Electronic Supplementary Material (ESI) for RSC Advances.

Supplemental Information

Tunable surface wettability and pH-responsive 2D structures from amphiphilic and amphoteric protein microfibrils

Xingchen Liu and You-Lo Hsieh

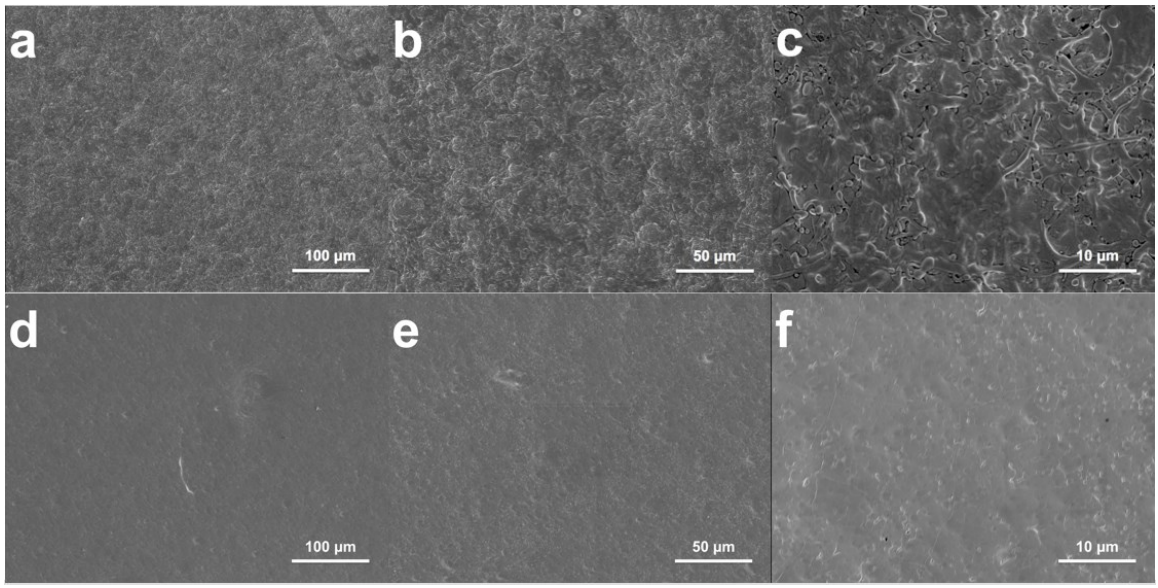


Figure S1. SEM images of film #1 (21 °C): a-c. top surface; d-f. bottom surface.

