First-principles molecular dynamics simulations of MgSiO$_3$ glass: Structure, density, and elasticity at high pressure

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ABSTRACT

We report a first-principles molecular dynamics study of the equation of state, structural, and elastic properties of MgSiO$_3$ glass at 300 K as a function of pressure up to 170 GPa. We explore two different compression paths: cold compression, in which the zero pressure quenched glass is compressed at 300 K, and hot compression, in which the liquid is quenched in situ at high pressure to 300 K. We also study decomposition and associated irreversible densification. Our simulations show that the glass at the zero pressure is composed of primarily Si-O tetrahedra, partially linked with each other via the bridging O atoms (present in 35%; the remaining being the non-bridging O atoms). With increasing pressure, the mean Si-O coordination number gradually increases to 6, with fivefold and subsequently sixfold replacing tetrahedra as the most abundant coordination environment. The Mg-O coordination comprising of a mixture of four-, five-, and sixfold species at zero pressure picks up more high-coordination (seven- to ninefold) species on compression and its mean value increases from 4.5 to 8 over the entire pressure range studied. Consistently, the anion-cation coordination numbers increase on compression with appearance of oxygen tri-clusters (three silicon coordinated O atoms) and mean O-Si coordination eventually reaching 2. Hot compression produces greater densities and higher coordination numbers at all pressures as compared with cold compression, reflecting kinetic hindrances to structural changes. On decompression from 6 GPa, the glass regains its initial uncompressed structure with almost no residual density. Decompression from 27 GPa produces significant irreversible compaction, and the peak-pressure of decompression significantly influences the degree of density retention with as high as 15% residual density on decompression from 170 GPa. Irreversibility arises from the survival of high coordination species to zero pressure on decompression. With increasing pressure, the calculated compressional and shear wave velocities (about 5 and 3 km/s at the ambient conditions) of MgSiO$_3$ glass increase initially rapidly and then more gradually at high pressures. Our results suggest that hot-compressed glasses perhaps provide closer analog to high-pressure silicate melts than the glass on cold compression.

Keywords: Silicate glasses, first-principles study, structure, equation of state, high pressure

INTRODUCTION

Silicate melts played a crucial role throughout the mantle pressure regime in the chemical differentiation of the Earth in its early stages of accretion (Tonks and Melosh 1993; Labrosse et al. 2007). Melts are also thought to exist in the present day Earth at pressures well beyond the shallow magma genetic zone, including at the top of the transition zone (14 GPa, Song et al. 2004; Mookherjee et al. 2008) and at the core-mantle boundary (136 GPa, Williams and Garnero 1996; Stixrude and Karki 2005). To understand the origin and stability of these deep melts, and the role of high-pressure melts in the earliest evolution of the Earth, knowledge of the physical properties of the silicate melts at the relevant pressure-temperature conditions is essential. A key quantity is the density of the silicate melt at high pressure, which determines whether the liquid is positively or negatively buoyant with respect to coexisting crystals. There is now substantial evidence that liquids can become denser than coexisting crystals at several crossover points throughout the mantle pressure regime, with important implications for chemical differentiation (Stolper et al. 1981; Agee and Walker 1988; Stixrude and Karki 2005; Sakamaki et al. 2006).

Silicate glasses have long been studied as experimentally accessible analogs of high-pressure silicate liquids (Williams and Jeanloz 1988; Kubicki et al. 1992; Meade et al. 1992). Silicate glasses can be studied at the ambient temperature, and have been examined spectroscopically to pressures beyond 100 GPa (Sato and Funamori 2010; Murakami and Bass 2010). However, because of the lack of data on silicate liquids, it is not known how closely the behavior of silicate glasses corresponds to that of liquids at high pressure. Experimental data on silicate liquids is still very limited at lower mantle conditions (Asimow and Ahrens 2010), and measurements of silicate liquid structure are nonexistent beyond a few GPa (Yamada et al. 2011).