Supplementary Information for

Mechanical stability of phase-segregated multicomponent lipid bilayers enhanced by PS-*b*-PEO diblock copolymers

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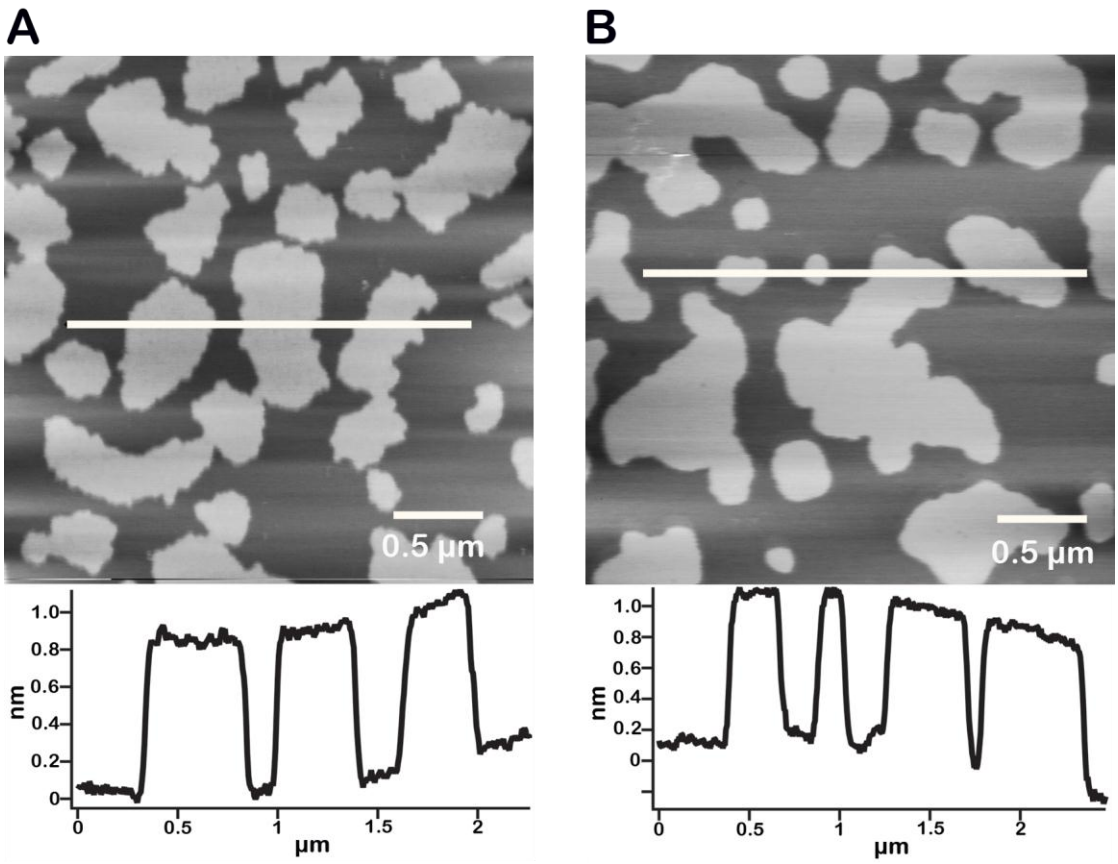

Figure S1 AFM height images and line profiles of DOPC/ESM/Chol with 0.05 mol% of PS(3.6)-*b*-PEO(16.6) (A) and PS(3.8)-*b*-PEO(6.5) (B). The strips indicate the position of the line profiles. The height scale is 0–4 nm in each AFM image.

