

Supporting Information:

Coacervate Formation Studied by Explicit Solvent Coarse-Grain Molecular Dynamics with the Martini Model

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1 Supporting Figures

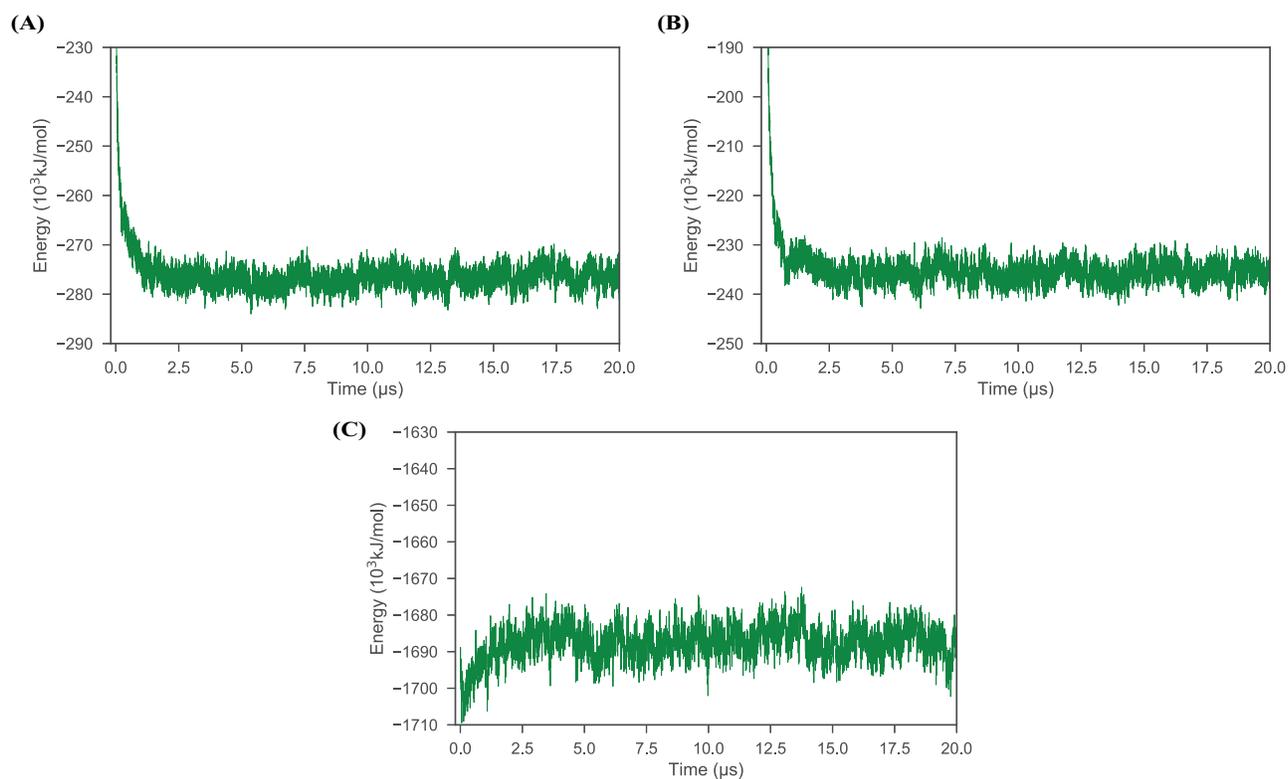


Figure S1: Equilibration of different coacervate systems. Polypeptide-polypeptide non-bonded interactions calculated for the systems composed of 100 Lys₃₀ and 100 Glu₃₀ peptides **(A)** with 0.17 M **(B)** with 0.9 M NaCl concentration. **(C)** Polypeptide-polypeptide non-bonded interactions calculated for the system composed of 600 Lys₃₀ and 600 Glu₃₀ peptides with 0.17 M NaCl concentration.

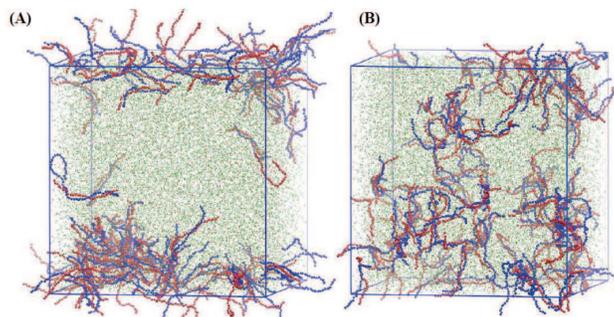


Figure S2: Breaking the coacervate by osmotic shock. **(A)** Snapshot of the coacervate formed with 0.17 M NaCl concentration, with NaCl added up to 1.3 M at 0 μ s. **(B)** Snapshot of the system after 1.6 μ s showing dissolution of the condensate.

