

Modifying Complex Coacervates Through pH Adjustment

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In solution, oppositely charged polyelectrolytes (PEs) can form a liquid-liquid phase separation known as complex coacervation. Coacervates are used in various fields from drug delivery to cosmetics where their ability to respond to stimuli allows for engineering materials with dynamic interfaces. Complex coacervates of the polycation, poly(L-lysine), (PLys), and the polyanion poly(D,L-glutamic acid), (PGlu), were created and studied under different pH conditions. Changes in pH act as a stimulus by protonating or deprotonating weak PEs, thus strengthening or weakening the electrostatic interactions that drive coacervation. Alkaline environments deprotonate the ammonium group on PLys while acidic environments protonate the carboxylate ion on PGlu. We explore changes in size and surface charge through dynamic light scattering and zeta potential. Ratios of 2:1 and 1:2 of PLys:PGlu are studied, and we apply a core-and-shell model to understand the heterogeneity in the coacervate drop where the more abundant PE forms a shell by dominating the surface and thus dictating the sign of the charge. For the 2:1 coacervate system, as pH increases, the charge on PLys and the zeta potential decrease, while the hydrodynamic diameter increases. This indicates that as the PE of the shell loses charge, the absolute value of the zeta potential decreases. The electrostatic interactions between PLys and PGlu weaken, resulting in an increase in hydrodynamic diameter. In the 1:2 system, as pH decreases, size increases and charge becomes less negative.

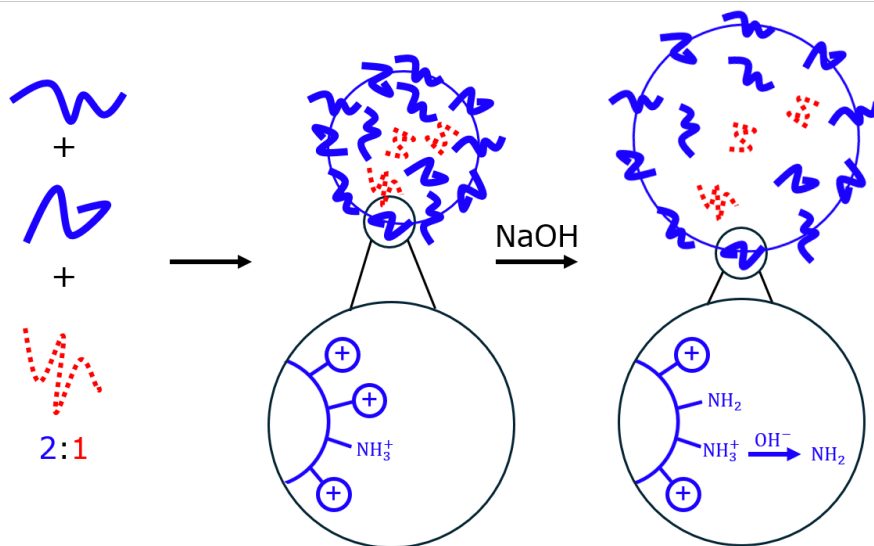


Figure 1: Formation and subsequent alkaline adjust of 2:1 coacervates. The blue chains represent the cation, PLys, and the red chains represent the anion, PGlu.