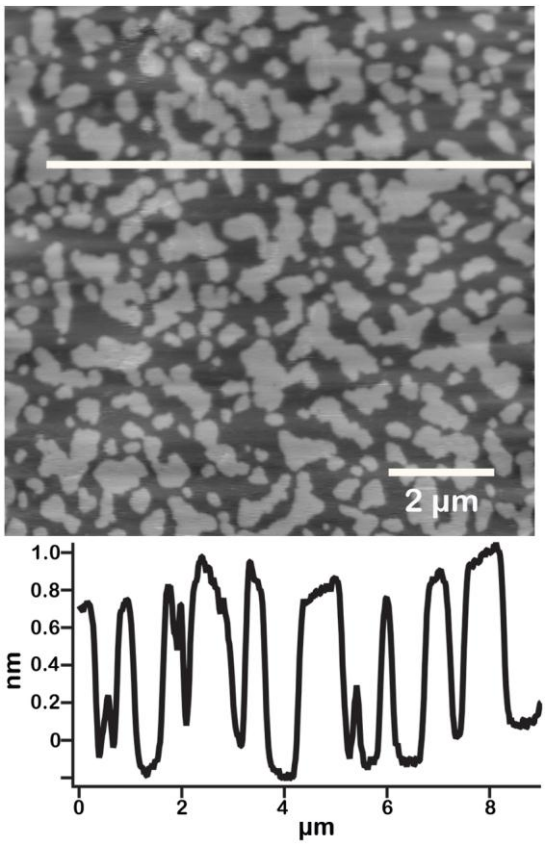

Figure S2 AFM height image and line profile of pure DOPC/ESM/Chol with 0.05 mol% PS(19)-*b*-PEO(6.4). The strip indicates the position of the line profiles. The height scale is 0–4 nm in the AFM image.

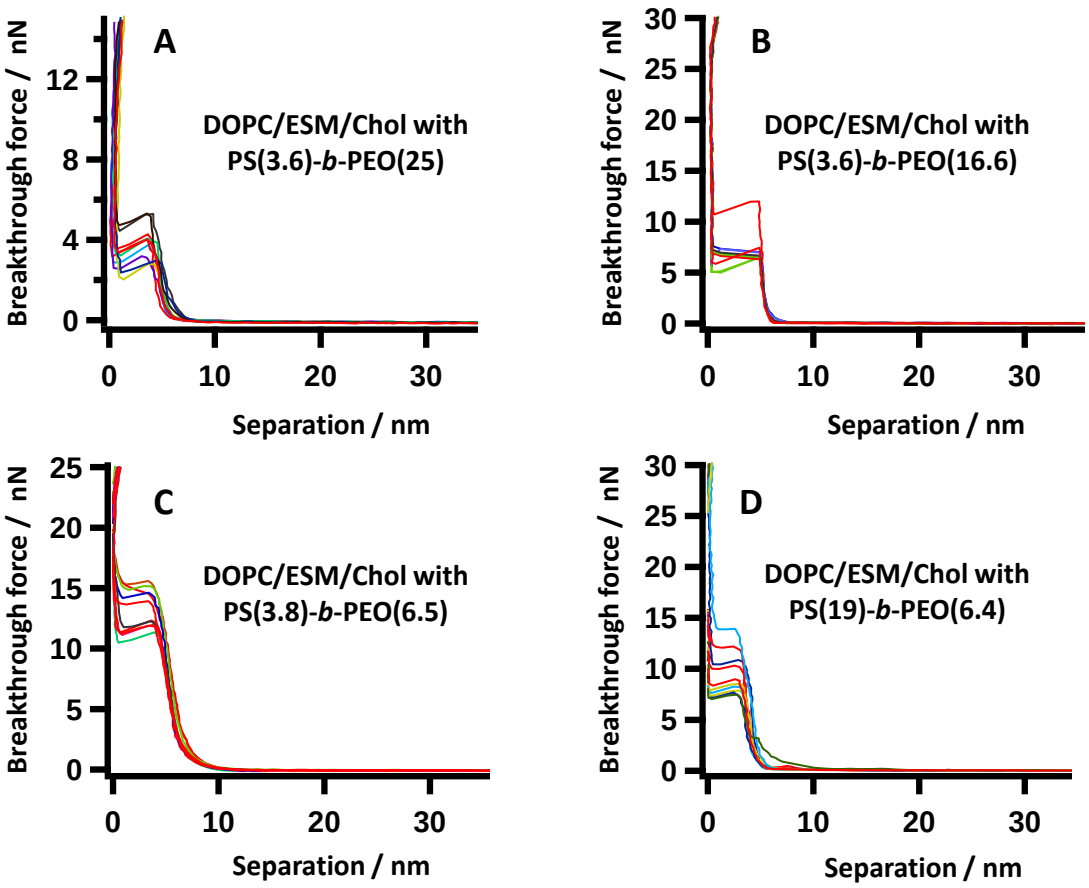

Figure S3 Ten overlapping force-distance curves of pure DOPC/ESM/Chol in the presence of 0.05 mol% PS(3.6)-*b*-PEO(25) (A), PS(3.6)-*b*-PEO(16.6) (B), PS(3.8)-*b*-PEO(6.5) (C), and PS(19)-*b*-PEO(6.4) (D). The block copolymer is expected to insert more densely when the PEO block is short — because of lower aqueous partition coefficient. When the sample is rinsed, and fresh water is on the outer surface, PS-*b*-PEO is partially dissolved out of phospholipid, but not from the substrate side (Scheme 2). In contact mode, the ordered phase appeared higher, but in compression (breakthrough) the disordered phase is thicker (see Table S1). As expected, this effect is more pronounced in the presence of PS(3.8)-*b*-PEO(6.5), where the PEO chain is shortest (Fig. S3C). The higher thickness of the bilayer with added PS(3.6)-*b*-PEO(6.5) is due to the inserted copolymer giving rise to the long-range repulsion in the force curves. This observation is

consistent with what has been observed for tethered lipid bilayer where the increased in the bilayer thickness is attributed to the presence of the polymer spacer.[1] Hence, the pattern of heights suggests that the polymer inserts preferentially into the disordered phase, and (after rinsing with water) is found mainly between the bilayer and the substrate.

