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## **Electronic Supplementary Information (ESI)**

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## Self-Assembled pH-Responsive Films Prepared from Mussel Anchoring Threads

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Table S1: Amino acid composition of the byssus thread and of the BPH extract (residues/1000). Results are expressed as average  $\pm$  standard deviation for N=3.

	Byssus	BPH extract
Asx	83 ± 1	107 ± 5
Glx	$64.0 \pm 0.8$	$89 \pm 4$
OH-Pro	37 ± 3	21 ± 5
Ser	63 ± 1	61 ± 2
Gly	$230 \pm 10$	149 ± 17
His	$27 \pm 3$	18 ± 2
Arg	$58 \pm 3$	61 ± 2
Thr	37 ± 2	42 ± 1
Ala	$98.5 \pm 0.6$	$82 \pm 6$
Pro	66 ± 2	$69 \pm 6$
L-DOPA	4 ± 2	$0.05 \pm 0.02$
Tyr	26 ± 2	$32 \pm 3$
Val	47 ± 1	$66 \pm 6$
Met	$10.0 \pm 0.5$	15 ± 1
Cys	$3.5 \pm 0.7$	$1.6 \pm 0.5$
lle	$29.9 \pm 0.1$	48 ± 5
Leu	47 ± 2	$59 \pm 5$
Phe	25 ± 1	$32 \pm 4$
Lys	44.7 ± 0.1	$46 \pm 4$



Figure S1: Representative tensile mechanical testing curves of the BPH films treated at various pH.



Figure S2: FTIR spectra of freeze-dried BPH solutions with different concentrations (a, d: 10 mg/mL, b, e: 20 mg/mL, and c, f: 30 mg/mL). Solutions a-c were quickly frozen, while solutions d-f were aged at room temperature for one week before being freeze-dried.



Figure S3: FTIR spectra of the three macroscopic sections of a byssus single fiber (distal part, proximal part and plaque), the BPH, and a film as prepared.