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Electronic Supplementary Information for the Manuscript Entitled

Tunable hydrogel morphology via self-assembly of amphiphilic pentablock copolypeptides

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Submitted to Soft Matter

Electronic Supplementary Information

Table S1 Molecular weights and segment lengths of select $K_aL_bK_cL_dK_e$ pentablock copolypeptides. DP = degree of polymerization. x = determined by GPC/LS. y = determined by NMR and GPC/LS data.

Sample	1st segment (K_a)[x]		Found lengths (DP) of individual segments[y]				
Predicted Composition	M_w/M_n	M_n (kDa)	K_a	L_b	K_c	L_d	K_e
$K_{60}L_{20}K_{10}L_{20}K_{60}$	1.23	15.9	60	19	11	23	59
$K_{60}L_{20}K_{40}L_{20}K_{60}$	1.27	16.1	61	18	41	22	71
$K_{60}L_{20}K_{60}L_{20}K_{60}$	1.19	16.6	63	20	62	21	62
$K_{60}L_{20}K_{100}L_{20}K_{60}$	1.21	15.3	59	17	101	24	64
$K_{60}L_{20}K_{200}L_{20}K_{60}$	1.20	17.2	66	19	216	22	53

Table S2 Minimum Gelation concentrations of $K_{60}L_{20}K_xL_{20}K_{60}$ pentablock copolypeptides.

Sample	Gelation Concentration (mM)
$K_{60}L_{20}K_{10}L_{20}K_{60}$	N/A
$K_{60}L_{20}K_{60}L_{20}K_{60}$	0.18
$K_{60}L_{20}K_{100}L_{20}K_{60}$	0.15
$K_{60}L_{20}K_{200}L_{20}K_{60}$	0.03

