Supporting information

A high throughput strategy. Bubble inflation strain engineering was realized through PDMS devices that were fabricated according to standard photolithography and soft lithography techniques. We have fabricated a series of PDMS devices of different base shapes (Fig. S1). The shining wrinkling areas were easily recognizable above the lumen of the devices.

Fig. S1. PDMS devices with lumen of various shapes. Digital camera pictures of a) a device with circle-shape lumens. b) a device with triangle-shape lumens. c) pool of devices. d) a device with square-shape lumens. e) optical microscope image of the enlarged area of d).

Orientations of patterns within moon-shape wrinkling areas. Moon-shapes are typical that have medium $\eta$ value in which herringbones and strips dominate the whole areas. And by studying moons with different $\eta$ values, we can detect the evolution from herringbones to strips. In Fig. S2, there are two moon-shapes, nearly waxing moon and waning moon. In waxing moon, herringbones predominate and strips only exist in the indent part. While in the waning moon, strips dominate almost the whole area and herringbones only appear in convex part.
Fig. S2. a) optical microscope images of typical herringbone-orientation patterns and d) typical strip-orientation patterns. b) and c) were waxing moon and waning moon respectively, the areas bounded by red lines featured strips and the areas bounded by green lines featured herringbones.

**Orientations of patterns within complex wrinkling areas:** We mentioned that labyrinthine and herringbones existed in sharp protrudes of complex wrinkling areas. For example, in the elbow angle part of zigzag areas and the tail of letter “a”, there are analogous strains in more than one direction, hence labyrinthine-orientation wrinkles form which are shown in Fig. S3.
Fig. S3. a-c) optical microscope images of patterns form within zigzag area. b) details of the area bounded by red rectangle in a), and c) is details of the red rectangle area of b), strips-herringbones-labyrinthine evolution is clearly shown in c). d) optical microscope image of letter “a”. e) SEM image of the red rectangle area in d), in this area where three sets of parallel strip patterns of different directions meet, they twist into labyrinthine through the process of herringbones as transition.
Formation of Cracks. In the edges of wrinkling area, the pre strain in certain direction is negative, this direction is in compression before recovery, so after Au sputtering and strain relief, cracks form perpendicularly to the originally compressed direction because the Au layer is stretched apart by the recovery of the compressed substrate. For example, in the edges of strip and annular wrinkling areas, $\varepsilon_1 < 0$, $\varepsilon_2 = 0$, hence cracks vertical to $\varepsilon_1$ form in these areas (Fig. S4a-b). Also in the edge of circle-shape wrinkling area, $\varepsilon_1 < 0$, $\varepsilon_2 > 0$, so in this condition, cracks vertical to $\varepsilon_1$ while wrinkles vertical to $\varepsilon_2$ come into being (Fig. S4c-d).

Fig. S4. Cracks formed in the edges of wrinkling areas. a-b) cracks vertical to the negative pre strain $\varepsilon_1$. c-d) cracks and wrinkles formed in the edges of circle-shape areas, cracks vertical to $\varepsilon_1$ and wrinkles vertical to $\varepsilon_2$. Red arrows show the locations of the cracks and black arrows show the direction of the pre strain. Broken line means negative strain.
Calculation

Calculation of $\varepsilon_1(\theta)$:

$$
\varepsilon_1(\theta) = \lim_{\Delta \theta \to 0} \left[ \frac{1}{\sin \alpha} \frac{\Delta \theta}{\tan (\Delta \theta + \theta) - \tan \theta} - 1 \right]
= \frac{1}{\sin \alpha} \lim_{\Delta \theta \to 0} \left[ \frac{\Delta \theta}{\tan (\Delta \theta + \theta) - \tan \theta} - 1 \right]
= \frac{1}{\sin \alpha} \frac{1}{\tan \theta} - 1
= \frac{\cos^2 \theta}{\sin \alpha} - 1
$$

Calculation of $\varepsilon_2$ of spherical expanding surfaces: through the expanding process, circle (1) in originally unstretched film expands into circle (2) in stretched film, and the change is even in the circle track (Fig. S5), so at any point in the circle, the strain is equal to the total strain of the circle, hence we can give:

$$
\varepsilon_2 = \frac{P(2) - P(1)}{P(1)} = \frac{R(2) - R(1)}{R(1)} = \frac{R(2)}{R(1)} - 1 = \frac{R \cos \theta}{R \sin \alpha} - 1 = \frac{\cos \theta}{\sin \alpha} - 1
$$

$P(1)$ and $P(2)$ are perimeters of circle (1) and circle (2) respectively; $R(1)$ and $R(2)$ are radius of circle (1) and circle (2) respectively.

Fig. S5: Parameters of spherically expanding PDMS cover