

Supporting Information for

**Quantitative Turbidimetric Characterizations of Stabilized Complex Coacervate Dispersions**

Advait Holkar,<sup>a</sup> Shang Gao,<sup>a</sup> Kathleen Villaseñor,<sup>a</sup> Michael Lake<sup>b</sup> and Samanvaya Srivastava<sup>a,b,c,d,\*</sup>

<sup>a</sup> *Chemical and Biomolecular Engineering, University of California, Los Angeles, Los Angeles, CA 90095, USA.*

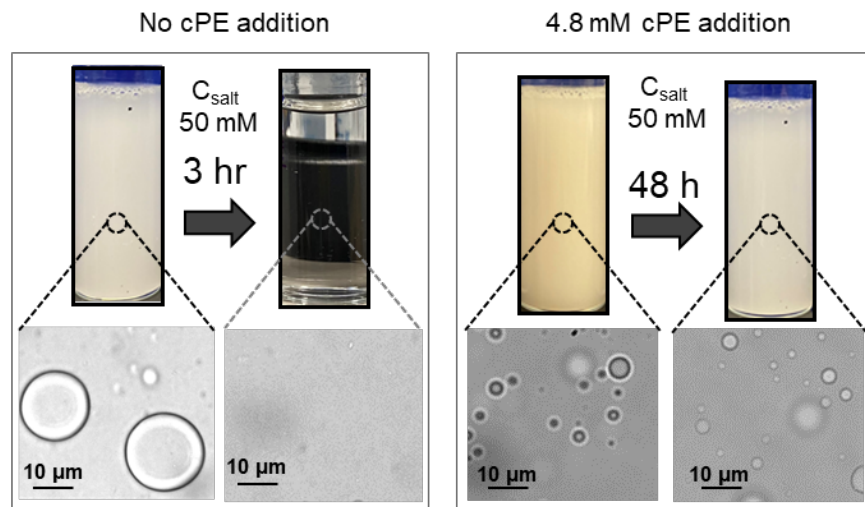
<sup>b</sup> *NSF BioPACIFIC MIP, University of California, Los Angeles, Los Angeles, CA 90095, USA.*

<sup>c</sup> *California NanoSystems Institute, University of California, Los Angeles, Los Angeles, CA 90095, USA.*

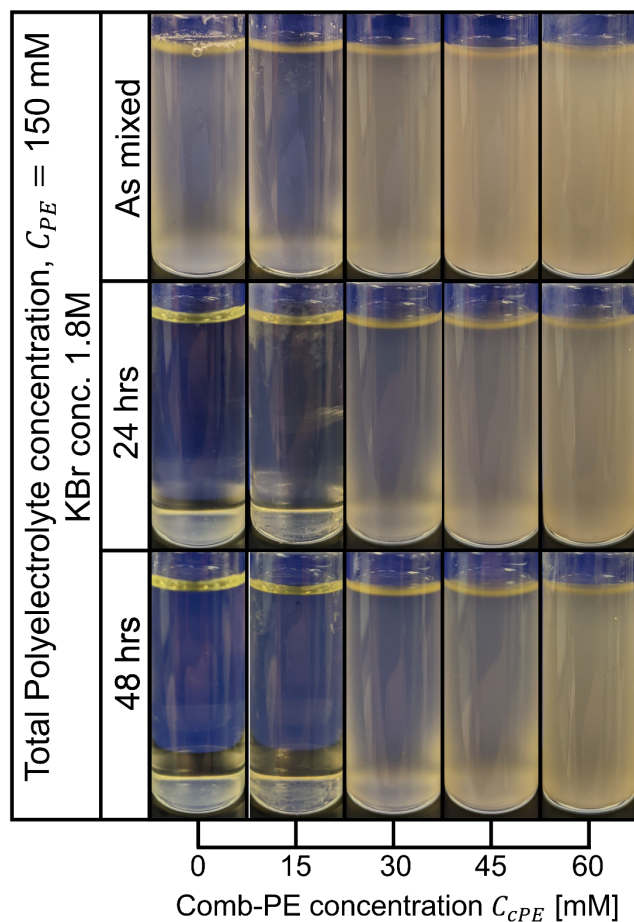
<sup>d</sup> *Institute for Carbon Management, University of California, Los Angeles, Los Angeles, CA 90095, USA.*

