

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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Figures and captions

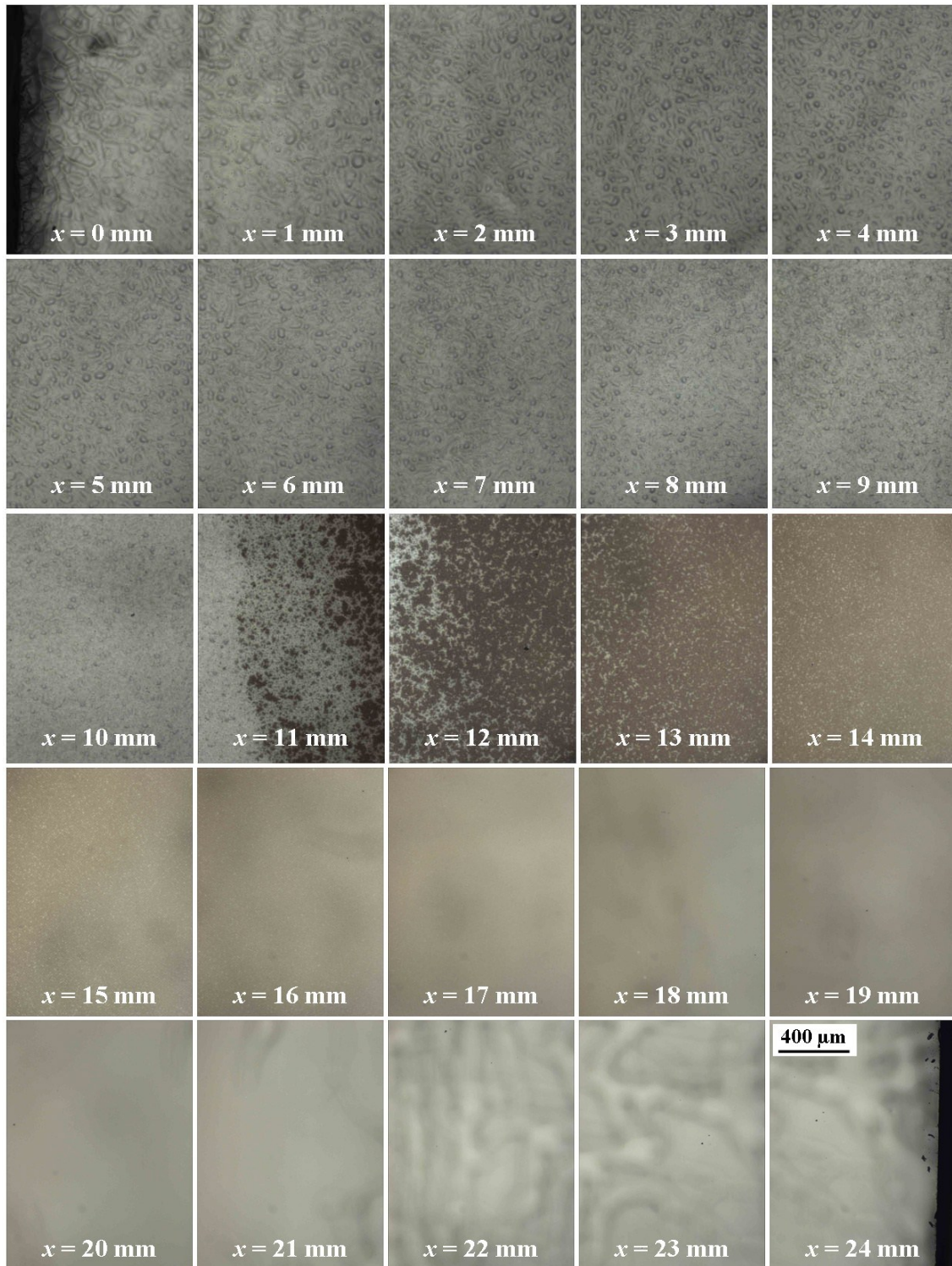


Figure S1. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 4$ sec taken by the optical microscopy ($\times 50$). The distance interval between the neighboring images is 1 mm. All images have the same size of $1044 \times 1390 \mu\text{m}^2$. From $x = 11$ to 14 mm, folds transit to wrinkles gradually.

