Organic-inorganic-hybrid-polymer microlens arrays with tailored optical characteristics and multi-focal properties

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Abstract: Plano-convex microlens arrays of organic-inorganic polymers with tailored optical properties are presented. The fine-tuning of each microlens within an array is achieved by confining inkjet printed drops of the polymeric ink onto pre-patterned substrates. The lens optical properties are thus freely specified, and high numerical apertures from 0.45 to 0.9 and focal lengths between 10 μm and 100 μm are demonstrated, confirming theoretical predictions. Combining nanoimprint lithography approaches and inkjet printing enables using the same material for the microlenses and their substrates, improving the optical performances. Microlens arrays with desired specifications are printed reaching yields up to 100% and high lens reproducibility with standard deviations of the apparent contact angle under 1° and of the numerical apertures and focal lengths under 6%. Microlens arrays involving lenses with different characteristics, e.g. multi focal length, and thus focal planes separated by only few microns are printed with the same reproducibility.

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1. Introduction

The demand of micro optical elements, such as microlens arrays (MLA) with specific and precise lens features, has increased in the last decades in very broad fields of application such as Light-Emitting Diode (LED) displays [1], Shack-Hartmann wave front sensors, photolithography [2], biomedical technologies [3], optical interconnects [4] and more recently optical security features [5]. Usually these applications rely on regular MLAs having lenses with the same geometry and characteristics. However, recently, some specific applications, such as multi-focal imaging with focal depth increase, require microlens arrays integrating lenses with different characteristics, i.e. different focal lengths ($f$) within the array [6]. The fabrication for curved micro-optical structures is truly challenging and traditional fabrication methods have no cost- and time-efficient solution to address this need. A standard fabrication process of plano-convex MLAs usually starts by a thermal reflow process of positive tone photoresist (PPR) [7], typically followed by a pattern transfer into the designated MLA material, i.e. by reactive ion etching (RIE) into glass [8,9], whereby the lens profile has been thoroughly investigated [3]. Subsequent lens replications also allow inversing the polarity obtaining thus concave profiles [10]. These MLAs fabrication paths allow large scale parallel processing but fine tuning of each individual lens independently remains a challenge. In addition, the aforementioned approach requires replica molding for optically good microlenses, aspect-ratio limitations or additional process steps to reach MLAs with different lenses [3,11]. Other alternatives are direct laser writing of microlenses, focused ion beam, two photon polymerization or laser ablation, but none of these methods is suitable for direct integration into functional systems without affecting the other parts of the device.

Inkjet printing (IJP) as an additive manufacturing technique was proposed for microlens fabrication and gained interest due to its simplicity [12–14] and to its compatibility with Roll-to-Roll (R2R) techniques. As a local and on-demand deposition technique, IJP works with very low material consumption permitting the use of hazardous or costly solutions and inks [12]. The fabrication of plano-convex spherical microlenses by IJP is very efficient due to surface energy minimization, resulting in extremely low lens surface roughness [15]. Furthermore, as a non-contact mode, IJP can be applied to a broad range of substrates, such as non-planar, previously structured [16] or even already functional devices requiring focal adjustment to optimize their performance without affecting neighboring areas - i.e. LED displays [17,18]. In addition, setting and specifying concrete optical performance can be done by confining the spherical microlens footprints [19,20]. This confinement is possible either chemically based on local surface energy modification, such as by micro-contact printing [21] or topographically based on pre-patterned substrates [22]. Based on IJP dispensing, the minimum rim height of protrusive platforms with vertical side walls allowing liquid confinement has been demonstrated to be slightly lower than 1 $\mu$m [23]. This principle has been successfully applied for the fabrication of arrays of plano-convex lens-like structures with controlled spherical profiles [22,24]. Nevertheless, the associated literature still suffers from gaps, such as the fabrication of MLAs with different specific lens characteristics into the same MLA with high optical-quality and the analysis of their optical behavior. The use of more sophisticated contours enlarges even further the possible microlens shapes and corresponding foci, and thus the range of applications [16].