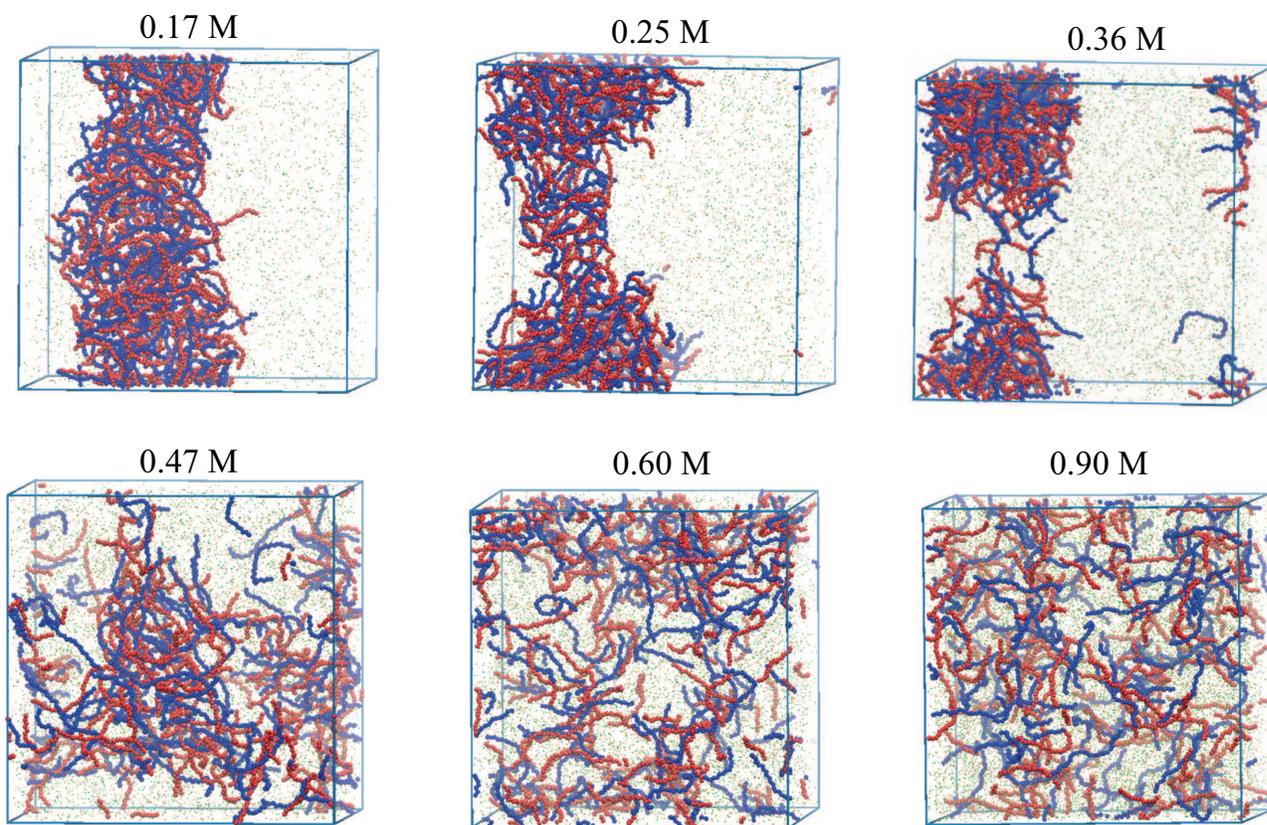


Figure S3: Salt-dependent coacervation. Snapshots of the final state of 100 Lys₃₀ and 100 Glu₃₀ polymers after 10 μ s of CG MD simulations using the final version of Martini 3.0 force field at different salt concentrations. Color code: Glu₃₀ and Lys₃₀ polyelectrolyte chains shown in blue and red respectively. The different salt solutions are shown with orange and green for sodium and chloride ions respectively in a 30x30x30 nm³ box of water, water not shown. The blue box indicates periodic boundary conditions.

