

Development and Implementation of a Rotating Nanoimprint Lithography Tool for Orthogonal Imprinting on Edges of Curved Surfaces – August 18, 2021

Uniform molding and demolding of structures on highly curved surfaces through conformal contact is a crucial yet often-overlooked aspect of nanoimprint lithography (NIL). This study describes the development of a NIL tool and its integration into a nanopositioning and nanomeasuring machine to achieve high-precision orthogonal molding and demolding for soft ultraviolet-assisted NIL (soft UV-NIL). The process was implemented primarily on the edges of highly curved plano-convex substrates to demonstrate structure uniformity on the edges. High-resolution nanostructures of sub-200-nm lateral dimension and microstructures in the range of tens of microns were imprinted. However, the nanostructures on the edges of the large, curved substrates were difficult to characterize precisely. Therefore, microstructures were used to measure the structure fidelity and were characterized using profilometry, white light interferometry, and confocal laser scanning microscopy. Regardless of the restricted imaging capabilities at high inclinations for high-resolution nanostructures, the scanning electron microscope (SEM) imaging of the structures on top of the lens substrate and at an inclination of 45° was performed. The micro and nanostructures were successfully imprinted on the edges of the plano-convex lens at angles of 45°, 60°, and 90° from the center of rotation of the rotating NIL tool. The method enables precise imprinting at high inclinations, thereby presenting a different approach to soft UV-NIL on curved surfaces.

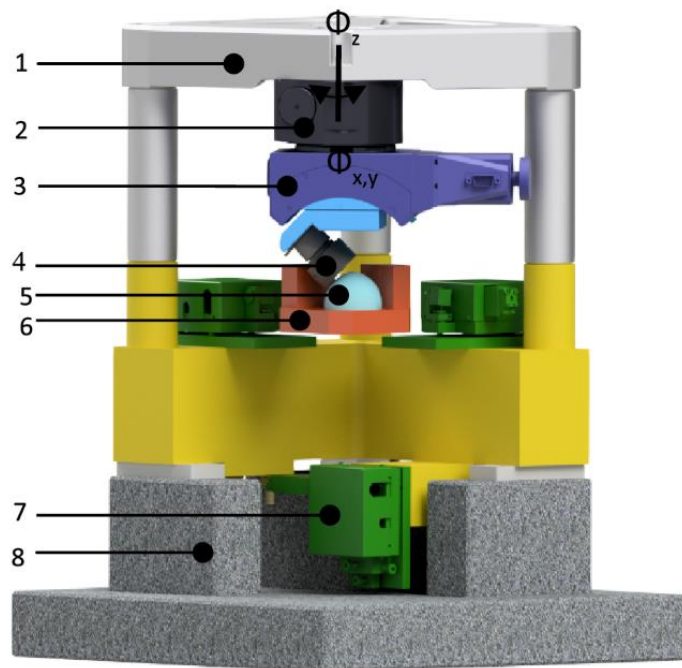
Introduction

The patterning on curved or non-flat surfaces has gained prominence in recent years because of its rapidly increasing applications in the fields of optics, photonics, biomedicine, flexible electronics, solar cells, and others [1, 2]. Over the years, different techniques, such as the use of gasbag pressure chambers and preshaping films [3, 4], have been adopted for imprinting on non-planar surfaces.

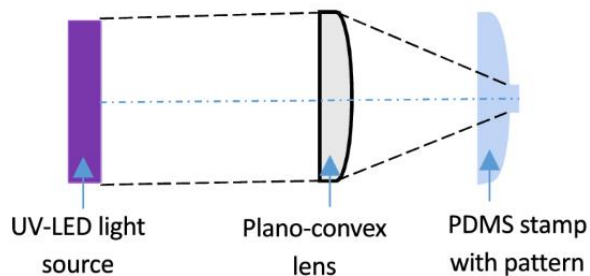
However, the stamps in these processes are bent to conform to the curvature of substrates. Therefore, the molding and demolding are not orthogonal to the surface. One of the major factors responsible for a successful nanoimprint lithography (NIL) process is demolding [5, 6]. Demolding is a crucial step during the solid phase or the post-ultraviolet (UV) curing phase of the NIL process. A problem arises when shear stress is induced during the separation of stamps and resist-coated substrates. Such stress can lead to structure distortion and pattern collapse, particularly in cases involving structures with high aspect ratios. For high-resolution structures with a sub-100-nm dimension, the high internal stress generated in their small cavities can weaken the rigidity of polymers and lead to deformation. Internal stress is therefore an inevitable part of the demolding process. Thus far, a few methods have been implemented to reduce stress by reducing surface adhesion; such methods include coating stamps with anti-sticking layers or using alternative stamp materials with low surface energy that makes the stamp surface hydrophobic [5].

