Supporting Information

Stabilizing Coacervate by Microfluidic Engulfment Induced by Controlled Interfacial Energy

Kyoung Duck Seo,[†,⊥,#] Sara Shin,[‡,#] Hee Young Yoo,[§] Jiafu Cao,[‖] Suyoung Lee,[§]

Jin-Wook Yoo,[‖] Dong Sung Kim[*,†,] and Dong Soo Hwang[*,‡,§]

† Department of Mechanical Engineering, Pohang University of Science and Technology (POSTECH), Pohang, 37673, Republic of Korea

‡ Division of Environmental Science and Engineering, Pohang University of Science and Technology (POSTECH), Pohang, 37673, Republic of Korea

§ Division of Integrative Biosciences and Biotechnology, Pohang University of Science and Technology (POSTECH), Pohang, 37673, Republic of Korea

‖ College of Pharmacy, Pusan National University, Busan, 46241, Republic of Korea

# K.D.S. and S.S. contributed equally to this work.

Corresponding Authors

* E-mail: smkds@postech.ac.kr (D.S.K.).

* E-amil: dshwang@postech.ac.kr (D.S.H.).

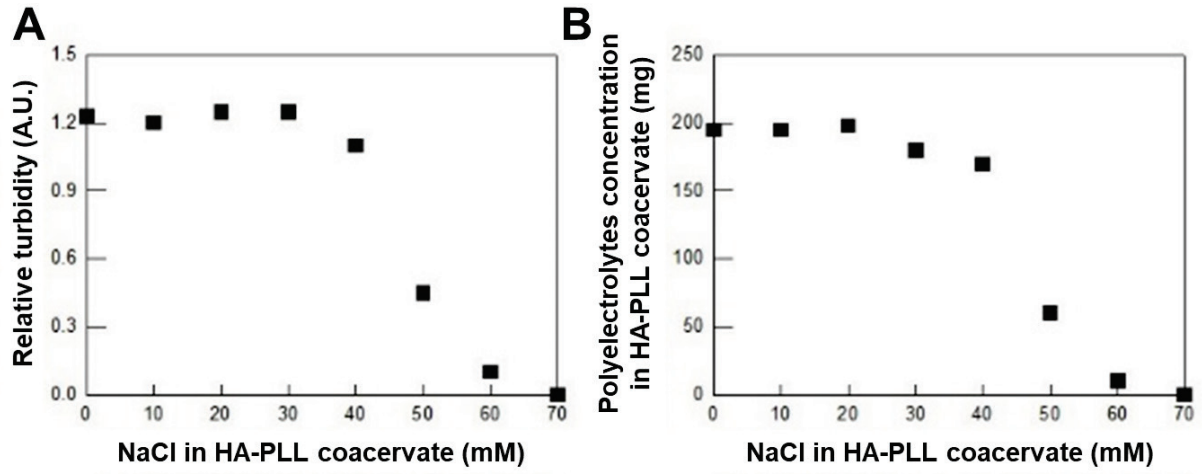

Figure S1. (A) Turbidity (at 600 nm) of HA-PLL coacervate (0.1 M, pH 5.0 sodium acetate with the addition of sodium chloride (mM) and (B) Polyelectrolyte concentration in the coacervate dense phase as a function of additional NaCl in 0.1 M sodium acetate (pH 5.0).

The HA-PLL coacervate is unstable against changes in salt concentrations. **Figure S1** shows the instability of HA-PLL coacervate against changes in salt concentration.

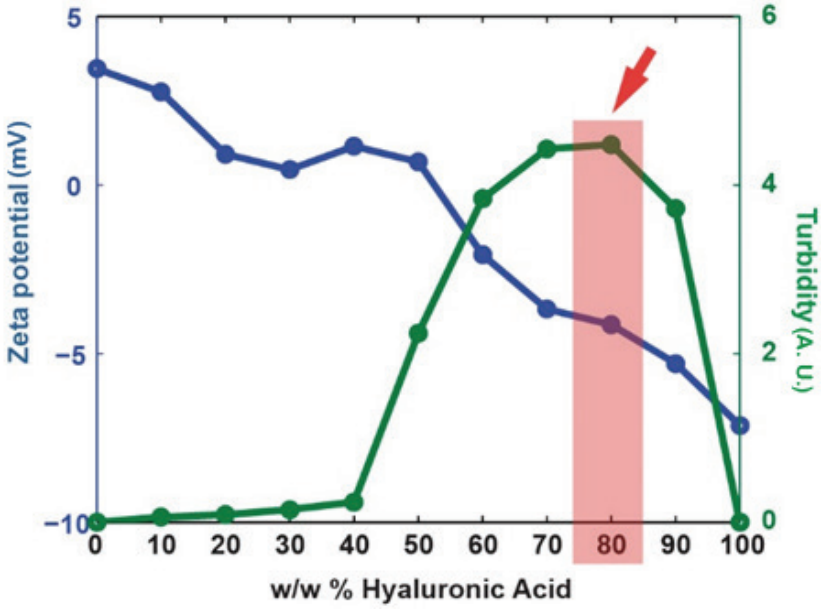

Figure S2. Left axis (blue): Zeta potential measurement for the coacervate of HA and PLL. Each point represents the average of duplicate measurements. Right axis (green): Relative turbidity measurement for the coacervate of HA and PLL. Each point represents the average of triplicate measurements.