Recently, these technological advances have been accompanied by the development of various kinds of inks optimized for IJP technology and having excellent optical properties, such as InkEpo or InkOrmo (micro resist technology GmbH) [25,26]. After cross-linking, InkOrmo, the IJP-compatible version of OrmoComp® exhibits excellent transparency in the near UV and visible wavelength range with an absorption coefficient below $1 \cdot 10^{-4}$ $\mu$m$^{-1}$ in the visible range [27]. In combination with its non-yellowing behavior, its high thermal, mechanical and chemical stability makes InkOrmo more suitable for permanent applications.
than epoxies, used for similar purposes. The unique compatibility of the UV-curable hybrid polymer OrmoComp® to several micro- and nanofabrication technologies, including nanoimprint lithography (NIL) [28] and IJP, is an essential benefit since it allows combining these manufacturing paths for improving further optical performances and achieving higher throughput.

In this paper, arrays of polymeric plano-convex microlenses with different defined lens profiles are shown and optically characterized. Experimental results are compared to theoretical predictions. Substrates of OrmoComp® and polydimethylsiloxane (PDMS) containing regular arrays of circular confinement platforms are replicated by UV-based NIL (UV-NIL) approaches. Then, controlled volumes of InkOrmo are locally deposited and confined onto such platforms by drop-on-demand IJP to form the microlenses. This allows a fine control of each microlens profile, thus of their optical specifications, achieving MLAs with highly repetitive characteristics from lens to lens. Furthermore, it is also proven that lenses with different concrete specifications can be printed on the same MLA with high optical quality and low technological complexity.

2. Fabrication

2.1 Substrate fabrication

In this work, circular platforms of PDMS and OrmoComp® (micro resist technology GmbH), with diameters ranging between 50 and 300 µm, and a height of 7 µm (Figs. 1(a) and 1(b)) are utilized as confinement structures [22]. Firstly, a master containing elevated platforms is fabricated by photolithography (Fig. 1(a), Step1). The master is then replicated by cast molding into OrmoStamp® (micro resist technology GmbH) stamps (Fig. 1(a), Step2) which are then coated with an anti-sticking layer of self-assembled fluoroalkyl-based silanes. OrmoComp® or PDMS final substrates are subsequently replicated from the stamps, resulting in replicas of the original structures with the same polarity (Fig. 1(a), Step3). The replication into PDMS is simply done by cast molding. On the other hand, for a conformal replication of the platforms into OrmoComp®, a UV-imprint process is done. The surface energy of the final substrates is reduced by means of an anti-sticking layer, in order to increase the contact angle (θ) of the printed drops and thus the aspect ratio of the final lenses.

2.2 Inkjet printing process

The microlens shape control is achieved by depositing ink drops onto round protrusive platforms. The ink is pinned at the platform rims and by increasing the ink volume on the platform, the apparent contact angle, herein called edge angle (ν), grows at the platform edge from the standard contact angle value until reaching a maximal value, the critical apparent advancing angle [29]. An IJP station is used to locally deposit the optical polymeric solution onto the platform array. A stable and reproducible drop generation is achieved by using InkOrmo. According to the design, a pre-determined number of drops is printed on each platform. The in-flight drop volume is 47 ± 3 pL, allowing to precisely control the final lens volume. This results in the growth of microlenses on the confinement platforms with freely specified profiles (Fig. 1(b)); within a range defined by the platform rim shape and substrate surface energy [22]. After solvent evaporation by a soft-bake step, the OrmoComp® microlenses are cross-linked by UV exposure. A final post exposure bake is applied in order to increase the polymer homogeneity and thus the optical performance. Prior to inkjet printing, the ink volume to be printed on each platform for a specific microlens profile can be predicted taking into account the volume loss occurring during the soft-bake, the exposure and the final hard-bake as described in [22].
Fig. 1. Schematic illustration of the fabrication process: a) Substrate replication; Step 1: master (photoresist on top of Si wafer) done by spin-coating, bake, exposure and development, Step 2: Stamp (of OrmoStamp®) done by casting, exposure and separation and Step 3: Replication done by UV-imprint/casting, exposure/bake of the final OrmoComp®/PDMS substrates. b) Step 4: Microlens growth by inkjet printing, bake and UV-exposure induced cross-linkage reaching specific characteristics schematized here with three types of microlenses (I, II and III).

