Electronic Supplementary Material (ESI) for Soft Matter.

Achieving high aspect ratio wrinkles by modifying material network stress

Supporting information

1. Materials

The materials used to make the substrate and the films in this study. Their thickness and the Young's moduli are listed in Table S1.

Table S1. The thickness and Young's moduli of the materials used.

Molding experiments		Post-curing experiments			Living silicone experiments	
Film	Substrate	Film	Substrate (under-cured)	Substrate (post-cured)	Film	Substrate
PAH/PSS	Sylgard 184 40:1	Sylgard 184 5:1	Sylgard 184 40:1	Sylgard 184 40:1	Momentive LSR 7070	Living silicone
80~120 nm	~5mm	2.5 ± 0.5 μm	~5mm	~5mm	8 ± 0.6 μm	~2mm
300 ± 30 MPa[1]	40 ± 10 kPa[a]	~1 MPa[b]	6 ± 2 kPa[a]	40 ± 10 kPa[a]	5.5 MPa[c]	389 ± 19 kPa[2]

a. Obtained with an Instron tensile tester. The material is molded into a 30x5x3 mm dogbone shape, and the test is performed at a displacement rate of 100 mm/min.
b. Calculated using equation 1, with λ = 60 μm, $\bar{E}_s$ = 6/(1-0.5^2) kPa, t = 2.5 μm, and $\bar{E}_f$ = E_f/(1-0.5^2), where E_f is the Young's modulus and taking Poisson's ratio as 0.5.
c. Obtained from the data sheet provided by Momentive Inc.

1. A. J. Nolte, M. F. Rubner and R. E. Cohen, Macromolecules, 2005, **38**, 5367–5370.
2. P. Zheng and T. J. McCarthy, *J. Am. Chem. Soc.*, 2012, **134**, 2024–7.

2. Aspect ratio as a function of applied strain for bare wavy substrate without top films

The wavy substrate without the top film localizes at relatively small applied strain. The surface localizes and forms a crease-like topography at low applied strains (~0.1).

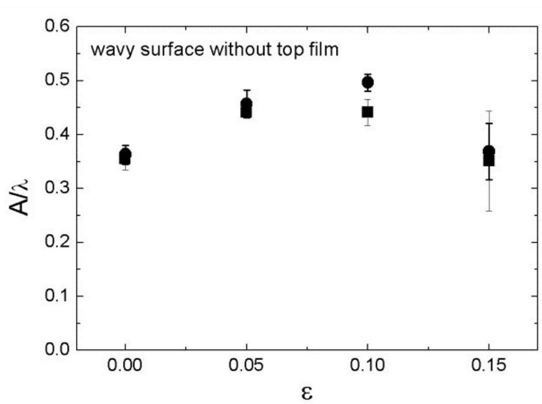

Figure S1. The aspect ratio as a function of strain for the bare substrate without the top film.

3. Wrinkle wavelength and the number of layers

In the molding experiment, the top film thickness is controlled so that the wavelength matches the wavelength of the pre-molded surfaces. The wavelength obtained on 40:1 PDMS and 30:1 PDMS is proportional to the number of layers as shown in figure S1.

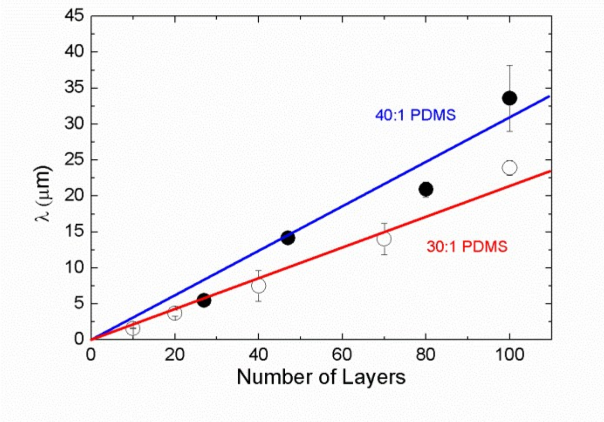

Figure S2. Wavelength as a function of layer number. This plot is used to determine how many layers are to be used for a given wavelength.

4. Different A_m/λ_m used in Fig. 6 for the post-curing experiment.

Table. S2. Amplitude and wavelength used in Fig. 6 for the post-curing experiment

A_m	λ_m	$(A_m/\lambda_m)^2$	A_F	λ_F	$(\lambda_F/\lambda_m)(A_F/\lambda_F)^2$
0	60.0*	0	25.3	46.2	0.23
9.3	43.1	0.046	17.5	25.1	0.28
9.6	43.2	0.049	19.5	29.3	0.30
12.3	37.9	0.11	19.0	27.9	0.34
27.0	54.0	0.25	34.1	39.0	0.55

*obtained by applying a strain slightly above the critical strain.

