DYNAMICS OF COLLOIDAL GLASS-FORMERS

Erin Teich, University of Michigan
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EXECUTIVE SUMMARY

Glass formation is a well-known outstanding mystery in the physical sciences. Although it has been an aspect of daily life for millennia, it still lacks a canonical thermodynamic explanation. We utilized our Blue Waters allocation to help shed light on this murky topic by investigating the structure and dynamics of assembly failure in a family of monodisperse colloidal systems composed of particles of related polyhedral shapes. We found that assembly failure arises from a competition between local structural motifs that are preferred in ordered structures composed of particles of similar, but not identical, shapes. Our work demonstrates the power of considering families of related systems when exploring phase behavior, and additionally probes the long-sought nature of the relationship between structure and dynamics in glass-forming systems.

RESEARCH CHALLENGE

Colloidal systems are capable of self-assembling into a wide variety of ordered structures ranging from the simple to the exceedingly complex [1]. Often, however, no such assembly occurs and the system instead remains disordered, displaying dynamical signatures characteristic of glass-formers. An understanding of why systems sometimes avoid crystallization is crucial, both for developing robust methods of colloidal self-assembly and for the development of glass-based technologies including rewritable data storage devices [2] and fiber optic networks. Despite the importance of this understanding, the underlying mechanism of the glass transition remains in contention. This is due in large part to the significant slowing down of any system as it approaches the glass transition [3], requiring investigations of glass formation to resolve system dynamics on time scales that vary by orders of magnitude. We were able to tackle this problem in studying glass-forming dynamics in a model colloidal system by using our highly parallel simulation package, HOOMD-blue [4], and access to both the Cray XK7 nodes hosted on Blue Waters and its very generous storage capabilities.

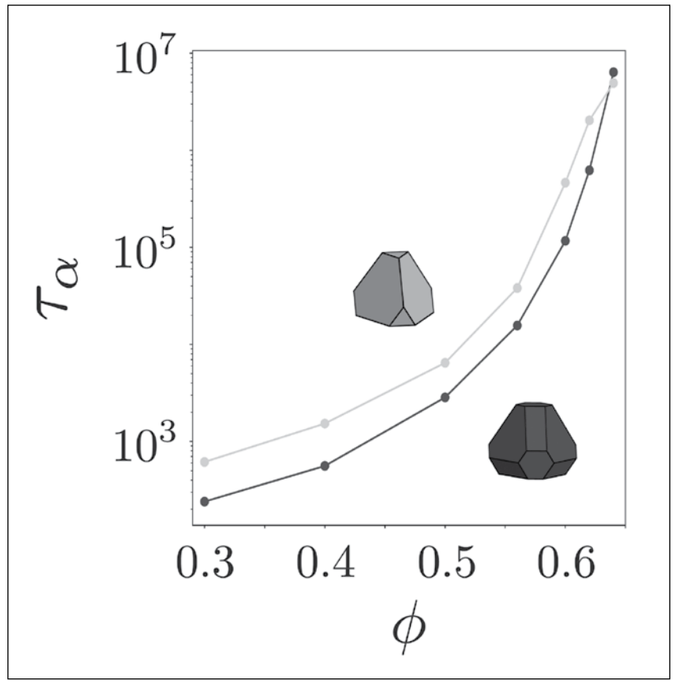

Figure 1: Relaxation time as a function of system density for two model glass-formers comprised of particles of slightly different polyhedral shapes. Note that the fragilities of these systems, or the slopes of these curves, are dependent on particle shape.

METHODS & CODES

We performed hard particle Monte Carlo (HPMC) [5] simulations of model glass and crystal-formers comprised of hard polyhedra contained in the spheric triangle invariant 323 family [6]. These are a set of convex polyhedra formed by truncating the vertices and edges of a tetrahedron by sets of planes at varying radial distances from the polyhedron center. In HPMC simulations, particles have no interactions aside from those of excluded volume, and systems are therefore entirely entropically mediated. It was discovered previously [7] that systems of particles in certain regions of this shape space assemble into a rich variety of colloidal crystals, while systems in other regions of this shape space fail to assemble into any ordered structure at any density studied. We used Blue Waters to corroborate this previous discovery and to verify that these systems do in fact display canonical dynamical signatures that are characteristic of glass-formers.

RESULTS & IMPACT

To probe the structural and dynamical properties of suspected colloidal glass-formers, we simulated systems of 4,096 particles on a single GPU at a variety of densities, and subsequently measured structural and dynamical information. To gather trajectory information at a wide range of time scales spanning six orders of magnitude, we wrote our system trajectories to disk frequently—every 10 Monte Carlo (MC) sweeps—and collected data for the far longer period of 100 million MC sweeps. We produced trajectories as large as 3.25 TB per simulation, each containing about 10 million simulation frames. We were able to store these extraordinarily large trajectories on Blue Waters' generous file system, using our 500 TB allocation, and we analyzed various dynamical signatures associated with canonical glass formation. We found that our colloidal glass-formers exhibited these signatures, including plateaus in the mean-squared displacement and the real part of the self-intermediate scattering function that indicates caging, and peaks in the non-Gaussian parameter [8] as well as the self-part of the four-point susceptibility [9] that indicates dynamical heterogeneity associated with relaxation events. We were also able to extract relaxation times for our simulated systems and to plot those relaxation times as functions of system density in a variation of the canonical Angell plot [3]. Fig. 1 shows these relaxation times for two example systems and demonstrates that glass-former fragility is dependent on particle shape.

Additional work on other systems demonstrated a structural connection to the dynamical arrest we quantified on Blue Waters: We found that assembly failure in each system we studied arises from a competition among local structural motifs that are preferred in ordered structures composed of particles of similar (but not identical) shapes. Our work, facilitated by the resources of Blue Waters, helps to provide insight into the relationship between structure and dynamics in colloidal glass-formers.

WHY BLUE WATERS

As previously mentioned, to gather trajectory information at a wide range of time scales spanning six orders of magnitude we wrote our system trajectories to disk frequently—every 10 Monte Carlo (MC) sweeps—and collected data for the far longer period of 100 million MC sweeps. We produced trajectories as large as 3.25 TB per simulation, each containing about 10 million simulation frames. We were able to store these extraordinarily large trajectories within our 500 TB allocation on Blue Waters' generous file system; this was essential for our research and analysis.

PUBLICATIONS & DATA SETS:

Teich, E.G., G. van Anders, and S.C. Glotzer, Identity crisis in alchemical space drives the entropic colloidal glass transition. Under review (2018).

Erin Teich is a sixth-year PhD student in applied physics. She is working under the supervision of Sharon C. Glotzer at the University of Michigan and planned to graduate in August 2018.