Following this process, polymeric MLA on both PDMS and OrmoComp® substrates with microlenses having defined specifications are printed. An array composed of 100 microlenses with 100 μm in diameter printed onto a pre-patterned PDMS substrate is shown in Fig. 2. Typically a yield between 95% and 100% can be achieved with a R&D setup as the one used in this work. However, this yield can still be improved by implementing this process in a high throughput industrial IJP equipment. In the specific case of the array of Fig. 2(a), only 2 microlenses slightly spread out of the confinement platforms leading to a success of 98%. The first overflow occurred for the first printed microlens, due to a well-known phenomenon, the so-called “first drop problem” [30]. The second one occurred most probably for one of the two following reasons (1) a dust or an imperfection in the confining platform rims preventing an appropriated pining or (2) a particle (i.e. dust) passing through the nozzle while printing and deviating some of the ejected drops. Usually, these issues only affect the lenses which have overflowed, keeping the other ones safe; further failure cases have been described in [31]. Furthermore, the flexibility to print microlenses only on desired location is also shown since only half of the platforms composing the square lattice have been covered with a lens.

In Fig. 2(b) a magnified line of the microlenses is presented. This image shows the reproducible lens shape. To reach the profile shown here, ν of 99.7 ± 0.8°, 30 drops were printed onto each platform leading to a total volume of 1.4 ± 0.1 nL / platform. In addition, the perfect alignment of the microlens vertex is a result not only of the stage and inkjet printing precisions but also of the self-alignment enabled by the confinement [20]. In this specific case, the limitation is thus expected to be coming only from the UV-based lithographically prepared substrate master.

Fig. 2. SEM micrographs of an inkjet printed and cross-linked MLA on PDMS platforms with lenses of identical curvatures: a) Full MLA with (b) a zoom into a few microlenses showing their overall spherical profile and high reproducibility. Both scale bars are 100 μm.

In this work, different pitches are shown, but high filling factors are not investigated. It is foreseen that the pitch can be increased and the platform diameters reduced until a level where two limitations may take place. Firstly, the diameter of the microlens shortly increases,
beyond the platform rims, when a drop is being printed, as explained in [15]. This implies a minimum spacing between the platforms preventing ink merging with the neighboring microlenses. This aspect takes a high importance when targeting high profiles, i.e. hemispherical or higher ($\nu \geq 90^\circ$); meaning lenses with high numerical apertures (NA). For such lenses, the diameter of the lens is larger than the diameter of the platforms, such as clearly shown in [23] which obviously imposes higher platform spacing. Secondly, the precision and accuracy of the printing position and the one of the motion stages imposes a minimum platform diameter slightly under 40 µm and a maximum value of the filling factor which depends on the lens profile targeted [31].

3. Microlens specifications

The microlens optical performances can be predicted by theoretical expectations and compared with optical measurements. The radius of curvature $R$, at the vertex of the lenses, is directly calculated from Eq. (1), where the factor $K$ is related to the lens type-of-profile; $K = 0$ for spherical and $K = -1$ for parabolic profiles [3] and $D$ is the lens diameter. The heights at the vertex of the microlenses $h$ are calculated from both the measured and the predicted microlens profiles.

$$R = (K + 1) \frac{h}{2} + \frac{\left(\frac{D}{2}\right)^2}{2h}$$  \hspace{2cm} (1)

For a material with a refractive index $n$, the NA and $f$ are calculated from Eq. (2) and Eq. (3) respectively allowing comparing the theoretical expectations with optical measurements as shown in Fig. 3 for a wide range of lens profiles. This calculation is done under the assumption of a parabolic profile despite the overall spherical shape. The high NAs - up to 0.9 - are enabled by the high microlens profiles ($\nu$ from 40° to 120°). In case finer NA or $f$ discrete steps than the ones demonstrated herein are required, values in-between the steps can be obtained either by reducing the IJP in-flight drops size [32] or by changing the proportion of the ink solvent [25].