\* Correspondence: [samsri@ucla.edu](mailto:samsri@ucla.edu)

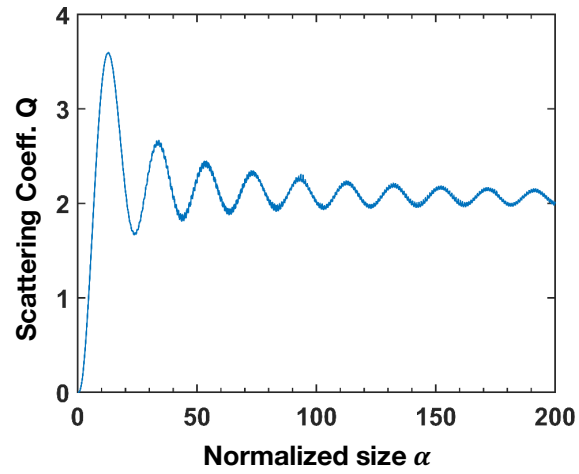
**Supplementary Figures S1 – S11**



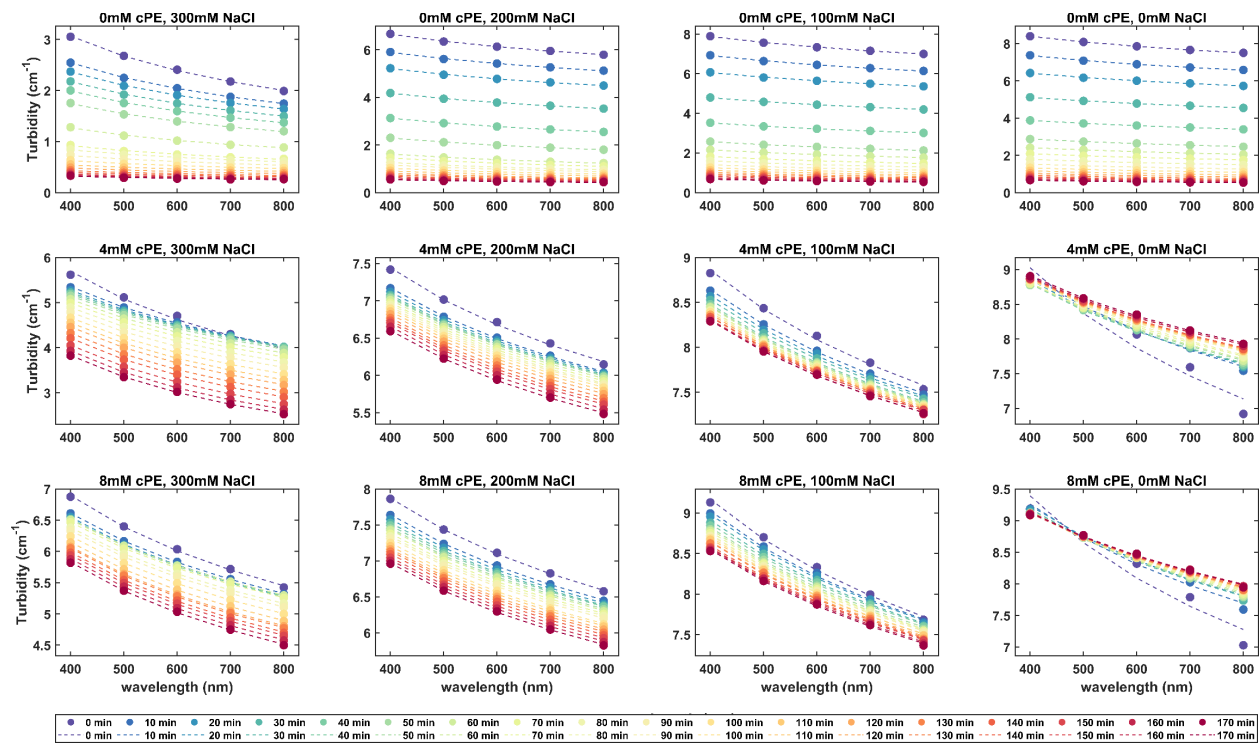
**Figure S1: Comb polyelectrolytes stabilize complex coacervate dispersions even in the presence of salt.** Complex coacervate microdroplets coalesce and phase separate within 3 hours at a low salt concentration of 50 mM while they remain dispersed with cPE addition for up to 48 hours.