The composition of hyaluronic acid (HA) required to form coacervate was chosen based on the values of zeta potential and turbidity. The preferred zeta potential was between ± 5 mV. At 80% of HA, the turbidity was at its maximum, thus the ratio of 5:1 for HA:PLL (w/w)% was chosen as the coacervate forming ratio.

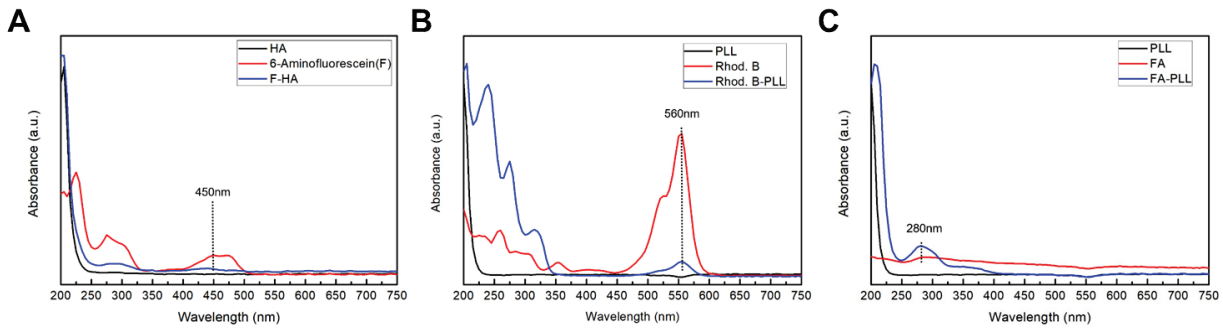

Figure S3. UV/vis spectra of HA and PLL before and after chemical conjugation. (A) 6-aminofluorescein-HA (F-HA) (B) Rhodamine B-PLL (Rhod. B-PLL), and (C) Folic acid-PLL (FA-PLL).

λ_{max} for 6-aminofluorescein (fluorescein; F), Rhodamine B (Rhod. B), and folic acid (FA) are ~440 nm, and ~560 nm, ~260 nm, respectively. The UV/vis absorbance spectra of HA and PLL before and after chemical conjugations could support the conjugation of F, Rhod. B, or FA on HA and PLL. F-HA, Rhod. B-PLL, FA-PLL have a λ_{max} at 450 nm, 560 nm, and 280 nm, respectively, after the conjugation.

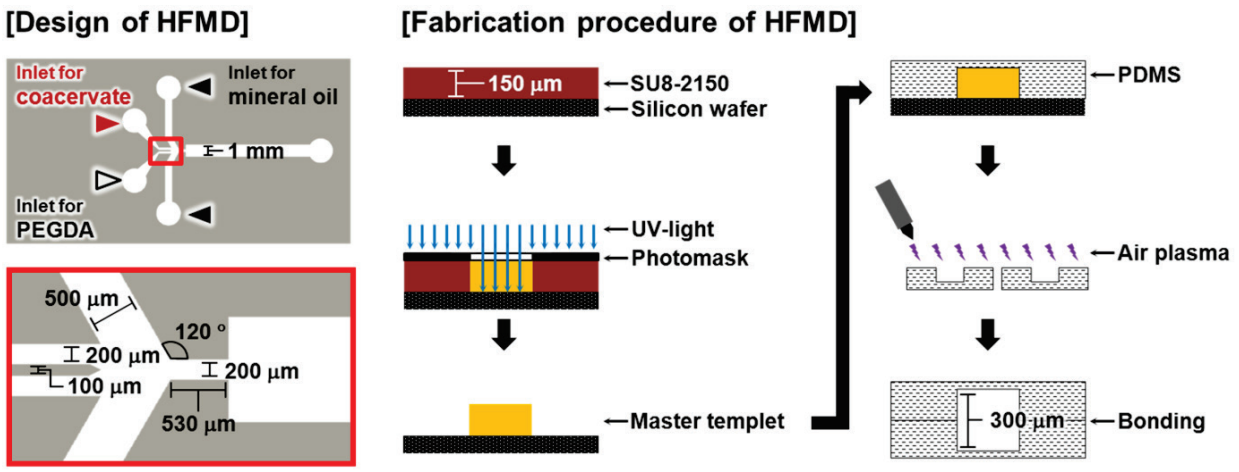

Figure S4. Design parameters and fabrication procedure for HFMD.

