Microlens arrays prepared via colloidal microsphere templating

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A simple and efficient templating method in combination with hot embossing technique is developed for fabricating large-area two-dimensional (2D) microlens arrays (MLAs) with uniform shape. By utilizing a modified microchannel method, a 2D large-area hexagonal close-packed (HCP) array of silica colloidal microspheres is prepared and serves as a template in the following hot embossing treatment to create a polycarbonate (PC) microcavity array. Then, with the obtained PC microcavity structure serving as a mold, a hot embossing process is applied to finally achieve a polymethylmethacrylate (PMMA) MLA. The effect of annealing time during the mold preparation process on the dimensions and shapes of the prepared microlens is investigated. The imaging performances of the prepared PC concave microcavities and PMMA convex microlenses are characterized by carrying out projection experiments. Our method provides a rapid and low cost approach to prepare large-area MLAs.

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Microlens has been studied for decades due to its important applications in imaging, optic communication, and biochemical detection[1−7]. For example, compact all-fiber zone plate microlens fabricated at the end of a mode-expanded hybrid fiber can help focus emergent light remarkably[8,9]. With a pair of microlens arrays (MLAs), pupil densification can be achieved with improving the imaging performance of the hyper telescope[10]. Moreover, multidirectional asymmetrical MLAs have been fabricated and served as light-control films for enhancing the imaging quality of refractive liquid-crystal displays[11]. Up to now, many different approaches have been reported to fabricate MLAs with particular shape and dimension, including lithography-assisted thermal reflow process[12−19], gray-scale lithography[20,21], all-liquid techniques[22−24], and mask-assisted focus-ion-beam lithography[25,26]. However, the aforementioned methods more or less require complex experimental processes or expensive equipments. By contrast, the self-organization technique, commonly used for preparing masks or templates[27−29], has provided a new way for fabricating MLAs with low cost and simple procedure. For example, Yabu *et al.*[30] have reported the preparation of hexagonally packed micropores structure by using self-assembled condensed water droplets as templates. Spherical MLAs have been successfully fabricated by directly molding from the resulting 'honeycomb' structure.

In this letter, we present a simple and efficient templating method to fabricate large-area MLAs with uniform shape. Our method comprises two processes which are mold preparation and subsequent hot embossing. We show that the size control of the fabricated microlens can be achieved by varying the annealing time during mold preparation process or the size of the colloidal particles. Imaging performances of both polycarbonate (PC) concave and polymethylmethacrylate (PMMA) convex microlens prepared are further characterized and the image resolution is improved through tuning the dimensions of microlens using our method.

The schematic of our fabrication method is shown in Fig. 1, which involves several steps. Firstly, a PC substrate was cleaned with distilled water and then immersed into a sodium dodecyl sulfate aqueous solution (1.0 wt.-%) to make its surface more hydrophilic. In our experiment, PC chips were chosen as the mold material since it had a relatively high glass transition temperature ($T_g = 149$ °C), excellent deformation resistivity, and low elasticity. Afterwards, monodisperse silica microspheres with diameter of 1.59 μm (size dispersion of 3%, Duke Company, USA) or 5 μm (size dispersion of 12%, Duke Company, USA) were self-assembled to a hexagonal close-packed (HCP) array on the surface of the PC substrate, utilizing a modified microchannel method[31−33]. In brief, a small volume of aqueous solution of colloidal dispersion with a suitable concentration was injected into a channel that was formed from the PC substrate and a glass slide separated by a U-shaped spacer. After drying in air, highly ordered silica colloidal crystals were grown within the channel under capillary force. Subsequently, the PC substrate together with silica colloidal crystal was put into an oven and heated at 155 °C (above T_g of PC) for certain time to let the silica template embed into the PC substrate surface. After cooling down to room temperature, the silica microspheres were dissolved by using a hydrofluoric acid (HF, 40%) solution, leaving an array of segmented spherical microcavities on the PC surface. Then, a PMMA film was brought into close contact with the patterned side of PC mold and the sample was sandwiched by two separate glass slides. To flatten the distribution of pressure, a piece of filter paper could be added on the back surface of the PC mold. The clamped whole structure was put into the oven again and heated up to 120 °C for 30 min to ensure an adequate pattern transfer. As this annealing temperature is above T_g of PMMA ($T_g = 105$ °C) but below that of PC, the as-prepared PC substrate can serve as a rigid mold. After cooling down to room temperature and carefully peeling off the PC mold, a HCP replica of hemispherical PMMA