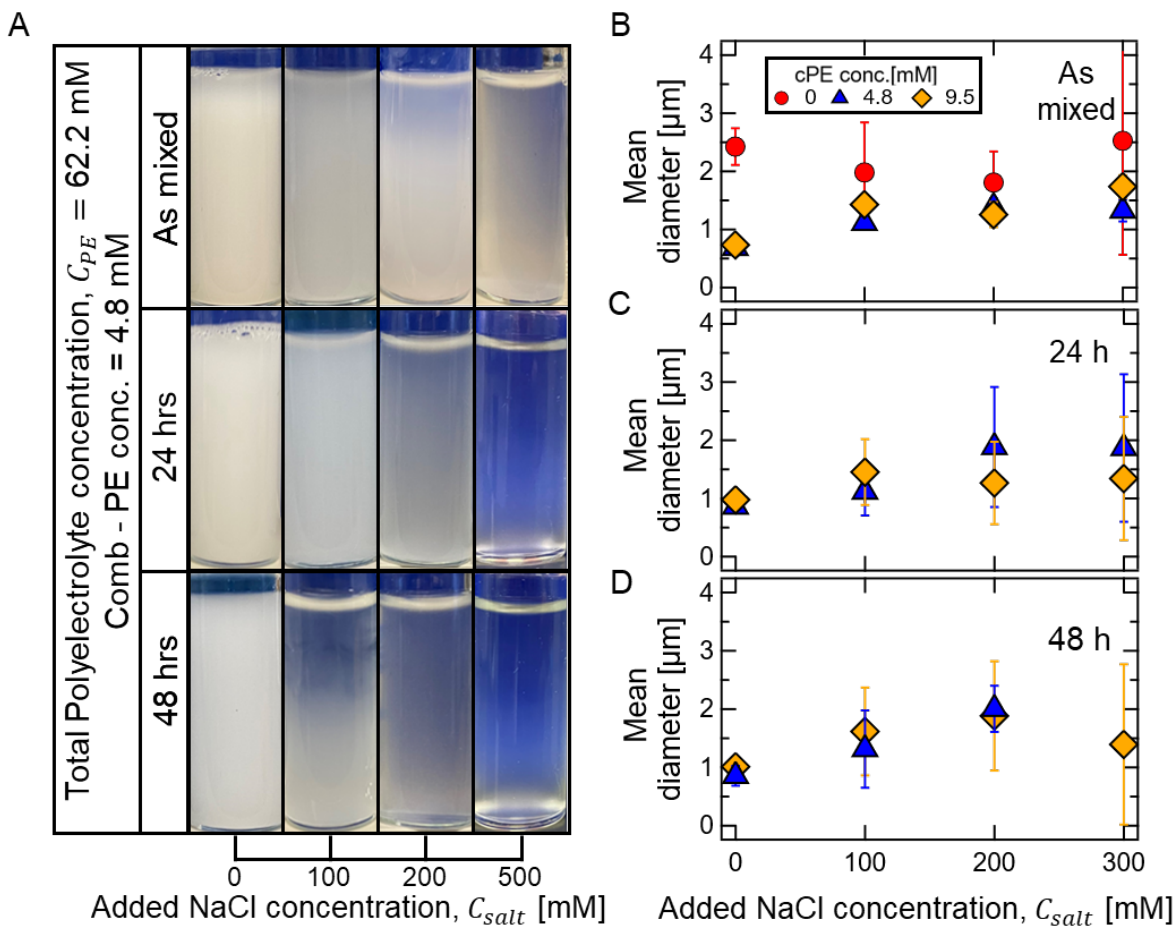
**Figure S2: Strong polyelectrolyte pair coacervate microdroplets formed at high concentrations of KBr can be stabilized by comb polyelectrolytes.** Electrostatic interactions between strongly and oppositely charged polyelectrolytes PDADMA and PSSNa were screened by introducing salt KBr (concentration of 1.8M); this plasticized the complexes formed to yield liquid-like coacervates instead of solid-like precipitates. Dispersions of coacervates were prepared and stabilized by the addition of increasing concentration of anionic comb-PE (cPE) from 0-60 mM charge concentration. Photographs of these taken at different times after preparation, indicated as *as mixed*, 24 hours and 48 hours. Turbidity in samples indicates the presence of coacervate microdroplets, and its persistence at different times shows that the dispersions are stable. Turbidity is maintained at high cPE concentration after 48 hours despite high KBr concentration. Hence, stabilization of coacervates using comb-polyelectrolytes is a versatile strategy that is effective for high ionic strengths, distinct salt identities, and different polyelectrolyte strengths.



