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Biophysical and Macromolecular Chemistry

Our research focuses on understanding and controlling the effects of additives on the self-assembly and transport properties of molecules of interest in water. Our two main research topics are discussed below.

Condensation of proteins and dendrimers in aqueous solutions.

Proteins and dendrimers (see Fig. 1) are globular macromolecules that find applications in the fields of pharmacology, biotechnology, catalysis and materials science.

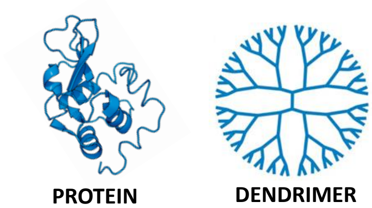

Figure 1. Structure of protein and dendrimer.

These applications often involve their aqueous mixtures. In solution, macromolecules are subject to processes such as coacervation, crystallization, self-assembly, aggregation and gelation. These transformations are denoted as "condensation" processes because they lead to the formation of a new phase rich in protein or dendrimer (see Fig. 2).

Understanding and controlling these processes is important for improving the physical stability of drug formulations, developing new nanomaterials, enabling crystallization and identifying strategies for curing diseases. Our research group focuses on understanding how additives such as salts, osmolytes, linear polymers and denaturants affect the condensation of proteins and dendrimers in aqueous media. Research projects in our laboratory involve the experimental characterization of condensation processes in the presence of additives, the development of theoretical models explaining experimental findings and examining the impact to the fields mentioned above.

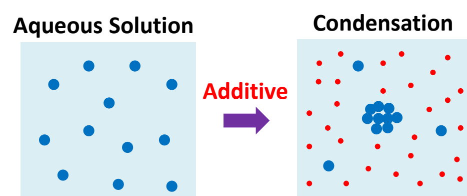

Figure 2. Condensation of globular macromolecules.

One phenomenon we are currently investigating is the formation of dendrimer nanospheres (see Fig. 3) through a process denoted as oligomerization-induced liquid-liquid phase separation.

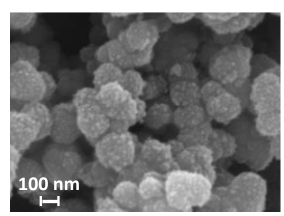

Figure 3. Dendrimer nanospheres with radius of ≈100 nm

Another phenomenon we are investigating in our laboratory is macromolecular crowding. This is the condensation of proteins induced in the presence of polymeric additives that mimic the crowded environment of living systems.

Salt-induced migration of proteins and polymers in aqueous solutions.

Understanding the molecular motion of proteins, polymers and ions in water is central to many chemical, physical and biological processes including dialysis, adsorption, crystal growth, transport within microfluids, reactions, transport inside living systems, separation technologies, and the controlled release of therapeutic molecules. Our research group focuses on understanding diffusion, the transport phenomenon describing molecular motion in the presence of concentration gradients. The intrinsic diffusion (Brownian motion) of macromolecules in aqueous media has been extensively investigated due to its importance in the transport processes mentioned above. However, in the presence of other components such as salts, macromolecules are subjected to another type of diffusion phenomenon beyond Brownian motion. Specifically, the concentration gradient of salt ions or other additives can induce the migration of a protein or a polymer in water. This phenomenon, which is not well understood, is denoted as diffusiophoresis or cross-diffusion.

Our research group has reported the first experimental characterization of salt-induced protein diffusiophoresis (see Fig. 4) using a very precise interferometric method.

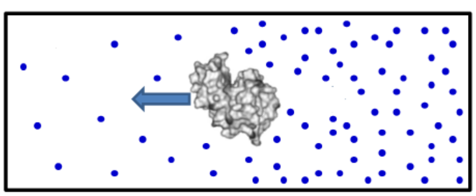

Figure 4. Protein diffusiophoresis induced by a concentration gradient of salt ions (●).

We have also shown that salt-induced diffusiophoresis of macromolecules correlates with the Hofmeister series, the ranking of salt ions according to their effectiveness (salting-out strength) in promoting macromolecular condensation (see Fig. 5). For example, strong diffusiophoresis of polyethylene glycol is observed in the presence of concentration gradients of sodium sulfate in water.

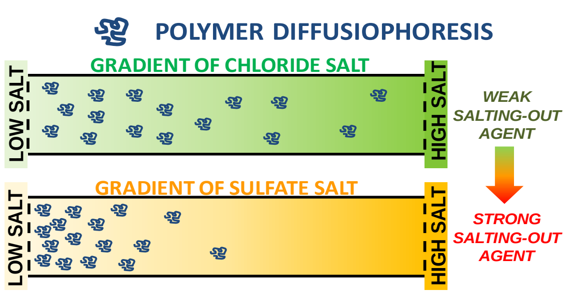

Figure 5. Accumulation of polymer towards low salt concentration due to diffusiophoresis in the presence of a weak (top) and strong (bottom) salting-out agents.

Research projects in our laboratory involve the experimental characterization of diffusiophoresis, the development of theoretical models explaining experimental findings and assessing the role of this transport process in diffusion-based mass transfer problems such as controlled release from porous media, adsorption onto surfaces, crystal growth and separation of macromolecules.

Selected Publications

- V.C.P. da Costa, and O. Annunziata, *Phys. Chem. Chem. Phys.*, **17**, 28818–28829 (2015).
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- M.S. McAfee and O. Annunziata, *Langmuir*, **30**, 4916−4923 (2014).
- O. Annunziata, D. Buzatu and J.G. Albright, *J. Phys. Chem. B*, **116**, 12694-12705 (2012).
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- O. Annunziata**,** A. Payne and Y. Wang, *J. Am. Chem. Soc.*, **130**, 13347-13352 (2008).
- H. Zhang and O. Annunziata, *Langmuir*, **24**, 10680-10687 (2008).
- Y. Wang and O. Annunziata, *Langmuir*, **24**, 2799-2807 (2008).
- Y. Wang and O. Annunziata, *J. Phys. Chem. B*, **111**, 1222-1230 (2007).

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