



Dependence of local order on cooling rate investigated in a colloidal glass



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ABSTRACT

The cooling rate with which liquid is cooled tremendously affects the properties of amorphous solids, but little information on underlying mechanism for this dependence is available, mainly due to the lack of clear characterization on microstructural variation. We built a colloidal glass to directly investigate the effect of cooling rate on local order. By quantifying effective cooling rate and structural properties, we show how local order changes with cooling rate. Furthermore we demonstrate that free-volume distribution results in local-order evolution.

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1. Introduction

An amorphous solid is normally formed by supercooling a viscous liquid fast enough to avoid crystallization [1]. The transition from a liquid state to an amorphous solid state is essentially kinetic, resulting from a “falling out of equilibrium” of the liquid as the structural relaxation time increases with decreasing temperature and exceeds the time scale of the experiment [2]. Thus the resulting amorphous solid is not in thermal equilibrium and its properties strongly depend on the thermal history, such as the cooling rate with which the liquid was cooled [3–5]. Such dependencies exist indeed has been shown in various experimental and computer simulation results in which the thermal, dynamic, and mechanical properties of amorphous solids are found to be sensitive to their cooling rates [6–11]. Although much is known about the effects of cooling rate on the macroscopic properties of amorphous solids, little information on the underlying mechanism for this link is available. This is mainly due to the lack of clear characterization on the microstructural variation induced by different cooling rates, since the properties of amorphous solids are essentially related to their microstructures [12,13]. Thus, figuring out the difference in microstructures of amorphous solids produced with different cooling rates is the key to understanding the dependencies of properties on the cooling rate. However, it is prohibitively difficult to directly investigate the microstructural changes induced by varied cooling rates within atomic and molecular solids. This is because atoms are too small and move too fast to visualize directly. Although significant progress has been achieved in microscopy techniques, it is still a huge challenge to directly explore the microstructure of amorphous solids with disordered atomic packing.

To study the atomic-scale problems that cannot be directly approached in atomic and molecular glasses, an effective way is to use large physical spheres to construct a structure similar to a glass, in which the glassy structure can be seen in real space. This approach was pioneered by Bernal and Mason [14], who used steel ball bearings to build a random packed structure and identified the structural commonalities of disordered materials based on this system. In addition, Argon and Kuo [15] have successfully used disordered bubble raft as an analog of metallic glass to investigate the plastic flow of atoms. Recently, with the development of confocal microscopy, colloids have increasingly been used to mimic many aspects of atomic and molecular systems [16–20]. Colloidal particles can be tracked spatially in three dimensions with great precision over large time scales. This is because the larger size and concomitant slower time scale of colloidal particles make them much more experimentally accessible. Colloidal glass of micrometer-sized spherical particles, perhaps the simplest system with a glass transition [21], can serve as an excellent experimental model to explore the problems of amorphous materials [22–26]. Inspired by these works, we have now followed the formation process of a colloidal glass by using a high-speed confocal microscopy to directly study how cooling rate influences the microstructures. We determine the positions of colloidal particles in three dimensions accurately by the subsequent image analysis, which can provide an accurate picture of three-dimensional configuration of colloidal glass [21].

2. Experimental methods

We used 1.55- μm diameter colloidal silica particles with a polydispersity smaller than 3.5% to prepare a colloidal system with more than 4×10^9 colloidal particles. The silica particles were suspended in a mixture of deionized water and dimethyl sulfoxide. To make the particles