![Image](image_url)

Fig. 3. Microlens optical characterization for a) 50 µm, b) 100 µm and c) 200 µm -footprint microlenses: experimental measured results of the NAs and $f$s by increasing the volume – expressed by the number of printed drops per platforms – compared with theoretical calculated evolution. The experimental results are measured from optical CCD images (empty points) and from geometrical profiles (bulk points shown with their standard deviations).
\[
NA = \frac{D / 2}{\sqrt{\left(\frac{D}{2}\right)^2 + f^2}} \\
f = \frac{R}{n-1}
\]

4. Microlens arrays

4.1 Microlens array with single lens characteristics

Figure 4(a) shows a dense array composed of 181 microlenses printed on an OrmoComp® substrate containing 100 µm-diameter platforms with a pitch of 176.8 µm (in x and y directions, see Figs. 4(a) and 4(b)). In this case 20 drops have been printed per platform, meaning a total volume of 0.9 ± 0.1 nL and achieving a \( \nu \) of 89.7 ± 0.7°. A CCD image of the focal plane of this array in which the focal points of the lenses are clearly visible is shown in Fig. 4(b). The NA and the \( f \) of this array have been experimentally measured, giving a NA of 0.56 ± 0.02 and an \( f \) of 74 ± 4 µm. In addition, the pitch accuracy of this array is also measured at the focal plane by the relative distances between the microlens focal points, Fig. 4(b), leading to a pitch to pitch distance of 176.8 ± 0.4 µm. The quality of the lens focus is also evaluated and an average Full Width at Half Maximum (FWHM) of 1.4 ± 0.2 µm is measured, as shown Fig. 4(d) for one concrete lens with a FWHM of 1.39 ± 0.03 µm. The surface roughness of the microlenses has also been measured by AFM at the lens vertex showing a roughness root mean square (rms), \( \delta \), from 1.63 ± 0.07 nm (averaged over 3 scan lines per lens and 3 lenses per array). This enables to calculate the total integrated scattering (TIS) of the microlenses for a given wavelength \( \lambda \), Eq. (4) [3], evolving from 0.3% to less than 0.1% for the visible range as can be seen in Fig. 4(e).

\[
TIS = \left(\frac{4\pi\delta}{\lambda}\right)^2
\]

4.2 Microlens array with multi lens characteristics

MLAs containing microlenses with different characteristics can also be printed by means of the proposed method, without increasing the technological complexity. Figure 5 presents a show-case MLA composed of 9 lenses in which each of them has different defined characteristics. As show in Fig. 5(a), each position is printed with an increasing number of drops, starting with 10 drops in position 1 and incrementing it by 10 drops per platform, reaching 90 drops onto the platform in position 9. Hence, this small array contains lenses with 9 different profiles precisely positioned and designed, forming thus a “3D-optical spiral”. Optical top and side views of the fabricated array are shown in Figs. 5(b) and 5(c) respectively.

In Fig. 6, a MLA composed of two different lens types is presented. The two different kinds of lenses are distributed as schematized in Fig. 6(a). The lens type I is composed of 30 drops while the lens type II is composed of 15 drops also shown in the colored SEM image of Fig. 6(b); dark and light blue respectively. The optical characterization of the array shows that the focal planes of lens type I and type II are separated by 13 ± 1 µm. As it is shown in Fig. 6(c), only the corresponding lenses are on focus in their respective focal plane. Properties of both lenses are summarized in Table 1.
Fig. 4. MLA on OrmoComp® platforms with single characteristics: a) SEM image of a part of the cross-linked MLA. b) Optical image of the focal plane showing the individual microlens focal points (pitch of 176.8 ± 0.4 µm). c) Intensity distribution of one of the lenses in the array; d) TIS of the microlens array calculated based on the microlens surface roughness, and a small inlet schematizing the AFM measurement. The scale bars are 100 µm.

