Beyond sixfold coordinated Si in SiO$_2$ glass at ultrahigh pressures

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We investigated the structure of SiO$_2$ glass up to 172 GPa using high-energy X-ray diffraction. The combination of a multichannel collimator with diamond anvil cells enabled the measurement of structural changes in silica glass with total X-ray diffraction to previously unachievable pressures. We show that SiO$_2$ first undergoes a change in Si–O coordination number from fourfold to sixfold between 15 and 50 GPa, in agreement with previous investigations. Above 50 GPa, the estimated coordination number continuously increases from 6 to 6.8 at 172 GPa. Si–O bond length shows first an increase due to the fourfold to sixfold coordination change and then a smaller linear decrease up to 172 GPa. We reconcile the changes in relation to the oxygen-packing fraction (OPF) by calculating FSDP positions showing that oxygen packing decreases at ultrahigh pressures to accommodate the higher than sixfold Si–O coordination. These results give experimental insight into the structural changes of silicate glasses as analogue materials for silicate melts at ultrahigh pressures.

The structure of silicate melts and glasses at high pressure is of great interest not only in physics, geophysics, and materials science, but also in engineering and industry. Their physical properties at high pressures and temperatures have fundamental influences on present-day magmatic processes and the evolution of the early Earth (1, 2). Seismological heterogeneities at the top of the transition zone (3, 4) and at the core–mantle boundary (5) have been interpreted with the potential presence of silicate melts at very high pressures in the Earth’s interior. At ambient conditions, polymerized silicate melts and glasses are characterized mainly by networks of Si–O tetrahedra (6). Experimental investigations (7–10) and molecular dynamics (MD) simulations (11, 12) have shown that Si–O coordination number (CN) increases from fourfold to sixfold between 15 and 40 GPa, resulting in a network of Si–O octahedra (13). At pressures >40 GPa, MD simulations indicate a continuous increase in Si–O CN in SiO$_2$ glass (11) and SiO$_2$ melt (12), whereas existing experimental data on SiO$_2$ glass indicate a plateau of sixfold coordinated Si–O up to 100 GPa from total X-ray diffraction analysis (9) and up to 140 GPa inferred from shear soundwave velocities (14).

A recent compilation of glass and melt structure data (15) found that evolution of network-forming structural motifs can be rationalized in terms of oxygen-packing fraction (OPF). A plateau region for fourfold coordinated A atoms (A = Si, Ge, Al, etc.) is found for an OPF of 0.4–0.6. The transformation to sixfold coordination occurs rapidly as OPF approaches 0.64. The highest experimental CN measured for SiO$_2$ so far is 6 at 100 GPa (9), indicating another plateau for sixfold CN. However, recent data for GeO$_2$ glass, which serves as a structural analogue for SiO$_2$ glass, show a CN larger than sixfold, up to 7.4 at 92 GPa (16). These data follow the trend seen from MD simulations, where it has been shown that CN continuously increases with pressure after the relatively sharp fourfold to sixfold transition has taken place.

We have conducted measurements of the SiO$_2$ glass structure factors up to 172 GPa using angular dispersive X-ray diffraction using a combination of diamond anvil cells (DACs) with a multichannel collimator (MCC) (17). The MCC, which was originally developed for Paris–Edinburgh Presses, effectively reduces the background scattering of the surrounding DAC, thus enabling total X-ray diffraction up to very high pressures and extending the pressure range for total X-ray diffraction with an angle dispersive setup by more than a factor of 3 (previously 50–60 GPa; e.g., ref. 7) for relatively low Z elements.

Selected structure factors $S(Q)$ and pair distribution functions $g(r)$ of the SiO$_2$ glass up to 172 GPa are shown in Fig. 1. The $S(Q)$ shows strong changes up to 33 GPa, whereby the main new feature is a peak rising between 2 and 3 Å$^{-1}$ and a decrease in the intensity of the first sharp diffraction peak (FSDP). From 33 to 172 GPa, the $S(Q)$ changes very smoothly with no apparent new features.

