



**Zimbitas, G. and Forsyth, C. and Sefcik, J. (2018) Concentration and pH dependence of colloidal scale solute clustering within aqueous solutions of small organic molecule. In: 32nd Conference of the European Colloid and Interface Society, 2018-09-02 - 2018-09-07, University of Ljubljana. ,**

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## **Concentration and pH dependence of colloidal scale solute clustering within aqueous solutions of small organic molecule**

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Aqueous solutions of well soluble molecules are usually assumed to be essentially homogenous systems with some degree of local structuring due to specific interactions on the sub-nanometre scale, involving the creation of, e.g. molecular clusters, hydration shells, etc., usually not exceeding several solute molecules. These small molecular structures (molecular clusters) are commonly observed (experimentally and theoretically) in aqueous solutions of many organic and inorganic systems.

The presence of colloidal scale (mesoscale) structures has been well established in aqueous solutions of various small molecules [1] and these clusters have been proposed to be involved in non-classical crystal nucleation mechanisms [2,3]. Clusters in solutions of amino acids have been reported to be thermodynamically stable and can be present in both undersaturated and supersaturated solutions with respect to the solid-liquid equilibrium [4,5].

We investigated concentration and pH dependence of mesoscale clustering in aqueous solutions of small organic molecules, including amino acids (such as glycine) and amines (such as triethylenetetramine), which are well soluble in water and have a range of charged states that can be adjusted by changing solution pH. We used Dynamic Light Scattering (DLS) and Brownian Microscopy/ Nanoparticles Tracking Analysis (NTA) in order to measure size distributions and number concentrations as well as scattered intensities and optical contrast of mesoscale clusters.

Mesoscale clusters were present in undersaturated solutions at solute concentrations well below the solid-liquid equilibrium (saturation) concentration at a given temperature, with mean diameters within 200-400 nm. Scattering intensities and number concentrations increased with increasing solute concentrations, while the mean size of mesoscale clusters remained approximately constant.

When pH was varied in amino acid solutions, values away from the isoelectric point resulted in a decrease of the number concentration of mesoscale clusters. While the mean size of mesoscale clusters remained approximately independent of pH, scattering intensities decreased sharply as solution pH moved more than 3 units away from the isoelectric point in either direction. This change was closely correlated with the speciation of charged species, whereby the zwitterions predominate near the isoelectric point and positively and negatively charged species predominate at lower and higher pH, respectively.

We note that the mesospecies are not to be seen as a separate phase and the system is better described as a thermodynamically stable mesostructured liquid containing solute-rich domains (mesoscale clusters) dispersed within bulk solute solution [6]. At a given temperature, solute molecules in such a mesostructured liquid phase are subject to equilibrium distribution between solute-rich mesospecies and the surrounding bulk solution.

Keywords: soluble molecules, aqueous solutions, small molecular structures, molecular clusters

Accepted manuscript of the following research output: Zimbitas, G., Forsyth, C., & Sefcik, J. (2018). *Concentration and pH dependence of colloidal scale solute clustering within aqueous solutions of small organic molecule*. Presented at 32nd Conference of the European Colloid and Interface Society, Ljubljana, Slovenia.