Maskless fabrication of concave microlens arrays on silica glasses by a femtosecond-laser-enhanced local wet etching method

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Abstract: A simple and efficient technique for large-area manufacturing of concave microlens arrays (MLAs) on silica glasses with femtosecond (fs)-laser-enhanced chemical wet etching is demonstrated. By means of fs laser in situ irradiations followed by the hydrofluoric acid etching process, large area close-packed rectangular and hexagonal concave MLAs with diameters less than a hundred of micrometers are fabricated within a few hours. The fabricated MLAs exhibit excellent surface quality and uniformity. In contrast to the classic thermal reflow process, the presented technique is a maskless process and allows the flexible control of the size, shape and the packing pattern of the MLAs by adjusting the parameters such as the pulse energy, the number of shots and etching time.

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1. Introduction

Microlenses arrays (MLAs) are important optical devices due to their diverse applications in
optical systems, micro-manufacturing and biochemical systems [1–4]. Numerous researchers
have poured into lots of enthusiasm to the manufacturing of MLAs. However, the majority of
well-established techniques, such as thermal reflow [5,6], hot embossing [7], droplet method
[8], gray-tone (GT) photolithograph and laser direct writing (LDW) [9,10], are not suitable for
the fabrication of concave or negative MLAs, which have extensive applications such as
diffusers, compound refractive lens for focusing X-rays and carriers of DNA chips [11–13].

During the last decade, some techniques for the fabrication of concave MLAs have been
proposed. For example, a concave refractive microlens array was fabricated in solgel glasses
by a proximity-effect-assisted reflow technique, which was proposed in 2004 [14]. Later on,
Ruffieux and associates developed a two-step process for fabrication of diffraction limited
concave microlens arrays [11]. First, cylindrical holes were produced by the photolithograph
method followed by a melting step preformed at 150°C for half an hour; the melted structures
were then filled by a second spinning step of photoresist. Other techniques such as 3D diffuser
lithograph and “breath figures” method [15,16], which were employed to fabricate concave
molds, can also fabricate the concave MLAs. Moreover, through the reversal replication
technique, the concave MLAs could be produced by the convex MLAs. We notice that most
approaches to the fabrication of concave MLAs are based on the process using the
photomasks, which are very expensive. The maskless processes such as LDW are complex
and inefficient, which are not suitable for the large area fabrication of concave MLAs. Herein,
a simple, high-efficient maskless technique for the concave MLAs is developed using a
femtosecond (fs) laser-induced crater arrays followed by a chemical etching process. This
method simplifies the classic laser etching process [17,18], improving the fabrication

efficiency significantly. In addition, it allows direct manufacturing of various concave MLAs
on glasses, which have better physical and chemical properties than the photore sist,
poly(methylmethacrylate) (PMMA) or poly(dimethylsiloxane) (PDMS), and more importantly,
the reflection loss of lights caused by the interfaces between the polymer layers and substrates
is not exist. Other advantages, such as simple process, facile processing environment and
flexible control of the size, the packing pattern and the shape of the MLAs by adjusting some
parameters, are also demonstrated.

As an example, a rectangular and a hexagonal microlens array are fabricated and the
fabrication process is described in section 2. The morphology and three-dimensional profiles
of the MLAs are evaluated by a field-emission scanning electron microscope (FE-SEM, JEOL
JSM-7000F) and a laser confocal scanning microscope (LCSM, Olympus LEXT OLS4000) respectively.
The optical properties of the MLAs, such as the focal length and imaging
abilities, are also tested. The results are presented in Section 3. In Section 4, the relationship between the profile of the microlenses and the processing parameters such as chemical etching time, \( \tau \), the pulse energy, \( E \), and the number of shots, \( N \), is discussed. Finally, the conclusion is given in Section 5.

