

Controlling line defects in wrinkling: A pathway towards hierarchical wrinkling structures

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Determination of characteristic wrinkle properties:

Characteristic values like wavelength and amplitude were computed with a self-programmed Python 3.0 script. The measured topographical images (resolution of 512 × 512 pixels) were scanned line by line perpendicular to the wrinkle direction. An algorithm searched for local height minima and maxima. Depending on the size of the image and the number of wrinkles, it was possible to get mean values and deviations of the wavelength and amplitude, as well as their changes. The wavelength was calculated as arithmetic mean of all lateral distances between local adjacent minima and the amplitude was calculated as vertical distances between local adjacent minima and maxima height values for all detected local extrema. Figure S1 shows an example of wavelength and amplitude quantification based on a topographical wrinkle image.

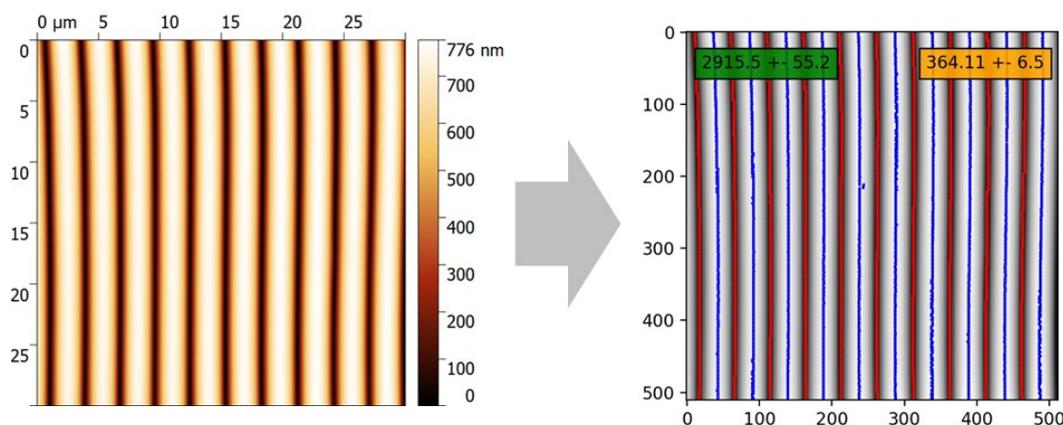


Figure S1: a) Topographical image of a wrinkled surface, b) visualization of detected minima (red lines) and maxima (blue lines) with the calculated wavelength (green box: arithmetic mean and deviation) and amplitude (orange box: arithmetic mean and deviation)