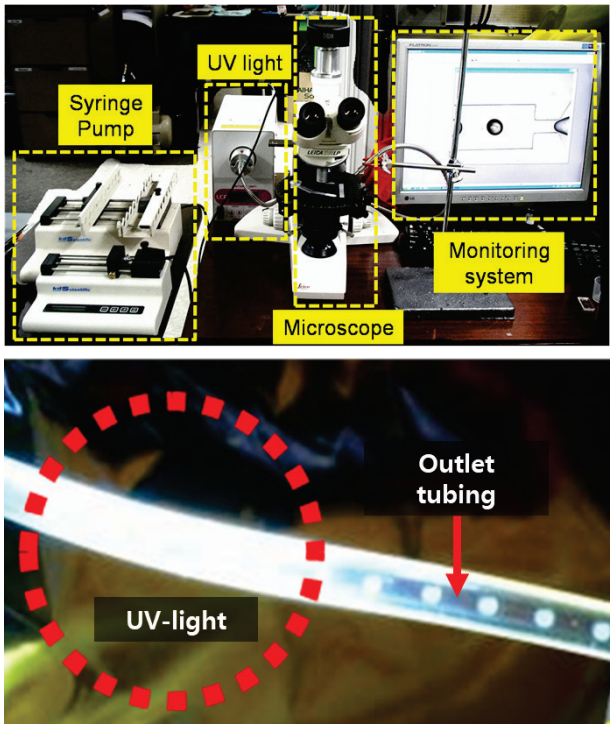

Figure S5. Actual picture of experiment setup composed of syringe pump, UV light, microscope, and monitoring system. UV polymerization was done at outlet tubing.