In the age of interdisciplinary research, the integration of multidisciplinary fields allows the exploration of and experimentation on different approaches to addressing challenges. The simplicity of the soft UV-NIL process makes it suitable to be integrated into a nanopositioning and nanomeasuring machine (NMM-1). The NMM-1 is a coordinate measuring machine that has a positioning resolution of less than 0.1 nm and a working range of 25 mm × 25 mm × 5 mm, [7, 8, 9]. To achieve nanometer accuracy, the machine utilizes the three-dimensional Abbe comparator principle. On the basis of this principle, the machine performs position measurement with three laser interferometers that are virtually crossing one point of a thermally and geometrically stable mirror corner made of Zerodur (Fig. 1a). The accurate positioning capability of the NMM-1 tool allows the precise control of the effect of stamp deformation during the imprinting procedure.

Fig. 1
(a)



(b)



a Basic model of nanopositioning and nanomeasuring machine with additional rotating nanoimprint tool: **1** metrological frame of NMM-1; **2** rotation stage (ϕ_z); **3** goniometer stage ($\phi_{x,y}$); **4** NIL tool; **5** substrate; **6** mirror corner of NMM-1 (moved in x, y, z); **7** interferometer of NMM-1; **8** metrological frame of NMM-1. **b** Detailed schematic of developed NIL tool

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Unlike conventional lithography tools, soft UV-NIL is preferable because it is not dependent on complex machines, strict environmental conditions, or optical limitations. In the current work, a compact and adjustable soft UV-NIL process was initially designed, assembled, and integrated into the NMM-1 [10]. The successful implementation of the fundamental process in the NMM-1 created a foundation for making an NIL tool to be combined with a rotating device and integrated into the NMM-1. The combination enables five degrees of freedom of motion for orthogonal molding and demolding on edges of curved substrates.

Device and Process Development

Rotating Nanoimprint Tool

The positioning of a substrate is based on three independent linear movements in a Cartesian coordinate system of the NMM-1. Moreover, a fixed nanoimprint tool limits the addressable substrate geometries. An enhancement is introduced through the implementation of the ultraprecise rotational movements of the NMM-1 tool while keeping the precision within the nanometer range. Kinematics with a high degree of fulfillment consider a common instantaneous center of rotation in the tool center point (TCP). The integrated kinematic is based on the serial combination of an L-611 precision rotation stage (2018: L-611.9ASD, PhysikInstrumente GmbH & Co. KG) and a WT-90 motorized precision goniometer (2018:WT-90(65509201), PhysikInstrumente GmbH & Co. KG)(Fig. 1a).

The additional tool rotations of 360° around Φ_z and 90° around $\Phi_{x,y}$ of the NMM-1 enable the hemisphere to be positioned orthogonally to its surface. As the TCP corresponds to the intersection of the three interferometers of the NMM-1, the Abbe comparator principle is adopted for all axes. This approach avoids first-order positioning errors and linear correction movements for tool rotation and enables orthogonal nanoimprinting on nearly free-formed surfaces.