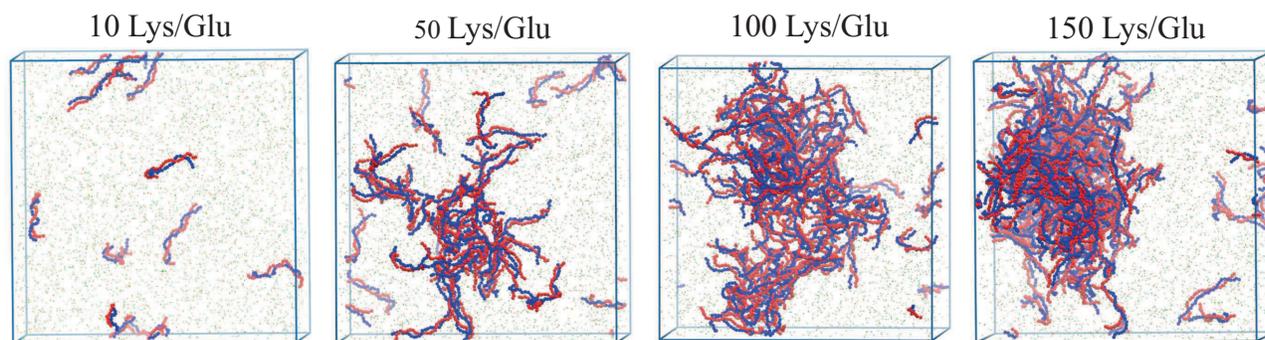


Figure S4: Snapshots of the final configurations of systems in different polymer concentrations with 0.17 M NaCl concentration.

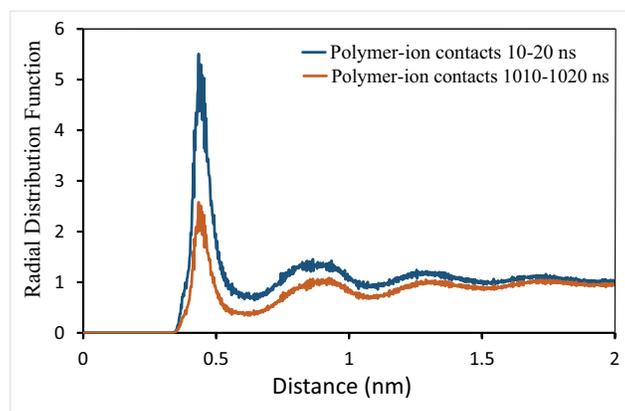


Figure S5: Peptide-ion radial distribution functions averaged over 10 ns during the beginning of the simulation (10-20 ns) before formation of the coacervate, and at the end (1010-1020 ns) when the system has phase separated. The system is composed of 100 Lys₃₀ and 100 Glu₃₀ peptides with 0.17 M NaCl concentration.

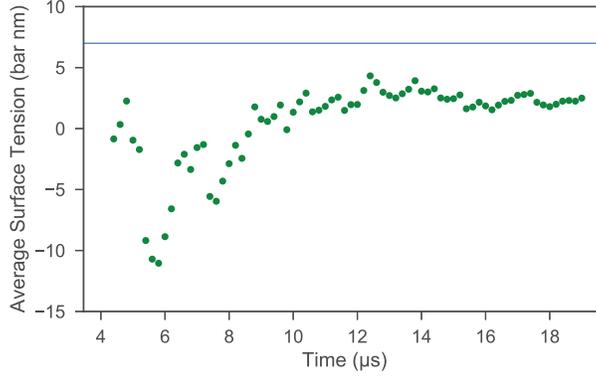


Figure S6: Cumulative average surface tension for the system with 0.17 M NaCl concentration in green. The blue line shows the experimental value for the system in 0.16 M NaCl concentration ($\gamma = 7 \text{ bar}\times\text{nm}$).