**Figure S3:** Scattering coefficient  $Q$  computed using the Mie Theory as a function of  $\alpha = \pi d/\lambda$ , where  $\lambda$  is the wavelength of incident light and  $d$  is the particle size, at a relative refractive index of 1.17.



**Figure S4:** Evolution of turbidity of different dispersions measured at different wavelengths at 10-minute intervals for 170 minutes. Solid markers represent measured values, and dotted lines represent fits.



**Figure S5:** (A) Photographs taken at different times for dispersions with a PE concentration of 62.2 mM and cPE concentration of 4.8 mM, indicated as *as mixed*, 24 hours, and 48 hours after mixing show that the presence of cPEs enables dispersion stability. (B - D) The evolution of the mean diameter of the complex coacervate microdroplets in dispersions comprising salt are shown *as mixed*, after 24 hours, and after 48 hours for cPE concentrations of 4.8 mM and 9.5 mM. The mean size of the comb polyelectrolyte stabilized coacervate microdroplets increases with increasing salt concentration but remains nearly constant with time up to 48 hours. The microdroplet diameters were measured using dynamic light scattering.

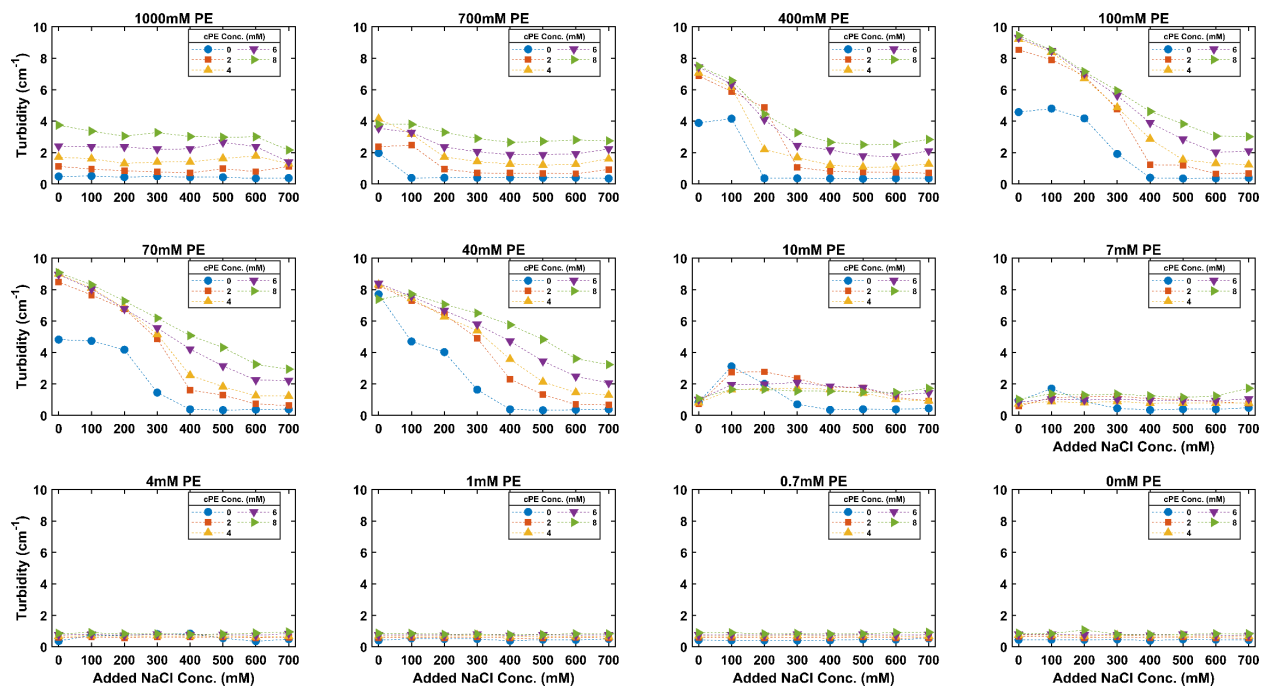


Figure S6: Turbidity of different *as mixed* complex coacervate dispersions measured at 400 nm.