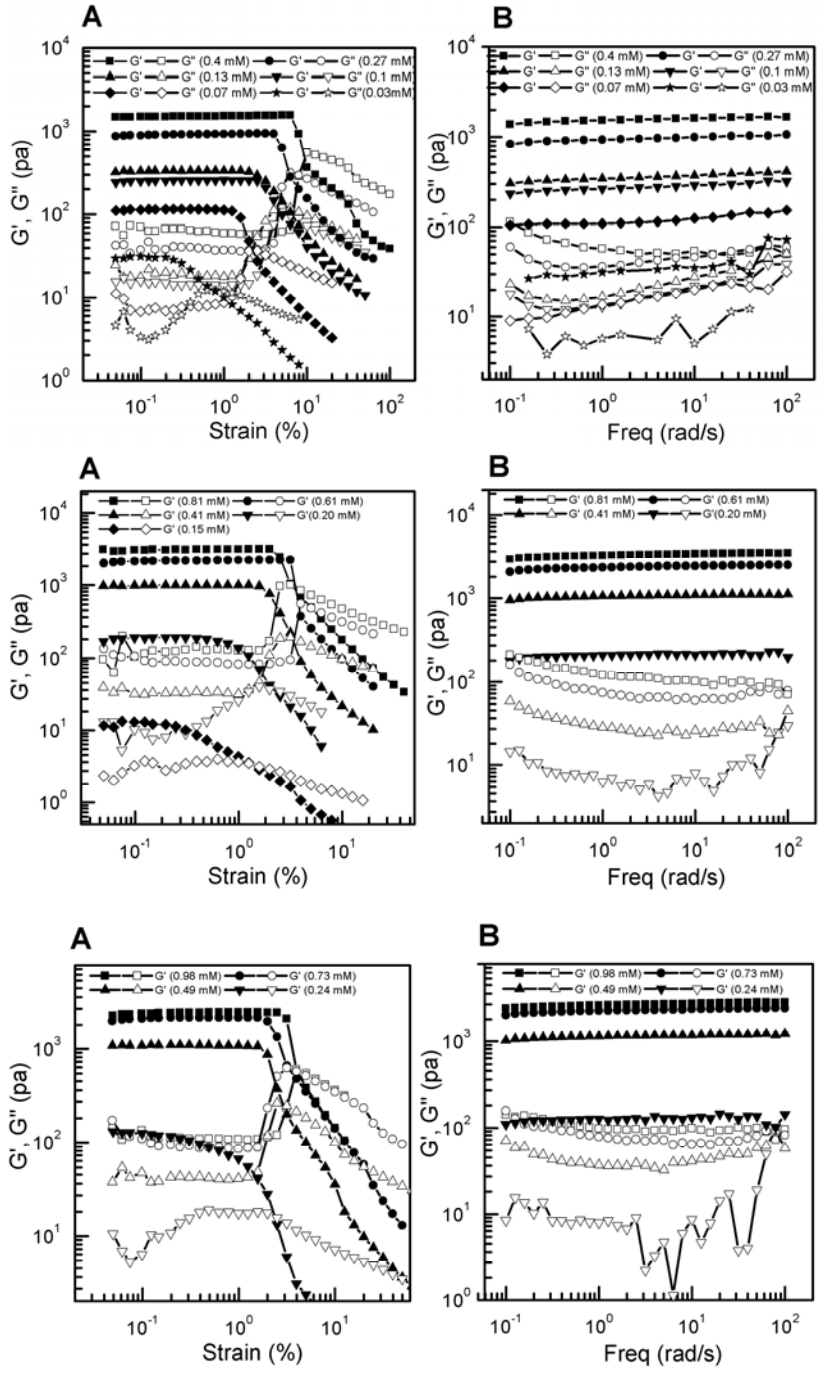

Figure S1 Storage modulus (G′) and loss modulus (G″) of (**A,B**) $K_{60}L_{20}K_{200}L_{20}K_{60}$, (**C,D**) $K_{60}L_{20}K_{100}L_{20}K_{60}$, and (**E,F**) $K_{60}L_{20}K_{60}L_{20}K_{60}$ pentablock hydrogels at different concentrations measured as functions of (**A,C,E**) strain amplitude (γ_0) at an angular frequency of 1 rad/s, and (**B,D,F**) angular frequency (ω) in the linear regime with γ=0.1.

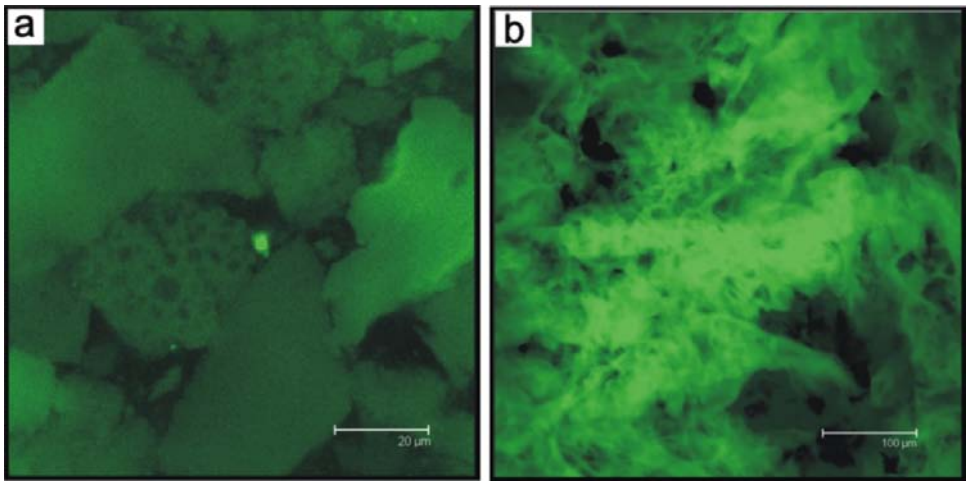

Figure S2 Laser scanning confocal microscopy images of $K_{60}L_{20}K_{200}L_{20}K_{60}$ pentablock hydrogels at 0.13 mM showing microporous structure. A small fraction (3 mol%) of the polyK amine group side-chains were fluorescently labeled with fluorescein isothiocyante. Scale bars = 20 µm.