1. X. Wang, M. M. Shindel, S.-W. Wang and R. Ragan, *Langmuir*, 2010, **26**, 18239.

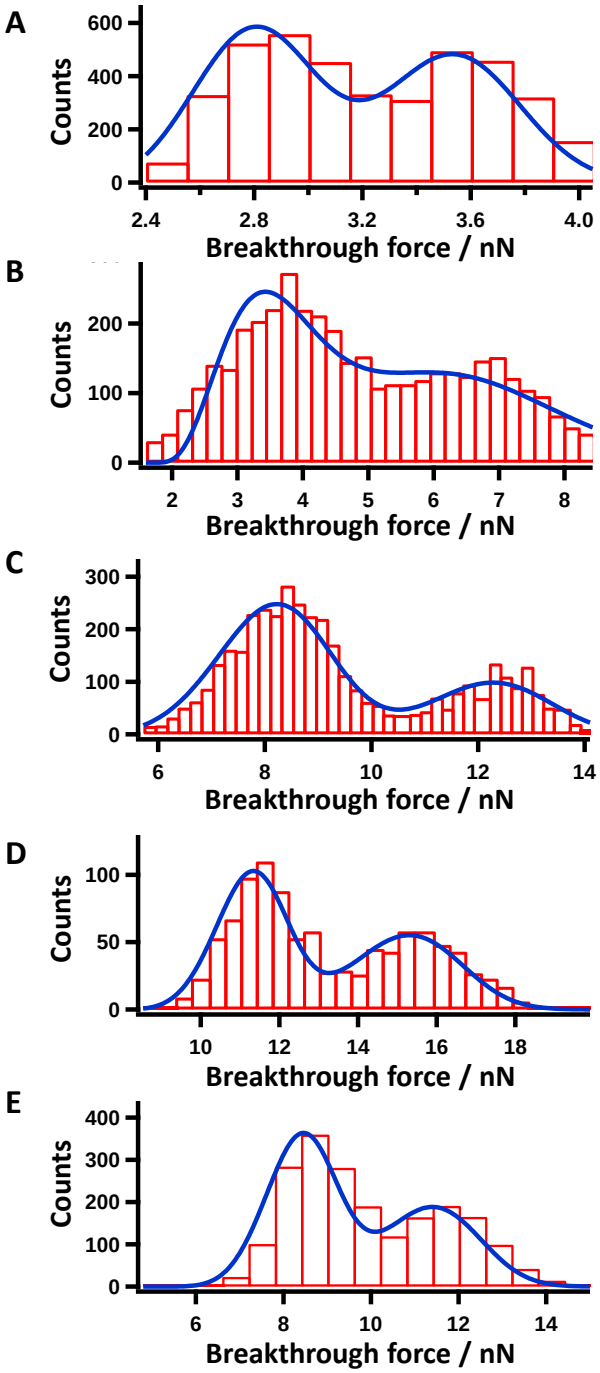

Figure S4 The spreading pressure and line tension fits to the breakthrough force histograms of pure DOPC/ESM/Chol (A), and in the presence of 0.05 mol% PS(3.6)-*b*-PEO(25) (B), PS(3.6)-*b*-PEO(16.6) (C), PS(3.8)-*b*-PEO(6.5) (D), and PS(19)-*b*-PEO(6.4) (E).

Table S1 Bilayer thickness of pure DOPC/ESM/Chol and with added asymmetric diblock copolymers, PS-*b*-PEO.

DOPC/ESM/Chol w/ and w/o diblock copolymers	**Bilayer thickness (nm)**	
	L_o	**L_d**
Pure lipid	6.10 ± 0.26	6.14 ± 0.24
with PS(3.6)-*b*-PEO(25)	6.47 ± 0.54	6.53 ± 0.52
with PS(3.6)-*b*-PEO(16.6)	5.92 ± 0.23	6.48 ± 0.28
with PS(3.8)-*b*-PEO(6.5)	8.94 ± 0.32	9.8 ± 0.31
with PS(19)-*b*-PEO(6.4)	5.87 ± 0.40	6.26 ± 0.48