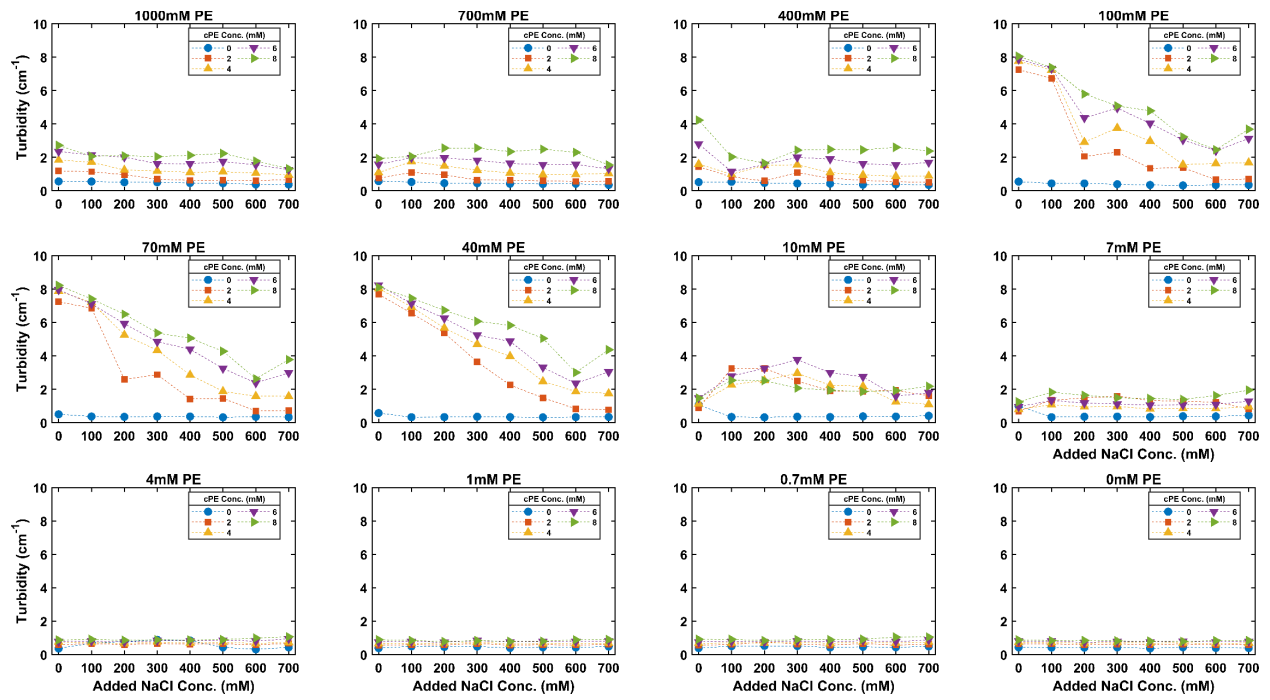
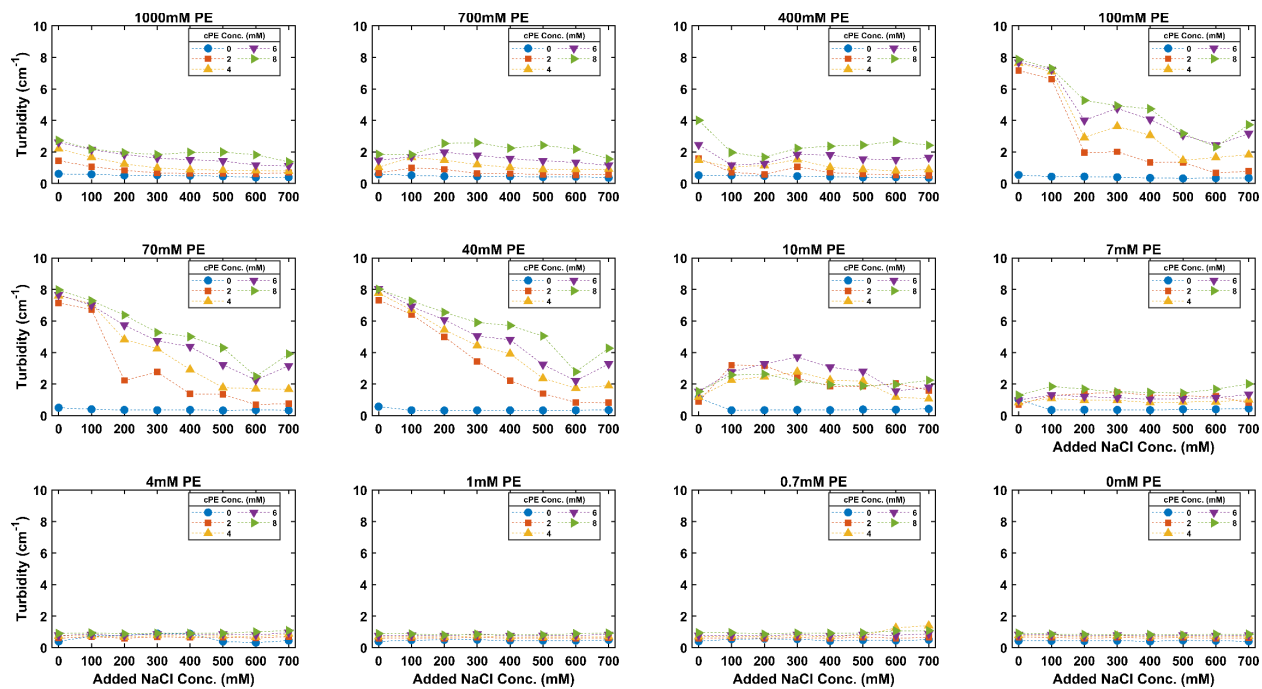
