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Supplementary Information

Experimental Section

Materials: The ELP gene $[(VPGVG)_{14}(VPGKG)]_8[VPGVG]_{40}$ was oligomerized using a recursive directional ligation procedure and transformed into *E. coli* strain $BLR(DE)_3$ (Novagen) for expression. The ELP was purified using an inverse transition cycling method and collected by freeze-drying. Glutaraldehyde, cottonseed oil, Rhodamin B isothiocyanate and FITC-dextran (70 kDa) were purchased from Sigma (Germany). Petroleum ether, acetone, and ethanol were acquired from Samchun Chemicals (Pyeongtaek, Korea).

Thermal Characterization of the ELP: To characterize the T_t of ELP, the optical density (OD_{350nm}) of an ELP solution (PBS, 30 mg 225μL^{-1}) was monitored relative to temperature using a Cary 100 Bio UV-visible spectrophotometer (Varian) equipped with a temperature controller. To characterize the phase separation properties of the ELP, the particles' hydrodynamic diameters were determined using dynamic light scattering (DLS) (Zetasizer Nano, Malvern) as related to temperature.

Observation of the ELP Phase Separation Inside a Emulsion Droplet: Firstly, ELPs were conjugated with Rhodamin B isothiocyanate (ELP-Rho) and purified with an inverse transition cycling method and subsequent freeze-drying. Next, 30 mg ELP-Rho was dissolved in 225 μL PBS at 4°C. The ELP-Rho solution was dispersed in an oil phase composed of 25 mL petroleum ether and 25 mL cotton seed oil at 4°C. The emulsion containing ELP-Rho was observed using fluorescence microscopy (Olympus, Japan).

Preparation of the Crosslinked Microstructures: ELP microstructures were prepared by water-in-oil emulsion method. Firstly, 30 mg freeze-dried ELPs were dissolved in 225 μL PBS at 4 °C. Subsequently, this solution was added dropwise to an oil phase composed of 25 mL petroleum ether and 25 mL cottonseed oil at 4°C. After magnetically stirring the emulsion for 1 h at various temperatures (4, 30 and 45°C), 50 μL glutaraldehyde (25% aqueous solution) was added to the emulsion to crosslink microstructures. The crosslinked microstructures were rinsed with petroleum ether, acetone and ethanol (in the listed order) to remove any cottonseed oil or residual crosslinkers; the material was collected by centrifugation and redispersed in ethanol for storage. The steps were finished in a thermostatic chamber.

Characterization of the ELP Microstructures: CLSM images of samples in deionized water (DW) solution were acquired using LSM510 (Carl Zeiss, Germany). All samples were excited at 488 nm, and the fluorescent images were obtained at 510-540 nm (green). The epi-fluorescent images were also acquired using fluorescence microscopy (Olympus, Japan). High-resolution morphological images were obtained with a SUPRA 55VP field-emission scanning electron microscope (FE-SEM Carl Zeiss, Germany). To observe the shape and surface morphology, the samples were immersed in DW at various temperatures until an equilibrium swelling state was reached; the particles were immediately dipped into liquid nitrogen and freeze-dried. Freeze-dried ELP samples were coated with sputtered gold before observation.

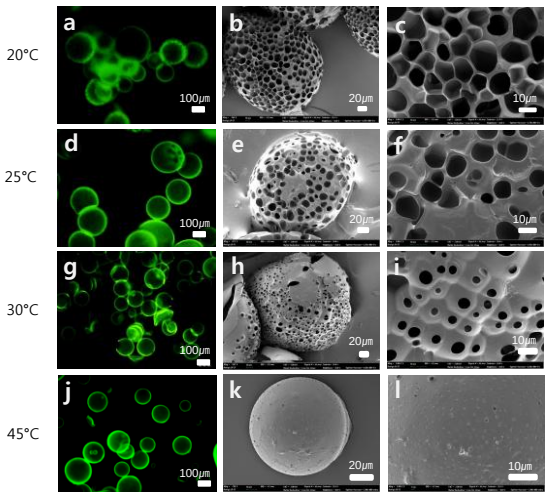

Fig. S1. Morphological variations between the ELP microcapsules prepared at different crosslinking temperatures. Microcapsules were prepared at (a-c) 20°C, (d-f) 25°C, (g-i) 30°C and (j-l) 45°C. (a, d, g, i) Confocal images, (b, e, h, k) FE-SEM images and (c, f, I, l) enlarged FE-SEM images of microcapsules.

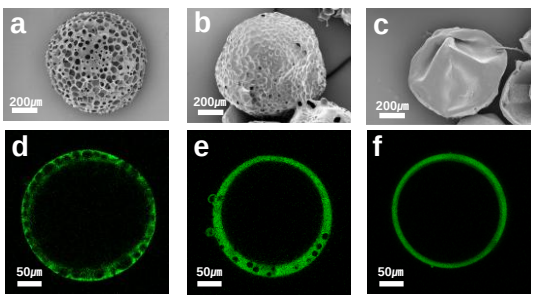

Fig. S2. Morphological variations of the ELP microcapules prepared at different maturation time. FE-SEM images of microcapsules prepared after the maturation for (a) 15 min, (b) 30 min and (c) 60 min. (d-f) Confocal images of a-c. Microcapsules were prepared at 45°C.