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Structural and Dynamic Characteristics of Densified Silica Glass: A Computational Analysis

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ABSTRACT

One significant and difficult problem in condensed-matter physics is the pressure-induced alteration of the structure and kinetic characteristics of noncrystalline materials, such as liquids and glasses. The microscopic image of changes in the structural and dynamic properties of densified silica glass was examined in this study using molecular dynamics (MD). Based on the effective interatomic potential, the simulations are performed. As a function of density, changes in the pair distribution function's first sharp peak's height and location as well as the liquid's dynamics were examined using the mean squared displacement (MSD) calculation. The average Si-O bond length at normal density is 1.62 Å. According to the pair-distribution function, and at high density it grows linearly to 1.67 Å. where the glass meets the stishovite at a high density. The MSD for SiO_2 glass, or the species that moves the slowest, illustrates the behavior of the liquids. temperatures at which the samples in the NPT ensemble are in equilibrium. The MSD exhibits a plateau at intermediate times, indicating that the dynamics are already somewhat glassy at these temperatures. The system has equilibrated and the particles have transitioned into the diffusive phase when the MSD is a linear function of time. These findings clearly show that the system diffusively outperforms the high density ($4.2 g/cm^3$) at normal density ($2.2 g/cm^3$).

Keywords:
Silica glass,
Molecular dynamics
Simulation,
High Pressure,
Oxide glass

INTRODUCTION

A common material with significant uses in geology, chemistry, and industry is silica (SiO2). The essential component of the three-dimensional framework structure present in minerals is this. The structures under ambient conditions are based on a comparatively open configuration of tetrahedral units that share corners and have a central silicon atom surrounded by four oxygen atoms (Du et al., 2022). Due to its numerous polymorphs, silica (SiO2) is especially intriguing. These include coesite, stishovite, cristobalite (cubic), quartz (trigonal), and hexagonal (quantz) (Brazhkin et al., 2011; Klinger, 2013; Machon et al., 2014).

In its crystalline state, silica can take on a range of polymorphic shapes and densities. According to several theoretical and experimental investigations (Grimsditch, 1986; Hong & Newville, 2020; Kapoor et al., 2017; Polian & Grimsditch, 1990), SiO2 experiences a variety of structural changes as a function of temperature and pressure. Low-temperature quartz (α-quartz), for instance, structurally changes into high β-quantz. Quartz transforms into coesite at room temperature when the pressure is slightly increased.

As the pressure increases more, coesite changes into stishovite. Unlike crystal transitions, the structural order of amorphous SiO_2 can be separated into interatomic distance between silicon and surrounding oxygen atoms, or intermediate-range order (IRO), and short-range order (SRO) in the glass.

This is in contrast to transitions in crystals and occurs when the interatomic distance between silicon and surrounding oxygen atoms changes. Corner-sharing $Si(O_{1/2})_4$ tetrahedral in normal density ($2.2 g/cm^3$) is a well-known property of silica glass under ambient circumstances.

Under pressure, the network structure of SiO_2 glass changes significantly. Several experimental techniques such as Brillioun (Grimsditch, 1986), Raman (Hemley *et al.*, 1986), infrared (Williams & Jeanloz, 1988), neutron and x-ray scattering (Meade *et al.*, 1992; Susman *et al.*, 1991), have been used to investigate the influence of pressure on silica glass. Results obtained reveal an irreversible densified structure in glass samples recovered after being subjected to a pressure of 17 GPa.