### 2. Fabrication process

The rectangular and hexagonal-packed concave MLAs are fabricated by a three-step process, as depicted in Fig. 1. Initially, ablation-induced craters with diameters of a few micrometers are induced on polished silica glass chips (10 \( \times \) 10 \( \times \) 1 mm\(^3\), China Daheng Group Inc., GCL-1202) using a 30-fs and 800-nm laser pulses at a repetition of 1 kHz (the laser source is a Ti:sapphire pulsed laser oscillator-amplifier system). The femtosecond laser, owing to its advantages of negligible thermal and shockwave-induced damages [19], when focused by an objective lens (NA = 0.5), can easily induce craters on transparent materials such as silica glasses without melting-ejections and cracks which will impact on the morphology of the fabricated microlenses. The diameter of the focal spot is about 1.4 \( \mu \)m (1/e). The pulse energy can be varied by a variable neutral density filter and the number of shots is controlled by a shutter. More details of the setup used here can be found in Ref [20]. Subsequently, the samples with craters are treated in 5% hydrofluoric (HF) acid solution assisted by an ultrasonic bath at 23°C. During this process, the chemical etching velocity is accelerated in the laser-induced craters and the concave spherical surfaces begin to form; the MLAs are consequently fabricated in tens of minutes. Finally, the samples are cleaned by the ultrasonic bath in acetone, alcohol and deionized water for 15 minutes, respectively, and dried in ambient air.

![Fig. 1. Schematic of the fabrication process. First, the laser pulses are focused by an objective lens, inducing crater arrays on the sample surface. Then, the sample is treated by ultrasonic-assisted HF etching. Finally, the sample is cleaned by the ultrasonic bath in acetone, alcohol and deionized water, respectively. The inset presents the 3D profile of the rectangular microlens array.](image)

The employed processing parameters for the rectangular and hexagonal MLAs, including the pulse energy, \( E \), the number of shots, \( N \), the chemical etching time, \( \tau \), and the distance between adjacent microlenses, \( L \), are listed in Table 1. We note that the size, the packing pattern and the profile of the MLAs are close-related to these parameters, which will be discussed in Section 4.
Table 1. Processing parameters used for the rectangular (Rec.) and hexagonal-packed (Hex.) MLAs

<table>
<thead>
<tr>
<th></th>
<th>E</th>
<th>N</th>
<th>L</th>
<th>t</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rec.</td>
<td>1.5 μJ</td>
<td>500</td>
<td>60 μm</td>
<td>90 min</td>
</tr>
<tr>
<td>Hex.</td>
<td>0.6 μJ</td>
<td>500</td>
<td>30 μm</td>
<td>45 min</td>
</tr>
</tbody>
</table>

3. Results

In the experiment, the whole fabrication process is monitored by an optical microscope (OM) equipped with a CCD camera. Figure 2 shows the evolution of the rectangular and hexagonal MLAs changing from the laser-induced crater arrays. In the beginning, the isotropic chemical etching occurs in the laser treated points [black dots shown in Figs. 1(a) and 1(d)], producing circular-shaped concave structures in the rectangular [Fig. 1(b)] or hexagonal [Fig. 1(e)] patterns, which can serve as the circular MLAs. Then the aperture diameter of the circular microlenses expands gradually with the chemical etching, and eventually, the adjacent ones “overlapped” with each other, resulting in the formation of the tetragonal and hexagonal shaped microlenses, as shown in Figs. 1(c) and 1(f). It demonstrates that the packing pattern and the shape of the microlenses can be easily controlled by the arrangement of the laser irradiated points and the chemical etching time. The areas of the fabricated rectangular and hexagonal MLAs are about $3 \times 3 \text{ mm}^2$ and $1.5 \times 1.5 \text{ mm}^2$, respectively, and the whole processing time is about 3 hours, which is more efficient than the LDW process [20]. Figures 3(a) and 3(b) show the SEM images of the rectangular and hexagonal MLAs, respectively. They visually express the excellent surface quality and uniformity of the MLAs.

Fig. 3. FE-SEM images of (a) the rectangular microlens array and (b) hexagonal microlens array.