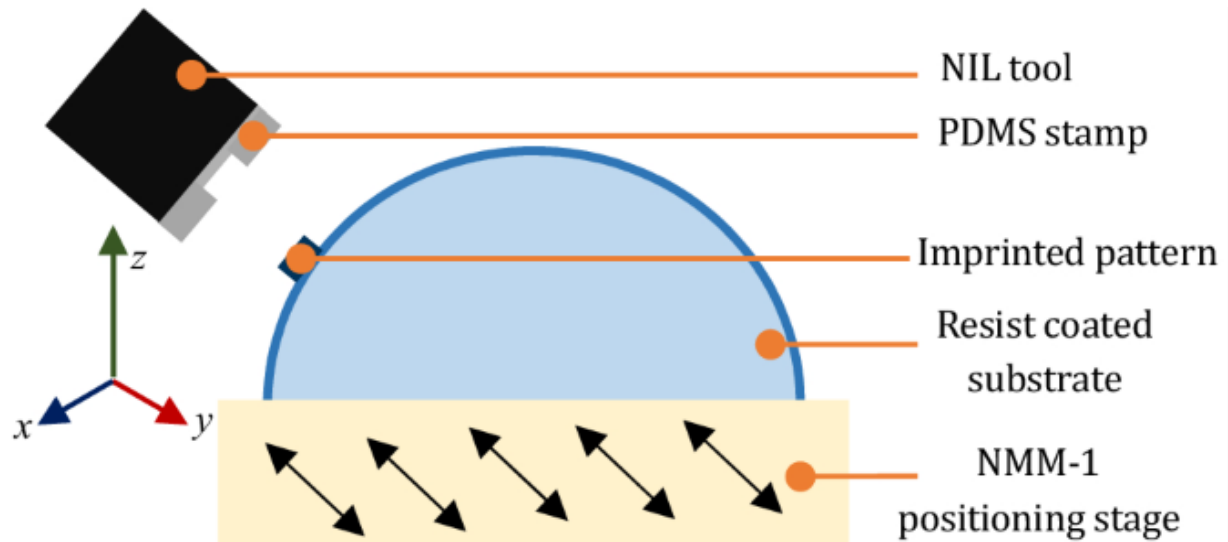
The 5-degree-of-freedom (DOF) combination of rotational bearings (rotary stage) and segmental rotational bearings (goniometer) provides an optimal solution for miscellaneous challenges. One such challenge is the combination of two rotational axes whose crossing under 90° is not possible because of working space limitations. An additional crossing angle would lead to the non-independent behavior of the two axes. The combination of a segmentally rotational bearing mounted on a rotational bearing fulfills all requirements without any restrictions in the measurement volume. Therefore, a goniometer stage is mounted on a rotary stage herein [11].

The goniometer, which now forms an integral part of the entire rotating mechanism, enables rotation in a precise angular position. Additionally, the goniometer stage provides $\pm 45^\circ$ rotation in the x- and y-directions by using a curved, profiled rail guide and a precision worm gear driven by a servo motor. In combination with the rotary stage, which is active at 360° in the z-axis, the goniometer stage enables ultrahigh resolution for small ranges of motion in micro- and nanopositioning. Thus, the combined motions, i.e., three translational motions of the NPMM and two rotational motions of the goniometer and rotation stage, enable 5-DOF motions.

The optimization of the rotation tool to address a hemisphere orthogonally is realized by determining several possible kinematic variants and then analyzing them by using a parameter-based evaluation system. A high degree of fulfillment is shown by these variants for the rotation of the tool. However, given the unknown structure of the substrate, linear corrections need to be made to address the entire surface of the substrate. If the sample fits the dimensions derived from the given Cartesian volume, then any hemisphere within the characteristic volume can be measured.

The integrated rotating NIL tool is developed on the basis of the basic components of soft UV-NIL (Fig. 1b). It consists of a UV light-emitting diode (LED) light source (*Lumitronix LED Technik GmbH*) with a wavelength of 365 nm and an intensity of 148 mW/cm^2 . A fused silica plano-convex lens is used to focus the light on an area of approximately 20 mm^2 . The focused light is transmitted through the patterned area of 25 mm^2 on the polydimethylsiloxane (PDMS, Sylgard 184 Silicone elastomer) stamp. The stamp has a diameter of 20 mm and is fixed to the edge of the NIL tool. The developed process is implemented with a resist filling time of 1 min for the filling of resist into the cavities of the micro- and nanostructures during molding. Then, resist curing by exposure to UV light for 1 min is performed. Demolding is subsequently carried out. Imprinting can be implemented multiple times on a single substrate at different angles of rotation. The imprinting process involves the synchronization of the movements of the rotating NIL tool and mirror corner so as to successfully implement the diagonal molding and demolding process (Fig. 2). The control of both tools is realized manually at this stage to ensure coordination.