On the modelling side, atomistic MD simulations demonstrated the existence of density maximum (Jin, 1995; Jin, *et al.*, 1992; Kien, *et al.*, 2020; Wu, *et al.*, 2012). Molecular dynamics (MD) simulation is a computational technique used to study the behavior of

atoms and molecules over time (Rapaport, 2004). Moreover, studying a material's microscopic structure can help anticipate its macroscopically. Simulation techniques offer this capability. Material physical attributes can be used to model and simulate materials at microscales, allowing for testing and observation of the materials' macro-physical characteristics (Fitriana, Iwan, & Ernik, 2019). We can more easily study the relationship between the interaction and the macroscopic properties by examining the numerical integration algorithm in molecular dynamics simulation. This allows us to analyze the microstructure, particle motion, and the macroscopic relationship between the material and the particles (Jundong, 2017). Numerous studies on silica glass have made extensive use of it due to its advantages for comprehending atomic structure and mechanical characteristics. Using MD modeling, Murakami et al. (Murakami et al., 2019) recently determined the static structural factor and pair distribution of SiO2 glass exposed to pressure. The First Sharp Diffraction Peak (FSDP) shifts to higher values of charges q and decreases in height as pressure increases. The initial acute FSDP almost disappears at 42 GPa. At 28 GPa of pressure, a peak may be seen at 3.18Å.

The structural changes in densified silica glass were examined in this work using MD simulations. The BKS potential was chosen because, in comparison to experiments, it could replicate a number of silica glass structural changes at high temperatures and at manageable densities. At different densities, the pair distribution function was computed. The bond angle distribution was then examined at each location.

Lastly, we go over the main structural process that works at high densities.

MATERIALS AND METHODS

Since the study is simulation therefore the following software's were used. LAMMPS package version 2022 as one of the materials in this study used as a software in simulating the system, Ovito 1.9 also known as Open Visualization Tool is a scientific software for atomistic and particle simulation data and JMP Pro13 is also a software for plotting graph in this study.

Details of the Computation

Interatomic Potential and Simulation Procedure

A potential that can accurately describe the geometry is required in order to run a molecular dynamics simulation. The popular BKS (van Beest, Kramer, and van Santen) potential has been shown to accurately characterize amorphous silica in the context of SiO_2 glass. The BKS potential in this study was chosen to help the researchers to gain additional insight into the reactions of silica glass to mechanical and thermal processes, such as compression and thermal expansion. It has also been applied to study phase transitions and structural changes that occur in densified silica glass under extreme conditions. According to van Beest et al. (1990), the BKS potentials s follows:

$$V(r_{ij}) = \frac{q_i q_j e^2}{4\pi\varepsilon_o r_{ij}} + A_{ij} e^{-rij/Bij} - \frac{C_{ij}}{r^6 ij} \quad (1)$$

where qi represents the partial charge of atom i and r_{ij} denotes the distance between atoms i and j. Shown are A_{ij}, Bij, and C_{ij}. Equation 2.6 is the product of a Buckingham term and a long-range Coulombic term. The Coulomb component means that these potentials must be evaluated using methods such as the Ewald summation, which makes applying them in large systems more time-consuming (Allen and Tildesley, 2017). Even though the simulations are running at a relatively sluggish pace, the algorithm was written on large scale LAMMPS, which uses reciprocal space fixes to compute the long-range interactions. A popular program for simulating molecular dynamics is called LAMMPS. The acronym LAMMPS stands for Large Scale Atomic/Molecular Massively Parallel Simulator. Newton's equations of motion are integrated into a piece of software known as LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator) for an interacting group of atoms. The program provides some details on the interatomic potentials of different kinds of materials to explain this interaction. For computational performance, LAMMPS uses neighbor lists to keep track of nearby particles (Bridgman & Simon, 2021). One possibility to avoid this problem is to use the Wolf summation method proposed by Wolf et al. (Wolf, *et al.*, 1999), in which the Coulomb term is replaced by:

$$\frac{qiqje^2}{4\pi\varepsilon_{or_{ij}}} \rightarrow \left\{ \frac{qiqje^2}{4\pi\varepsilon_o} \left[\left(\frac{1}{r_{ij}} - \frac{1}{r_c} \right) + \frac{r_{ij} - r_c}{r^2 c} \right] r < r_c \right\} \quad (2)$$

where the cutoff distance for Coulomb interactions is denoted by rc. By smoothly truncating the potential, the effective long-range potential in this form becomes short ranged and, hence, significantly more computationally efficient. Hartree-Fock computations of a single SiO4 tetrahedral, charge saturated by four hydrogen atoms, served as the basis for the BKS model's parameterization.