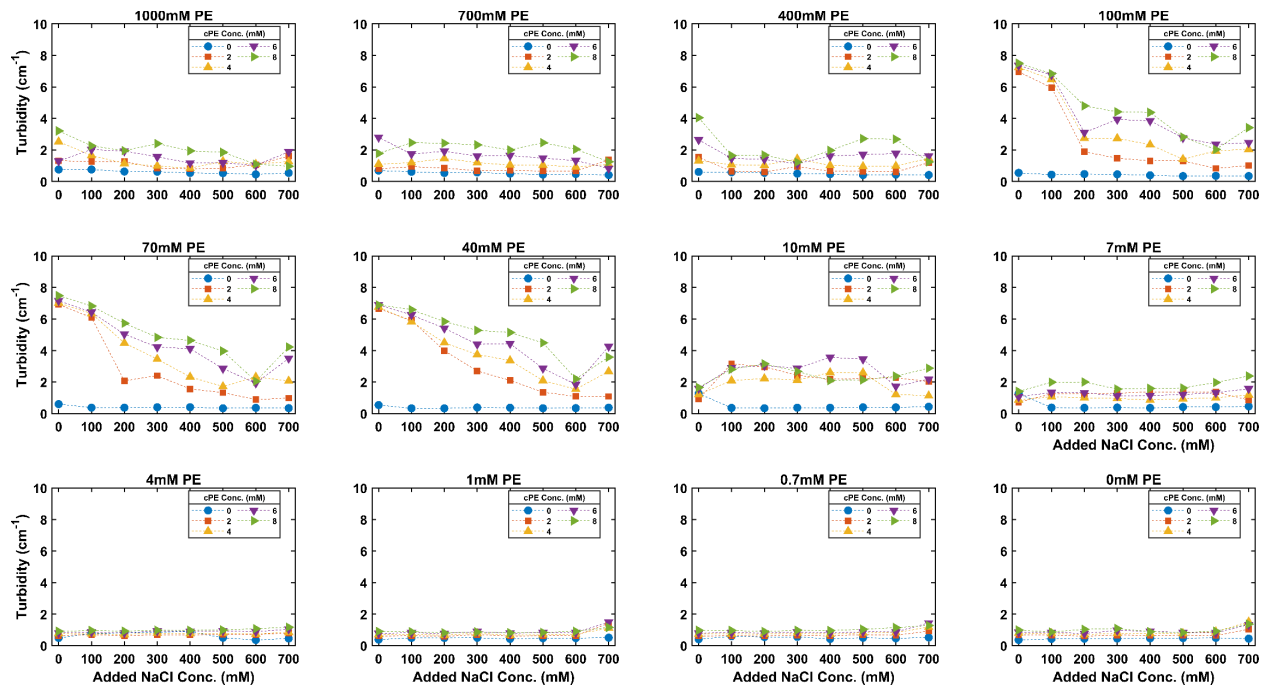


