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Coupling of gelation and glass transition in a biphasic colloidal mixture—from gel-to-defective gel-to-glass¹ HE CHENG, DI JIA, CHARLES HAN, Chinese Academy of Sci (CAS) — The state transition from gel to glass is studied in a biphasic mixture of polystyrene core/poly (*N*-isopropylacrylamide) shell (CS) microgels and sulfonated polystyrene (PSS) particles. At 35 C, the interaction between CS is due to short-range Van der Waals attraction while that between PSS is from long-range electrostatic repulsion. During variation of the relative ratio of the two species at a fixed apparent total volume fraction, the mixture exhibits a gel-to-defective gel-to-glass transition. When small amounts of PSS are introduced into the CS gel network, some of them are kinetically trapped, causing a change in its fractal structure, and act as defects to weaken the macroscopic gel strength. An increase of PSS content in the mixture promotes the switch from gel to defective gel, *e.g.*, the typical two-step yielding gel merges into one-step yielding. This phenomenon is an indication that inter-cluster bond breakage coincides with intra-cluster bond fracture. As the relative volume fraction of PSS exceeds a critical threshold, the gel network can no longer be formed; hence, the mixture exhibits characteristics of glass. A state diagram of the biphasic mixture is constructed, and the landscape of the different transitions will be described in future studies

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He Cheng
Chinese Academy of Sci (CAS)

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