Fig. 2



Orthogonal molding and demolding on edges of curved surface enabled by the diagonal movement (shown by *arrows*) of the positioning stage

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Spray Coating for Plano-Convex Lens Substrate

Spray coating is performed using EVG 101 spray coater (EV Group). In the case of non-planar surfaces, such as the plano-convex lenses used in this work, a multiple-layer coating is required to achieve good homogeneity and coverage. Before starting the spray coating process, the lens is cleaned using acetone and isopropanol and then placed on a hotplate for 10 min at 115 °C to remove any moisture from the surface of the lens. It is then coated with an adhesion promoter (TI Prime, Microchemicals GmbH) by using spin coating at 4000 r/min and baked for 2 min at 115 °C on the hotplate.

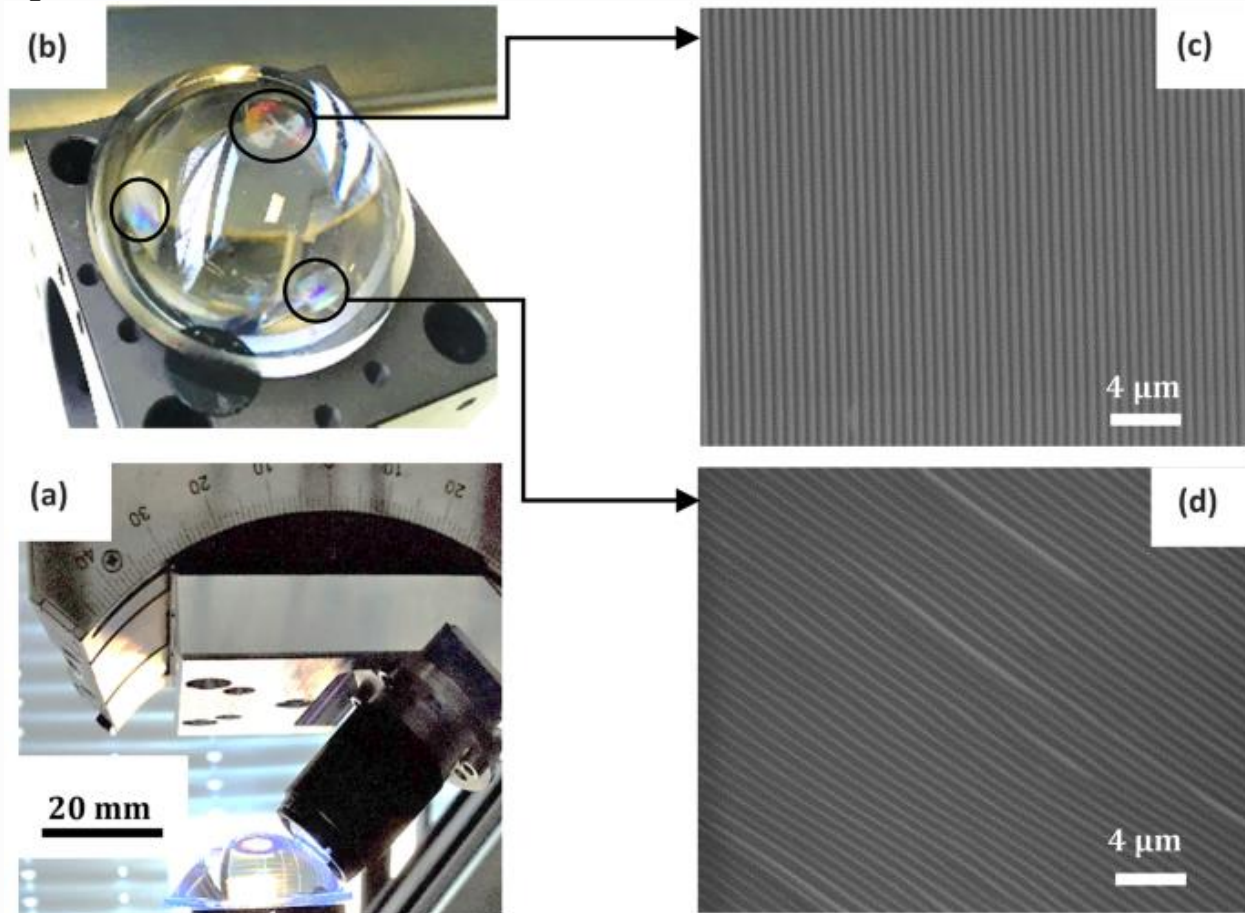
Before starting the process of spray coating using the spray coater, the ultrasonic nozzle to be used is cleaned using acetone. Then, the resist AMONIL MMS4 (AMO GmbH) is filled into the syringe pump of the spray coater. After filling the syringe with the resist, the parameters of the spray coater are adjusted. Additional tests on varying geometries show that the individual parameters of the spray coater need to be optimized according to the geometries (length, width, height, and diameter). Two layers of spray coating are completed, and the dispense rate and nozzle flow are changed for each layer. Meanwhile, the high-frequency power and spin speed are kept constant at 0.7 W and 50 r/min, respectively. For the first layer, a dispense rate of 6 $\mu\text{L/s}$ and nozzle flow rate of 180 dL/min are set. After the coating of the first layer, a soft bake is performed for 15 s at 100 °C. The second layer of coating is completed at a dispense rate of 11 $\mu\text{L/s}$ and a nozzle flow rate of 150 dL/min. Thereafter, the lens is again soft baked on the hotplate at 100 °C for 45 s.