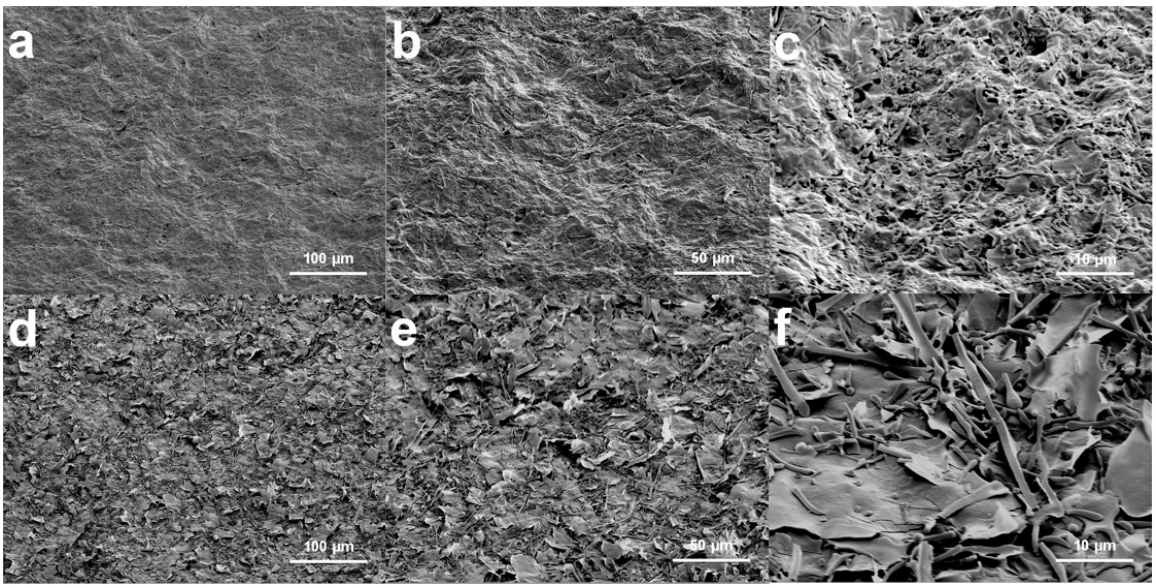


Figure S2. SEM images of film #2 (21 °C, vacuum): a-c. top surface; d-f. bottom surface.

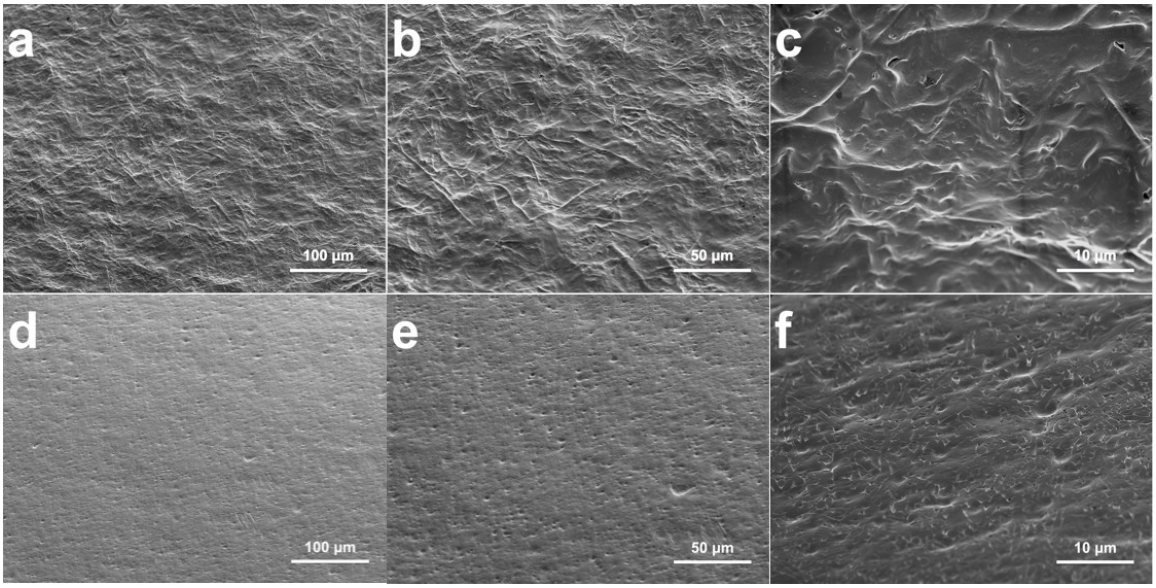


Figure S3. SEM images of film #3 (21 °C, vacuum, moisture): a-c. top surface; d-f. bottom surface.

