Multi Particle Tracking Microrheology Probing Structure and Flow of Heterogeneous Complex Fluids

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Multi particle tracking (MPT) optical microrheology using fluorescent tracer particles with diameters be- tween 100 nm and 1000 nm is a versatile tool to study the structure and dynamics of heterogeneous complex fluids. The particle size and also the inter-particle distance are the length scales on which structural information is obtained. The method can be applied to study transparent systems like protein filament networks, polymeric thickener solutions or dilute clay suspensions, but it is also applicable to turbid, highly concentrated colloidal dispersions.

Comparing the mesh size of intermediate filament (IF) networks - the key structural components of metazoan cells - as obtained from MPT with their bulk modulus, reveals a strong entropic contribution of stretched filament configurations to G_0. Attractive interactions due to hydrophobic amino acid clusters in the central part of the proteins prevent stretched filaments between adjacent cross-links from thermal equilibration and thus provide the high modulus values.

Aqueous solutions of three different commercial acrylic thickener solutions have been investigated. The polymer with the highest thickening efficiency exhibits a pronounced heterogeneity on the micrometer length scale and the degree of heterogeneity strongly increases with increasing polymer concentration. Predominantly elastic and viscous regions within the solution can be identified based on the slope of the mean squared displacement (MSD) of individual particle trajectories. This heterogeneity is directly imaged using Voronoi diagrams and characteristic length scales varying from 5 to 20 μm are found. Probe size variation additionally reveals that the elastic regions themselves are heterogeneous on the length scale < 500 nm.

MPT reveals unique information about kinetics of structure formation and aging of clay suspensions in the repulsive glassy and attractive gel-like state. The gel-state is characterized by a pronounced spatial heterogeneity resulting in a more than an order of magnitude variation of MSD values at the macroscopic sol-gel transition. The structure of attractive gel-like states continuously refines and the crossover of G' and G'' in MPT experiments occurs later when smaller particles are tracked. This gradual refinement of the structure corresponds to a pronounced aging on the macroscopic scale. In contrast, glassy systems are spatially homogeneous down to 200 nm.

Highly concentrated, turbid, aqueous colloidal dispersions have been studied systematically covering the full range of the phase diagram. Fluid suspensions with short range repulsive interactions at particle loading $\phi < 0.5$ exhibit perfect agreement

between viscosity from bulk rheometry and microrheology.

Hard sphere type crystallizing dispersions in the liquid/crystalline coexistence regime with and without added non-adsorbing polymer show a large variation in MSD of different tracer particles with slopes $\partial MSD / \partial \tau$ between 0 and 1. The heterogeneity of the samples can be directly imaged based on this rheological contrast. The broadening of the coexistence region due to weak attractive depletion forces induced by added polymer is directly proven by MPT.

Tracer particle mobility systematically varies as the colloidal glass transition ϕ_g = 0.58 is approached. The change of particle mobility and the variation of sample heterogeneity is also discussed in the so-called re- entry regime at $\phi > \phi_g$ where the system transitions from glassy to fluid and from fluid to gel-like when attractive interaction controlled by non-adsorbing polymer increases from 0 to about 10 kT.