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Multiarm Star Polymers as Model Soft Colloids

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Over the last decade, star polymers emerged as a useful model colloids that interpolate between polymers and hard sphere colloids. Together with microgels, they represent two benchmark soft colloidal systems, their internal structure being the key difference. Indeed, in the case of stars with open structure, the arms can interpenetrate in dense suspensions. The latter feature, that can be probed experimentally, is responsible for a number of interesting structural and dynamic properties of star polymers that set them apart from microgels. In this talk we present the basic properties of star polymers and focus on their extraordinary behavior in the highly concentrated regime, which is typically glassy. Our rheological and scattering experiments demonstrate unique features of the star glasses. Here we discuss two major ones: (i) Aging after pre-shear (the so-called rejuvenation) proceeds via a two-step process, associated with a fast arm engagement and a slow cooperative (cage) rearrangement. Remarkably, at extremely long times a steady state is observed and the terminal time in these systems can be experimentally accessible (and hence tailored at molecular level), as a consequence of the arms fluctuations. (ii) Multiple glassy states can be obtained when mixing stars with polymers or with other stars. Simultaneous theoretical and simulations work suggests that the softness is at the core of this unexpected behavior where depletion gives rise to glass melting and eventually re-entrant glasses are formed. Construction of a state diagram suggests kinetic pathways for tailoring the flow of soft colloids. These examples outline the importance of particle architecture on colloidal properties. Stars are a representative of a large class of hairy particles. The parallel important developments in mode coupling theory and its verses provide much needed predictive tools and rationalization for a number of phenomena such as those discussed here, as well as the complex rheological response. A wide range of applications in this exciting, fast growing field appear to emerge. Parts of this work reflect collaboration with M. Cloitre (ESPCI), B. Erwin (FORTH/ESPCI), C. N. Likos (Duesseldorf), G. Petekidis (FORTH), F. Sciortino (Rome), E. Stiakakis (FORTH), and E. Zaccarelli (Rome). Synthesis of particles by N. Hadjichristidis (Athens), M. Gauthier (Waterloo) and J. Roovers (NRC).