Electronic Supplementary Information for

Importance of interfacial adhesion in the buckling-based mechanical characterization of materials

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PS (Mw=35,000g/mol, Sigma Aldrich)



PS film on 10:1 PDMS





Young's modulus of PDMS from the slope

	Young's modulus (MPa)	R-square
10:1	1.0978	0.9977
5:1	2.3472	0.9979

Fig. S1 Mechanical moduli of PDMS substrates by the buckling method, using PS as a sensor film. The moduli of PDMS substrates (5:1 and 10:1) were extracted from the slope of λ vs. *h*(PS film thickness) plot. Again, the modulus of 5:1 PDMS is much larger than that from microtensile test.



Fig. S2 Electron microscopy images of PDMS substrates, by (a, b) SEM and (c, d) TEM, respectively. Note here that there is no noticeable difference in internal micro/nano structures in PDMS substrates having different mixing ratio.



PS (Mw=35,000g/mol, Sigma Aldrich), Thickness of PS film = 187nm

Fig. S3 OM images of the PS film morphology on elastomeric PDMS substrates for (a) 10:1 and (b) 5:1, as a function of mechanical compression and release. B, C, and D in each image denote buckling, crack, and delamination, respectively. Note that the cracks/delaminations occur after the wrinkling on 10:1 PDMS while cracks/delaminations/wrinkles occur simultaneously on 5:1 PDMS.



PS (Mw=350,000g/mol, Sigma Aldrich), Thickness of PS film = 230nm

Fig. S4. (a) Surface evolution of 10:1 PDMS by optical profiler, images and their corresponding height distributions under each image. The Gaussian fit of the experimental height distribution is plotted as solid red lines. The bimodal height distribution for the last image contains deconvoluted (in green) and their sum (in red) peaks. The sample was compressed infinitesimally (so, exact strain values could not be determined) until uniform buckling was obtained.



PS (Mw=350,000g/mol, Sigma Aldrich), Thickness of PS film = 230nm

Fig. S4. (b) Surface evolution of 5:1 PDMS by optical profiler, images and their corresponding height distributions under each image. The Gaussian fit of the experimental height distribution is plotted as solid red lines. The bimodal height distribution for the last 3 images contains deconvoluted (in green) and their sum (in red) peaks.



Fig. S4 (c) Evolution of RMS surface roughness as a function of compression.



Fig. S5. AFM height (left) and phase (right) images of PS film on PDMS without any external strain. It should be noted that the display range for height image is 1.55nm and RMS roughness is ~0.4nm, which means that the nanoscale bumps observed by optical profiler under compression (Fig. 4 in the main text) is the interfacial debonds, rather than intrinsic film defects or nonuniformities.

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Fig. S6 Mechanical buckling of ultrathin PS films, thinner than 100nm, on PDMS substrates. The wavelength data from these images (OM & AFM) were used to plot Fig. 5a in the main text.



PS (Mw=35,000g/mol, Sigma Aldrich), Thickness of PS film = 170nm

Fig. S7 Thermal buckling of PS film on 10:1 PDMS substrates. The PS film was transferred onto pre-heated PDMS substrate at 50°C, then naturally cooled down to room temperature. (a) OM images of wrinkled PS surface and (b) buckling wavelength and Young's modulus, as a function of time, respectively.