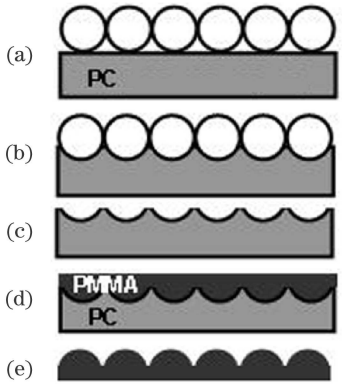

Fig. 1. Schematic of the fabricating processes. (a) Self-assembly of silica microspheres on the surface of the PC substrate; (b) embedding of silica colloidal crystal under heat treatment; (c) forming of PC microcavity arrays after dissolving the silica microspheres by a HF solution; (d) a pattern transfer achieved by using hot embossing technique; (e) a HCP replica of hemispherical PMMA convex MLAs obtained after peeling off the PC mold.

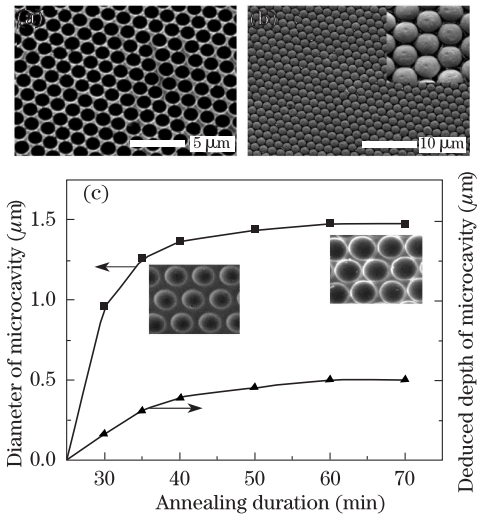

Fig. 2. SEM images of (a) PC microcavity arrays and (b) PMMA MLAs, both fabricated from 1.59-μm-diameter silica microspheres. The inset in (b) shows a magnified image of the fabricated PMMA MLAs, which demonstrates the uniform hemispherical shape. (c) Measured diameter and deduced depth of the microcavities as functions of annealing time during mold preparation process under the annealing temperature of 155 °C. The insets are the SEM images of the microcavity arrays formed on PC surface after annealing the sample for 35 min (left) and 60 min (right), respectively.

MLAs was finally obtained. Note that only silica colloidal crystal was sacrificed during the fabrication processes and the PC mold could be repeatedly used without any deformation or contamination owing to the "self-cleaning" characteristic of hot embossing process.

Figure 2 shows the scanning electron microscope (SEM, FEI Philips XL-30, Holland) images of PC microcavity arrays and PMMA MLAs which are fabricated from 1.59-μm-diameter silica microspheres. It is clearly seen from Fig. 2(a) that the periodicity and uniform shape of the original template are well reserved in the PC mold and the fabricated PMMA MLAs preserve all the structural features from the PC mold except for a slight shape deformation for some microlenses, which may be attributed to the local shrinkage[34] of the PC and PMMA during cooling process, as shown in Fig. 2(b).

In our method, the dimensions of PC microcavities (or the corresponding PMMA microlenses) can be tuned by monitoring several parameters. Firstly, larger size silica microspheres result in microlenses with the larger diameter and height. Secondly, the annealing temperature during mold preparation process is also an important parameter to control the microlens structural parameters. Since very high annealing temperature causes severe deformation of PC mold, the annealing temperature was kept at 170 °C or below. In such cases, the maximal diameter and depth that microcavities can reach are rather limited and actually independent of the annealing duration. In other words, in order to form a microcavity with certain diameter and depth on PC surface, slight falling of the annealing temperature could only be compensated by tremendously increasing the annealing duration, which is unreachable in our experiment. Therefore, we choose to tune the annealing time during mold preparation process, under a moderate annealing temperature of 155 °C to achieve the size control of microcavities and microlenses.

