Electronic Supplementary Information

Harnessing fold-to-wrinkle transition and hierarchical wrinkling on soft material surfaces by regulating substrate stiffness and sputtering flux

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**Figure S1.** Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 4$ sec taken by the optical microscopy ($\times 50$). The distance interval between the neighboring images is 1 mm. All images have the same size of $1044 \times 1390 \ \mu m^2$. From $x = 11$ to 14 mm, folds transit to wrinkles gradually.
**Figure S2.** (a-p) Morphological evolution of the gradient crosslinking PDMS surface with $t = 4$ sec in the fold-dominated region taken by the atom force microscopy (AFM). The data appearing in the bottom-left corners represent the distance $x$. All images have the same size of 80×80 μm$^2$. The scale bar in (p) is applied to all the AFM images. (corresponding to Fig. 2 in main text)
Figure S3. (a-e) Detailed transition from folds to wrinkles taken by the AFM. The sputtering time $t = 4$ sec. All images have the same size of $40 \times 40 \mu m^2$. The scale bar is applied to all the AFM images. (corresponding to Fig. 4 in main text)
Figure S4. (a-n) Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 4$ sec in the wrinkle-dominant region taken by the AFM. All images have the same size of $10 \times 10 \, \mu m^2$. The scale bar is applied to all the AFM images. (corresponding to Fig. 5 in main text)
Figure S5. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 7$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $10 \times 10 \, \mu m^2$. The scale bar is applied to all the AFM images. Note that the wrinkles have spread all over the PDMS surface and only a few isolated folds can be observed in the vicinity of the left edge for this sample. (corresponding to the first row of Fig. 7 in main text)
Figure S6. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 11$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of 20×20 μm$^2$. The scale bar is applied to all the AFM images. Note that in the range of $x = 20 \sim 22$ mm, a hierarchical wrinkle pattern composed of G1 and G2 wrinkling can be observed. (corresponding to the second row of Fig. 7 in main text)
Figure S7. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 20$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $20 \times 20 \mu m^2$. The scale bar is applied to all the AFM images. Note that in the range of $x = 18 \sim 20$ mm, the hierarchical wrinkles can be observed. (corresponding to the third row of Fig. 7 in main text)
Figure S8. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 60$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $30 \times 30 \mu m^2$. The scale bar is applied to all the AFM images. Note that in the range of $x = 16 \sim 19$ mm, the hierarchical wrinkles can be observed. (corresponding to the fourth row of Fig. 7 in main text)
Figure S9. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 180$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $40\times40$ $\mu$m$^2$. The scale bar is applied to all the AFM images. Note that in the range of $x = 13 \sim 16$ mm, the hierarchical wrinkles can be observed. (corresponding to the last row of Fig. 7 in main text)
Figure S10. Comparisons of wrinkle profiles in the gradient crosslinking PDMS samples with varied sputtering times. (a) $t = 7$ sec; (b) $t = 11$ sec; (c) $t = 20$ sec; (d) $t = 60$ sec; (e) $t = 180$ sec. The profile lines are shifted artificially along the vertical coordinate for clarity.
Figure S11. Evolutions of the wavelength $\lambda$ of G1 and G2 wrinkles (a) and RMS surface roughness $R_q$ (b) with the distance $x$ for the gradient samples with varied sputtering times.
**Figure S12.** Typical hierarchical pattern composed of G1 and G2 wrinkles with different dimensions on the dielectric gel surface by sputtering of molybdenum atoms with $t = 360$ sec. Optical micrographs with (a) low magnification ($\times100$) and (b) high magnification ($\times500$).