Different component ratios of PDMS and their influence on the wrinkling patterns

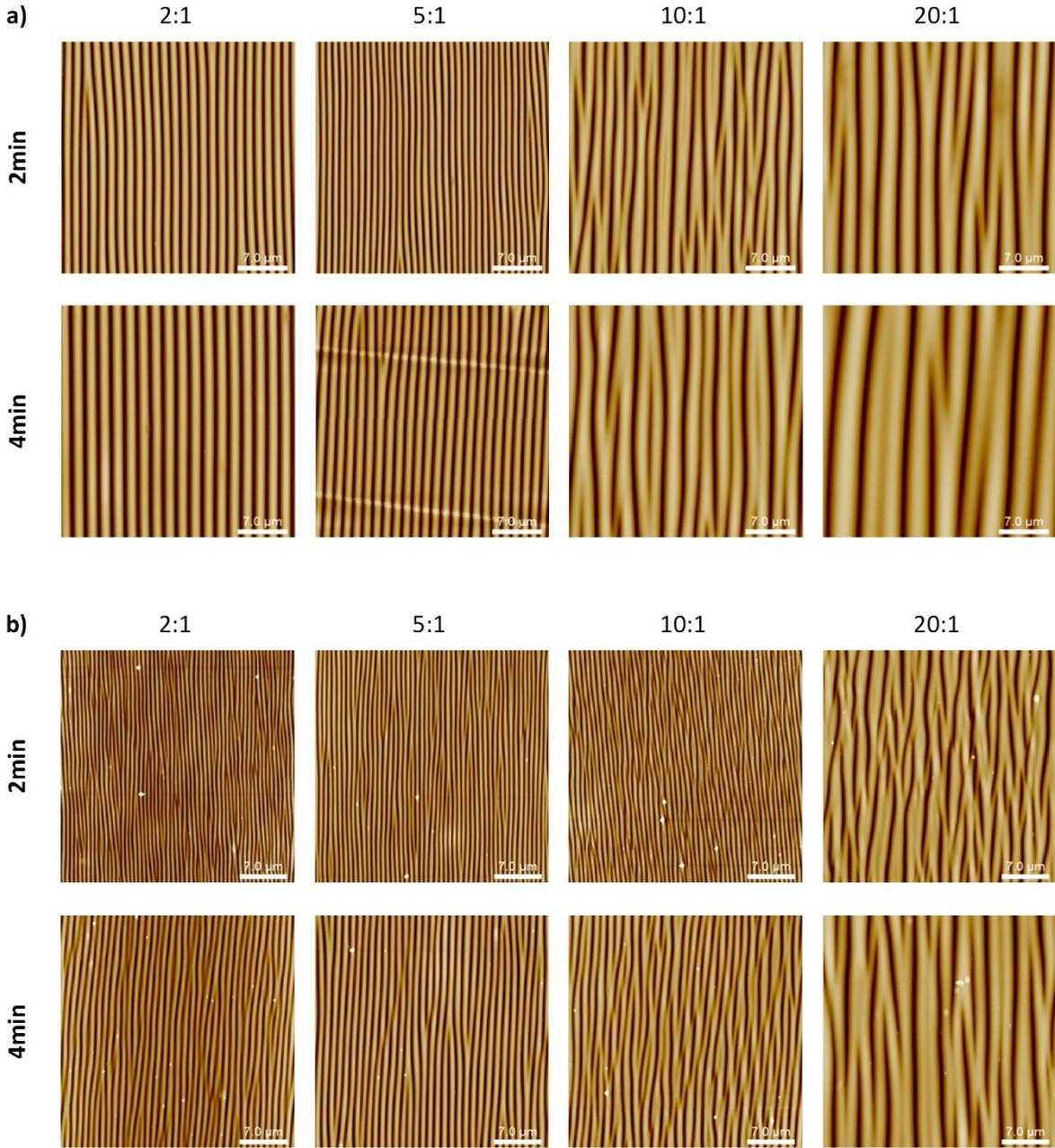


Figure S2: Topographical wrinkle images of the component ratio screening with N₂ plasma treatment at 30% strain for component ratios of Sylgard 184 varied from 2:1 to 20:1 (base polymer to hardener component) and two treatment times for a) substrate curing condition 4h at 80°C and b) substrate curing condition 4h at 80°C + 48h 165°C (scale bar is 7μm, dark color represent the topographical minimum and the light color the topographical maximum)