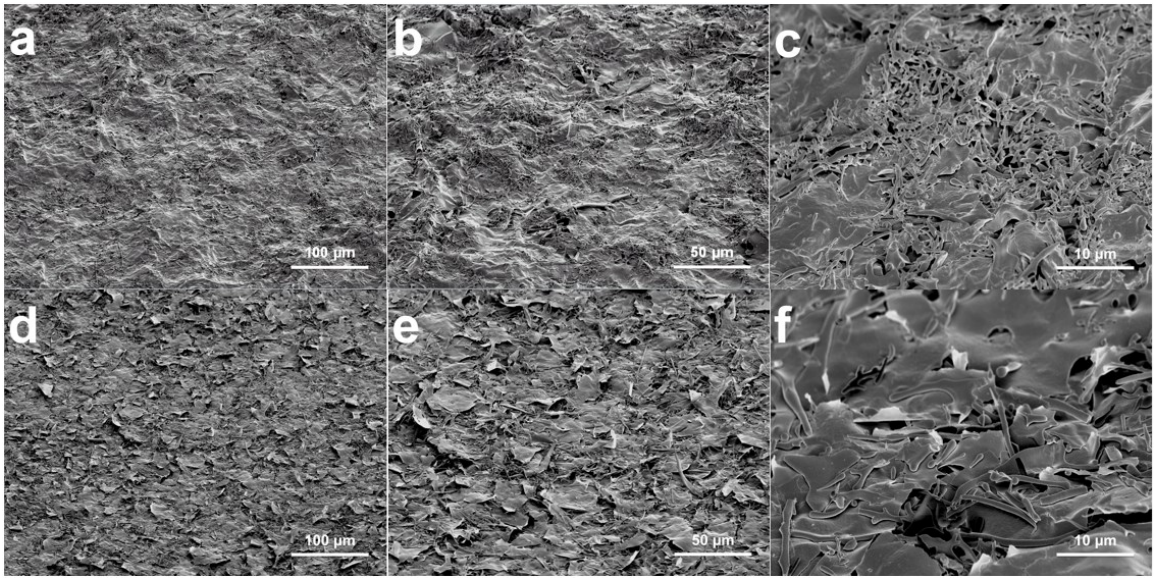


Figure S4. SEM images of film #4 (65 °C): a-c. top surface; d-f. bottom surface.

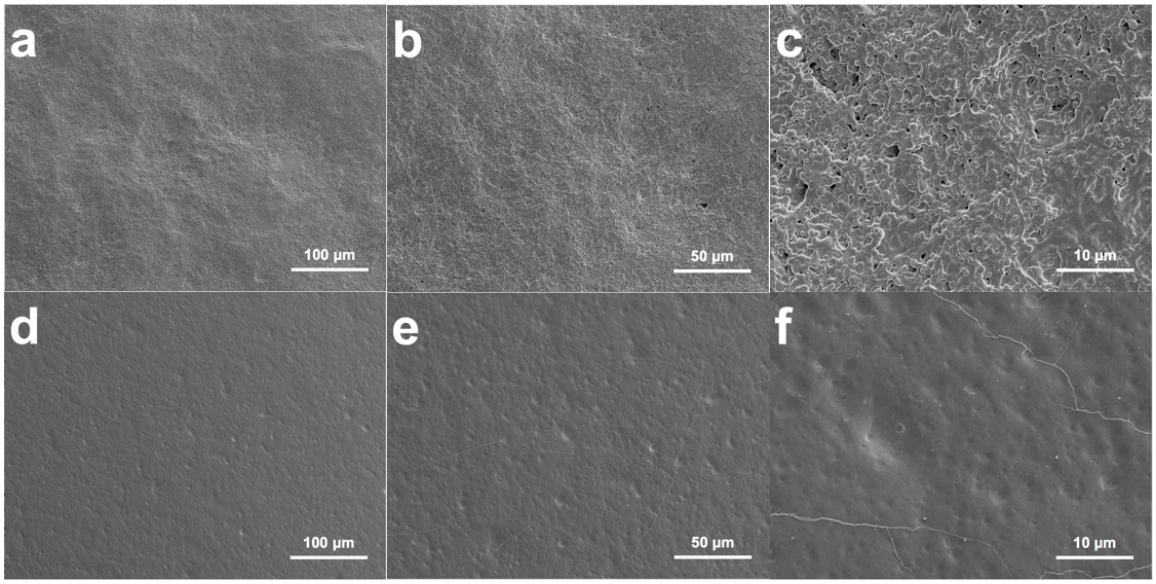


Figure S5. SEM images of film #5 (65 °C, vacuum, moisture): a-c. top surface; d-f. bottom surface.