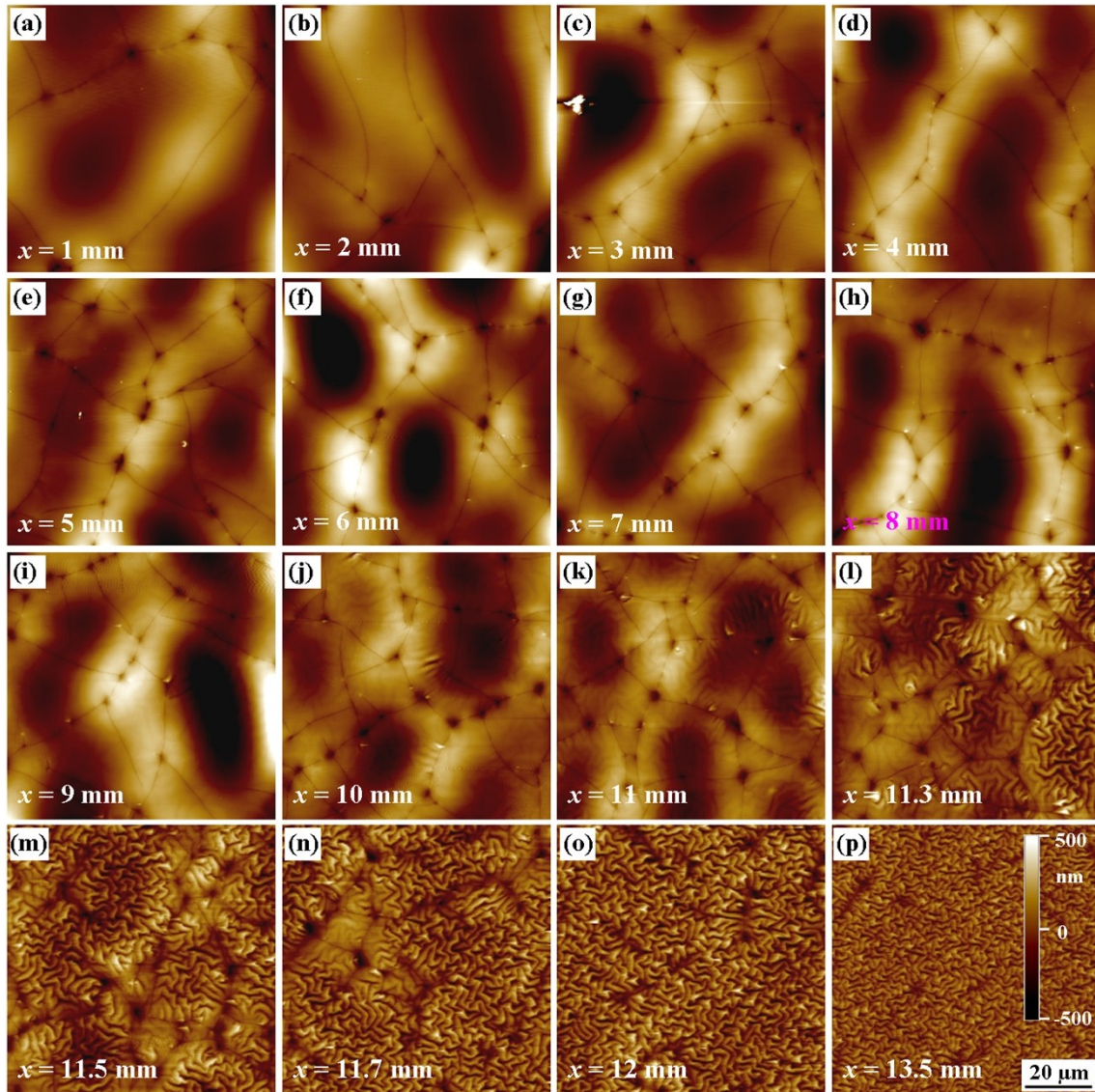


Figure S2. (a-p) Morphological evolution of the gradient crosslinking PDMS surface with $t = 4$ sec in the fold-dominated region taken by the atom force microscopy (AFM). The data appearing in the bottom-left corners represent the distance x . All images have the same size of $80 \times 80 \mu\text{m}^2$. The scale bar in (p) is applied to all the AFM images. (corresponding to Fig. 2 in main text)

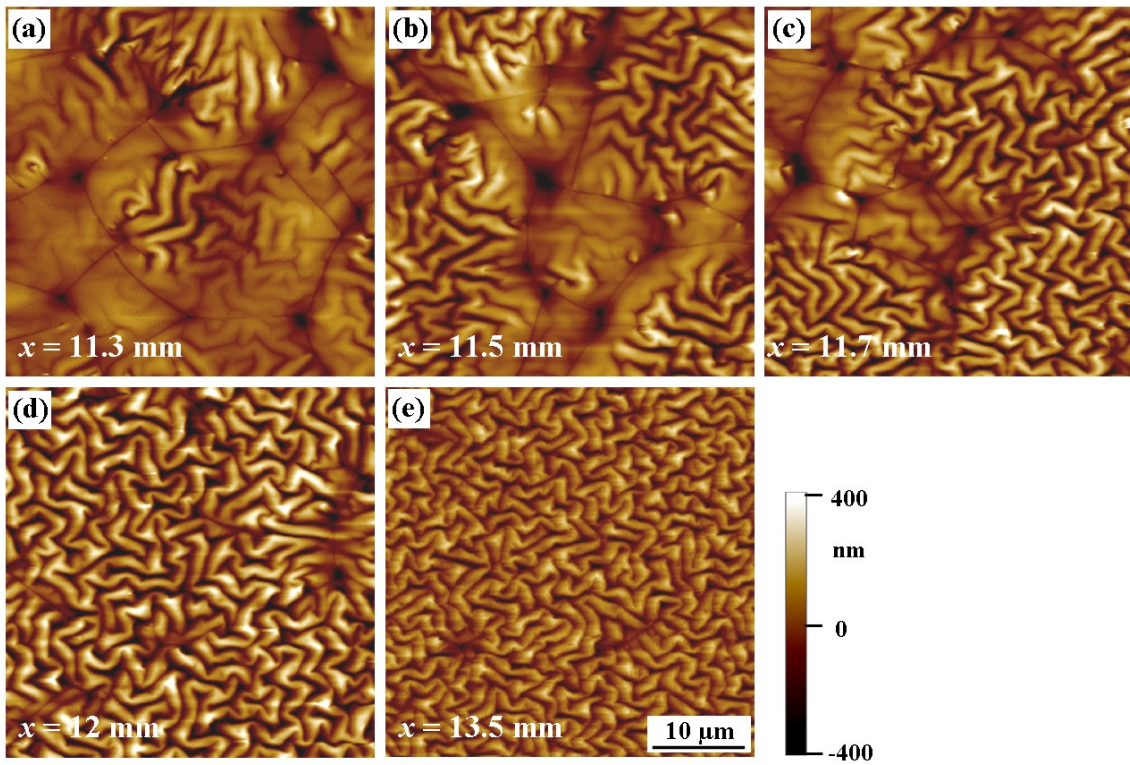


Figure S3. (a-e) Detailed transition from folds to wrinkles taken by the AFM. The sputtering time $t = 4$ sec. All images have the same size of $40 \times 40 \mu\text{m}^2$. The scale bar is applied to all the AFM images. (corresponding to Fig. 4 in main text)