In Fig. S3, We provide the aspect ratio changes for different A_m/λ_m used. It is very likely that the post-curing step introduce thermal stress to the material. Thus, the aspect ratio increases by the post-curing step. The aspect ratios before and after the post-curing step are both included in the plot. Note that the aspect ratio before post-curing is used as A_m/λ_m.

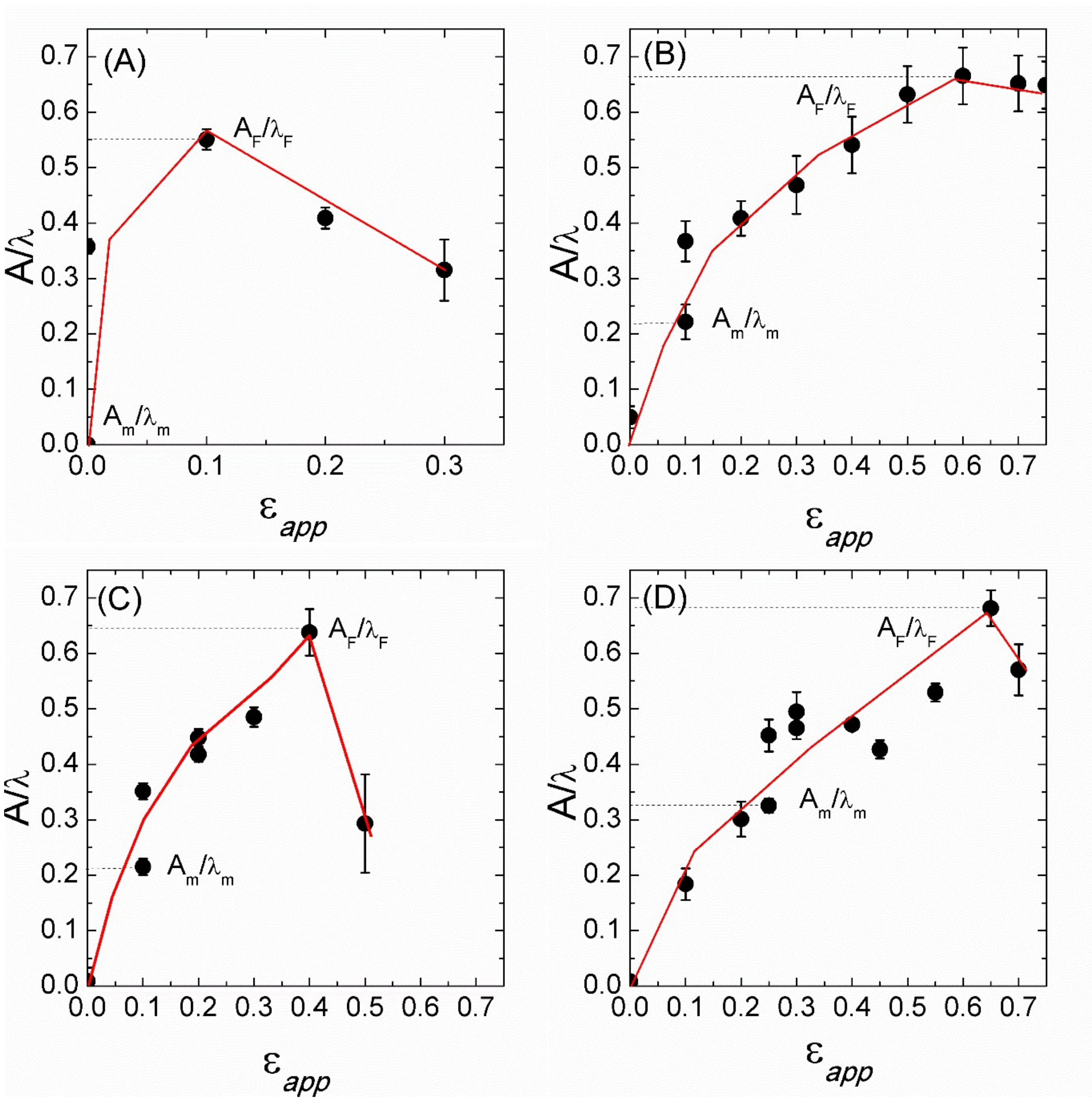

Figure S3. Post-curing experiments of different values of A_m/λ_m. The red solid lines are only for guiding of eyes.