The three-dimensional (3D) morphology of the MLAs is observed by the LCSM. Figure 4 presents the 3D and the cross-section profile of the rectangular [Figs. 4(b) and 4(a)] and the hexagonal MLAs [Figs. 4(c) and 4(d)]. The measurement results show that the diameter, $D$, of...
the former one is 67.05 µm, and the sag depth, \( h \), is 10.68 µm. For the latter one, \( D \) and \( h \) are 30.54 µm and 3.35 µm, respectively. By a primary equation: 
\[ R = \frac{h^2 + r^2}{2h} \] 
where \( r \) is the radius of the microlens (\( D/2 \)), the values of the curvature radius, \( R \), can be figured out and the results are 57.96 µm and 36.48 µm for the rectangular and hexagonal MLAs, respectively. In addition, the arithmetic-mean surface roughness value, \( R_A \), is measured in an area of 258 × 258 µm² for both rectangular and hexagonal MLAs and the results are 71 nm and 48 nm, respectively. The values of surface roughness obtained in this work are higher than those fabricated by the reflow techniques, but are much smaller than the microlenses fabricated by the femtosecond laser ablation, which was about 200 nm [20].

![Fig. 4. The results of 3D measurements of the MLAs. (a) and (b), the cross-section and the 3D profiles of the rectangular microlens array. The aperture diameter, \( D \), and the sag height, \( h \), of the microlens array are about 67.05 µm and 10.28 µm, respectively. (c) and (d), the 3D and cross-section profiles of the hexagonal-packed microlens array. \( D = 30.54 \) µm, \( h = 3.35 \) µm.](fig4.png)

![Fig. 5. Schematic of the optical system for the measurement of the focal length. The inset (bottom) is the CCD images of the sample surface (\( z = 0 \) µm) and the focal point (\( z = 84 \) µm).](fig5.png)
To evaluate the focal length of the MLAs, \( f_{\text{exp}} \), an optical system equipped with a He-He laser (633 nm), a computer-controlled stage, a lens and a CCD camera, is built up, as shown in Fig. 5. Moving the computer-controlled stage along the direction parallel to the laser beam (z-axis), the position of the top surface and the focal point can determined by the images captured by the CCD camera. The values of \( f_{\text{exp}} \) are obtained by equation, \( f_{\text{exp}} = L - h \), where \( L \) is the distance between the focal point and the top surface of the MLAs. The focal length of the rectangular and the hexagonal MLAs is 125.0 ± 5 \( \mu \)m and 84.0 ± 5 \( \mu \)m, respectively. These values match well with the calculated focal length, \( f_{\text{cal}} \), which are obtained by equations:

\[
f_{\text{cal}} = \frac{R}{n - 1}
\]

where \( n \) denotes for the refractive index of the silica glass at wavelength of 633 nm. Considering \( n = 1.45 \), the values of \( f_{\text{cal}} \) are 128.80 \( \mu \)m and 81.06 \( \mu \)m for the rectangular and hexagonal MLAs. Furthermore, the values of numerical aperture, \( \text{NA} \), for both MLAs are calculated by equation: \( \text{NA} = \frac{D}{2f} \), and the results are 0.26 and 0.19, respectively. The experimental results are summarized in Table 2.

### Table 2. The measurement results of the MLAs

<table>
<thead>
<tr>
<th></th>
<th>( D (\mu m) )</th>
<th>( H (\mu m) )</th>
<th>( R (\mu m) )</th>
<th>( f_{\text{cal}} (\mu m) )</th>
<th>( f_{\text{exp}} (\mu m) )</th>
<th>( \text{NA} )</th>
<th>( \text{Ro} (\text{nm}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rec.</td>
<td>67.05</td>
<td>10.68</td>
<td>57.96</td>
<td>128.80</td>
<td>125.0 ± 5</td>
<td>0.26</td>
<td>71</td>
</tr>
<tr>
<td>Hex.</td>
<td>30.54</td>
<td>3.35</td>
<td>36.48</td>
<td>81.06</td>
<td>84.0 ± 5</td>
<td>0.19</td>
<td>48</td>
</tr>
</tbody>
</table>

Figure 6 shows the imaging ability of the rectangular [Fig. 6(a)] and the hexagonal MLAs [Fig. 6(b)]. A mask with letters, “FE”, was fabricated by the focused femtosecond laser pulses on a glass-based 100-nm Au thin film. Then it was inserted between a tungsten light source and the MLAs. Consequently, reduced false images were captured by an objective lens (NA = 0.3) and the CCD camera which are placed on the other side of the MLAs, as shown in Fig. 6. The entire width of the images for rectangular and hexagonal microlens array is 8.9 \( \mu \)m and 11.7 \( \mu \)m, respectively.