Figure 2(c) shows the measured diameter and deduced depth of PC microcavities created using 1.59-μm-diameter silica microspheres, under the annealing temperature of 155 °C as a function of the annealing duration. It can be seen that no embedding happens when the annealing duration is less than 25 min. However, the dimensions of microcavities increase rapidly in the next 15 min. When the annealing time is set to 40 min, the diameter and depth of the microcavities can reach 1.37 and 0.39 μm, respectively, forming a non-close-packing array of microcavities. Further increasing this duration to about 60 min or longer, the structural parameters increase slowly, especially for the microcavity diameter (see the SEM image in the right inset of Fig. 2(c)). This can finally lead to a structure having hemispherical shape of the individual microcavity in a HCP arrangement. It is noted that the maximal embedding depth which can be achieved using the present method is equal to about half diameter of the silica microspheres (1.48 μm), which is independent of the annealing duration. Such an embedding behavior is not clear yet although similar embedding behavior of the gold nanoparticles into polymer surface has been reported by Teichroeb *et al.*[35] By using the viscoelastic contact mechanics (VCM) theory, Hutcheson *et al.* showed that the surface tension interactions between gold nanospheres and polymer surface, instead of the gravity or buoyant forces, contributed mainly to the semi-embedding behavior of gold nanospheres into polymer surface[36]. In our cases, however, the gravity of silica microspheres is much larger than that of gold nanospheres, and thus is not negligible. We think that the limited embedding behavior of silica microspheres could not be explained simply by the VCM theory, and the competition between the embedding driving forces and the resisted surface tension should be considered. Further investigation is still needed to fully understand this behavior.

In order to characterize the imaging performance of the fabricated microlens, a projection experiment was carried out using an optical microscope (Olympus BX51,

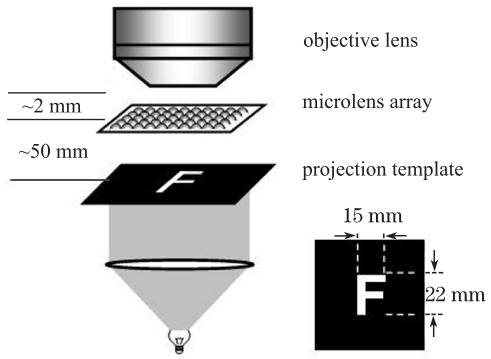

Fig. 3. Schematic of the projection experiment. The projection object is a transparent letter 'F' with a size of 15×22 (mm).

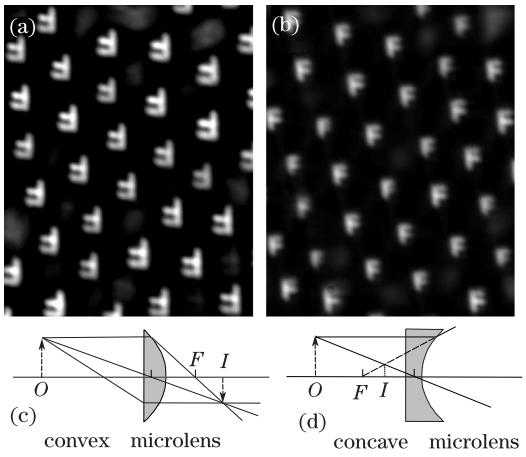

Fig. 4. Comparison of the optical micrographs of the images projected through (a) PMMA convex microlenses and (b) PC concave microlenses. (c), (d) Beam paths for (c) the convex lens and (d) the concave lens, respectively.