Fig. 5. MLA with 9 different microlens specifications: a) Scheme of the array, b) optical top view showing the microlens height varying with the volume increase from platform to platform and c) side view optical image showing the microlens different heights depending on the number of inkjet printed drops. The scale bar is 100 µm.

Table 1. - Lens type I and II properties

| Lens type | # Drops | Vol (pL) | θ | NA | f (µm) |
|-----------|---------|----------|---|----|--------|
| I         | 30      | 378 ± 25 | 100° | 0.608 ± 0.003 | 65.2 ± 0.5 |
| II        | 15      | 189 ± 12 | 75°  | 0.542 ± 0.007 | 78 ± 1 |

Comparison of the optical properties of the lens types I and II composing the array showed in Fig. 6.
Fig. 6. MLA with two focal lengths. a) Scheme of the array representing the two different microlens types onto the platforms; type I in dark blue and type II in light blue. b) SEM images of a part of the array artificially colored to highlight the two microlens types. c) Optical images of the focal planes of the lens type 1 (left) and type 2 (right) in B&W. The scale bars are 100 µm.

5 Discussions

Optical micro-components with extended freedom in their features as compared to previous works are demonstrated by the presented results; herein demonstrated with microlenses with single and multiple characteristics. Such structures have been successfully fabricated into polymeric materials by combining IJP and UV-NIL approaches, namely UV-replication. In particular, a broad range of NA and \( f \) values have been precisely and reproducibly achieved. Additionally, we have also demonstrated the capability to develop MLAs comprising up to 9 different microlenses that could be distributed in any arbitrary order. These results present the proposed method as an attractive alternative for the development of complex micro-optical systems, especially in applications where specifically tailored microlenses are required. Furthermore, the presented range of NAs and \( f \) can be easily extended. Increasing the surface energy – by simply avoiding the surface treatment before printing – allows reaching lower microlens profiles, and thus higher \( f \) and lower NAs. Different patterns – platform diameters and pitches – have been selected, but working with smaller pitches and thus densely packed microlenses and higher filling factor is also possible by starting with other initial designs.

The pitch-to-pitch repeatability shows a standard deviation under 0.5% (± 0.4 µm). This result is very probably over estimated as it also suffers from experimental imprecision - CCD detector and intermediate optical components’ quality - on the focal plan imaging. This is also the case for the FWHM measured on the MLA with single characteristics but still showing results better than other optical components of similar dimensions [21]. The pitch-to-pitch
and focus point quality exceeds the expectations and requirements for a myriad of applications. The average surface roughness, $\delta$, of 1.63 ± 0.07 nm and the TIS evolving from 0.3% to less than 0.1% for the visible range is comparable or better than other results reported in the literature for microlenses fabricated by thermal reflow and transferred into SiO$_2$ [3], proving the high optical quality of the fabricated microlenses. The use of the UV-curable hybrid polymer OrmoComp$^\text{®}$ in its well-established and inkjet printable version InkOrmo allowed combining both IJP and UV-NIL approaches reaching the generation of homogeneous complex patterns, not suffering from material transition. This point should not be underestimated, since backscattering reflection occurring at material transition having different index of refraction may reduce the optical efficiency and thus the device functionality.

In addition, although the inkjet printing set-up was performed using a single nozzle printhead, faster printing can be readily achieved by using printheads with up to 2048 nozzles and jetting frequencies between 5 and 10 kHz [33]. For high throughput, based on the compatibility of NIL and IJP with R2R, these steps can be integrated into large scale production apparatus [34,35].

6. Conclusion

In this work we present polymeric MLAs with precise and reproducible single and multiple lens characteristics developed by means of IJP. We demonstrate that by printing a specific ink volume on a pre-patterned platform, microlenses with targeted and specific geometries as well as optical characteristics could be printed. The emphasis has been set on microlenses with 50 µm to 200 µm diameter footprints and high NAs and short $f$, ranging from 0.5 to 0.9 and from 10 to 100 µm respectively. Based on theoretical predictions and experimental results, it has been demonstrated that despite overall spherical profiles, as intuitively expected, the optical behavior of the microlenses studied in near field follows parabolic profiles. Subsequently, the microlens quality has been evaluated based on their spot FWHM of 1.4 ± 0.2 µm and on their TIS evolving from 0.3% to less than 0.1% in the visible light range. These results show microlenses with improved qualities when compared the ones obtained based on thermal reflow and RIE transfer to SiO$_2$.