![Fig. 6. The false images captured by the OM system for (a) rectangular microlens array and (b) hexagonal microlens array.](image)

### 4. Discussions

It has been mentioned that the profile of the microlens can be controlled by the processing parameters, such as \( \tau \), \( E \) and \( N \). To investigate the profile revolution of the microlenses versus the chemical etching time, \( \tau \), groups of craters are induced on the silica glass at \( E = 1.1 \mu \)J and \( N = 500 \), and they are treated by the 5% HF acid solution for different times. For each group, the aperture diameter, \( D \), and the sag height, \( h \), are the average values calculated by ten microlenses. The etching time dependency of \( D \) and \( h \) are plotted in Fig. 7(a) and 7(b), respectively. It is easy to see that \( D \) and \( h \) increase with the increase of \( \tau \) and the increasing process consist of two different parts. For the further analysis, the two parts are fitted exponentially and linearly, and the curves are shown in Figs. 7(a) and 7(b). They can be expressed by Eqs. (4) and (5):
\[ D = -22.76\exp\left(-\frac{\tau}{11.69}\right) + 26.04 \text{ (\mu m)} \quad \tau \in [0, 20 \text{ min}) \]  
\[ D = 0.49\tau + 11.92 \text{ (\mu m)} \quad \tau \in [20 \text{ min}, +\infty) \]  
\[ h = \exp\left(-\frac{1.37}{\tau + 0.62} + 1.82\right) \text{ (\mu m)} \quad \tau \in [0, 20 \text{ min}) \]  
\[ h = 0.002\tau + 6.17 \text{ (\mu m)} \quad \tau \in [20 \text{ min}, +\infty) \]  

In addition, the variation ratio of the aperture diameter and sag height versus, \( V_D \) and \( V_h \), are given by the derivation of the \( D \) and \( h \):

\[ V_D = \frac{\partial D}{\partial \tau} = 1.95\exp\left(-\frac{\tau}{11.69}\right) \text{ (\mu m/min)} \quad \tau \in [0, 20 \text{ min}) \]  
\[ V_D = \frac{\partial D}{\partial \tau} = 0.49 \text{ (\mu m/min)} \quad \tau \in [20 \text{ min}, +\infty) \]  
\[ V_h = \frac{\partial h}{\partial \tau} = \frac{1.37}{(\tau + 0.62)^2}\exp\left(-\frac{1.37}{\tau + 0.62} + 1.82\right) \text{ (\mu m/min)} \quad \tau \in [0, 20 \text{ min}) \]  
\[ V_h = \frac{\partial h}{\partial \tau} = 0.002 \text{ (\mu m/min)} \quad \tau \in [20 \text{ min}, +\infty) \]  

From the figures and the equations, the etching processes for both \( D \) and \( h \) can be divided into the early stage (0-20 min) and the later stage (after 20 min) by the critical time \( \tau = 20 \text{ min} \). At the early stage, both \( D \) and \( h \) increases with the increase of etching time, but the variation ratio, \( V_D \) and \( V_h \), are decreasing. For example, when \( \tau = 1 \text{ min} \), the values of \( V_D \) and \( V_h \) is 1.79 \( \mu m/\text{min} \) and 1.38 \( \mu m/\text{min} \), respectively, which are much higher than that after 20 min. At the later stage, \( D \) increases linearly with \( \tau \) at a velocity of 0.49 \( \mu m/\text{min} \). This value is close to the etching velocity of untreated silica glasses in 5\% HF solutions at room temperature. However, the value of \( h \) stops increasing and kept to about 6 \( \mu m \) at this stage (\( V_h = 0.002 \mu m/\text{min} \)). This is because at the early stage, the etching velocity of the bottom of the microlenses is higher than that of the top surface, and \( h \), which is the distance between the top and the bottom surfaces, keep increasing. When \( \tau \geq 20 \text{ min} \), both velocities are identical; and therefore \( h \) keeps a constant. It means that the value of \( h \) for a microlens is determined by its early stage of etching process, and the prolongation of the etching process will enlarge the curvature radius of the microlens because the aperture diameter keeps increasing and sag height keeps constant.