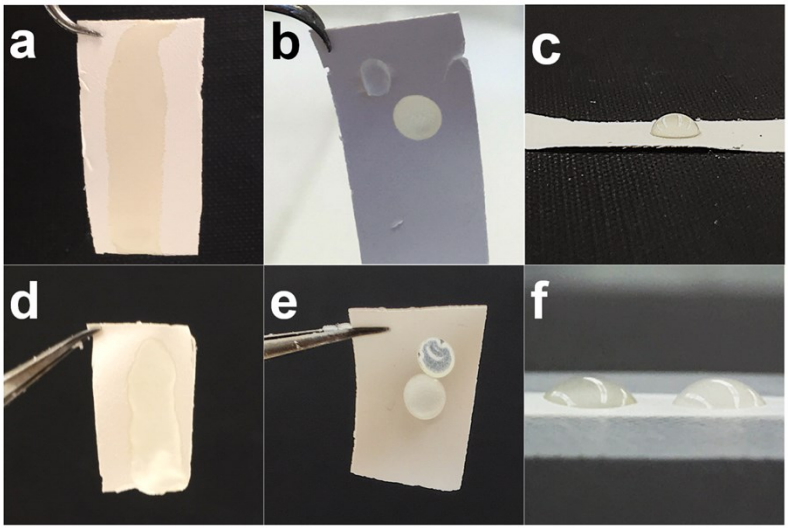
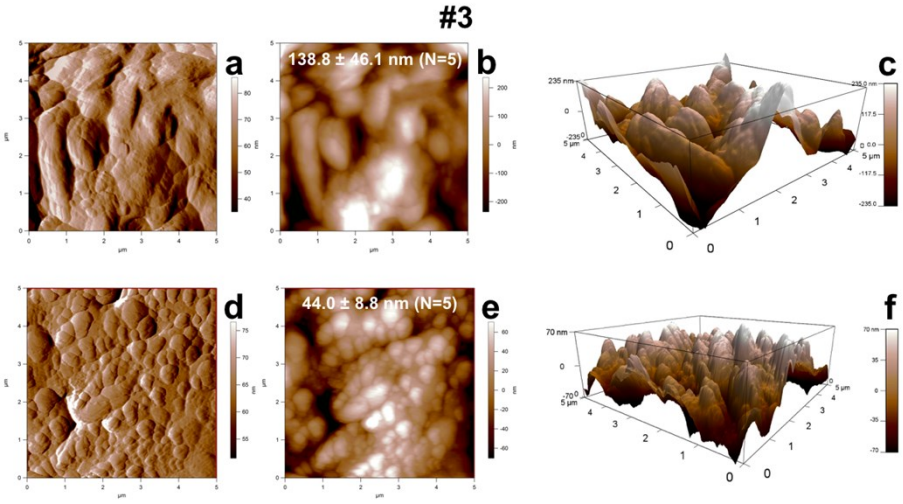


Figure S6. Water drops on top (a and d) and bottom (b-c and e-f) surfaces of film #2 (21 °C, vacuum, a-c) and #4 (65 °C, d-f) and: films were vertical in a-b and d-e and horizontal in c and f.