Figs. 2–4 show the pressure dependence of the FSDP position and the Si–O CN and Si–O distances, respectively. The FSDP position increases almost linearly with pressure up to ~20 GPa, followed by a gradual curvature to another linear regime with a much smaller slope from 50 to 172 GPa. Our FSDP positions are in good agreement with refs. 8, 9, and 18, whereas refs. 7 and 19 reported slightly lower values at >25 GPa. The data used in refs. 19 and 9 are the same, and the discrepancy can probably be explained by different methods for extracting FSDP positions. The large change in FSDP positions at low pressures can be associated with a strong change in intermediate-range order. The accompanied drop in intensity also indicates a large decrease in void space in the structure (20) and ring closure (21), which is

Significance

The combination of multichannel collimator and diamond anvil cells enabled the investigation of the real-space structure of an amorphous material >100 GPa. We have measured the structure of SiO$_2$ glass by angle-dispersive X-ray diffraction up to 172 GPa. Our results are in agreement with existing data up to 50 GPa showing a sharp change from fourfold to sixfold Si–O coordination number (CN). However, at higher pressures, CN continuously increases to values beyond 6 without sharp structural changes. The behavior of SiO$_2$ glass at high pressure serves as a model for more complex silicate glasses and melts. Thus, our results provide experimental insight into the structural evolution of silicate glasses and melts at ultrahigh pressures.

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supported by the rapid increase in density in this pressure region (10).

The Si–O CN (Fig. 3) remains constant at ∼4 up to 14 GPa and then rose sharply to ∼6 at 40 GPa, which is in agreement with previous experimental studies (7, 9, 21). The MD simulation data of ref. 11 shows an increase to higher CN at lower pressures. Above 50 GPa, our Si–O CN linearly increases with pressure to ∼7 at 172 GPa. Previous estimates reported a constant sixfold Si–O CN from 50 to 100 GPa (9). While our data are in agreement within the uncertainties of the data of ref. 9, our data, which cover a much greater pressure range, show a smooth linear increase (Fig. 3). A linear increase of Si–O CN after the fourfold to sixfold crossover is also observed in MD simulations of SiO$_2$ glass (11) and ab initio MD simulations of SiO$_2$ melt (12), although the absolute values are shifted. Ab initio MD simulations of SiO$_2$ glass (22, 23), on the contrary, show only a very slow increase of CN >6 at pressures >100 GPa. Also, the fourfold to sixfold crossover seems to be completed at higher pressures, at ∼70–90 GPa. This indicates that there is a general shift in cross-over pressures, which might be induced by the small size of the simulation cell, which is not giving enough statistical information. Thus, SiO$_2$ glass shows a similar behavior as GeO$_2$ glass, where a larger than sixfold CN has been reported (16).

The Si–O distance (Fig. 4) first remains constant at ∼1.62 Å up to 10 GPa and then almost linearly increases to ∼1.69 Å between 10 and 40 GPa. Above 40 GPa, the Si–O distance shows a linear decrease to 1.65 at 172 GPa. This behavior can be rationalized by means of two mechanisms, the effect of compression and change in CN. Below 10 GPa, the compression mechanism for SiO$_2$ glass is mostly due to collapsing void space, while the Si–O distance remains constant. The increasing distance between 10 and 40 GPa is due to the increase from fourfold to sixfold coordination characterized by longer Si–O bond distances. Above 40 GPa, the compression effect on the Si–O distance is larger than the small continuous increase in CN, resulting in a decreasing Si–O distance from 40 to 172 GPa. Below 30 GPa, the data are in good agreement with ref. 9, whereas >30 GPa, our Si–O bond distances were slightly lower. However, there was a considerable disagreement with the data of refs. 7 and 21. The discrepancy in Si–O distances between refs. 21 and 7 has been attributed to radiation-induced annealing (21) in the study of ref. 9 due to the use of white-beam, energy-dispersive X-ray diffraction. However, our data were also collected with monochromatic X-ray diffraction, as in ref. 7, and also collection times were relatively short. Therefore, this mechanism is not likely to be the main cause. Another possible factor might
be different degrees of anhydrostaticity. Sato et al. (24) have shown that differential stress results in large differential strain in SiO2 glass <20 GPa, whereas the strain decreases with higher pressures.

Our data show that the structure of SiO2 continuously changes with pressure from 50 up to 172 GPa. We do not see evidence for a stable plateau with a CN of 6 at higher pressures as reported for SiO2 glass (9) and GeO2 glass (16), but, rather, a continuous increase in CN up to ~7 at 172 GPa. Furthermore, we do not observe first-order structural changes at 140 GPa, as predicted from soundwave velocity measurements of SiO2 glass (14). If existent, the origin of those sound speed changes might lie in other mechanisms or be caused by the different experimental approaches. For example, it might be specific to the laser interaction with the glass and the very long collection time needed for a Brillouin spectroscopy measurement >140 GPa, resulting in possible structural changes and/or densification.