Simulation Procedure

The simulations were carried out in a (N, Ω, E) ensembles for 3000 atoms (1000 silicon and 2000 oxygen) in a cubic box of volume $\Omega = L^3$. For the normal density system, the lentgh of the MD cell (L) is chosen to be 25.391Å so that the mass density ρ_o is 2.20g/cm^3. Systems with cell length 20.122, 18.434, 18.012, 17.023 Å were also simulated. These correspond to mass densities of 2.64, 2.89, 3.56 and 4.28g/cm^3 respectively.

The melt-quench process produced the glass structure. A cubic box containing 2000 oxygen atoms and 1000 silicon atoms was the initial random configuration used in the simulation. The box's dimensions are set to

correspond with the glass's physical density. After heating the system to 8000 K in an NVT (constant number of atoms, volume, and temperature) ensemble and allowing it to stabilize for 100 ps, the sample was cooled to 300 K in an NPT (constant number of atoms, pressure, and temperature) ensemble using a temperature step of 2000 K. To relieve the thermal stress, the quenched sample was relaxed for 150 ps at 300 K and 0 bar. According to previous studies (Vollmayr, Kob, & Binder, 1996), the cooling rate had a great influence on the structure properties and a slow cooling rate was considered to be a better option. Therefore, the coolingrate was set to be 0.2 K/ps in this study. The pair distribution function (PDF) and the bond angle distributions (BAD) were calculated after running for six picoseconds at 293K and were accumulated every ten timesteps for three hundred timesteps.