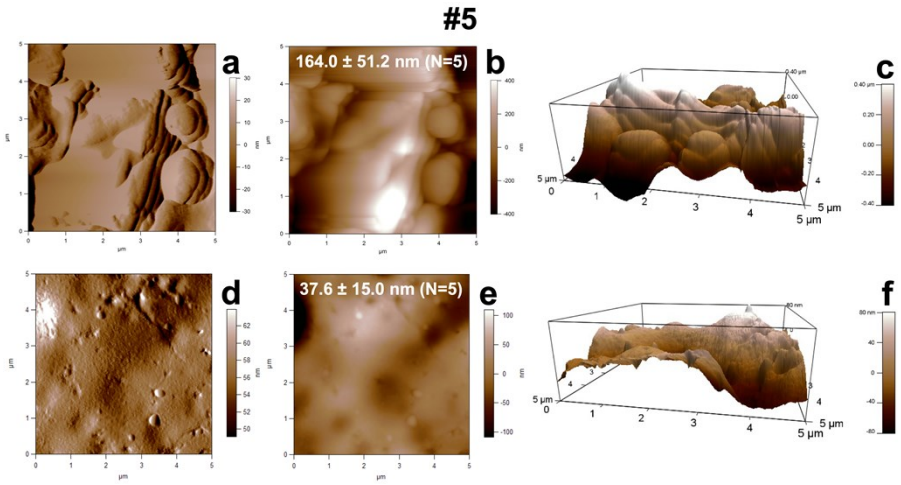
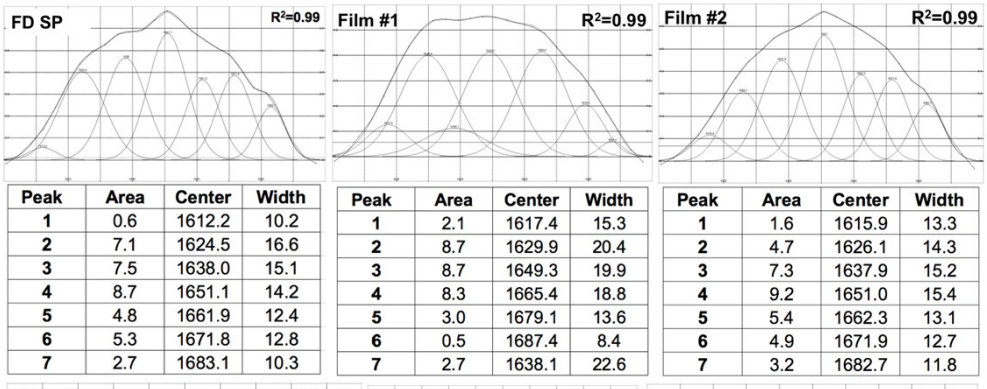


Figure S7. AFM amplitude (a, d), height (b, e) and 3D (c, f) images of the top (a-c) and bottom (d-f) surface of film #3 (21 °C, vacuum, moisture) and #5 (65 °C, vacuum, moisture).



Peak	Area	Center	Width
1	0.6	1612.2	10.2
2	7.1	1624.5	16.6
3	7.5	1638.0	15.1
4	8.7	1651.1	14.2
5	4.8	1661.9	12.4
6	5.3	1671.8	12.8
7	2.7	1683.1	10.3

Peak	Area	Center	Width
1	2.1	1617.4	15.3
2	8.7	1629.9	20.4
3	8.7	1649.3	19.9
4	8.3	1665.4	18.8
5	3.0	1679.1	13.6
6	0.5	1687.4	8.4
7	2.7	1638.1	22.6

Peak	Area	Center	Width
1	1.6	1615.9	13.3
2	4.7	1626.1	14.3
3	7.3	1637.9	15.2
4	9.2	1651.0	15.4
5	5.4	1662.3	13.1
6	4.9	1671.9	12.7
7	3.2	1682.7	11.8

Peak	Area	Center	Width
1	0.5	1610.0	6.8
2	3.2	1618.8	12.8
3	8.6	1629.3	16.9
4	12.7	1653.6	20.6
5	10.3	1669.7	19.5
6	2.8	1683.1	12.6
7	7.0	1641.0	17.7

