Controllable elastocapillary folding of silicon nitride 3D structures by through-wafer filling

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Abstract—We present the controllable capillary folding of planar silicon nitride templates into 3D micro-structures by means of through-wafer liquid application. We demonstrate for the first time hydro-mechanical, repeatable, actuation of capillary folded structures via addition or retraction of water on demand.

I. INTRODUCTION

A. Elastocapillary folding

Elastocapillary phenomena refer to the deformation of objects due to capillary effects of liquid droplets or films [1]. The final deformation is governed by the balance between bending and capillary forces. It is helpful to define an elastocapillary length, $L_{ec} = (B/\gamma)^{1/2}$ where B is the bending stiffness [N·m] and γ the surface tension [N·m⁻¹]. Typically, objects larger than the elastocapillary length are significantly deformed by capillary forces [2].

Previously, we have combined elastocapillary interactions with micro-machining to fabricate 3D silicon nitride microobjects, starting from flat patterns – a method we called elastocapillary folding or capillary origami [3], [4]. This more deterministic form of self-assembly of 3D objects enabled by a weak interaction (here, surface tension) can be generally defined as self-folding [5].

A 2D elastocapillary model has been developed and we showed that this simplistic model can be used to accurately describe the self-folding of oblong triangular prismatic structures. The final state of the folding (closed or reopened) is determined by a parameter which is closely related to the elastocapillary length [3].

The continuous demand for smaller functional elements in microtechnology brings the necessity to use all three spatial dimensions effectively. By combining the assets of maskbased fabrication and self-assembly, self-folding is a promising approach to reach the third dimension.

B. Repeatable folding

In our previous works on elastocapillary folding we manually deposited a droplet on the structures [3], [4]. This technique suffers from several disadvantages, such as difficult droplet application and lack of control of applied volume. Moreover, the structures can be folded only once, which considerably limits their potential applications.

Repeatable elastocapillary folding is typically performed on millimetre sized thin sheets, governed by the deposition and evaporation of a droplet of water. A variety of folded shapes such as tubes, tetrahedra and cubes can be attained depending on the geometry of the sheet template [6]–[8]. Control over the folding state is possible by adding and retracting liquid with a needle [9], but attachments like these restrict sub-millimetre miniaturization [10].

Apart from changing the volume of the liquid, its shape can be influenced with help of an external field. An electric field between the liquid and the folding material substrate flattens the droplet and unfolds the structure [8], [11]. The typical time-scale of actuation can be identified by using an alternating electric field [12]. Another approach is the use of ferrofluid in combination with an external magnetic field [13].

On the micro scale, repeatable folding is often achieved by generating differential stress along the thickness of a thin film structure. Changing the various properties of the surrounding fluid of the device determines the bending through expansion. Common stimuli are thermal, pH or chemical, while bending can either be distributed or concentrated by combining active and passive material [14], [15]. Although these structures can be tetherless, a disadvantage is the dependence of stimuli on diffusion and reaction rates. This strong connection limits the time-scale at which they can be actuated [14].

Therefore, as far as we know, repeatable elastocapillary folding has only been done on structures with dimensions above a millimetre. In this paper, we show the first demonstration of repeatable elastocapillary folding of micro-structures. The applied technique of through-wafer filling (explained in Fig. 1) is unique for elastocapillary folding and enables precise hydro-mechanical actuation. Along with the technical details on fabrication and experiments, an analytical study of the folding process is presented.

II. THEORY

The fundamentals developed for elastocapillary folding by front side droplet application [3] has to be extended when employing through-wafer filling. When the proceeding liquid meniscus meets the diverging section at the top of the tube, it is pinned. The overpressure needed to overcome the pinning effect and to advance the meniscus on top of the origami pattern is given by the Young-Laplace equation:

$$\Delta P_{\max} = -\frac{4\gamma\cos\left(\theta_{a} + \frac{\pi}{2}\right)}{D_{0}}$$

where γ is the surface tension of water, θ_a the advancing contact angle of water on silicon nitride and D_0 the maximum diameter of the tube [16]. In case of a practical fluidic system, which is not infinitely stiff, this overpressure causes a slight

increase of the system volume. Once the meniscus leaves the tube, the overpressure reduces and additional liquid is released due to system relaxation, resulting in an overshoot.

III. EXPERIMENTAL

A. Fabrication

Fig. 2 shows a concise outline of the fabrication process for the origami patterns used in the experiments. Fig. 3 shows the resulting structure. The last three lithographic steps (four in total) following the drilling of the tubes were performed using spray resist for an optimal protection of the edges of the tubes. Low stress silicon-rich nitride layers were deposited by LPCVD. The structures were cleaned in HNO₃ just before the last lithographic step and final cleaning was done by oxygen plasma.