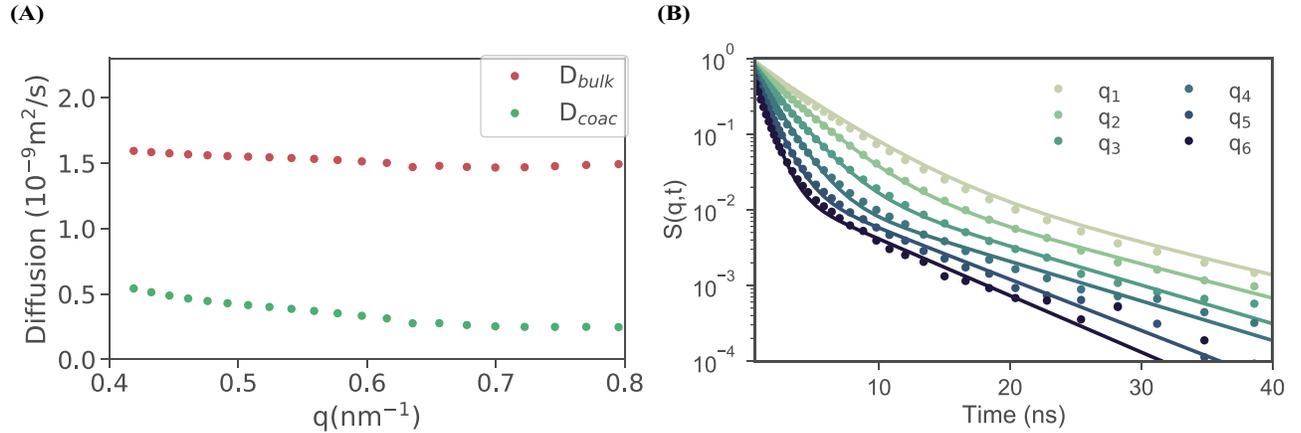


Figure S7: **(A)** Diffusion coefficients for the bulk and for the water inside the coacervates, indicated as D_{bulk} and D_{coac} respectively. For q values above ~ 0.64 (i.e. a length scale lower than 2.8 particle diameters), we find that the diffusion coefficients plateau at values of $D_{bulk} = 1.56 \pm 0.01 \times 10^{-9} \text{ m}^2/\text{s}$ and $D_{coac} = 0.240 \pm 0.02 \times 10^{-9} \text{ m}^2/\text{s}$. **(B)** Fitted the Incoherent Scattering Function of 0.17 M system for different q values. The apparent diffusion coefficients for D_{coac} are growing with decreasing q . This is likely related to the fact that for increasing probed length scales the water beads exchange with the bulk and hence display an increased diffusion.