Peak	Area	Center	Width
1	0.6	1610.0	8.8
2	2.3	1618.5	11.1
3	2.6	1626.2	10.6
4	3.5	1635.0	11.4
5	3.2	1644.7	10.5
6	2.5	1652.6	8.9
7	2.5	1660.2	8.9
8	2.3	1668.0	9.2
9	1.4	1675.4	8.2
10	1.1	1682.3	7.6
11	0.5	1689.5	6.7
12	0.1	1694.5	3.6

Peak	Area	Center	Width
1	1.7	1615.7	13.0
2	4.7	1625.8	13.4
3	4.2	1634.9	11.8
4	4.1	1642.8	11.4
5	5.5	1651.0	11.8
6	5.7	1660.4	12.8
7	5.5	1670.8	13.6
8	3.0	1682.4	12.3

Figure S8. Curve-fitted amide I region (1600–1700 cm^{-1}) with secondary structure determination of FD SPs and films.

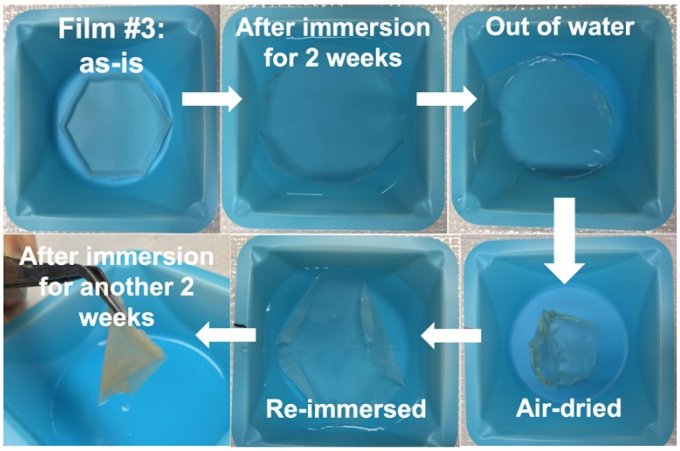


Figure S9. Film #3 was immersed in water for two weeks, air-dried and re-immersed in water for another two weeks.

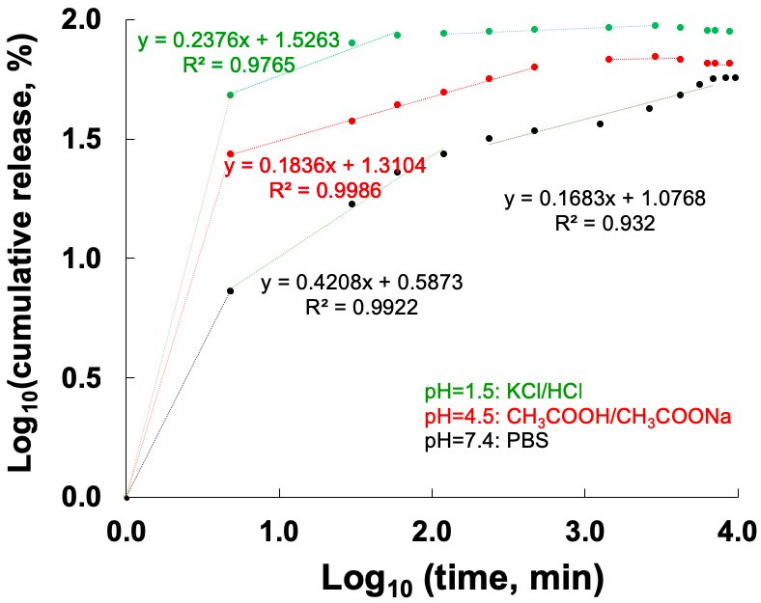


Figure S10. Korsmeyer-Peppas model fitting of MB release in three buffers.

