Insights into competing colloidal interactions in polymer encapsulation of iron oxide nanoparticles via flash nanoprecipitation

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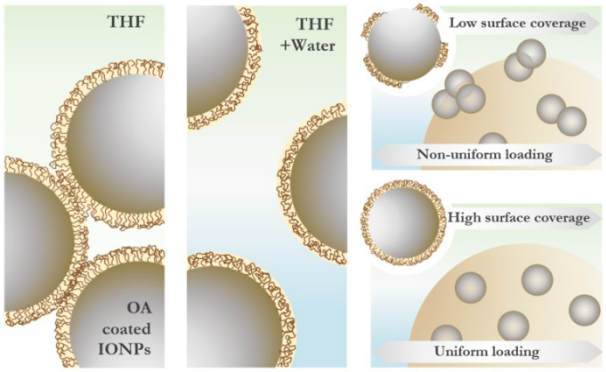
The control over synthesis conditions is necessary to ensure high loading in polymer encapsulated iron oxide nanoparticles (P-IONPs) to enable magnetic-field-guided localization for biomedical applications. While flash nanoprecipitation (FNP) is a robust method to fabricate P-IONPs, very little is known regarding the response of hydrophobic IONPs to the change in solvent quality upon water addition and its effect on the nature of encapsulation. In this work, we first probe the cluster sizes of oleic acid (OA) coated IONPs at varying composition of water in THF(tetrahydrofuran)-water mixture through kinetic measurements using dynamic light scattering (DLS). It was found that the OA-IONPs exist as large micron sized clusters in THF, which broke into smaller clusters as water composition increased. A free energy model of clustering is presented and fitted to qualitatively describe the competing repulsive and attractive interparticle interactions driving the equilibrium cluster sizes (d*) as a function of water composition. This effect was further pronounced when an aqueous Tween80 solution was added to the dispersion, resulting in individually dispersed IONPs at high . These interpretations from DLS measurements were confirmed using cryo-electron-microscopy. The d* showed direct co-relation with the nature of P-IONPs produced using FNP at aqueous to organic flow rate ratio, resulting in complete encapsulation when IONPs were individually dispersed, and corona like assemblies around polymer particle surface at slightly larger d*. Finally, a systematic study was performed to investigate the effects of concentration of polymer and IONPs on the extent and nature of encapsulation of IONPs with varying OA surface coverage. The IONPs with low surface coverage and high IONP concentration resulted in maximum encapsulation with non-uniform distribution of IONPs in the polymer particle matrix. Although, IONPs of similar sizes with higher surface coverage resulted in moderate encapsulation, the distribution inside polymer matrix was found to be highly uniform. These understandings on competing interactions during the course of encapsulation would enable rational design of P-IONPS using FNP, with high and uniform loading.



Schematic of IONP clustering and surface coverage driving encapsulation of IONPs