In comparison with the glass at high pressure, crystalline SiO2 undergoes a succession of phase transformations with increasing pressure, depending on the state of the starting material (25). Experimental investigations have shown that, for example, α-quartz transforms to coesite at ~2.5 GPa, to a rutile-type structure (stishovite) at ~10 GPa, to a CaCl2-type structure at ~60 GPa, to α-PbO2 structure (seifertite) at 120 GPa, and a polymorph with Pa3 space group >270 GPa, which has been suggested to be a pyrite like structure due to the same space group (26). Many of those phase transformations are not readily occurring at room temperature and need heating at the relevant pressures to overcome kinetic barriers. Room temperature compression of different SiO2 starting materials has been shown to lead to a variety of other metastable polymorphs (25, 27) or even amorphization at high pressures (28).

Beyond current experimental achievable pressure, a cotunnite SiO2 structure has been predicted at >750 GPa (29). However, a later detailed search for different structure types at ultra-high pressures has found the Fe2P-type structure to be more stable than the cotunnite structure (30). This structure type was predicted to be stable at >650 GPa. Lyle et al. (31) further predicted an I4/mmm structure at >10 TPa. It is remarkable that in almost all of those high-pressure structure types, except for the predicted structures >650 GPa, the first-neighbor maximum Si–O CN is 6. Only in the very high-pressure structures, Si was either 9-fold coordinated in the cotunnite-type and Fe2P-type structures or 10-fold coordinated in the I4/mmm structure. The steady increase of the Si–O CN with pressure >50 GPa in the glass observed here suggests that amorphous and crystalline SiO2 exhibit different densification mechanisms at very high pressures.

However, a closer look at the second Si–O neighbor distances in crystalline SiO2 shows a decrease with each subsequent structural transformation. While it was 2.952 Å for CaCl2-type structure at 70 GPa, it was only 2.372 Å for the suggested pyrite structure of the Pa3-type polymorph at 270 GPa. In fact, the pyrite type structure can be either described as 6- or 8-coordinated Si with six small and two longer Si–O bonds. Thus, it is reasonable that the randomly distorted structure of the SiO2 glass could reach higher Si–O CN than 6 at high pressures. If we assume linear increase of the CN with pressure, extrapolation of the Si–O CN to higher pressures reaches a value of 8 at ~325 GPa, which is only slightly higher in pressure than the observed transition to the pyrite structure in crystalline SiO2 at 270 GPa (26).

The changes of CN with pressure in oxide glasses and oxide melts have been shown to be linked to the OPF (15). Due to the lack of glass and melt structural data with a CN >6, the systematics in Zeidler et al. (15) is reported only for CNs between 3 and 6. Kono et al. (16) reported the first data on above-sixfold Ge–O CN in GeO2 glass at high pressures. They extended the systematics by using a similar approach to calculate OPF for structures with above sixfold CN. The OPF ρ0 is calculated by ρ0 = V0ρCO2, with V0 = (4/3)πr03, where r0 is the oxygen radius, CO is the atomic fraction of oxygen, and ρ is number density. The number densities were taken from density measurements of the respective glasses at high pressures or by extrapolation of this data by using different equation of state (EOS) formalisms, when experimental density data are not available. The oxygen radii are more difficult to obtain, since the apparent O–O peaks in experimentally derived g(r) do not necessarily represent the shortest O–O distance. For example, by comparing the distances of the apparent O–O peak of the SiO2 glass at high pressures to the distances in crystalline analogue structures, it is clear that there has to be contributions with lower O–O distance than the peak position suggests. This can also be seen, for example, by the height increase of the minimum between the Si–O peak and the neighboring O–O peak in the g(r) with
For the SiO₂ glass data, we used the Fe₂P-type structure as endmember, and for the GeO₂ glass data, we used the cotunnite structure as endmember, since these have been predicted to be the next stable crystalline phases at pressures higher than the maximum pressure reached in each experimental study, respectively (30, 32). The results are shown in Fig. 5.