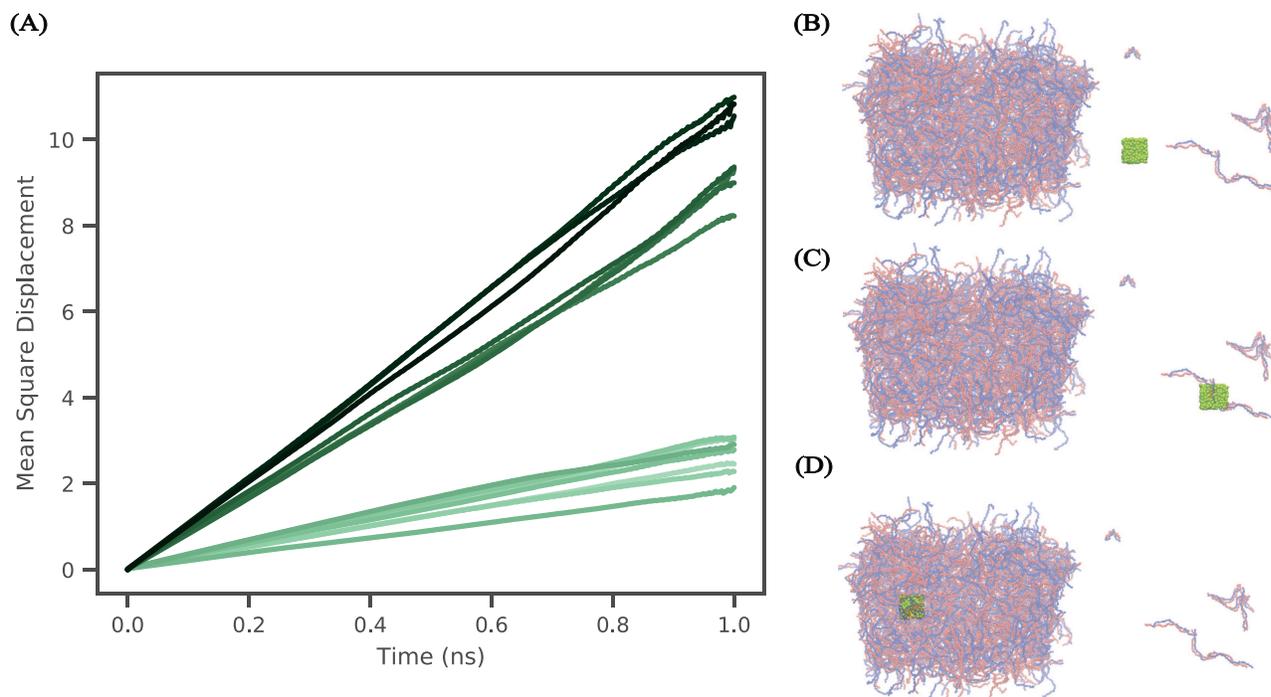


Figure S8: Mean square displacement (MSD). MSD calculations for seven different cubes of water molecules in the bulk phase (dark green) and for seven different cubes of water molecules in the coacervate phase (light green). We find the mean diffusion coefficients for the bulk and coacervate phase to be $D_{bulk} = 1.60 \pm 0.10 \times 10^{-9} \text{ m}^2/\text{s}$ and $D_{coac} = 0.43 \pm 0.04 \times 10^{-9} \text{ m}^2/\text{s}$ respectively. Snapshots of the biphasic system of 600 Lys₃₀ and 600 Glu₃₀ polyelectrolyte chains in a $24 \times 24 \times 176 \text{ nm}^3$ simulation box with 0.17 M NaCl concentration, with a cube of water molecules **(B)** in the bulk phase, **(C)** in the bulk phase and close to peptides and **(D)** in the coacervate phase.

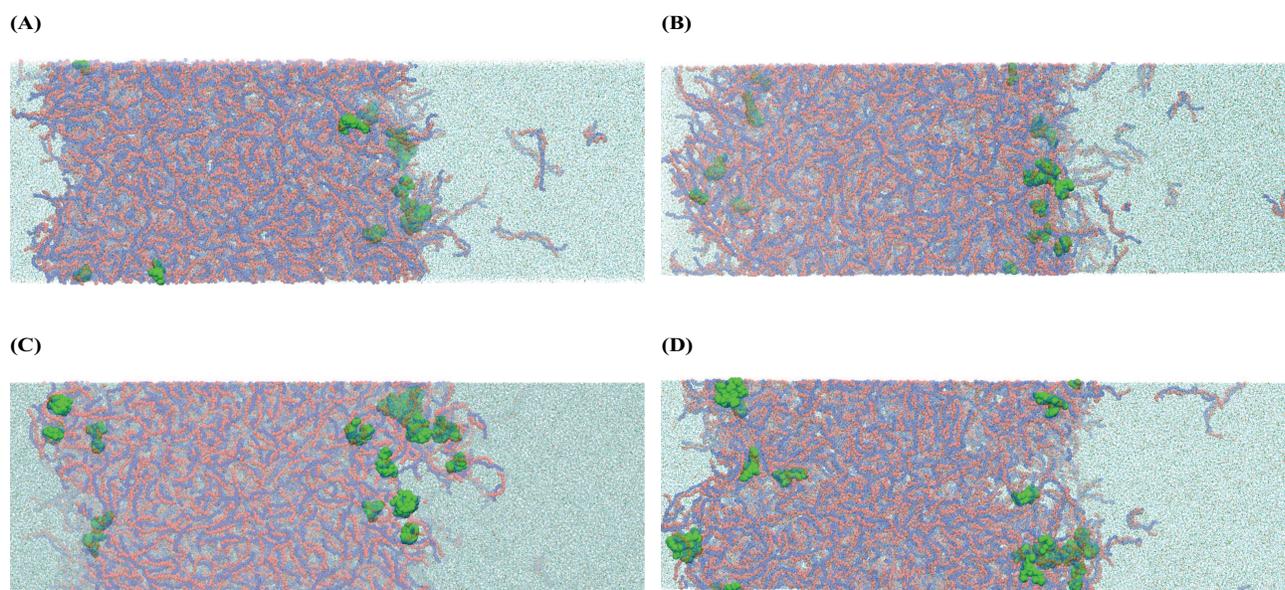


Figure S9: Partition of RNA. Final configurations of the extended systems of 581 Lys₃₀ (in red) and 581 Glu₃₀ (in blue) polymers with 20 5-mer ssRNA molecules (in green) with **(A)** 0.17 M and **(B)** 0.25 M NaCl concentration, and with 20 10-mer ssRNA molecules (in green) with **(C)** 0.17 M and **(D)** 0.25 M NaCl concentration.