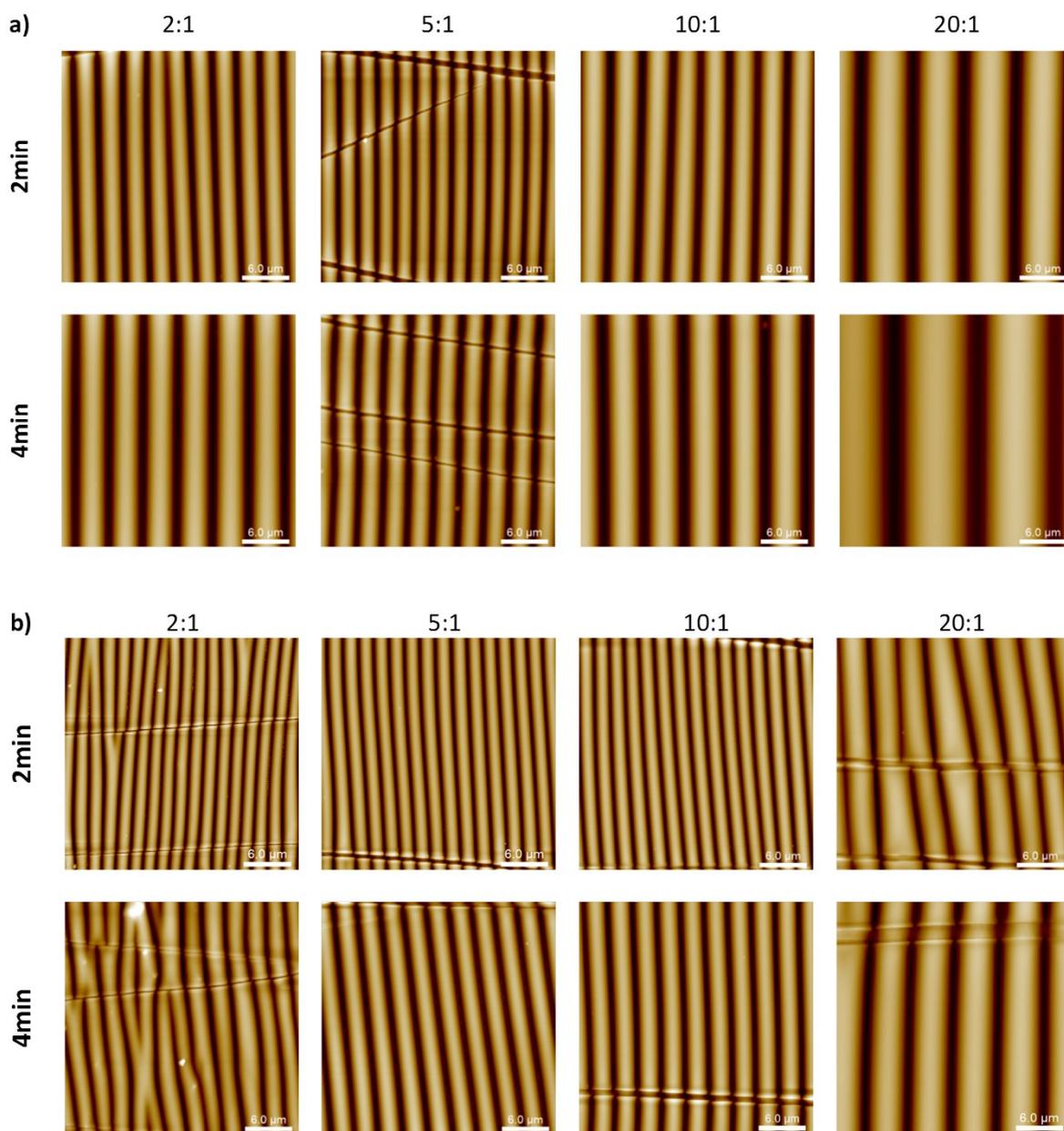
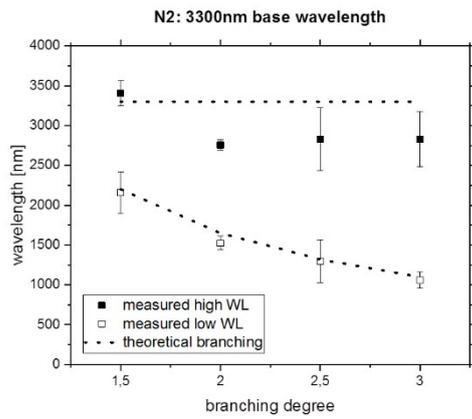