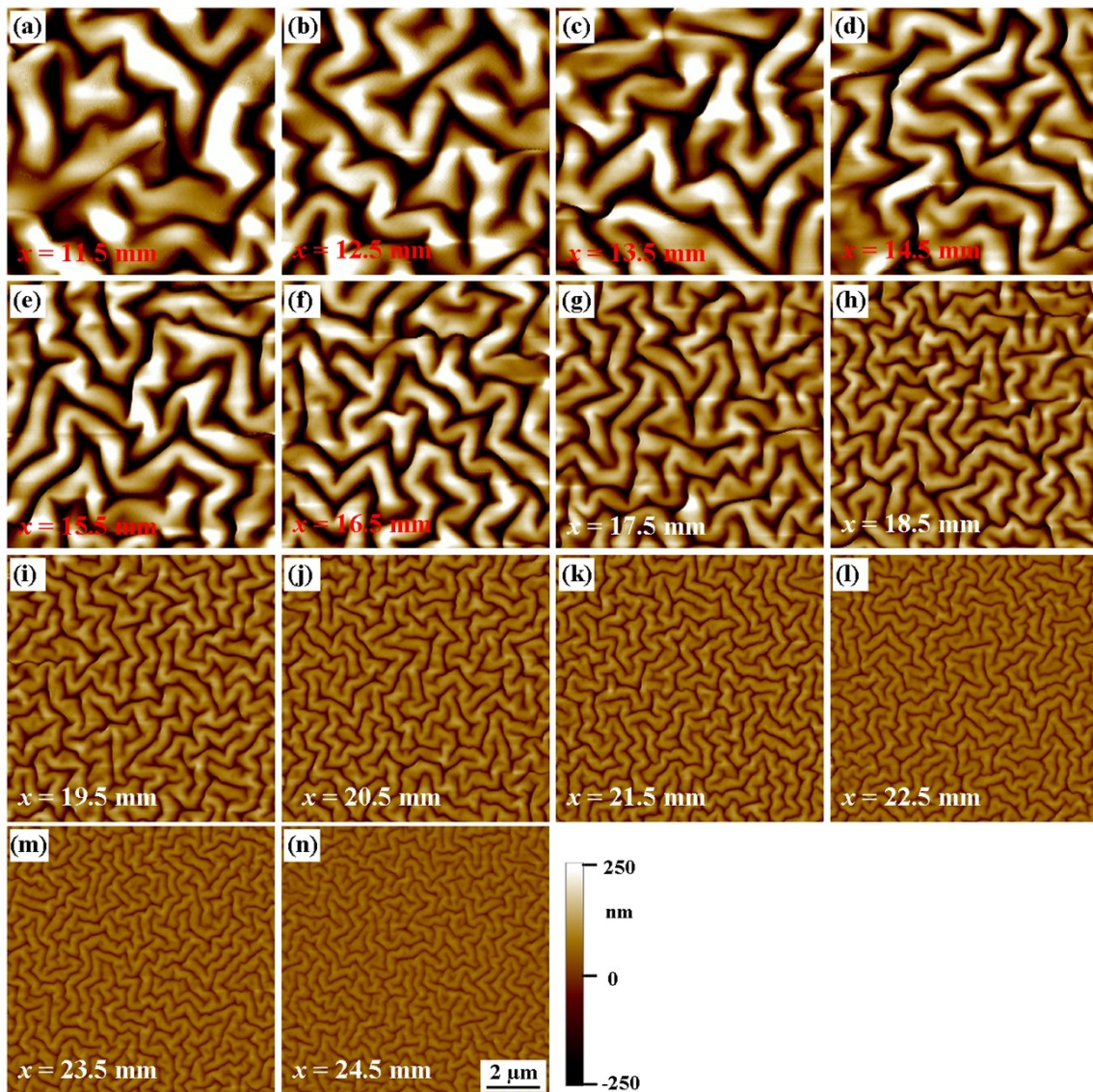


Figure S4. (a-n) Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 4$ sec in the wrinkle-dominant region taken by the AFM. All images have the same size of $10 \times 10 \mu\text{m}^2$. The scale bar is applied to all the AFM images. (corresponding to Fig. 5 in main text)