The η₀-CN systematics of SiO₂ glass up to sixfold coordination followed the trend in the compilation of ref. 15. The OPF η₀ for fourfold Si-O coordination ranged from 0.44 to 0.67. At this η₀, the Si-O CN increased to ~6. Above sixfold Si-O CN, η₀ actually decreased, rather than remaining more or less constant, as reported for GeO₂ (16). However, this is an expected behavior: A close-packed oxygen structure only has fourfold (tetrahedral) and sixfold (octahedral) gaps; thus, to increase the Si-O CN to a larger value, the structure has to depart from a closed packed oxygen arrangement. The increasing CN > 6 is caused by the increasing ionic radius ratio r₅/r₇ at very high pressures (Fig. S1). This results in an increasing Si-packing fraction by the expense of the OPF, while the total packing ηtotal of the glass structure remains more or less constant (Fig. S2). If we imagine a closed packed structure of an ideal AO₂ compound with r₅ = r₆, where both atoms are building the packing structure, η₀ would be 0.49 (2/3 of the KC). Projecting from this ideal AO₂ structure through the values of pre- 

Fig. 5. Dependence of OPF on A-O CN for SiO₂ glass from this study and GeO₂ glass from ref. 16. We compare experimental data to calculated values for the Fe₂P-type and cotunnite structural polymorphs for SiO₂ (red left/right triangles (30) and GeO₂ (white up/down triangles) (32). Hereby, different directions of the symbols are for different cutoff values (2.1 and 2.5 Å) for the O–O MEFIR distance calculations for these crystalline structures. The dashed blue line represents the Kepler conjecture (KC) marking the densest possible OPF (~0.74 (34)). The black diamond represents an "ideal" hypothetical close-packed AO₂ structure where both atoms have the same size and contribute to the close packing equally; thus, A-B coordination will be 12, but the OPF η₀ is only two-thirds of the KC. The shaded areas are linear extrapolations from the ideal AO₂ structure through the values of pre- 

cducted crystalline high pressure phases to sixfold coordination. The magenta crosses show the results of the ab initio MD (AIMD) simulation for different densities (7-16 g/cm³ at 4,000 K).

pressure (Fig. 1B). In Zeidler et al. (15), the A–O distance is used as reference distance, and the O–O distance is calculated from geometric considerations (e.g., r₀ = r₆/O−O/V² for sixfold coordination in octahedral geometry). There, O–O distances for structures with a CN between 3–4 and 4–6 are calculated as weighted averages from the corresponding polyhedra, assuming a linear dependence. Kono et al. (16) applied this principle for sixfold to ninefold CN by using the GeO₂ cotunnite-type structure predicted by first-principles calculations (32). They calculated the r₆/√r₅ ratio using average Ge–O and O–O distances up to 2.5 Å in the predicted cotunnite structure and applied this ratio for estimating the oxygen radius in the glass.

However, the atomic radii should be more closely represented by the smallest distances, whereas the larger distances are often induced by the specific medium-range structure of the material. To correct for this effect and obtain a more robust estimate, we used a different approach. We calculated the oxygen radius in the predicted ultrahigh pressure structures using the concept of mean fictive ionic radii (MEFIR) (33), which weighs shorter bond distances more than larger bond distances. On this basis, we recalculated r₆/r₅ = 0.593 and r₆/√r₅ = 0.589 for the predicted cotunnite structure of GeO₂ and SiO₂, respectively. For the predicted GeO₂ and SiO₂ Fe₂P-type structures, we obtained r₆/r₅ = 0.576 and r₆/√r₅ = 0.575, respectively. Furthermore, while the apparent Ge–O/Si–O CN for both structures, cotunnite-type and Fe₂P-type, is 9, the polyhedral of the cotunnite-type structure is much more distorted. To account for this distortion, Hoppe (33) introduced the concept of effective CN (ECOn), which counts the contribution from atoms deviating from the closest distance slightly less. This results in an ECOn of 7.7 for the cotunnite structure and of 8.5 for Fe₂P-type structure. Using these values, we have calculated the OPF-CN systematics for our SiO₂ glass data and recalculated the systematics for the GeO₂ glass data (16).
predicted high-pressure polymorphs (Fig. 5), thus, confirming the validity of our approach for the calculation of \( n_0 \)-CN systems for the SiO\(_2\) and GeO\(_2\) glasses for above sixfold Si–O coordination.

