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Enhanced gel formation in binary mixtures of nanocolloids with tunable short-range attraction

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We report a combined experimental, theoretical, and simulation study of the phase behavior and microstructural dynamics of concentrated binary mixtures of spherical nanocolloids with a size ratio near two and with a tunable, intrinsic short-range attraction. In the absence of the attraction, the suspensions behave as well mixed, hard-sphere liquids. For sufficiently strong attraction, the suspensions undergo a gel transition. Rheometry measurements show that the fluid-gel boundary of the mixtures does not follow an ideal mixing law, but rather the gel state is stable at weaker interparticle attraction in the mixtures than in the corresponding monodisperse suspensions. X-ray photon correlation spectroscopy measurements reveal that, in contrast with depletion-driven gelation at larger size ratio, gel formation in the mixtures coincides with dynamic arrest of the smaller colloids while the larger colloids remain mobile. Molecular dynamics simulations of the suspensions similarly observe gel formation driven by a structural arrest of the smaller particles that occurs at weaker strength of attraction than the gel point of the monodisperse suspensions. Characterization of the microstructure n the simulations indicates the arrest results from microphase separation that results from a subtle interplay of entropic and enthalpic effects and that drives the smaller particles to form dense regions. These observations suggest a potential new avenue for tailoring the gel-forming properties of colloidal suspensions.

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