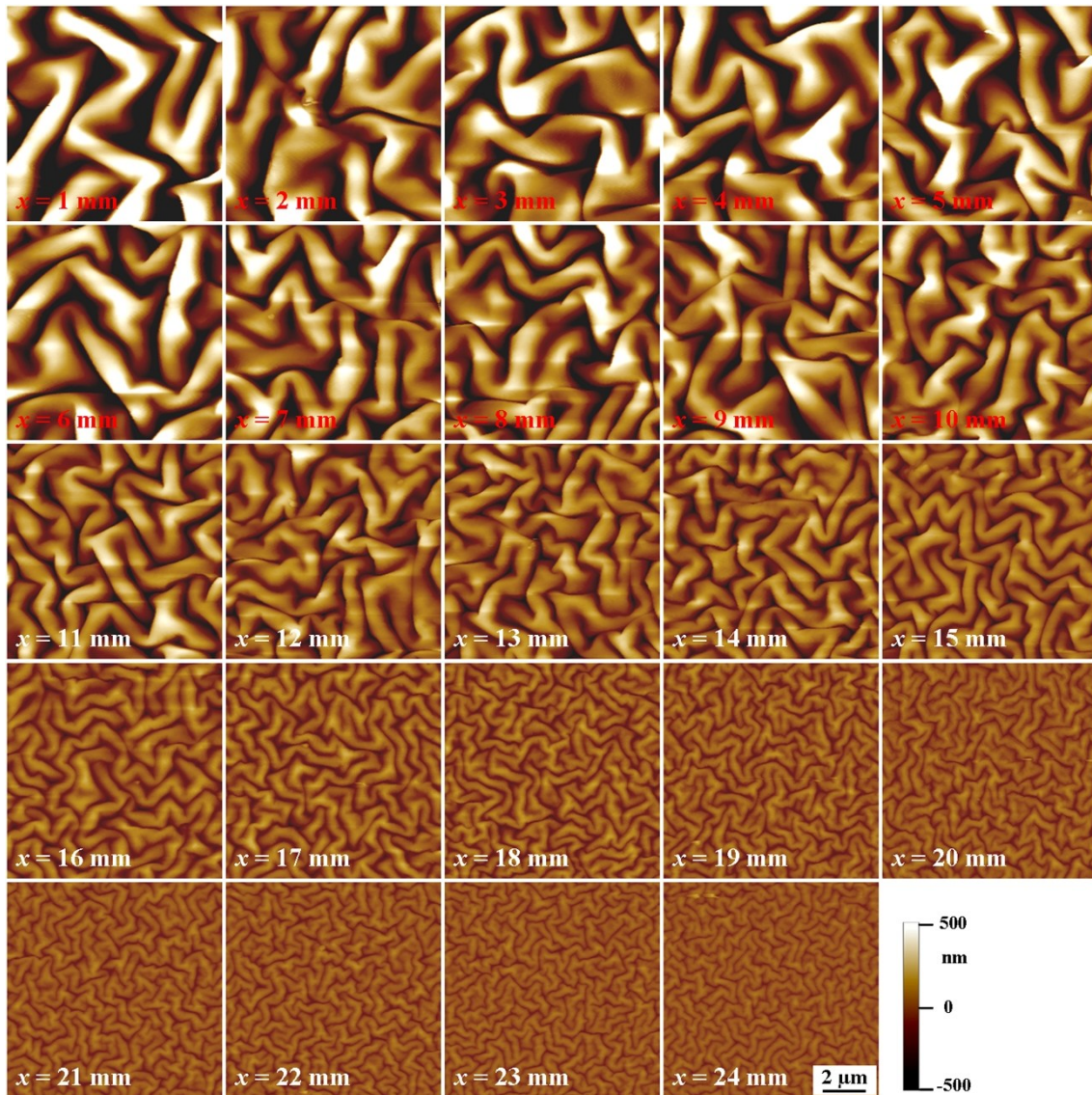


Figure S5. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 7$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $10 \times 10 \mu\text{m}^2$. The scale bar is applied to all the AFM images. 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)

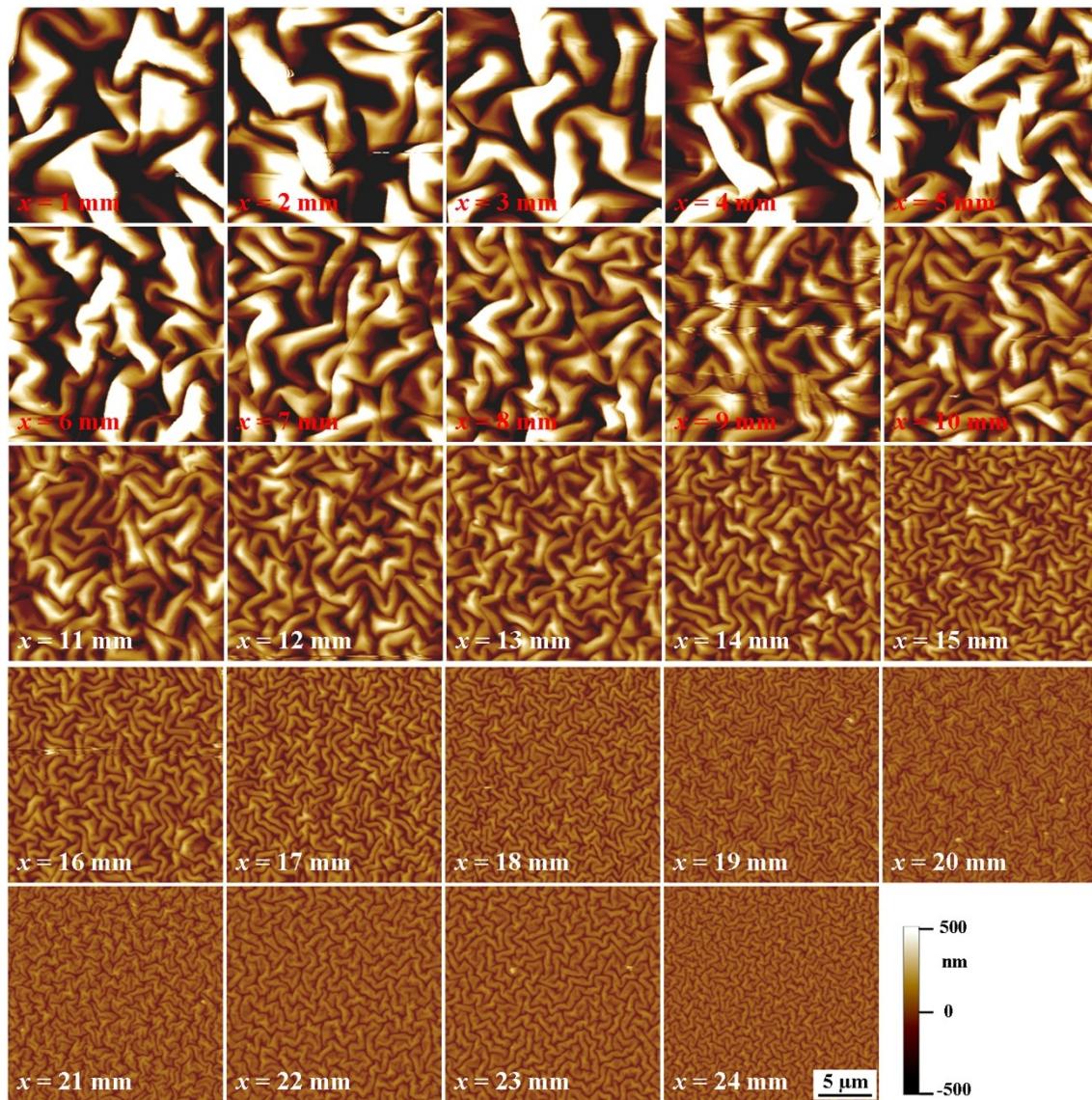


Figure S6. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 11$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $20 \times 20 \mu\text{m}^2$. The scale bar is applied to all the AFM images. 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)