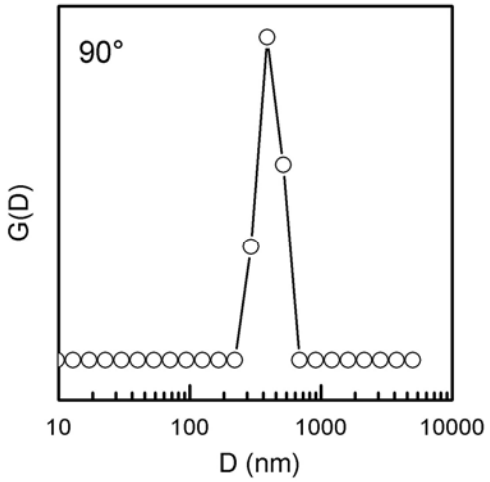

Figure S3 Size distribution (diameter) plot of $K_{60}L_{20}K_{10}L_{20}K_{60}$ assemblies (0.5 wt% in water) from DLS measurements.

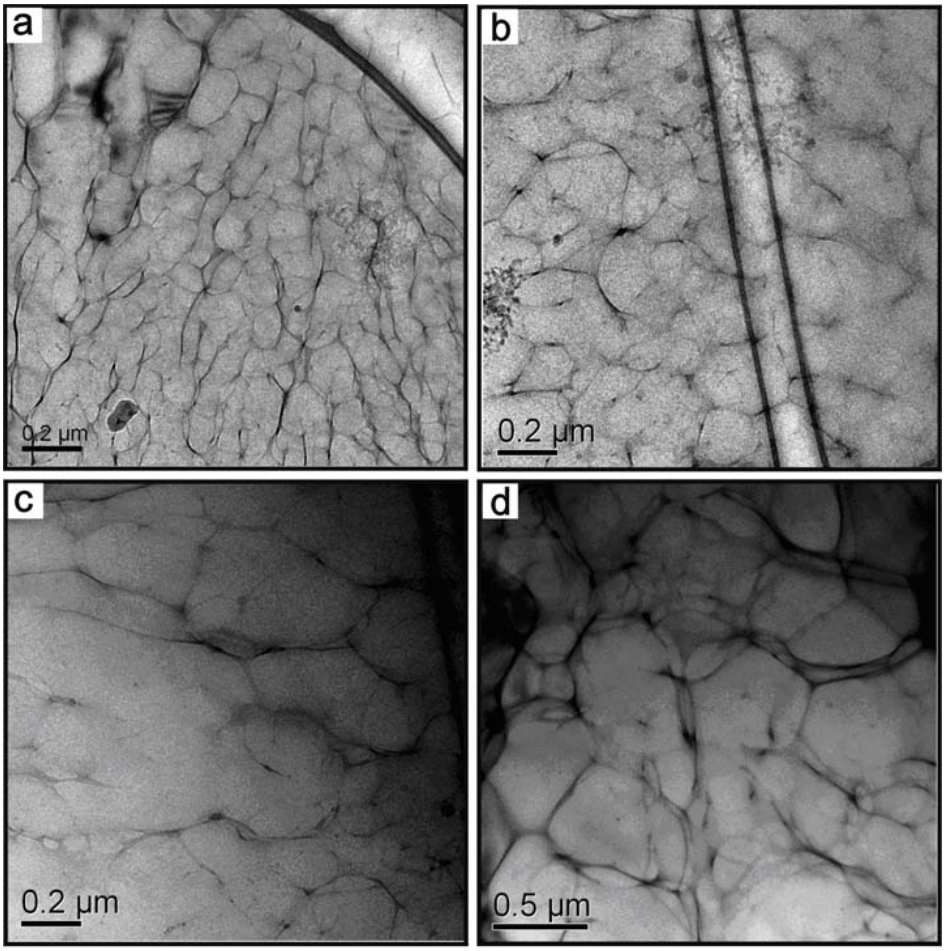

Figure S4 Additional CryoTEM images of $K_{60}L_{20}K_{200}L_{20}K_{60}$ hydrogels (0.25 wt%).