RESULTS AND DISCUSSION

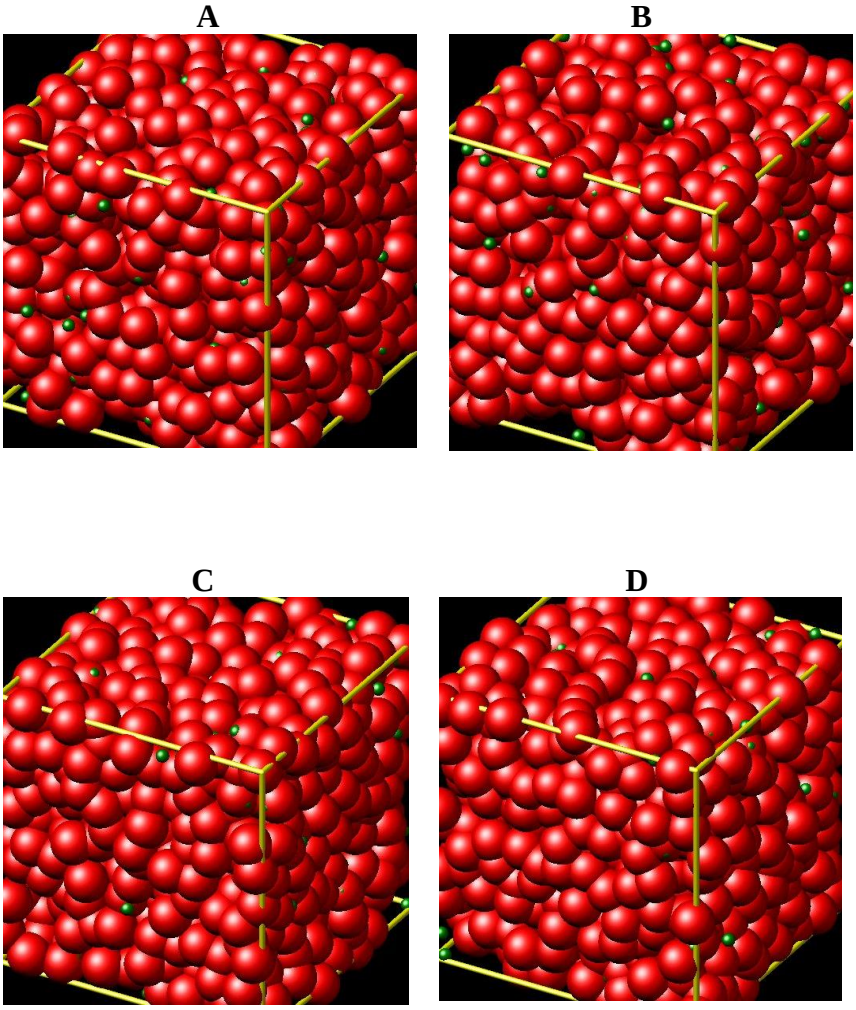

Figure 1(a, b, c and d): Structural evolution images of SiO_2 glass at (A) initial configuration, (B) melting, (C) quenching and (D) relaxation steps of simulation

Once the system (SiO_2) reached its initial equilibrium temperature of 4700KK, it was quenched down to 300K to chill and relax the system.

Three thousand atoms were housed in a cubic box at the start of the simulation, when the initial configuration was established. (Two thousand oxygen, red, and one thousand silicon, green). plate 4.1a above illustrate that. At the initial state, SiO_2 can be considered to be in its amorphous (glassy) form, where silicon (Si) and oxygen (O) atoms are arranged randomly in a disordered fashion, lacking a regular crystalline lattice structure.

Plate 4.1b demonstrates that SiO_2 is significantly over its melting point when atoms in a box are heated to 4700K in NVT. SiO_2 loses its structural integrity as it goes through a phase change from a solid to a liquid at such high temperatures. The amorphous SiO_2 undergoes a breakup of its Si-O bonds, resulting in a highly

disordered liquid with extremely energetic atoms. Plate 4.1c provided additional explanation on the quenching process. Rapid cooling occurs during quenching, which occurs from a molten state at 4700 K to 300 K. The SiO_2 is unable to reorganize into its crystalline structure due to the quick cooling rate. Rather, it becomes "frozen" in an amorphous state, effectively maintaining its amorphous, disordered form. At 2000 K, the amorphous SiO_2 maintains its chaotic atomic configuration from its liquid state.

The relaxation at 300 K (room temperature) and 0 bar (standard atmospheric pressure), as shown in plate 4.1d above figure 4.1d, indicates that the SiO_2 is now in a solid state but has the potential to progressively modify its structure over time. At this point, the atoms in the amorphous SiO_2 may rearrange to form a more stable, lower-energy state through a process known as gradual structural relaxation.

The time, temperature, and any impurities or flaws in the amorphous SiO_2 will all affect the degree and pace of relaxation. To sum up, an amorphous glassy structure is formed when SiO_2 is heated to 4700K and subsequently cooled to 300 K. The material can then be relaxed to 300 K and 0 bar, which allows it to progressively change its atomic arrangement, though it is most likely going to stay amorphous. The particular temperature history and conditions that were applied to SiO_2 resulted in this final structure, which will preserve some of the disorder and unpredictability of the initial glassy state.