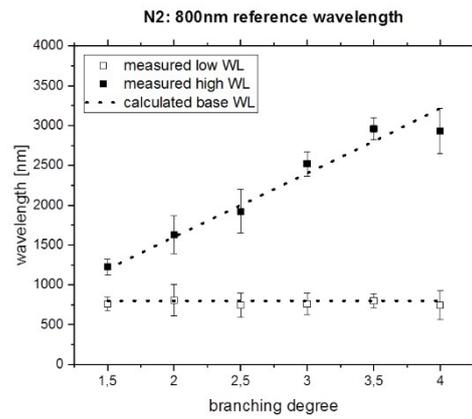
Figure S3: Topographical wrinkle images of the component ratio screening with H_2 plasma treatment at 30% strain for component ratios of Sylgard 184 varied from 2:1 to 20:1 (base polymer to hardener component) and two treatment times for a) substrate curing condition 4h at 80°C and b) substrate curing condition 4h at 80°C + 48h 165°C (scale bar is 6 μm , dark color represent the topographical minimum and the light color the topographical maximum)

Branching results for different plasma gases:

a)



b)



c)

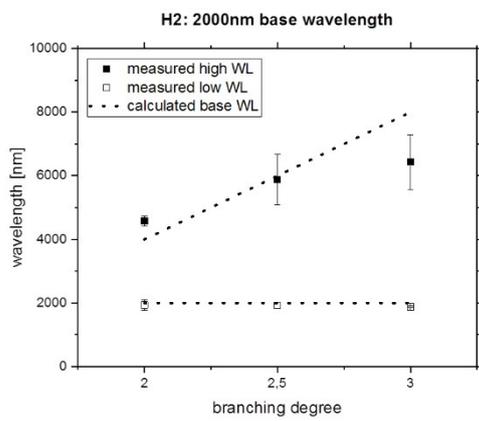


Figure S4 Results of different branching experiments: a) N_2 plasma gas with 3300nm base λ , b) N_2 plasma gas with 800nm base λ , c) H_2 plasma gas with 2000nm base λ

QNM measurements and determination of the influence of low-pressure plasma treatment:

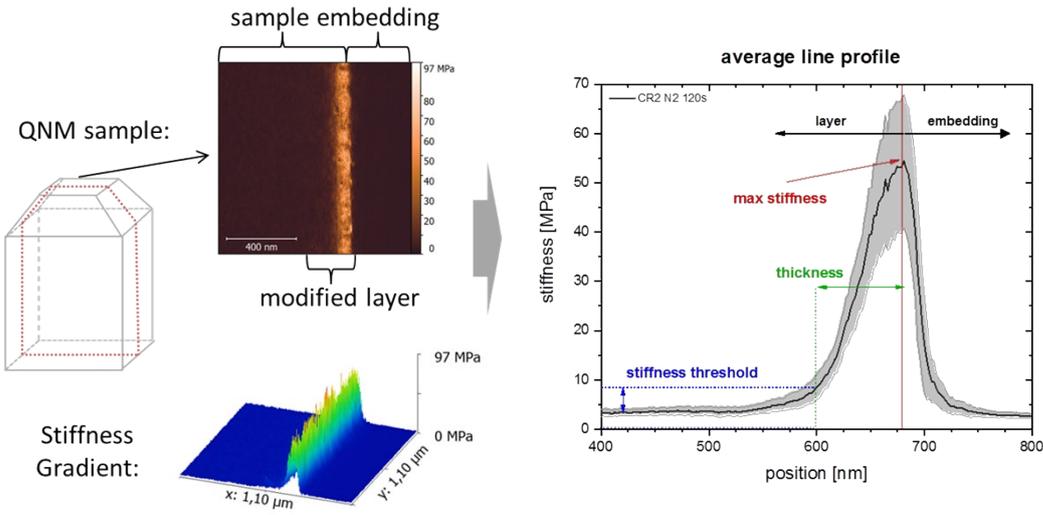


Figure S5: QNM sample preparation and determination of thickness and stiffness

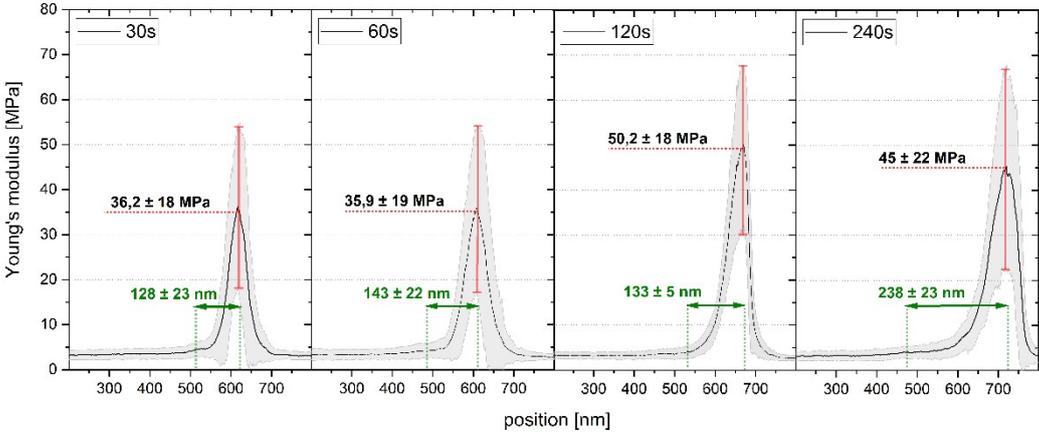


Figure S6: stiffness profiles with estimated thickness h_f and maximal stiffness $E_{f,max}$ for CR2 at different treatment times with N_2 process gas: h_f (green values) represents the film thickness from 25% increase of the measured stiffness to the stiffness maxima $E_{f,max}$

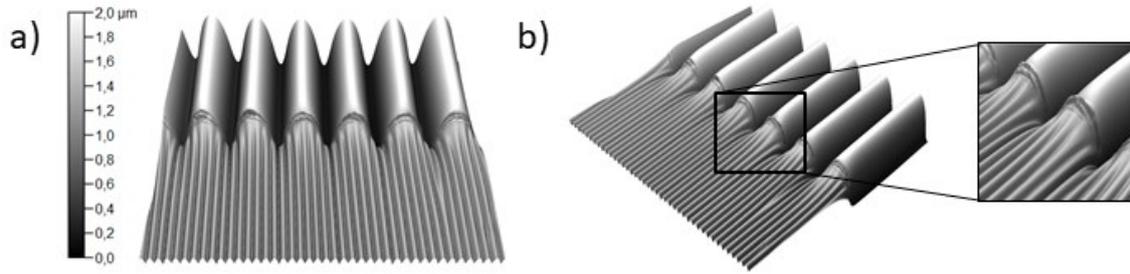


Figure S7: Detailed view and perspectives of branching degrees up to seven produced with a combination of process gases N_2 and H_2

Simulation Parameters:

C_{10}	$-1.67 \times 10^6 \text{ Pa}$
C_{01}	$1.94 \times 10^6 \text{ Pa}$
C_{20}	$2.42 \times 10^6 \text{ Pa}$
C_{02}	$6.52 \times 10^6 \text{ Pa}$
C_{11}	$-7.34 \times 10^6 \text{ Pa}$
k	$57 \times 10^6 \text{ Pa}$

Table S1: Mooney-Rivlin parameters for the substrate material CR2

description	variable	value upper half	value lower half	unit
<i>thickness</i>	h	2.385×10^{-7}	1.425×10^{-7}	m
<i>Lamé's first parameter</i>	λ	1.05×10^7	9.23×10^6	Pa
<i>Lamé's second parameter</i>	μ	4.55×10^6	5.19×10^6	Pa
	L_c		0.001	
	b_1, b_2, b_3		1	
	μ_c		0	

Table S2: Parameters for the Cosserat shell