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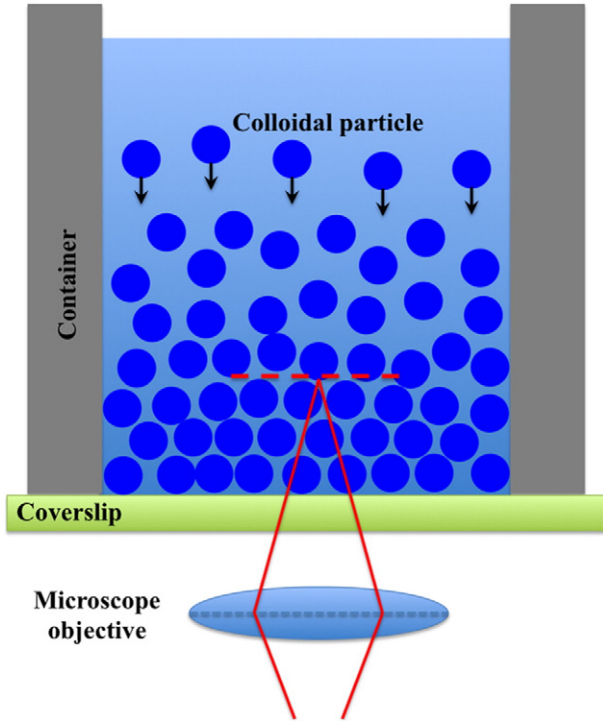


Fig. 1. Schematic showing the experimental set-up with colloidal glass.

appear as dark spots on a bright background under fluorescence microscopy, we dyed them with fluorescein-NaOH solution [27]. The schematic of the sample cell is shown in Fig. 1. Due to the density difference between colloidal particles and their surrounding fluid, the colloidal structures were constructed by sedimentation under gravity. By using a high-speed confocal microscope, we acquired three-dimensional scans of our sample yielding a $77 \times 77 \times 23 \mu\text{m}^3$ observation volume for each image stack. Each image stack takes 150 s. We identified particle positions in 3D with a horizontal accuracy of $0.03 \mu\text{m}$ and a vertical accuracy of $0.05 \mu\text{m}$ [22].

3. Results and discussion

In hard-sphere system, the viscosity approaching the glass transition varies with volume fraction can be described as $\eta = \eta_0 \exp[\nu\phi/(\phi_0 - \phi)]$ [28]. Correspondingly, the viscosity of a molecular liquid approaching the glass transition varies with temperature (T) can

be described by Vogel–Fulcher–Tammann (VFT) equation, $\eta = \eta_0 \exp[DT_0/(T - T_0)]$ [29]. The above two equations are similar in form, suggesting that ϕ plays the similar role in hard sphere system as T in usual liquids [30]. In other words, the control parameter of hard sphere system is ϕ rather than T , and the effective temperature of hard sphere system is $T_{eff} = 1/\phi$ [31]. Then the effective cooling rate (\dot{T}_{eff}) of the hard sphere system can be expressed as

$$\dot{T}_{eff} = \frac{1/\phi_1 - 1/\phi_2}{\Delta t} \quad (1)$$

where ϕ_1 and ϕ_2 are the volume fractions of colloidal system at time t_1 and t_2 , respectively; $\Delta t = t_2 - t_1$ is the time interval between t_1 and t_2 . As described above, we built the colloidal glass by sedimentation under gravity. Then along the height of the colloidal glass, there may be \dot{T}_{eff} difference. To check this assumption, we calculated the \dot{T}_{eff} at different heights. Fig. 2 shows a typical process of the sedimentation of the colloidal glass. Fig. 2 panels (a) and (b) are two reconstruct colloidal structures in $3\text{-}\mu\text{m}$ -thick x - z section centered at $y = 10 \mu\text{m}$ at $t = 0 \text{ s}$ and 1650 s , respectively. Along the z direction, the packing densities of the colloidal system vary a lot at the initial time ($t = 0 \text{ s}$), but no apparent difference can be detected after 1650 s , strongly suggesting that \dot{T}_{eff} is different with the height of colloidal glass. To determine the \dot{T}_{eff} more precisely, we measure the mean ϕ of colloidal system in $3\text{-}\mu\text{m}$ -thick x - y section centered at different height. We determine the ϕ from the relationship $\phi = nV_0$, where n is the number density of particles and $V_0 = 4\pi R_0^3/3$ is the volume of colloidal particle with radius R_0 . As shown in Fig. 2(c), the mean ϕ values of glass sections at different height are all increased with time firstly. After 1650 s , the mean ϕ values constant at about 0.60 , well into the colloidal glassy state [31]. We then chose the mean ϕ values at 0 s and 1650 s as ϕ_1 and ϕ_2 , respectively. Then the \dot{T}_{eff} of glass sections at different height calculated according to Eq. (1) are listed in Fig. 2(c). Clearly, the \dot{T}_{eff} gradually increases with the height increasing, from $0.76 \times 10^{-4} \text{ s}^{-1}$ to $8.72 \times 10^{-4} \text{ s}^{-1}$. This gradual variation of \dot{T}_{eff} along the height of colloidal glass offers us a good chance to study the effect of cooling rate on the microstructure.