Figure 2 displays the pair-distribution functions, g(r), for SiO2 glasses at high densities of 3.5 and 4.28 g/cm3 as well as conventional densities of 2.2 g/cm3. The average bond distances between Si and O, Si and Si, and O and O are represented by the first, second, and third peaks of g(r), respectively. The first peak in g(r)si-o for normal density glass indicates that the Si-O bond length is 1.61 Å. The range of g(r)si-o is 1.80 to 2.98 A, with a large peak at 4.16 Å and a tiny shoulder at 3.80 Å. Up to 3.53 g/cm3, the location of the first peak in g(r)si-o remains essentially constant as density increases. Nevertheless, the second peak moves to 3.98 Å at 3.53 g/cm3, and the gap in g(r)si-o narrows as density rises. The first peak in g(r)si-o occurs at 1.68 Å rather than 1.61 Å at a pressure of 43 GPa, where the glass density (4.28 g/cm3) reaches the density of stishovite. The Si-O bond lengths in crystalline stishovite are 1.76 and 1.81 Å. The second peak in g(r)sio in the glass at 4.2 g/cm3 is located at 3.15 Å, which is near the Si-O distance in stishovite that is the next closest neighbor (3.20 Å).

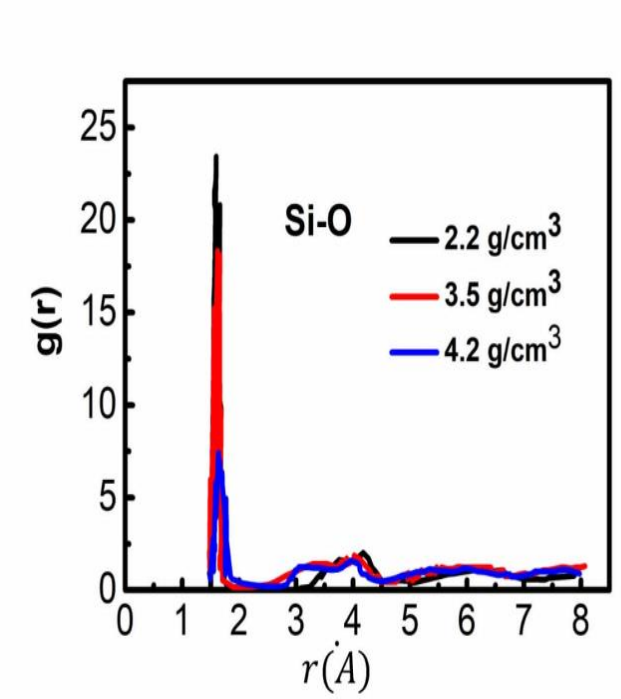

Figure 2: Si-O pair-distribution functions for SiO2 glasses at normal and high densities at 300 K. Sharp peaks in the figure at 4.2 g/cm3 correspond to pair-distribution functions for stishovite.

The Si-Si and O-O pair distribution functions' changes with densification are depicted in Figure 3(a,b). Si-Si and O-O distances for the normal density glass are 3.09 and 2.65 Å, respectively, based on the first peaks in g(r)si-si and g(r)o-o. The Si-Si and O-O lengths are 5.03 and 5.06 Å, respectively, based on the second broad peaks in g(r)si-si and g(r)oo.

The first peak in g(r)si-si separates into two peaks when the glass density rises to 4.28 g/cm3. In the stishovite, one of these peaks is situated at 2.59 Å, not far from the Si-Si distance (2.67 Å). In the stishovite, the second peak is located at 3.07 Å, which is near the Si-Si distance (3.24 Å). at a typical density. At 3.53 g/cm3, it rises to 10, and at 4.28 g/cm3, it reaches 12.