B. Fluidic setup

A schematic of the fluidic setup is shown in Fig. 4. The elasticity of the system was designed to be as small as possible to limit the amount of overshoot. Fluidic connection to the tube in the wafer is achieved by a tiny recessed o-ring in a metal part. The wafer sits on the metal part and the connecting force is predominantly transferred between metal and wafer. In this way we aim to limit the change in compression of the o-ring under liquid pressure variation. Other measures are the use of a Hamilton glass syringe and thick-walled PEEK tubing. Finally, the volume between the syringe plunger and the sample is kept as low as possible (around 10 μ L).

IV. RESULTS

A. Overshoot

To quantify the overshoot of the fluidic setup the syringe pump was quasi-statically actuated, starting with water in the through-wafer tube filled by capillarity. When water exceeded the edge of the tube and wetted the surface, actuation of the pump was stopped. The water droplet volume was derived







Fig. 2. Fabrication outline. I: Tubes are drilled through the wafer using a DRIE Bosch process. A layer of 1 μ m thick silicon nitride (SiN-1) is conformally deposited. II: The layer SiN-1 is dry etched in a plasma system after a second lithographic step. III: A second 100 nm thin layer is deposited (SiN-2). This layer is thin enough to be deformed by surface tension. IV: A third lithographic step is used to define the overall geometry of the structures and a fourth for the release of the flaps by semi-isotropic SF₆ silicon etch.

from snapshots of the captured video. Measurements on geometrical features of identical structures in SEM images were used as reference. Fig. 5 shows the snapshots that were used to determine the volumes. The left part shows the typical excess volume before revision of the fluidic setup, which translates to 93 ± 12 nL. The error is due to the uncertainty in identifying the features of the droplets in the images. After improving the setup (right part of Fig. 5), the overshoot reduced to 1.4 ± 0.4 nL. However, the tube diameter in the first case is $34 \mu m$, versus 50 μm in the second. After taking the overpressure difference into account, this results in a reduction of the overshoot volume by an approximate factor 50.

B. Elastocapillary folding

We experimented with two structures with perforated hinges (see Fig. 3) that have the shape of an oblong triangular prism when folded. The hinges of structure 1 have 50% more material than structure 2 by adjusting the amount of perforation, resulting in a difference in stiffness of 50%.



Fig. 3. SEM image of a silicon nitride origami template with two flaps, resting on a silicon pillar with a hollow silicon nitride tube. Length of the structure is 800 μ m and flap width is 80 μ m. The hinges are 7 μ m wide and diameter of the tube is 50 μ m. The hinges are perforated to reduce their stiffness. Note that the flaps are initially not aligned with the central part. This type of structure was used for the analysis presented in Fig. 6 and 8.



Fig. 4. Schematic of fluidic setup filled with ultra clean water. Fluidic connection to the wafer is made with a recessed o-ring in a metal part. Tubing is thick-walled (PEEK) and the syringe is made of glass. The angle α represents the rotation of the flaps, plotted in Fig. 6 and 8.

The evolution of folding was recorded during the evaporation of water, after being wetted by through-wafer filling. The angle of rotation of the flaps with respect to the central part (see Fig. 4) was extracted, along with the normalized water "volume" $[m^3 \cdot m^{-1}]$: the cross section of the liquid volume perpendicular to the long axis of the structure, normalized by w^{-2} , where w is the width of the flaps [3].

Fig. 6 shows that structure 1 folds until reaching the maximum folding angle, $\alpha_{max} = \frac{2\pi}{3}$, at which the object will stay closed because of sufficiently large stiction forces.

On the other hand, the stiffer structure 2 does not attain the maximal angle of rotation during folding. While the volume decreases due to the evaporation of water the surface energy lowers and, at a certain point, cannot sustain the increasing bending energy any more. The structure then reopens and the flaps come back to their initial position. This difference in behaviour is in agreement with the theory and results presented in our previous work [3].

C. Repeatable folding

The novelty of this through-wafer filling technique is the possibility to repeatedly open and close the structures. Fig. 7 shows the actuation of a cubic structure. Once the pinning of water at the edge of the tube is overcome, accurate addition and retraction of water is possible by using the syringe pump. Controlled reopening of structures was impossible using the



Fig. 5. Comparison of the amount of excess of liquid before (left) and after (right) the optimization of the setup. The yellow dashed line in the right hand picture denotes the contour of the drop; the height at the centre of the drop is approximately 40 μ m. This drop is resting on a large silicon nitride platform that cannot be seen here. By selecting rigid components and decreasing the internal volume, the overshoot was reduced by approximately a factor 50.



Fig. 6. Flap angle α versus liquid volume $[m^3 \cdot m^{-1}]$, for two structures similar to those shown in Fig. 3) but with a difference in spring constant. Structure 1 (red triangles) closes completely. The stiffer structure 2 (black circles) does not attain the maximal rotation and reopens.

top filling method that we employed in our previous work [3], [4]. Our structures were actuated up to 20 times without showing any signs of wear. However, endurance experiments to test the long-term stability have not been carried out yet.

