Experimental Report

15/01/2014

Proposal:	**9-11-1566**	**Council:**	10/2011

Title: Exploring the chemical architecture dependence of polyelectrolyte solutions on spatial distribution of counter-ion by SANS

This proposal is a new proposal

Research Area: Physics

Main proposer: **PORCAR Lionel**

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Samples: Polystyrene

Instrument	Req. Days	All. Days	From	To
D22	3	2	17/09/2012	18/09/2012
			19/07/2013	20/07/2013

Abstract:

Polyelectrolytes are unique among polymers because of their good solubility in aqueous solutions. Numerous commercial applications have been developed using such materials, mostly related to their capabilities of modifying flow and stability properties of aqueous solutions and gels. Generally, the small angle neutron scattering (SANS) intensity I(Q) obtained from polyelectrolyte solutions are characterized by the presence of an interaction peak. This characteristic peak is related to the correlation length and the liquid ordering of polyions in solution. In addition to these characteristic peaks, an upturn is observed in the low Q region. This low Q upturn in SANS spectra is repeatedly observed for all types of ionic macromolecule systems including polyelectrolytes, DNA and protein solutions. [2] It has been found that the upturn in I(Q) cannot be predicted by any existing liquid theories or computer simulations. As this upturn is related to the stability and structure formation of a variety of ionic macromolecules in aqueous media, understanding the origin of this phenomenon is critical for future development.

Experimental Report of D22

An active field of soft colloidal research is the investigation of the structure of a single soft colloidal particle. When concentration c is below the overlap concentration c*, results of scattering experiments suggest that their global size of soft colloid is independent of c. Therefore it is generally believed that the conformation of soft colloids in solution exhibits essentially no concentration dependence below the overlap concentration c*.

Combining two neutron scattering techniques and molecular dynamics (MD) simulation, we investigate the conformational properties of PAMAM dendrimers dissolved in water, an extensively studied soft colloid model system. MD simulation indicates the importance of invasive water for determining the dendrimer conformation. Via monitoring the evolution of invasive water distribution inside a dendrimer, Contrast variation SANS experiment carried out at D22 further demonstrates the occurrence of significant intra-particle dehydration upon increasing the concentration below c*.

The dynamics of soft colloids in solutions is characterized by internal collective motion as well as center-of-mass diffusion. Based on the complementary In15 study, the origin of this dehydration is found to be the competition between the relaxation processes associated with these two degrees of freedom.

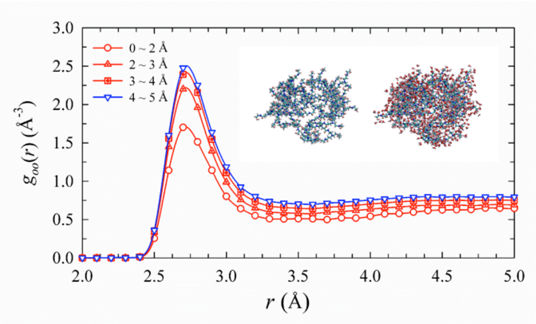

Figure 1. The averaged oxygen-oxygen pair distribution function goo(r) of TIP3P water around the vicinity of a G4 dendrimer. The insets give the snapshots of hydrocarbon components of a dendrimer (left) along with the surrounding invasive water (right).

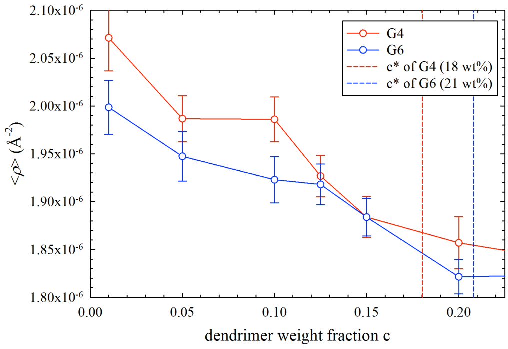

Figure 2. The average scattering length density of a single dendrimer <ρ> for G4 and G6 PAMAM dendrimer in water as a function of concentration. Upon increasing *c*, a progressive decrease in <ρ> is observed. Our data analysis demonstrates that this observation is the reflection of the decrease in intra-dendrimer hydration.