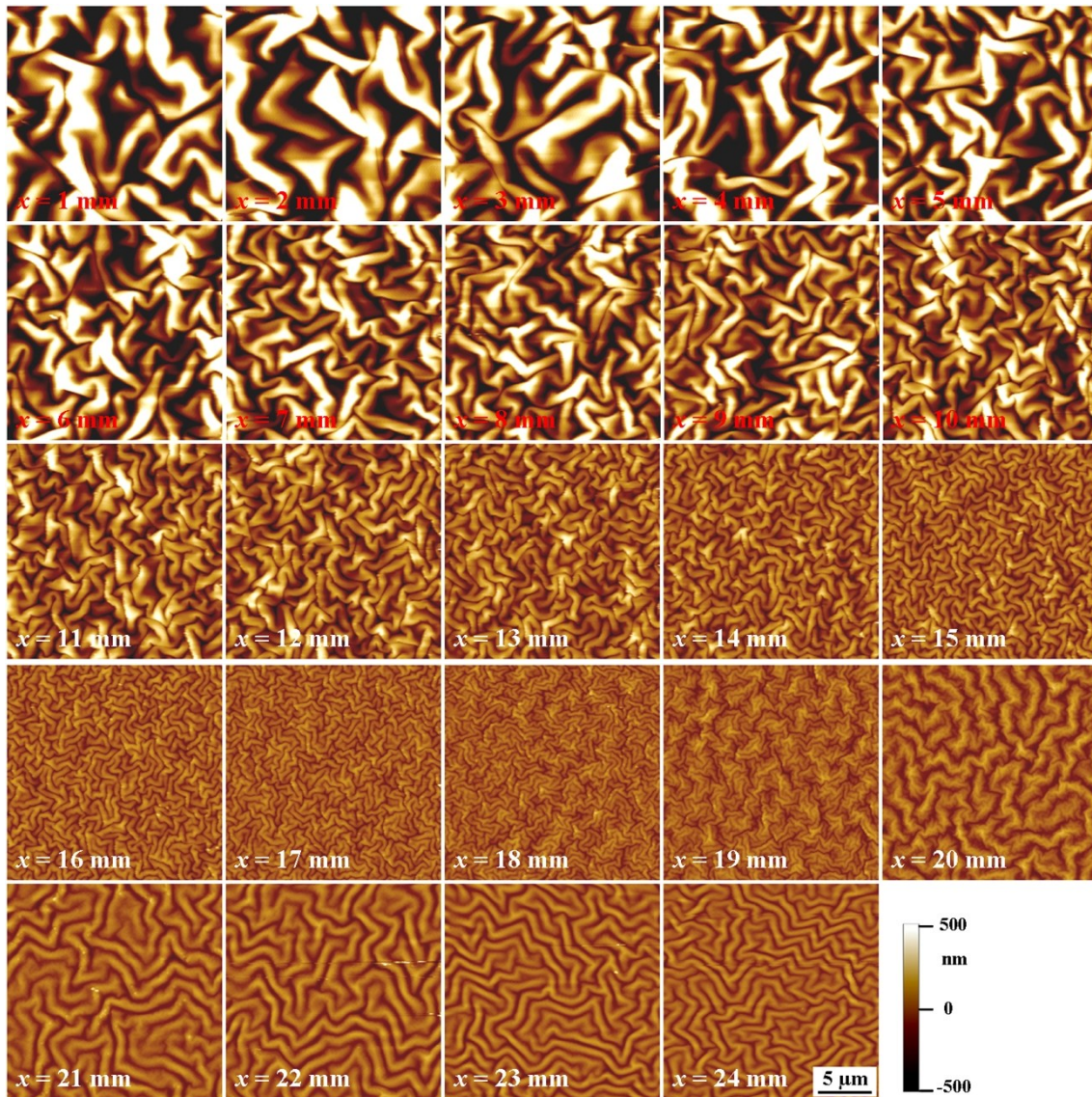


Figure S7. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 20$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $20 \times 20 \mu\text{m}^2$. The scale bar is applied to all the AFM images. 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)