This technique has been applied for printing MLAs with single lens characteristics reaching routinely fabrication yields of 95% to 100% and a lens profile standard deviation of less than 1° (measured by the microlens apparent contact angle). The optical characterization of the microlenses showed a high reproducibility with NA, $f$ and pitch standard deviations in the order of 4%, 5% and 0.2% respectively. Finally, MLAs with multiple lens characteristics within the same array have been printed with high optical quality, demonstrating up to 9 different microlenses in a single array. The optical performances of MLAs involving two different kinds of microlenses have been studied and showed focal planes separated by 13 ± 1 µm. Hence, materials and methods demonstrated herein are presented as a cost-efficient and potentially high throughput attractive additive manufacturing approach for microlenses and multi-focal lens arrays. These optical components with multiple lens characteristics may in addition open new routes for integrated optics and other challenging applications.

7. Experimental section

7.1 Substrate fabrication

The masters are prepared by photolithography on 4" Si wafers using a PPR (ma-P 1200 series). OrmoStamp$^\text{®}$ is used for the stamp replication and OrmoComp$^\text{®}$ for the final substrate containing the confining platforms. All these materials were obtained from micro resist technology GmbH. The substrates are replicated into OrmoComp$^\text{®}$ by an imprint at 1.5 MPa for 60 secs followed by a UV-imprint at 1.5 MPa for 56 secs in Obducat NIL-2.5 Nanoimprinter. The replication into PDMS (from Wacker Chemie AG) is done by direct
casting and 1 hour curing in an oven at 90 °C. All silanization processes are done at room temperature in vapor phase of fluoroalkyl-based silanes (C₈H₄Cl₃F₁₃Si (Tridecafluoro-1,1,2,2-tetrahydrooctyl)trichlorosilane; 97% purchased from abcr GmbH).

7.2 Microlens inkjet printing
MLAs IJP is done following the process described in [22] with the JetDrive III from MicroFab Technologies Inc. and a 50 µm aperture nozzle. The linear and rotational stages are from Newport Corporation. The optical ink printed is InkOrmo, which is a commercially available inorganic-organic hybrid polymer with excellent optical properties tailored for micro optical applications [25,26]. It is a UV-curable ink based on OrmoComp®, which has been modified for use in IJP by adding an organic solvent. The in-flight drop volume is 47 ± 3 pL, calculated from the in-flight drop diameter, which is of 45 ± 1 µm. After printing, the solvents are evaporated by a soft-bake on a hotplate at 60 °C for 30 mins. The InkOrmo microlenses are cross-linked by a UV flood exposure of 1000 mJ/cm². The hard-bake is done at 130 °C on a hotplate for 10 mins with a slow temperature ramp down.

7.3 Microlens shape characterization
The edge angle measurements are done from optical microscope side views with LBADSA, an ImageJ plugin developed at EPFL [36]. The heights are both calculated from the measured angles and measured using an optical profiometer in the Institute for Nanometre Optics and Technology at the Helmholtz-Zentrum Berlin (HZB). Both calculated and measured values are in the same range. The roughness is measured with an Autoprobe AFM tool from Park Scientific Instruments and averaged over 3 scan lines per lens selected from a 2 µm x 2 µm area from the top of 3 different lenses.

7.4 Microlens optical properties’ characterization
Focal plane of the MLA is recorded using a microscope objective (x10, Olympus) and a CCD camera (DCU223C, Thorlabs Inc). Precise relative displacement between of the lenses and the microscope objective is controlled by means of a x-y-z motorized micropositioner (MAX313D/M, Thorlabs Inc). The pitch to pitch distances of the MLAs is measured in X and Y directions with ImageJ analysis tools based on the imaged focal plane.

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