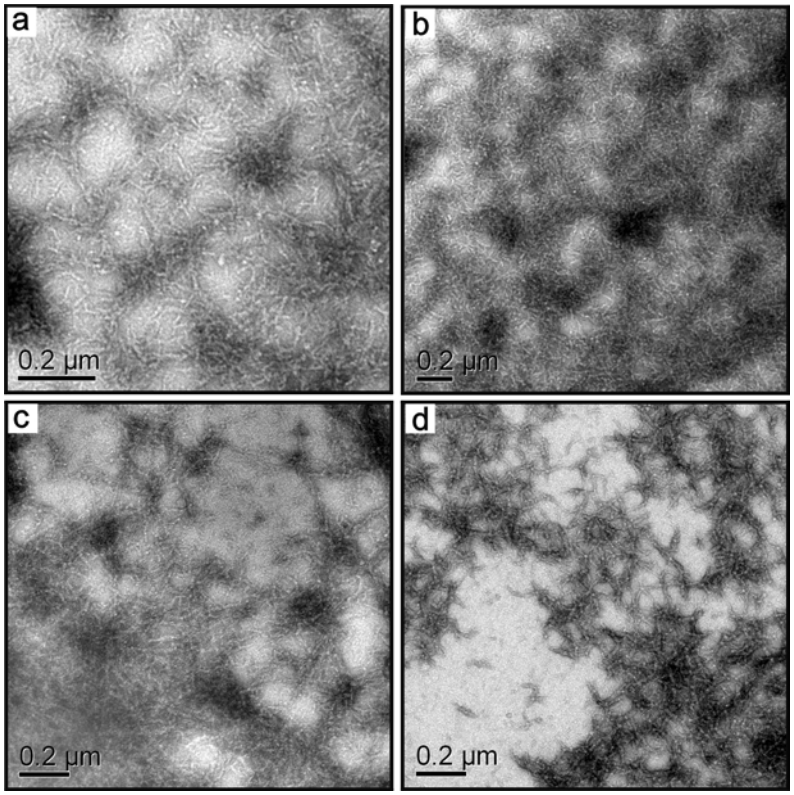

Figure S5 TEM images of $K_{60}L_{20}K_{200}L_{20}K_{60}$ hydrogels. The samples were negative stained with 1.5 wt% aqueous uranyl acetate.

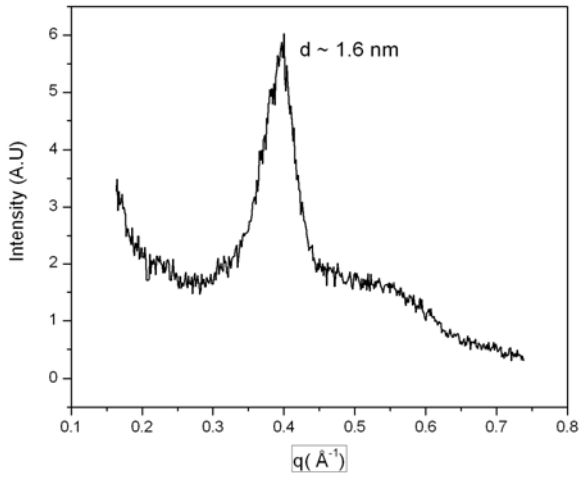

Figure S6 SAXS pattern of a freeze-dried $K_{60}L_{20}K_{200}L_{20}K_{60}$ hydrogel sample.