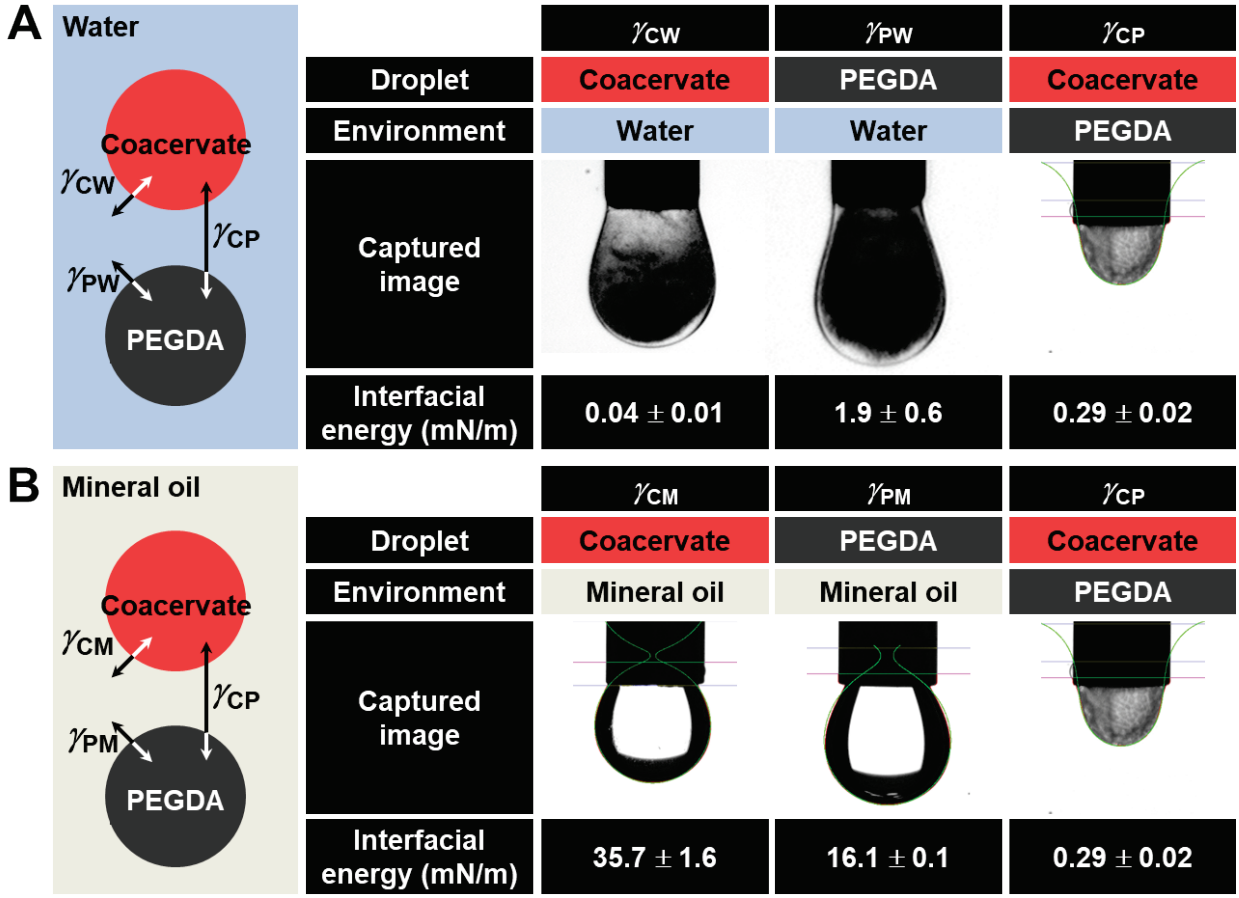

Figure S6. Interfacial energy (mN/m) of HA-PLL coacervate and PEGDA measured using pendant drop method in (A) water and (B) mineral oil with the surfactant as the medium.

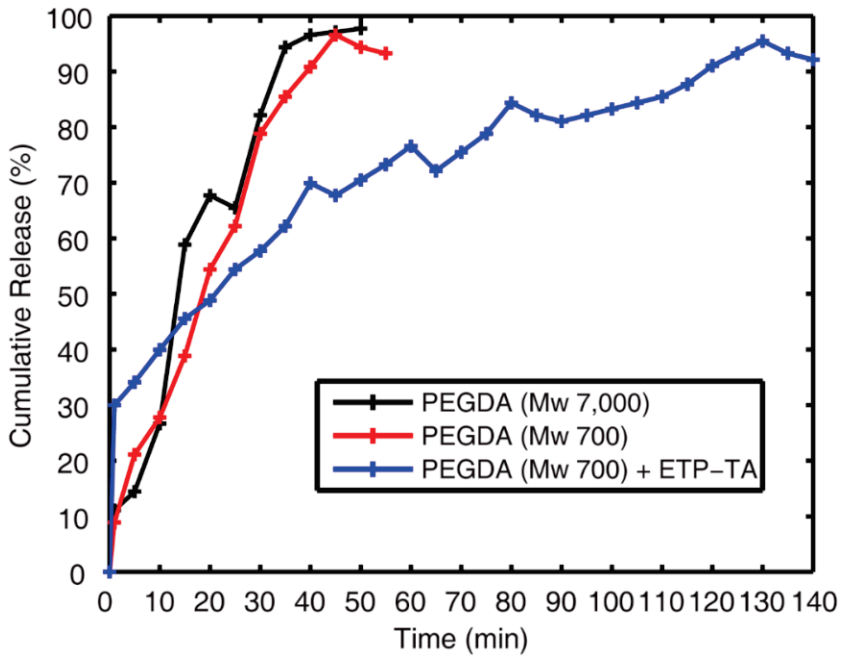

Figure S7. Release profile of the HA-PLL coacervate engulfed by PEGDA (M_w 700) in black, by PEGDA (M_w 7,000) in red, and PEGDA (M_w 700) with 1 (w/w) % ETP-TA in blue.

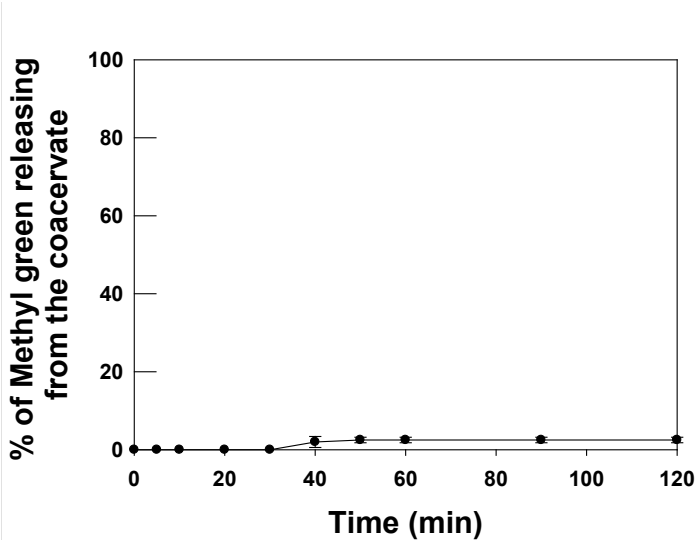

Figure S8. Dye (methyl green) release profile from the coacervate of HA and PLL in water.