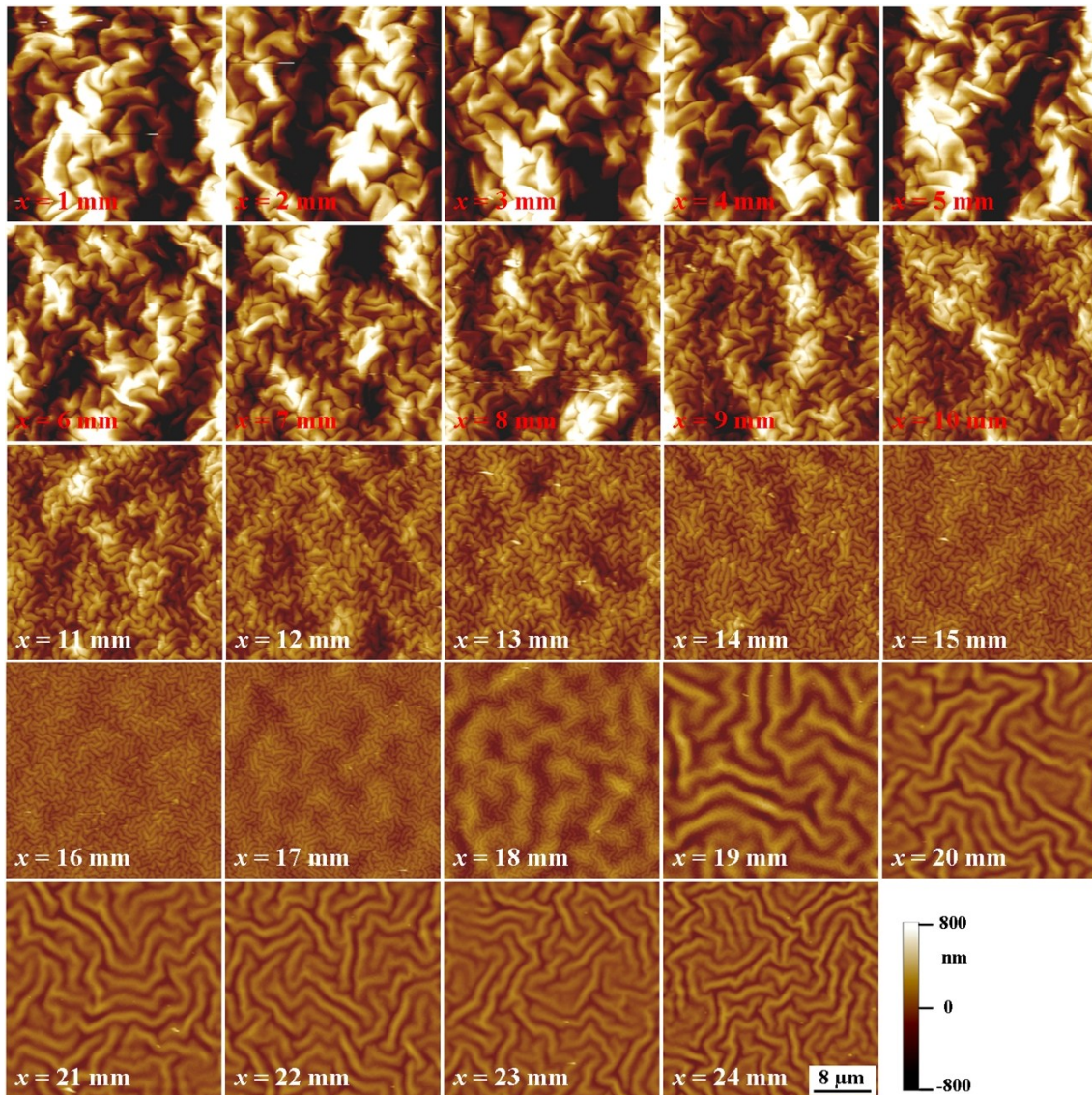


Figure S8. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 60$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $30 \times 30 \mu\text{m}^2$. The scale bar is applied to all the AFM images. 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)