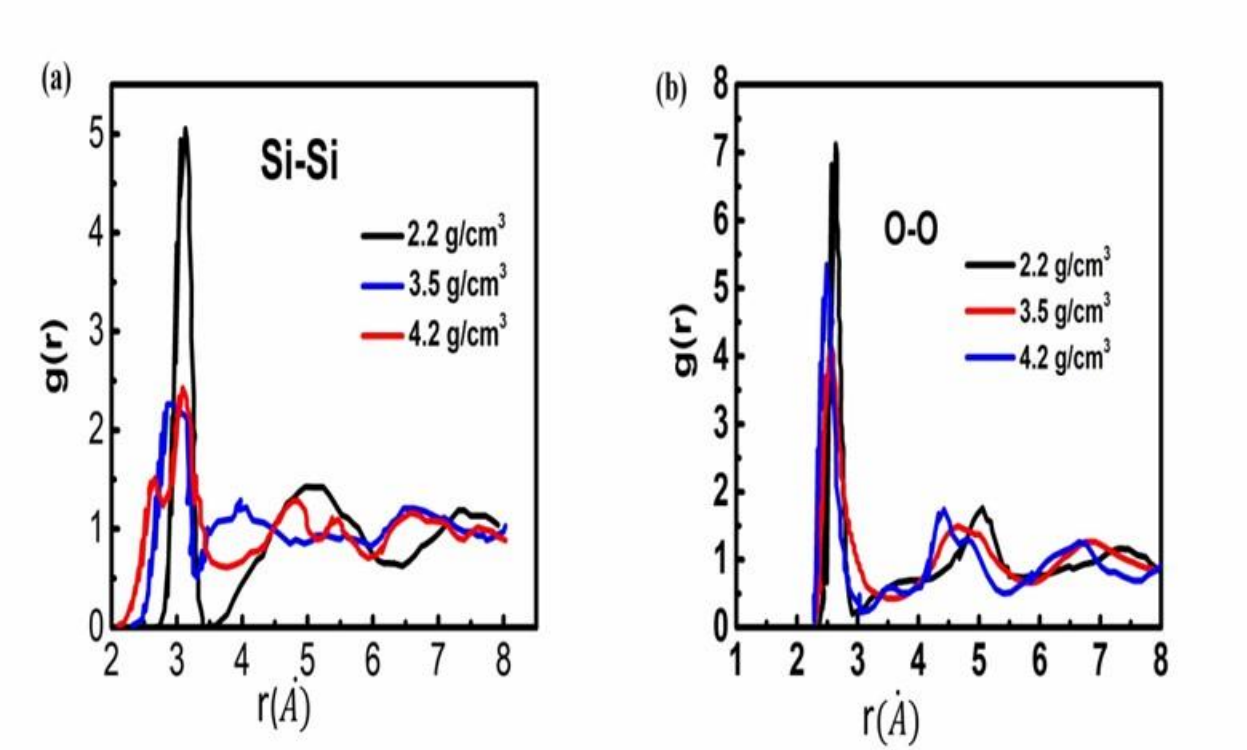

Figure 3: Si-Si and O-O pair-distribution functions for SiO_2 glasses at normal and high densities at 300 K.

The dynamics of the liquid was investigated by calculating the mean squared displacement (MSD).

Fig. 3 uses the species that moves the slowest, or the MSD for SiO_2 glass, to show how the liquids behave. These are the temperatures at which the NPT ensemble's samples reach equilibrium. At these temperatures, it is clear that the dynamics are already a little glassy because the MSD has a plateau at intermediate times. The graph also shows that, over long times, the MSD is a linear function of time, indicating that the system is equilibrated and that the particles have entered the diffusive phase. We find that, in agreement with previous findings, the diffusion constant and the MSD at long durations show a strong dependence on the potential. Since it can be expected that the activation energy for the diffusion constant also depends on the potential considered, the diffusion constant at lower temperatures will differ even more.