Figure S7: Turbidity of different complex coacervate dispersions measured 24 hours after preparation at 400 nm.

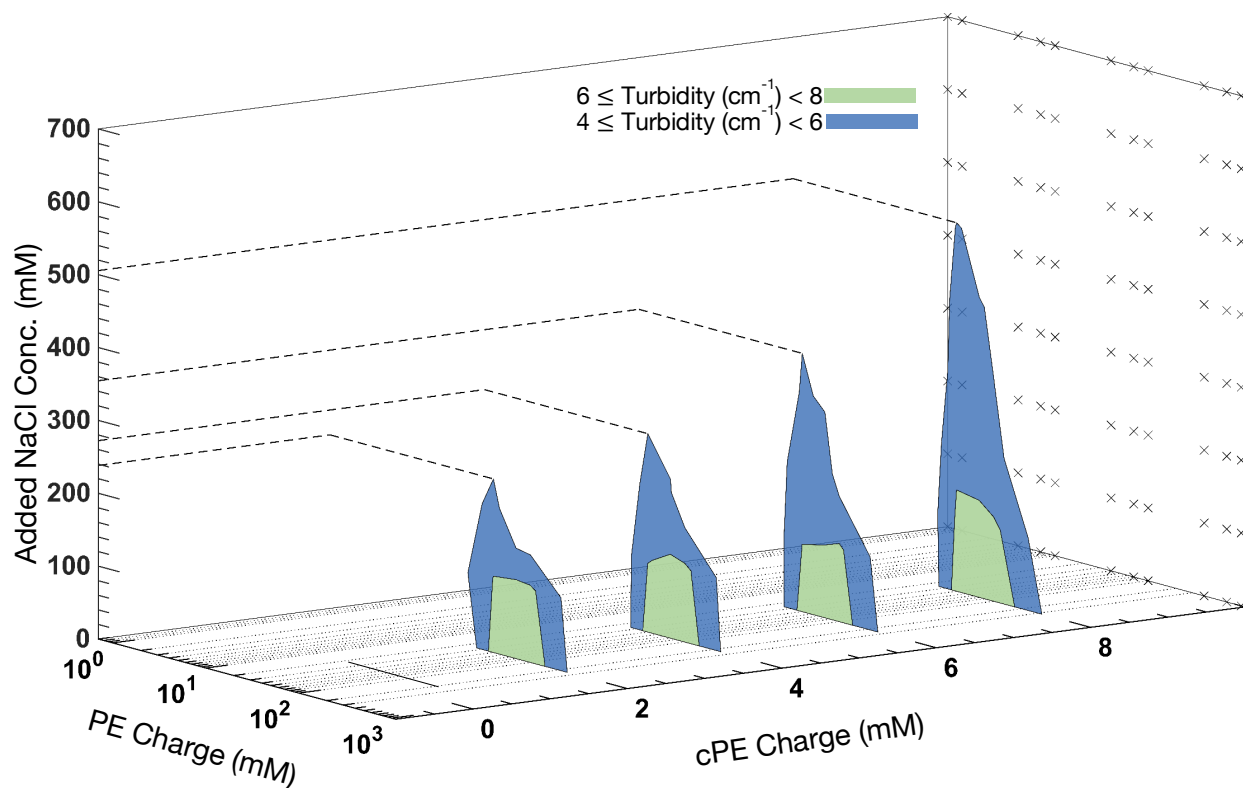




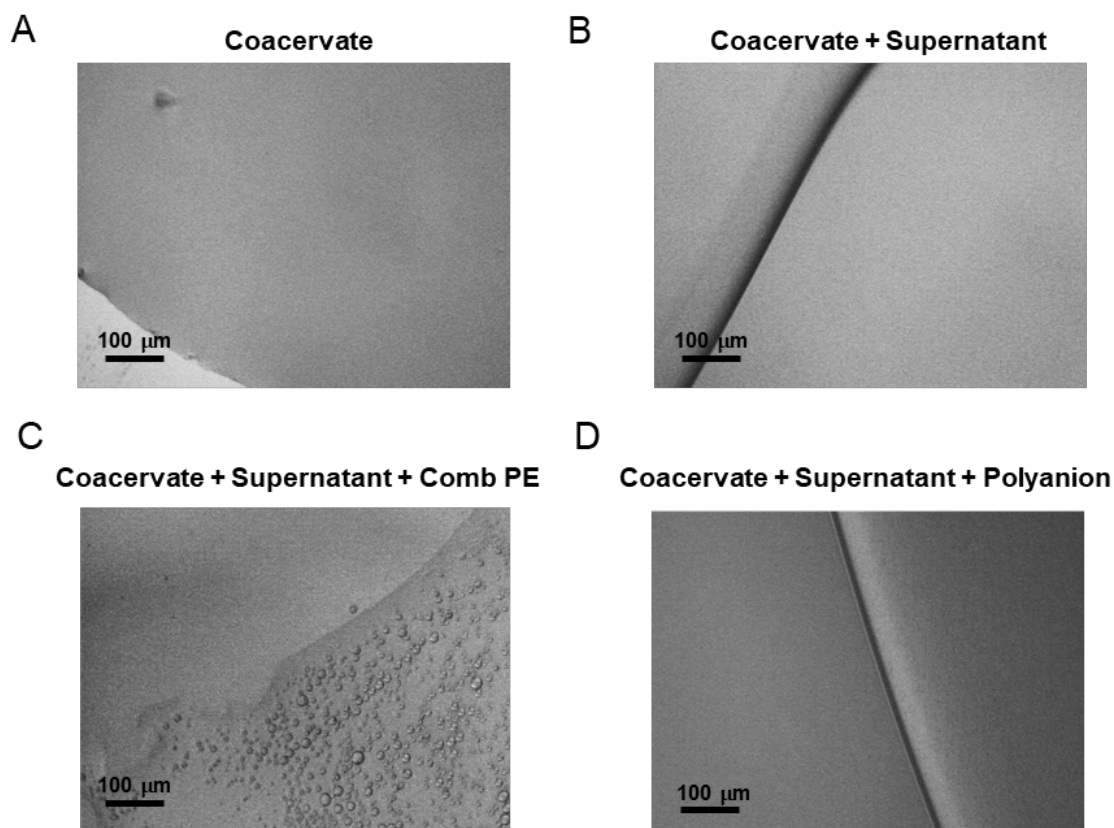
**Figure S8:** Turbidity of different complex coacervate dispersions measured 48 hours after preparation at 400 nm.



**Figure S9:** Turbidity of different complex coacervate dispersions measured 15 days after preparation at 400 nm.



**Figure S10:** Turbidity map for complex coacervate dispersions measured after 48 hours at 400 nm. The dotted lines highlight the maximum salt resistance, and the cross symbols on the back panel denote the concentrations of NaCl and PE in the dispersions.



**Figure S11: Comb polyelectrolytes significantly influence coacervate-supernatant phase equilibrium.** Coacervates were formed in the absence of cPE and were separated from the supernatant by centrifugation. The coacervate phase shows no droplets (A), and no microdroplets emerge when the supernatant is brought in contact with it (B). However, when cPE is added to the supernatant (final concentration 4 mM) and then the solution is brought into contact with the coacervate, microdroplets emerge spontaneously. (D) Coacervate and supernatant with 4 mM PAA show no microdroplet formation.