![Fig. 7](image_url)

**Fig. 7.** The evolutions of (a) the aperture diameter, \( D \), and (b) the sag height, \( h \), of the microlens versus the chemical etching time, \( \tau \).

We believe that the enhancement of the etching velocity for both \( D \) and \( h \) at the early stage of the chemical wet etching process is associated with the fs-irradiation-induced modifications of materials [21–23]. Figure 8(a) shows the morphology of an untreated crater irradiated by 500 pulses at \( E = 1.1 \mu J \). Nanostructures such as the parallel grooves and protrusions can be
observed in the crater. These structures and defects will increase the interface of the samples surface and HF solutions, accelerating the etching process in the crater. The modified regions will be gradually etched by the chemical corrosion. The diameter and depth of the concave structure are increasing and the surface roughness decreases until $\tau = 20$ min, when the modified materials are completely removed. At $\tau = 20$ min, as shown in Fig. 8(d), the roughness of the bottom surface is close to the top surface and the chemical etching process come into the later stage. When $\tau = 50$ min, a fine circular-shape concave microlens forms, as shown in Fig. 8(f). The eventual shape of the microlens seems to be independent with the original profile of the laser-induced craters. So to speak, rigorous demands for the morphology of the craters are unnecessary, which is facile to be obtained.

![Fig. 8. FE-SEM images of the laser-induced craters (a) before the HF treatment and treated for (b) 1 min, (c) 5 min, (d) 20 min, (e) 30 min and (f) 50 min. Note the different scale bars.](image)

Figure 9 presents the influence of the profiles of the microlenses on the pulse energy, $E$, and the number of shots, $N$. Here, $D$ and $h$ are also the average values obtained from ten microlenses treated by the 5% HF solutions for 120 min. We can see that $D$ and $h$ of the microlenses increase with the increase of $E$, as shown in Fig. 9(a) and 9(b) respectively, but they are not strongly dependent on $N$, which are present in Fig. 9(c) and 9(d). The curvature radiuses of these microlenses are figured out by the Eq. (1) and its dependency of $E$ and $N$ are spotted and fitted in Figs. 9(e) and 9(f). The maximum values of $R$ tend to be obtained at the intermediate pulse energies (3.0-4.5 $\mu$J), as demonstrated in Fig. 9(e). It is resulted from the different increase amplitude of $D$ and $h$ versus $E$. Take $N = 1000$ as an example, see Figs. 9(a) and 9(b), when $E = 0.36 \mu$J, the aperture diameter of the fabricated microlens is about $45 \mu$m, while $E$ increases to $7.39 \mu$J, $D$ increases to about $90 \mu$m correspondingly, thus the increase amplitude of $D$ is about 100%. However, this value is about 400% for $h$. According to the Eq. (1), the maximum $R$ for $N = 1000$ is obtained when $E$ is about $3.5 \mu$J. On the other hand, the values of $D$ and $h$ gradually reach saturation when the number of shots is more than 1000, as shown in Figs. 9(c) and 9(d), respectively. Consequently, the curvature radius of the microlenses has little relationship with $N$, as depict in Fig. 9(f). It is worth to mention that the increase of $N$ is benefit for reducing the impact of the energy instability between the laser pulses and improving the uniformity of the MLAs, but it will also increase the processing time. The employed number of shots, $N = 500$, is an optimized value in our experiments.

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5. Conclusion

To summarize, a femtosecond-laser-enhanced local chemical etching technique for efficient fabrication of concave MLAs on silica glasses has been demonstrated. Within 3 hours, a rectangular and a hexagonal microlens array with diameters of 67.05 µm and 30.54 µm were successfully manufactured in areas of 3 × 3 mm$^2$ and 1.5 × 1.5 mm$^2$, respectively. We believe that the femtosecond-laser-induced modification of the materials cause the enhancement of the chemical etching, resulting in the formation of concave structures. The profile of the microlens could be flexibly controlled by the chemical etching time. The diameter of the microlens, D, keeps increasing with the chemical etching process while the sag height, h, stops increasing at a critical time. Therefore, the curvature diameter of the microlens, R, increases with the chemical etching. In addition, D and h are also increase with the increase of
the pulse energy. Correspondingly, the maximum value of $R$ can be obtained at the intermediate energies.

Acknowledgments

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