency of above 80% of FTL. Thus, the diffractive optical elements fabricated using SG systems can have the high efficiency and the SGH film is suitable for various applications, including diffractive optical element and micro-optical components in optical communications.

In conclusion, the photosensitive SGH materials containing large amounts of photoactive molecule exhibited very high photosensitivity with large changes in both refractive index and volume on UV exposure. With the high photosensitive SGH materials, the simple fabrication of a high-efficiency FTL by PHD imaging is achieved. The PHD imaging is a noncontact method, which could reduce sample damage and simplify the fabrication process. Finally, the fabricated FTL showed good focusing performance and an efficiency as high as 85%.

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