We focus on the microstructure of the colloidal system at $t = 1650 \text{ s}$, when the stable colloidal glass just forms through verification and its microstructure may be affected by \dot{T}_{eff} more evidently. We calculate the RDF of colloidal glass ($t = 1650 \text{ s}$) in $3\text{-}\mu\text{m}$ -thick x - y section centered at different heights. Fig. 3(a) shows the overall RDF of the colloidal glass at various heights. Fig. 3(b) and (c) is the enlargement of the first and second peaks of the overall RDF. From Fig. 3(b) we can note that, with z moving from the top to the bottom part of colloidal glass, the

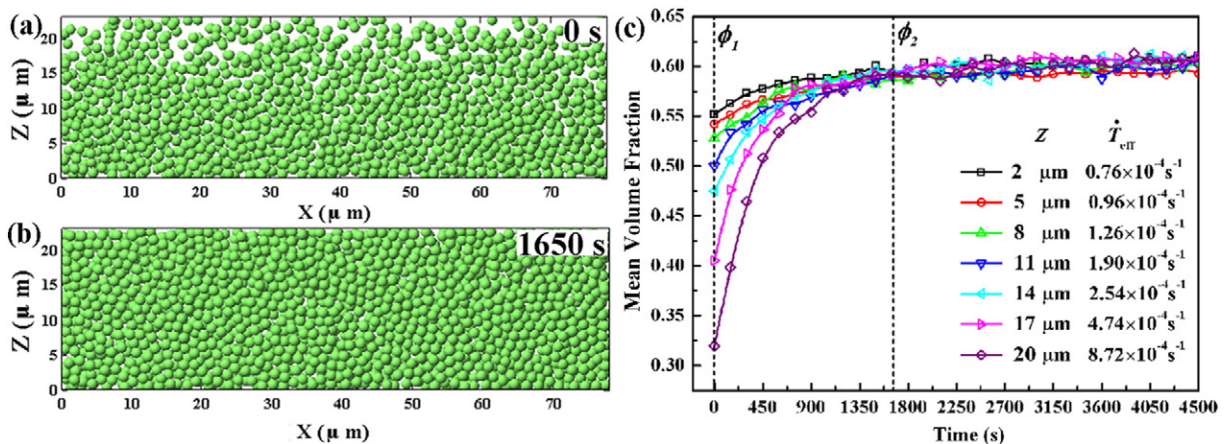


Fig. 2. (a) and (b) are $3\text{-}\mu\text{m}$ -thick x - z sections centered at $y = 10 \mu\text{m}$ at time (a) $t = 0 \text{ s}$ and (b) $t = 1650 \text{ s}$. (c) The mean ϕ of colloidal system in $3\text{-}\mu\text{m}$ -thick x - y section centered at $z = 2 \mu\text{m}, 5 \mu\text{m}, 8 \mu\text{m}, 11 \mu\text{m}, 14 \mu\text{m}, 17 \mu\text{m},$ and $20 \mu\text{m}$ at different times. The \dot{T}_{eff} of different heights calculated according to Eq. (1) is also listed.

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