Table S1. XRD parameters of SPs analyzed by peak fitting method

FD SP (CrI: 41.1%)	**2θ (°)**	9.0	17.1	19.4	22.3	25.2
	d_{hkl} (Å)	9.8	5.2	4.6	4.0	3.5
	Area (%)	**10.8**	**4.2**	**9.9**	**8.4**	**7.7**
#1 (CrI: 53.6 %)	**2θ (°)**	7.7	15.1	17.6	20.3	23.4
	d_{hkl} (Å)	11.5	5.9	5.0	4.4	3.8
	Area (%)	**9.6**	**7.1**	**17.5**	**12.2**	**7.2**
#2 (CrI: 49.8 %)	**2θ (°)**	8.9	17.5	19.4	23.7	28.3
	d_{hkl} (Å)	9.9	5.1	4.6	3.8	3.2
	Area (%)	**1.4**	**18.1**	**16.2**	**8.0**	**4.1**
#3 (CrI: 52.3 %)	**2θ (°)**	6.7	14.1	17.4	21.6	24.3
	d_{hkl} (Å)	13.2	6.3	5.1	4.1	3.7
	Area (%)	**1.1**	**18.1**	**17.3**	**7.5**	**4.9**
#4 (CrI: 55.6 %)	**2θ (°)**	8.6	16.3	19.1	22.6	27.1
	d_{hkl} (Å)	10.3	5.4	4.6	3.9	3.3
	Area (%)	**0.5**	**13.3**	**15.6**	**14.4**	**8.9**
#5 (CrI: 56.2 %)	**2θ (°)**	8.8	16.6	19.6	23.5	28.1
	d_{hkl} (Å)	10.0	5.3	4.5	3.8	3.2
	Area (%)	**1.5**	**12.4**	**17.5**	**14.6**	**8.1**

Table S2. Moisture content (%) of films under different conditions (N=3)

Sample	**150 °C, 0 % RH**	**21 °C, 30-35 % RH**	**21 °C, 65 % RH**	**21 °C, 85-90 % RH**
#1	-12.0 ± 1.4	0.0	4.9 ± 0.3	40.3 ± 1.1
#2	-9.0 ± 1.1	0.0	1.2 ± 0.9	53.0 ± 1.7
#3	-9.9 ± 0.9	0.0	6.8 ± 0.4	44.0 ± 1.0
#4	-8.0 ± 0.3	0.0	2.9 ± 0.8	51.4 ± 5.4
#5	-9.4 ± 0.6	0.0	4.1 ± 0.5	38.9 ± 1.3

Table S3. S_p of film #3 and 5 (N=3)

pH	**5 min**		**1 week**		**2 weeks**	
	#3	**#5**	**#3**	**#5**	**#3**	**#5**
0	1.50 ± 0.07	1.29 ± 0.03	1.49 ± 0.05	1.27 ± 0.03	1.31 ± 0.01	1.34 ± 0.01
1	2.24 ± 0.05	2.24 ± 0.04	2.28 ± 0.04	2.40 ± 0.03	2.24 ± 0.01	2.23 ± 0.03

3	1.17 ± 0.07	1.52 ± 0.01	1.27 ± 0.04	1.68 ± 0.03	1.17 ± 0.03	1.70 ± 0.01
7	1.38 ± 0.12	1.55 ± 0.09	1.57 ± 0.01	1.46 ± 0.03	1.57 ± 0.02	1.41 ± 0.04
10	1.87 ± 0.12	1.59 ± 0.01	4.13 ± 0.18	1.57 ± 0.04	4.63 ± 0.09	1.74 ± 0.01

Table S4. S_p of SP/MB films in buffers at 37 °C (N=3)

pH	5 min	30 min	1 h	17 h	48 h	120 h
7.4	1.23 ± 0.02	1.22 ± 0.03	1.16 ± 0.01	1.18 ± 0.04	1.19 ± 0.04	1.20 ± 0.04
4.5	1.18 ± 0.02	1.12 ± 0.05	1.14 ± 0.05	1.08 ± 0.00	1.08 ± 0.03	1.09 ± 0.06
1.5	1.38 ± 0.03	1.47 ± 0.04	1.47 ± 0.08	1.45 ± 0.04	1.49 ± 0.04	1.47 ± 0.05