We investigate the phenomenon of gravitational collapse in colloidal gels via dynamic simulation in moderately concentrated gels formed via arrested phase separation. In such gels, rupture and re-formation of bonds of strength $O(kT)$ permits ongoing structural rearrangements that lead to temporal evolution — aging — of gel structure and rheology [1]. The reversible nature of the bonds also allows the gel to transition from solid-like behavior to liquid-like behavior and back to solid-like behavior when forcing is removed. But such gels have also been reported to be susceptible to sudden and catastrophic collapse of the entire structural network, during which the gel sedimented into a dense layer, eliminating any intended functionality of the network scaffold. Although the phenomenon is well studied in the experimental literature, the microscopic mechanism underlying the collapse is not understood [2–17].

To study this behavior, we conduct large-scale dynamic simulation to model the structural and rheological evolution of a gel subjected to a range of strengths of gravitational stress. The model comprises 750,000 Brownian particles interacting via a hard-core repulsion and short-range attractive interactions that lead to formation of a gel; the gel is evolved quiescently over time, whereby its structure and rheology evolve via diffusive particle migration [1]. A range of attraction strengths and gel ages are studied. Particle positions and network stress are monitored throughout simulation, along with bulk strain of the gel.

We find that the bulk deformation of the gel agrees with experimentally reported behavior [3, 9, 14]: Three temporal regimes emerge: macroscopic slow, pre-collapse evolution; collapse and rapid sedimentation; and long-time compaction, each of which is connected to distinct phases of structural and rheological evolution. The detailed microstructural evolution during this process is reported, along with the dependence of the delay time and speed with attraction strength and magnitude of the applied stress relative to Brownian forces. The influence of the interplay between Brownian motion, attractive forces, osmotic pressure, and gravitational forces on collapse and subsequent restructuring is elucidated, and a connection to phase behavior is made.

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