In addition to the aforementioned parameters, the velocity profile of the nozzle is an important parameter influencing the homogeneity of resist coating on non-flat surfaces. The nozzle speed is divided into several indices that define the nozzle speed in different segments of the substrate. During the process, seven out of 15 indices are used on the basis of the geometry of the lens substrate. Pham et al. [12] describe the working mechanism of the spray coater and the effects of the variations of different spray coater parameters.

Results and Discussion

Figure 3 provides an overview of the implemented process. Here, Fig. 3a shows the rotating NIL tool in contact with the substrate during the UV curing stage. The different imprinted areas on the lens surface are shown in Fig. 3b. Figure 3c, d are the scanning electron microscope images of the imprinted nanostructures on top and at one of the edges of the lens, respectively. The nanostructures have a lateral resolution of 200 nm. The microstructures are imprinted to obtain an imprinted pattern profile and analyze the fidelity of the imprinted patterns on the edges of the lens by using a laser scanning microscope (LSM).

Fig. 3

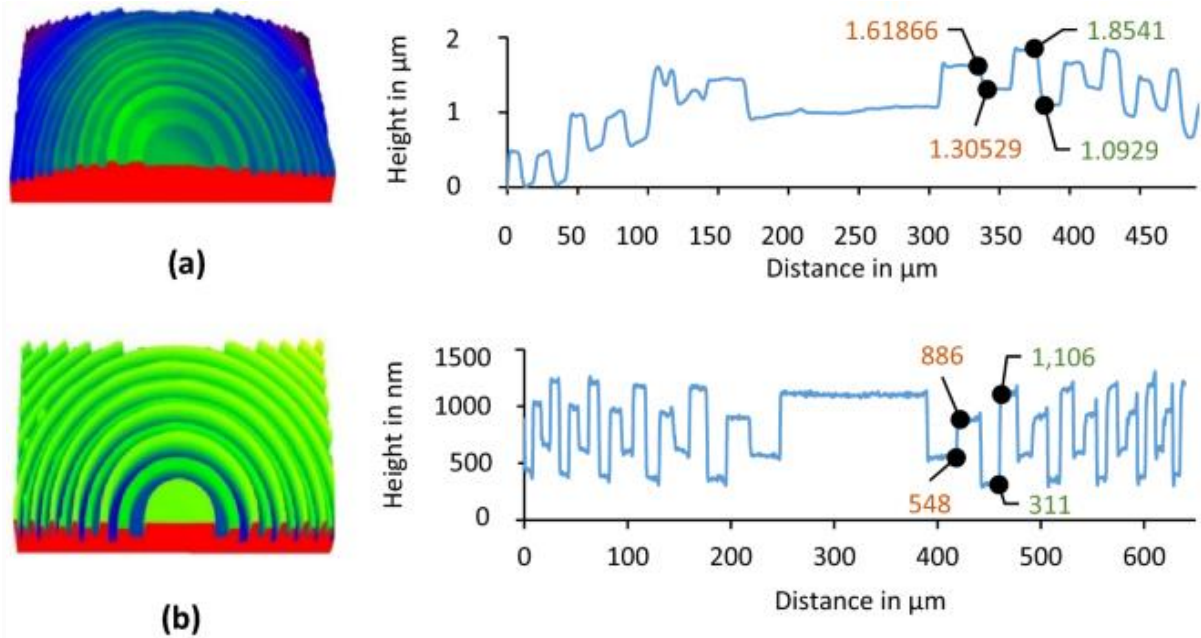


a Soft UV-NIL process at the edge of the plano-convex lens; **b** imprinted plano-convex lens at angles of **c** 90° (*top*) and **d** 45° (*edge*) of the rotating NIL tool

