# The Importance of Salt-Enhanced Electrostatic Repulsion in Colloidal Crystal Engineering with DNA

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Here, we show that solution ionic strength can be used to control the attachment kinetics and corresponding crystal growth kinetics of DNA-functionalized nanoparticles. Simulations and theory show that at high salt concentrations (1 M NaCl), the energy barrier for crystal growth increases by over an order of magnitude compared to low concentration (0.3 M), resulting in a transition from interface-limited to diffusion-limited crystal growth at larger crystal sizes. Therefore, although one might intuitively conclude that higher salt concentration would lead to less electrostatic repulsion and faster PAE-to-PAE hybridization kinetics, the opposite is the case, especially at larger inter-PAE distances. These observations provide important insight into how solution ionic strength can be used to control the attachment kinetics of nanoparticles coated with charged polymeric materials in general and DNA in particular.

## Significance and Impact

Quantitatively understanding the self-assembly of amphiphilic macromolecules at liquid–liquid interfaces is a fundamental scientific concern due to its relevance to a broad range of applications including bottom-up nanopatterning, protein encapsulation, oil recovery, drug delivery, and other technologies. Elucidating the mechanisms that drive assembly of amphiphilic macromolecules at liquid–liquid interfaces is challenging due to the combination of hydrophobic, hydrophilic, and Coulomb interactions, which require consideration of the dielectric mismatch, solvation effects, ionic correlations, and entropic factors.

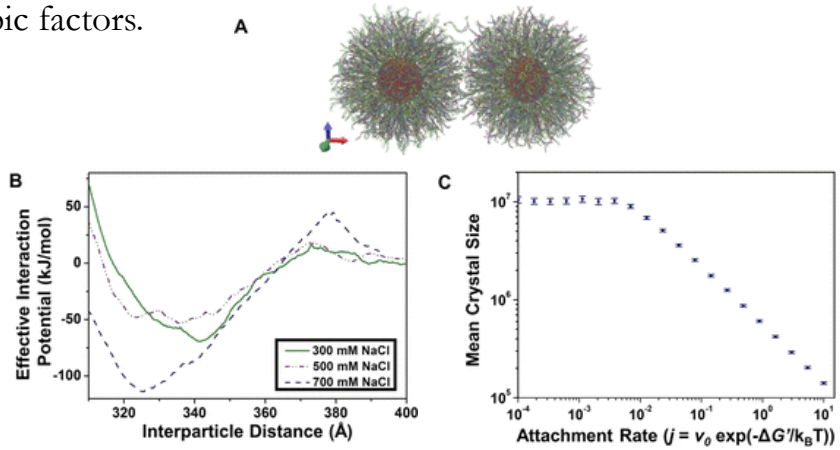


Figure 1. (A) A snapshot of two complementary PAEs at different salt concentrations in simulations. The centers of the 15 nm nanoparticles are 370 Å apart in this snapshot. (B) Pair potential energies between complementary PAEs were calculated across a range of interparticle distances. PAEs exhibit a well-defined equilibrium interparticle distance at potential minimum. At an interparticle distance of 380 Å, the attachment barrier peaks; this increases with increasing salt concentration. (C) Mean crystal size obtained without coarsening as a function of the attachment rate to the surface.

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