Japan) with a charge-coupled device (CCD) camera. As schematically shown in Fig. 3, a transparent letter 'F' with a size of 15 × 22 (mm) notched in a piece of black hardboard is taken as the projection object. The fabricated MLAs are placed on the sample stage of the optical microscope and illuminated with a light source through this letter 'F'. Miniaturized images projected through the microlenses can be captured by the CCD camera through objective lens.

As an illustration, we demonstrate the imaging performance of the hemispherical microlens fabricated from silica microspheres of 5 μm in diameter. Miniaturized images of the inverted letter 'F' projected through the PMMA convex microlenses are clearly identified in Fig. 4(a), and good imaging performance of the fabricated microclens is confirmed. On the other hand, both convex and concave shaped lenses can focus light beam (or its backward-extension line) emitting from a single spot light source, as the simple beam path shown in Figs. 4(c) and (d). Owing to the high luminousness and low haze, the PC microcavities can also serve as concave microlenses (with a negative curvature). From Fig. 4(b), miniaturized images of an upright letter 'F' with high contrast and clear profile are observed. It is noted that some miniaturized images are a little bit obscure due to the slight deformation

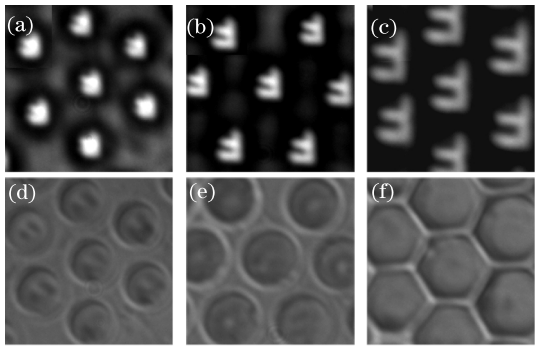

Fig. 5. Comparison of the inverted letters 'F' projected through the convex microlenses with diameters of (a) 3.25, (b) 4.12, and (c) 5.0 μm, respectively. (d)–(f) Optical micrographs of three microlenses corresponding to (a)–(c) where the unclear edges are due to the optical diffractions.

of polymer materials during mold preparation, hot embossing, and demoulding processes.

For a PMMA convex microlens, the focal length f is predicted from the following relation[37]:

$$f = \frac{n_{\rm a}}{n_{\rm L} - n_{\rm a}} \cdot R_{\rm C} = \frac{n_{\rm a}}{n_{\rm L} - n_{\rm a}} \cdot \frac{h^2 + (D/2)^2}{2h}, \quad (1)$$

where $R_{\rm C}$, D, and h are the radius of curvature, diameter, and height of the microlens; $n_{\rm L}$ and $m_{\rm a}$ are the refractive indices of lens material and ambient medium, respectively. In our case, the lens material is PMMA of which the refractive index is 1.49 and the ambient medium is air. Figure 5 shows the optical micrographs and imaging pictures of the three PMMA convex microlenses with different dimensions, all fabricated using 5-μm-diameter silica microspheres but under different annealing durations. An improvement in image resolution can be identified from Figs. 5(a)–(c), when the diameter of microlens was increased from 3.25 to 5.0 μm. In fact, the image resolution σ is determined by

$$\sigma \propto \frac{\lambda}{\rm NA}, \quad (2)$$

where λ is the wavelength of the projected light, and $\mathrm{NA} = n_{\rm L} \sin\theta = n_{\rm L} \frac{D/2}{\sqrt{(D/2)^2 + f^2}}$ is the numerical aperture of the microlens. According to Eq. (1), a focal length f is calculated to be about 5.1 μm for all the three microlenses. This is in good agreement with our experimental results (4.9–5.1 μm), which are measured by the optical microscope. Therefore, according to Eq. (2) and the expression of NA, microlens with bigger diameter D will have bigger NA, which results in an improvement in the image resolution.

In conclusion, we have successfully fabricated MLAs by using a simple templating method in combination with hot embossing technique. By varying the annealing duration, the size control of the fabricated MLAs is realized. Good imaging performances of both microcavities and microlens are demonstrated in experiment. Our method should provide an alternative approach towards large-area MLAs.

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