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The imprinted microstructures measured at an angle of 45° on the edge of the lens are found to be uniform and of good fidelity relative to the structure profile of the stamp (Fig. 4a and b). The imprinted structures are further scanned using a profilometer to obtain an enhanced profile of the structures. As a result of the use of different techniques, differences in the units of the scale bar can be observed in the graphs in Fig. 4a, b. The measured differences in the structure heights between the stamp and the imprint at two different steps of the structure profile are measured to be 25 and 33 nm, as derived from Table 1. These differences are calculated for two different levels of the microstructure, as marked in the graphs in Fig. 4.

Fig. 4



3-D laser scanning microscope images and corresponding structure profiles of (a) imprinted pattern using rotating NIL tool on the edge of the plano-convex lens (measured using a profilometer); b master pattern on PDMS stamp

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Table 1 Structure height comparison between the stamp and the imprint at two different steps

[Full size table](#)

The minimal difference in the structure heights implies the good filling of the resist into the structure cavities, with the pattern almost replicating the stamp. However, as a result of the strong curvature of the macroscopic substrates, their entire surface cannot be patterned uniformly with a single stamping process. Instead, the entire procedure must be divided into individual imprint processes, and the individual surfaces must be stitched together. For nanometer accuracy stitching over the entire surface of the strongly curved substrate, the ultraprecise positioning of the stamp relative to the substrate is necessary. In this case, the highest possible reproducibility of positioning in conjunction with a corresponding in situ measurement of position deviations is indispensable. This factor is realized for the three linear axes of the NMM-1 through the position detection of the mirror corner via laser interferometers and autocollimators. The maximum measured position repeatability of the TCP over the entire rotation range is ± 100 nm for the x - and y -directions and ± 150 nm in the z -direction [11] [13]. A high positioning accuracy requires the in situ position determination of the rotating tool. Further steps include the final assembly of such a developed system, the integration of the nanoimprinting tool, and the development of the stitching process.

Conclusions

The newly developed rotating NIL tool, in combination with the NPM machine, allows not only orthogonal imprinting on the edges of highly curved substrates but also imprinting on free-form surfaces with high accuracy and precision. The integration of the two tools results in a good combination of their advantages and provides a unique method for implementing the soft UV-NIL process. On the one hand, the low Young's modulus of the PDMS stamp enables conformal contact for curved surfaces and easy release from imprinted substrates. On the other hand, this factor limits the replication of high-resolution features. With the developed tool, a soft stamp can be used as a hard stamp by placing it orthogonal to the edge of the substrate. Such a setting eliminates the disadvantage of structure bending at high curvatures because a rigid stamp facilitates the replication of high-resolution features [14]. The previously established

methods for the NIL process on curved surfaces focus mostly on PDMS stamp formation or the modification of the actual process conditions. However, these processes are focused on small-sized substrates that are mostly in the micrometer or millimeter range. They also neglect the characterization of structures at edges because of the challenges arising from their complexities. With the development of the rotating NIL tool, these facets are explored.

Imprinting is performed on the edges of large plano-convex lens with a diameter of 48.7 mm and a corresponding height of 18.6 mm at angles of 45°, 60°, and 90° from the center of rotation of the rotating tool. Additionally, the placement of the substrate on the mirror corner of the NPM machine enables the highly precise diagonal motion of the substrate, which in turn allows orthogonal molding and demolding at different angles. The high resolution and accuracy of the positioning stage ensure a smooth demolding process without shear effects, which is one of the major issues in the NIL process. The imprinted high-resolution nanostructures with good fidelity at the edges of the high curvature substrates depict a desirable outcome given the intricacy of the high-resolution soft UV-NIL process, even on flat substrates.

Thus, based on fundamental concepts of the soft UV-NIL process, together with a high-precision positioning machine, the rotating tool with multiple degrees of freedom combines the best features of each and enables successful orthogonal imprinting process. The further optimization of the process can lead to the enabling of stitching for covering entire surfaces of the substrate.

Availability of Data and Material

Data available on request from the corresponding author.

Code Availability

Not applicable.

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