Electronic Supplementary Information (ESI)

Dynamic transformations of self-assembled polymeric microspheres

induced by AC voltage and shear flow

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Video captions in ESI

- **Video S1** Fusion behaviors of PEG-SB 22-74 MLMs in an open chamber under AC voltage (10 V_{pp} and 10 milli-Hz) in water. The video plays at 10 times speed.
- Video S2 Fusion and bursting behaviors of PEG-SB 22-74 MLMs in a closed chamber under AC voltage (10 V_{pp} and 100 milli-Hz) in water. The video plays at practical speed.
- **Video S3** Transformation behaviors of PEG-SB74 MLMs from microsphere to string-like structure under shear flow, and from string-like structure to MLMs transformation by removal of shear flow. Shear rate was 5,000 s⁻¹ and performed at 25°C. The shear flow started at 2 s and repeated flow and the stop was applied from 2 min 30 s. The video plays at double speed.
- Video S4 Transformation behaviors of PEG-SB74 MLMs from microsphere to string-like structure under shear flow, and from string-like structure to MLMs transformation by removal of shear flow. Shear rate was 1,500 s⁻¹ and performed at 25°C. The video plays at 5 times speed.

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Figure S1 Chemical structure of PEG-b-PDMAPS

Polymer	Polymerization degree*		Cloud point (°C)**	Size (µm)***
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22-37	22	37	23.2 ± 1.4	7.2 ± 1.2
22-56	22	56	39.6 ± 1.6	4.8 ± 0.5
22-74	22	74	48.5 ± 1.7	1.5 ± 0.2
22-87	22	87	54.1 ± 2.3	2.6 ± 0.7
110-73	110	73	42.0 ± 1.7	1.2 ± 0.1

Table S1 Characterization of PEG-b-PDMAPS

* Estimated by ¹H-NMR in D₂O containing 1M NaCI
** Determined by UV-Vis measurements at 50 % turbidity

*** Evaluated by microscopic observation (in MilliQ water)

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Figure S2 Schematic illustration of direct microscopic observation system under AC voltage



Figure S3 Wave form effect on size-growth rate of 22-74 MLMs. SIN: sine wave, TRI: triangle wave, SQU: square wave. The frequency was 100 mHz

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Figure S4 Schematic illustration of direct microscopic observation system for controllable shear flow