Materials and Methods

Experimental Details. The high-pressure X-ray diffraction experiments were performed at the GSECARS, 13-IDD beamline, Advanced Photon Source, Argonne National Laboratory. A BX90 DAC (36) was used for pressure generation, and all measurements were collected at room temperature. An incident monochromatic X-ray beam with an energy of 40 keV and 2.5 × 3 \( \mu \)m beam size was used. An MCC as described in ref. 17 with a 50-\( \mu \)m inner slit size and 200-\( \mu \)m outer slit size was used. Commercially available SiO\(_2\) glass was packed in the pressure chamber in a Re gasket; beveled diamonds with 120-\( \mu \)m culet size were used; and no pressure medium was used. A small piece of gold was added as pressure calibrant, whereby pressure was estimated before and after each measurement based on the known equation of state (EOS) of gold (37), with the reported pressures being the average, and the difference was used as error estimate. Diffraction data were collected with a Mar345 image plate detector, which was calibrated by using a LaB\(_6\) standard. The typical collection time was 300 s.

Data Analysis. Detector calibration, image integration, and intensity corrections for oblique X-ray to detector angle, c-BN seat and diamond absorption were performed by using the Dioptras software package (38). The resulting diffraction patterns were corrected for an additional diamond Compton scattering contribution, which was necessary because the background measurement before compression was measured with a thicker sample chamber than the compressed sample material at high pressure (Fig. S3). The thinner sample chamber resulted in more diamond in the volume of diffraction constrained by the MCC. Both the sample signal and the additional diamond Compton scattering contribution were corrected for the transfer function of the MCC (39).

The Faber–Ziman structure factors and pair distribution functions were calculated by using the Glassure software package (40). The data-processing steps included: (i) normalization to atomic units by fitting the high part of the wave vector \( Q \) to the calculated scattering factors of SiO\(_2\), (ii) extrapolation of the resulting 3\( Q \) to zero by using a second-order polynomial function with \( y = ax^2 + bx + c \), and (iii) optimization of the data using a Kaplow-type correction (41) with typically three iterations. The amount of extra diamond Compton scattering \( n_{\text{dia}} \) was optimized by minimizing the amplitude of the oscillations before the first peak in \( g(r) \) after Fourier transformation from 3\( Q \) to \( g(r) \) before any Kaplow-type correction (41). The resulting \( n_{\text{dia}} \) are shown in Fig. S4. No Lorch-modification (42) function was used, since it was found to be unnecessary. Number densities for calculation of \( g(r) \) and CNs were estimated by using two third-order Birch–Murnaghan EOSs for different pressure regions: (i) up to 45 GPa with \( n_0 = 2.212 \text{ g/cm}^3, K_0 = 18.88 \text{ GPa} \) and \( K' = 3.53 \), (ii) > 45 GPa with \( n_0 = 3.86 \text{ g/cm}^3, K_0 = 176 \text{ GPa} \) and \( K' = 4.5 \). These EOSs have been determined by X-ray absorption up to almost 100 GPa. Because our data did not show any sign of abrupt structural changes > 100 GPa, we assumed the estimated EOS parameters to be valid up to 172 GPa. Example analyses at 5 GPa and 172 GPa are shown in Figs. 5S and 5F, respectively.

CN Estimation. In principle CNs can be extracted from the calculated pair distribution functions \( g(r) \) by integrating over the first peak. This works directly for monatomic materials. However, for materials with two or more elements, there exists no direct solution, since \( g(r) \) is influenced by weighing the partial \( S_{\text{Si}}(Q) \) with the X-ray form factors of each element. One way to overcome this is by calculating pseudospatial structure factors through reweighing of the original \( S_{\text{Si}}(Q) \) using the X-ray form factor factors \( f(Q) \) for Si and O:

\[
f_{\text{Si}}(Q) = \frac{(c_0f_0 + c_Sf_S)^2}{Z_{\text{Si}}c_0f_0(Q)f_S(Q)}
\]

\[
S_{\text{Si,O}}(Q) = (S_{\text{Si}}(Q) - 1) \cdot f_{\text{Si}} + 1
\]

where \( f(Q) \) is the weighing factor, \( c_0 \) and \( c_S \) are the relative concentrations of each element (\( c_0 = 2/3 \) and \( c_S = 1/3 \)), \( f_0(Q) \) and \( f_S(Q) \) are the X-ray form factors for Si and O, respectively. By performing the Fourier transform of \( S_{\text{Si,O}}(Q) \), we obtain the pseudospatial pair distribution function \( g_{\text{Si,O}}(r) \):