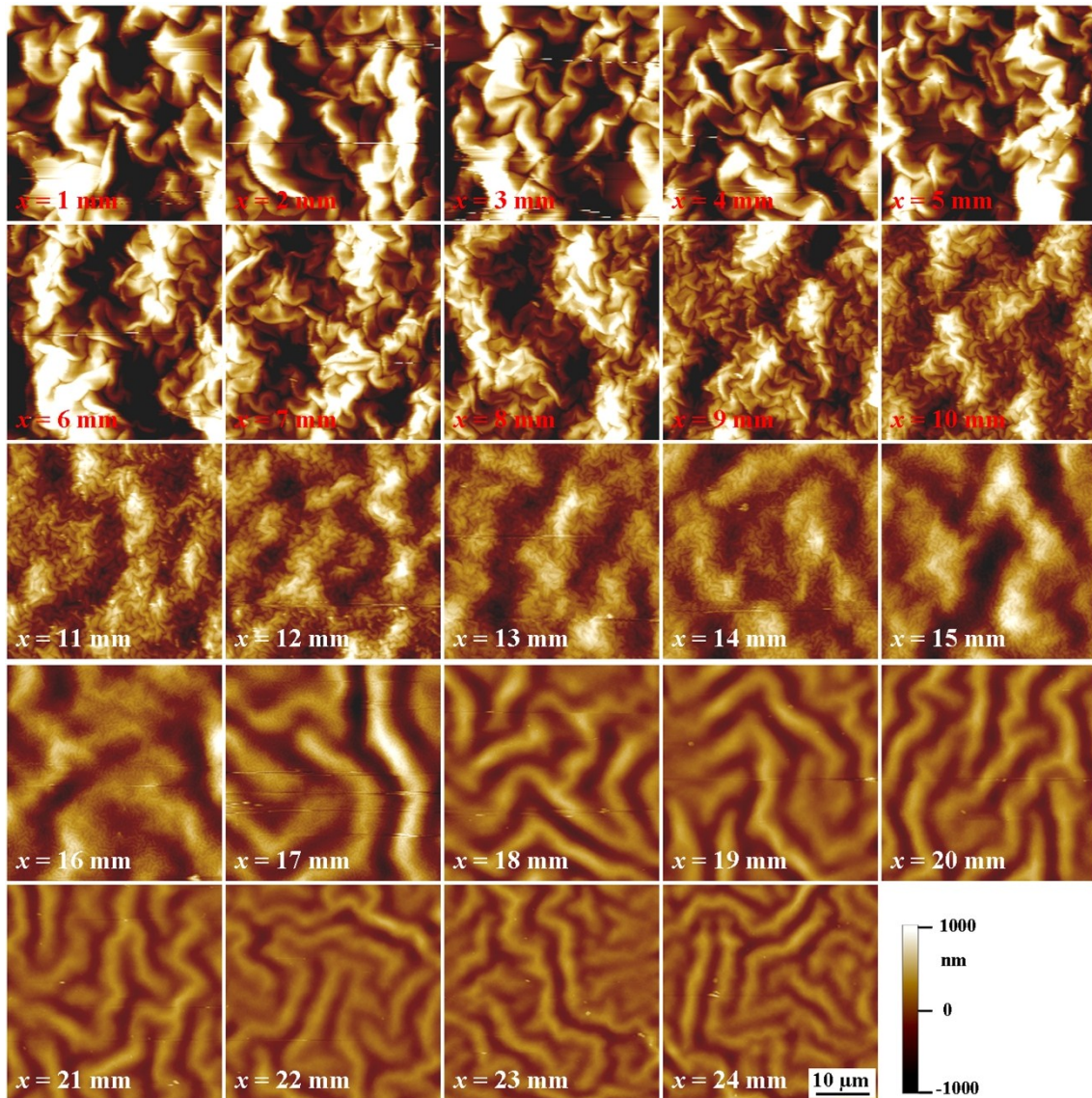


Figure S9. Morphological evolution of the gradient crosslinking PDMS surface with the sputtering time $t = 180$ sec taken by the AFM. The distance interval between the neighboring images is 1 mm. All images have the same size of $40 \times 40 \mu\text{m}^2$. The scale bar is applied to all the AFM images. 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)

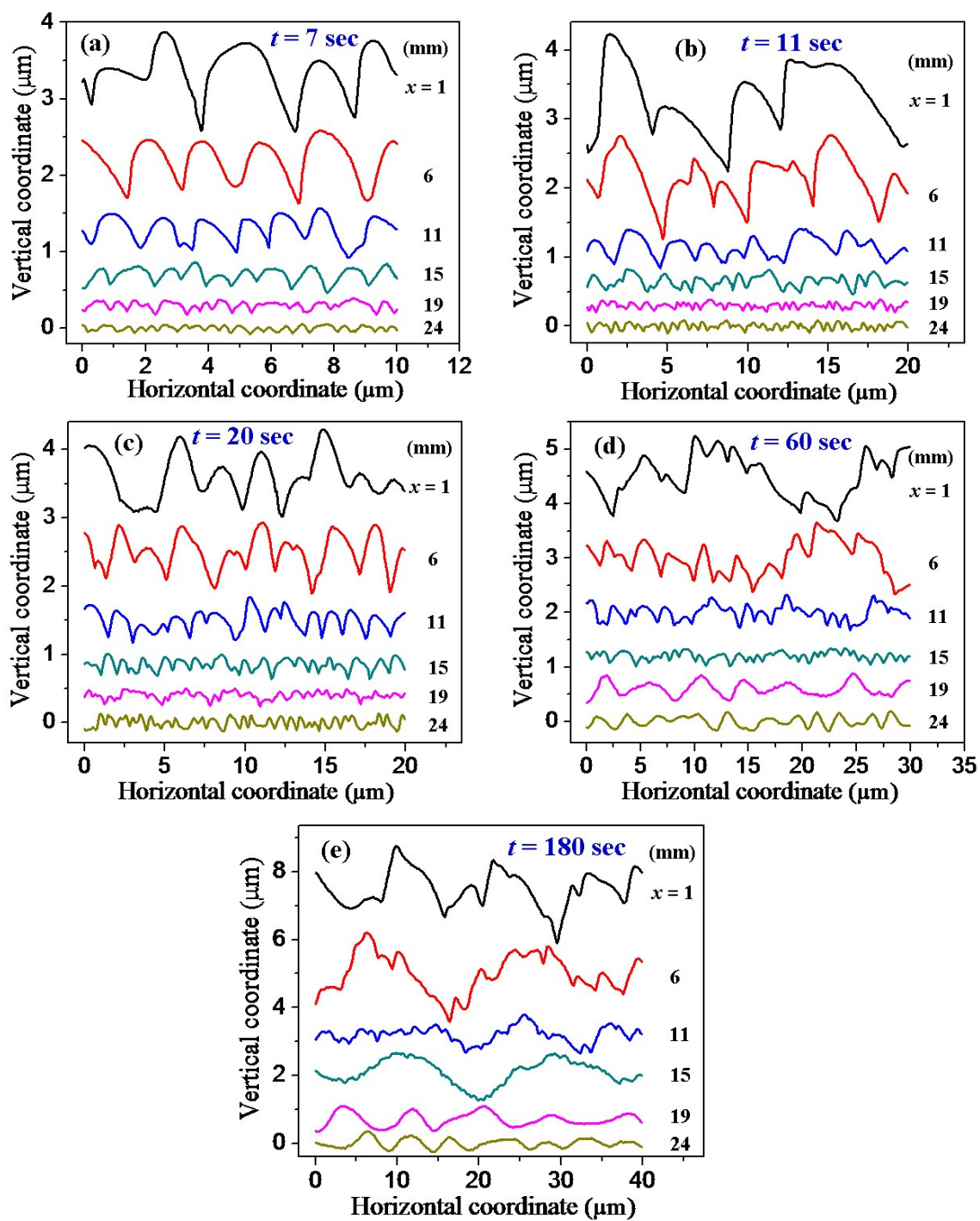


Figure S10. Comparisons of wrinkle profiles in the gradient crosslinking PDMS samples with varied sputtering times. (a) $t = 7 \text{ sec}$; (b) $t = 11 \text{ sec}$; (c) $t = 20 \text{ sec}$; (d) $t = 60 \text{ sec}$; (e) $t = 180 \text{ sec}$. The profile lines are shifted artificially along the vertical coordinate for clarity.