The folding of structures, which takes a few minutes when letting the water evaporate (Fig. 7 between b and d), can be considerably sped up to times less than a second by retracting water through the tube. Furthermore, by tuning the height of the water container (see Fig. 4) it is possible to set the applied pressure and "freeze" the folding structure for certain ranges of states. This situation evidently implies that water is added to compensate for the evaporation.

Surprisingly, a structure that undergoes several cycles of water deposition and evaporation shows a change in behaviour for every iteration, as shown in Fig.8. While the flaps reach an angle of 80° before reopening during the first folding sequence, the maximal angle of rotation drops to 40° during the fifth iteration. Two parameters could explain this behaviour: a change in stiffness of the hinges and a drop of surface tension through the actuation cycles.

D. Residues on the structures

Large amounts of residue were observed on the structures after their actuation, see Fig. 9. This residue seems to aggregate at the level of the hinges during repeated folding. The residue probably originates from the fabrication process, which contains a final release step that cannot be followed by a liquid cleaning step (see Fig. 2).

Whether these residues are diluted in the water during folding or stay on the hinges is not clear yet. Both situations explain the results presented in the previous section, however. Residues deposited on the hinges make them thicker and therefore stiffer, whereas the surface tension of water can possibly drop when polluted.



Fig. 7. (a-h). Chronological sequence of snapshots from a movie of the folding process of a five-faced cube with rib length of $100 \mu m$ (Time format: mm:ss). Between b) and d) the folding occurred by allowing the water to evaporate. In the time between d) and h), the cube was repeatedly opened and closed (20 times). One sequence of opening and closing is shown from e) to g).





Fig. 9. SEM picture of the top part of the silicon nitride template after several folding sequences, showing the accumulation of residue on the hinges.

Fig. 8. Flap angle versus volume $[m^3 \cdot m^{-1}]$ for the same structure measured for the first folding sequence (closed circles) and during the fifth sequence (open circles). The maximal rotation angle decreases with the number of iterations.

V. DISCUSSION

Our novel backside-filling elastocapillary folding method allows repeated assembly of 3D micro-structures, which has to our knowledge never been demonstrated before. Our optimized setup allow us to actuate these 3D objects in a well-controlled way. However, the fact that the structures become polluted during the actuation is a major drawback. We believe residue causes both the structure and the water to change. This makes a comparison to the existing model, presented in [3], impossible. While the folding curves look similar to theory, we cannot draw any conclusions. Further work on this subject will therefore focus on avoiding the residue and extending the analytical study of the folding behaviour.

Furthermore, after fabrication the flaps are generally oriented somewhat downward, which could be caused by a stress gradient in the hinge. However, it has also been observed in the SEM that the structures bend downward with an electron beam of sufficient intensity. Clearly, charge accumulation in the silicon nitride in combination with the good conducting silicon substrate causes the downward bending of the flaps. We contemplated whether this also happens in ambient conditions, where the breakdown electric field strength is lower than in vacuum.

A FEM model for this phenomenon was constructed using a simplified form of the cross section of the folding structure, with a 2D approach similar to the derived elastocapillary model in earlier work [3]. The electric field of a uniformly charged flap (and its surroundings) was simulated and the resulting moment on the hinge was calculated. With a standard formula relating the curvature to the moment applied to the free end [17], the deflection angle of the hinge was derived. A significant deflection of 10° is already reached when the global maximum of the electric field is $100 \text{ kV} \cdot \text{m}^{-1}$. The breakdown electric field strength of air at atmospheric pressure with the gap dimension in question ($\sim 40 \text{ }\mu\text{m}$) is on the order of $10 \text{ MV} \cdot \text{m}^{-1}$ [18]. This shows that electrostatic bending of the structures at ambient conditions is possible when charge accumulation is sufficient.

After the first folding cycle the flaps often return to a slightly upward state (see Fig. 3). Next to the electric field screening due to the liquid that was present on the structure, this could be caused by the residues on the hinges.

VI. CONCLUSION

We demonstrated controllable capillary folding of planar silicon nitride templates into 3D micro-structures by means of through-wafer liquid application. For the first time, hydromechanical, repeatable actuation of capillary folded structures via subsequent addition and retraction of water has been achieved.

Through-wafer filling requires a sufficiently high pressure to overcome pinning of the liquid meniscus at the edge of the tube. This high pressure in combination with the elasticity of the fluidic setup causes an overflow of water. Precise hydromechanical actuation can only be achieved by selecting rigid components for the fluidic setup. In our situation, the volume of excess liquid could be reduced to 1.4 ± 0.4 nL.

Folding of a structure that does not fully close was monitored during five repeated cycles of filling and subsequent evaporation of water. The maximum folding angle decreased with every cycle.

Our results demonstrate that repeatable elastocapillary folding on the micrometer scale is possible, which opens up a route towards more complex structures, micro-actuation and possibly, drug delivery.

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