\[
g_{\text{Si,O}}(r) = \frac{1}{2\pi^2} \int_0^{\infty} S_{\text{Si,O}}(Q) \sin(Qr) dQ
\]

where \( \rho \) is the number density in atoms per cubic \( \text{A}^3 \) (e.g., 0.0662 \( \text{A}^{-3} \) for SiO\(_2\) glass at ambient conditions). This partial pair distribution function can now be used to estimate the Si–O CN by integrating over the first peak, provided that the peak is only from an Si–O contribution. In this case, we do an integration over the full peak up to the first minimum in the \( 4\pi r^2 g(r) \) curve after the peak:

\[
CN_{\text{Si,O}} = 4\pi n_0 c_0 \int_0^{\min} r^2 g_{\text{Si,O}}(r) dr
\]

However, this method is only reliable if the peak is well separated and we are sure there is no other contribution (e.g., from O–O) for the SiO\(_2\) glass data at high pressures, this method works very well up to sixfold coordination at ~40–50 GPa. At higher pressures, it is known from crystalline polymorphs of SiO\(_2\) that the O–O distances can be as low as 1.9 \( \text{A} \)—for example, for \( \alpha \)-PbO\(_2\) type SiO\(_2\) at ~120 GPa using the EOS data of ref. 51, which will be well within the integration area (the usual \( n_{\text{dia}} \) is at ~2.1 \( \text{A} \)) and thus leading to erroneously high Si–O CNs. To avoid this issue, we can also assume that the peak is symmetric and integrate over the left half of the peak and double the intensity, whereby the \( n_{\text{max}} \) will be the maximum in \( r^2 g(r) \) of the first peak:

\[
CN_{\text{Si,O}} = 8\pi n_0 c_0 \int_0^{\max} r^2 g_{\text{Si,O}}(r) dr
\]

This maximum peak integration method is only reliable if the distribution is symmetric, which is often not the case for glasses. Depending on the type of asymmetry of the peak, the different methods will give upper and lower limits of the actual average CN present in the glass. For the analysis of the SiO\(_2\) glass presented here, we chose to use Eq. 1 for the CN up to 40 GPa, because the peak is well separated, and we can account for the asymmetric peak shape. Eq. 2 was used at higher pressures to avoid the artificially high CN due to the extra O–O contribution. Comparison between the extracted CN of both methods is shown in Fig. S7. The minimum method [Eq. 1] leads to very high CNs > 50 GPa, probably due to extra O–O contributions, and the peak maximum method leads to slightly higher CNs < 30 GPa, due to not representing the slight asymmetric nature of the Si–O peak and therefore overestimating the CN. The CN errors were calculated by the variation within the estimate of the \( r \)-space uncertainty used for the Si–O distances.

Computational Details. The AIMD simulations are based on a PBE exchange correlation functional (43) in conjunction with a Goedecker-type (44) pseudo-potentials as implemented in the CPMD code (45). Born–Oppenheimer simulations were performed in canonical ensemble with a Nosé–Hoover thermostat (46, 47) and with 264 atoms. An integration time step of 0.3 fs was used, and each density was equilibrated for at least 3 ps. Parameters from the simulations were extracted from a subsequent production run with a run time of at least 6 ps. A good compromise between accuracy and computation speed was found for the plane-wave expansion of the Kohn–Sham orbitals with a cutoff of 100 Ry. We calculated \( n_0 \) based on box size and number of O atoms in the simulation cell and the Si–O CN was estimated by integrating the first peak of the time averaged Si–O pair distribution function up to the first minimum after the peak (Eq. 1).

The starting configuration for the AIMD simulations was produced by classical MD (CMD) simulations using the DL_POLY Classic code (48) in combination with Morse potentials (49, 50) and a time step of 1 fs. The CMD simulations were run in the NPT ensemble with a Nosé–Hoover thermostat (46, 47). An initial randomized cell was created with 264 atoms and a number density of 0.066 atoms/\( \text{A}^3 \), whereby atoms were moved until all atomic distances were above reasonable cutoff distances (\( d_{\text{Si–O}} > 1.4 \text{ A} \) and \( d_{\text{O–O}} > 2 \text{ A} \)). This cell was then equilibrated at ambient pressure and 3,000 K for 100 ps. After this, the cell was equilibrated at 70 GPa and 5,000 K for 1,000 ps. This structure produced by the CMD simulations was further equilibrated by an AIMD run of >30 ps at 4,000 K.

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