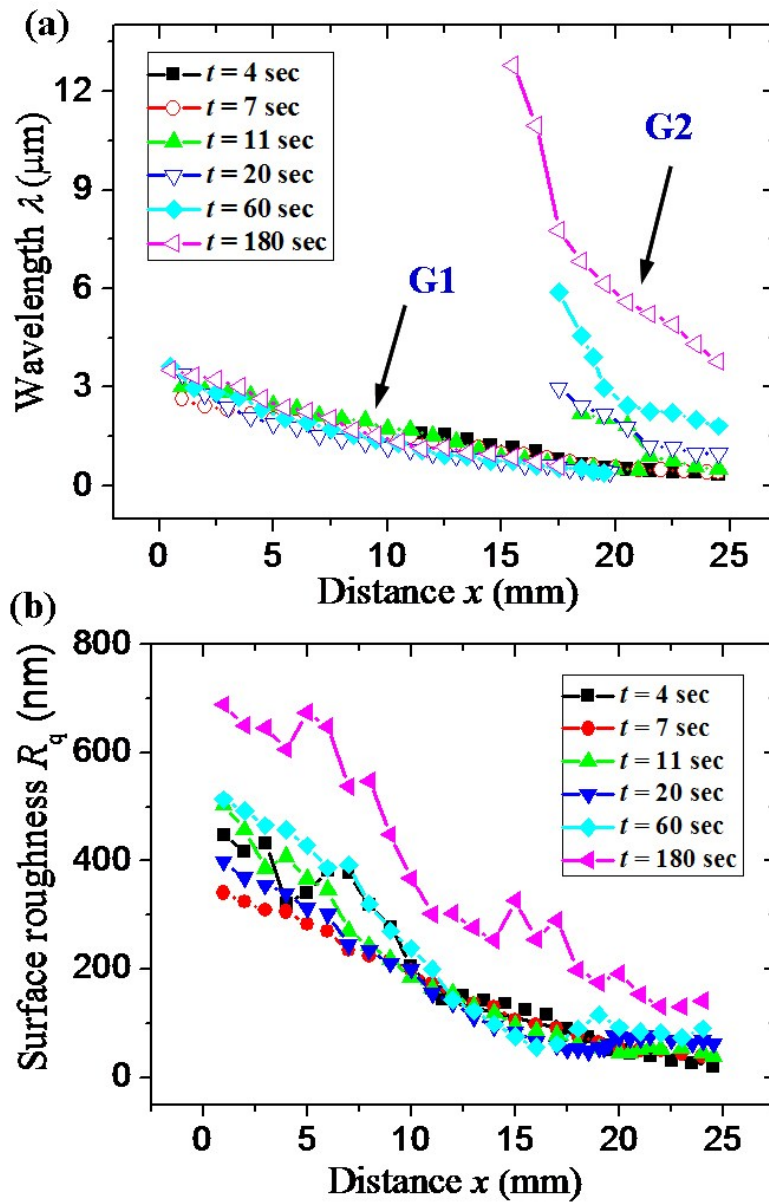


Figure S11. Evolutions of the wavelength λ of G1 and G2 wrinkles (a) and RMS surface roughness R_q (b) with the distance x for the gradient samples with varied sputtering times.

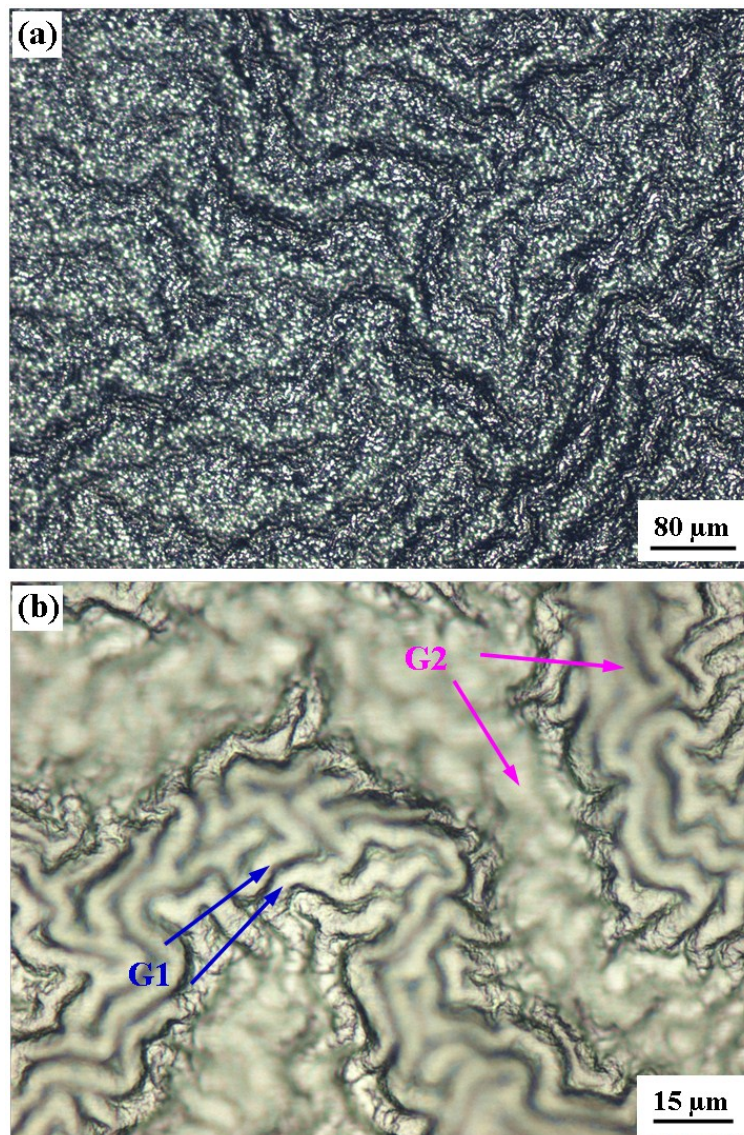


Figure S12. Typical hierarchical pattern composed of G1 and G2 wrinkles with different dimensions on the dielectric gel surface by sputtering of molybdenum atoms with $t = 360$ sec. Optical micrographs with (a) low magnification ($\times 100$) and (b) high magnification ($\times 500$).