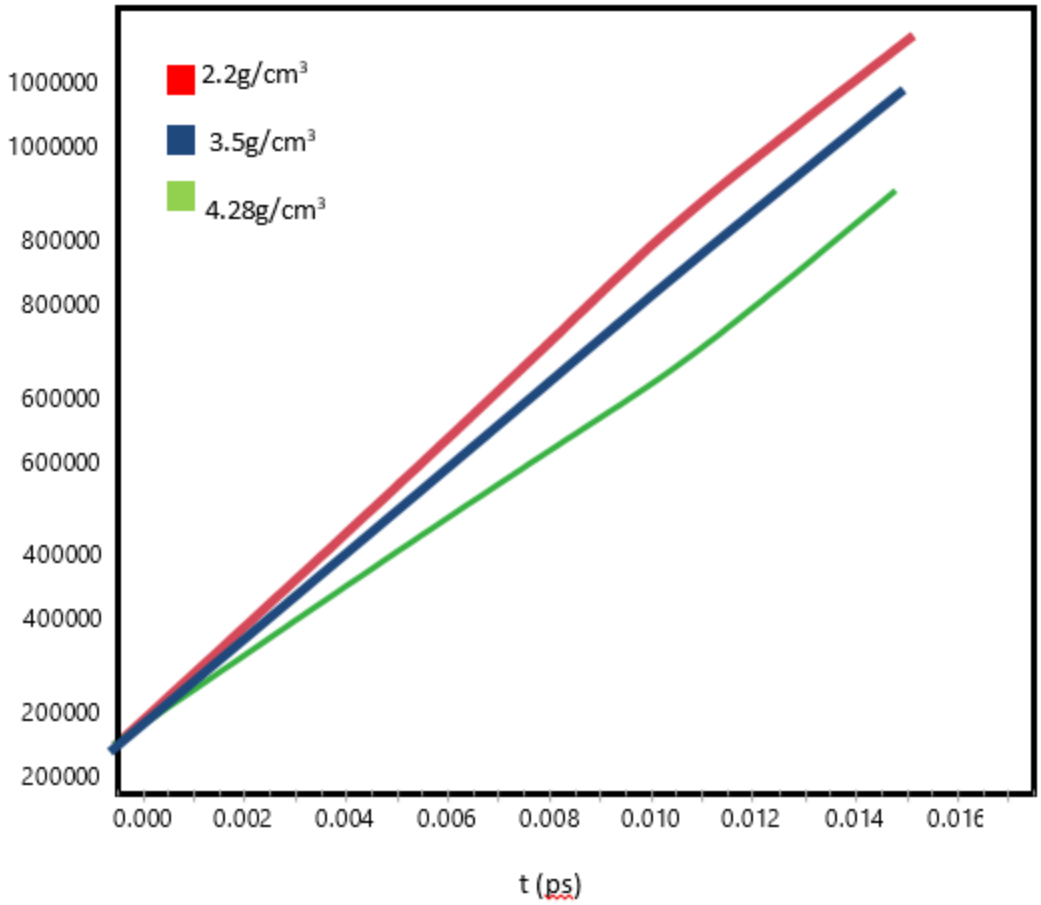

Figure 4: Graph of mean square displacement.

CONCLUSION

Molecular dynamic simulations were used to simulate the materials (SiO_2), and simulated results were extracted. In structural evolution heating SiO_2 to 4700K and finally cooling it to 300 K, which creates an amorphous glassy structure. Subsequently, relaxing the material at 300 K and 0 bar allows it to gradually adjust its atomic arrangement, but it is likely to remain in an amorphous state. This final structure is a product of the specific thermal history and conditions applied to SiO_2, and it will retain some of the disorder and randomness of the initial glassy state.

The microscopic structures in densified SiO_2 glasses were investigated using molecular dynamics simulations. The results of pair distribution functions and dynamic properties show that up to 60% densification the $Si(O_{1/2})_4$ tetrahedral structural unit remains intact and structural change is associated with bending and distortion of the tetrahedral. The bond length increases from 1.61 to 1.67Å at high pressure, where the glass density reaches the stishovite density. The first three peaks of the total PDF represent the Si-O, O-O, and Si-Si bond lengths, respectively. The O-O, Si-Si, and Si-O partial pair distribution functions' bond lengths in bulk silica glass structures were calculated using the PDFs from this study, and the results showed good agreement with previous MD simulations as well as observations. Because the silica tetrahedron is so stable, it has been found that Si-O bond lengths do not change as structural deformation increases, but the Si-Si and O-O partial pair distributions do tighten as the silica undergoes permanent densification.

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