Sound speed and refractive index of amorphous CaSiO₃ upon pressure cycling to 40 GPa

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ABSTRACT

Brillouin spectroscopy at room temperature and pressures up to 40 GPa documents nearly identical elasticity and refractive index of amorphous CaSiO₃ created by two different methods: temperature-quenching the melt at ambient pressure and pressure-amorphizing crystalline wollastonite at room temperature. We find reproducible hysteresis of 0 to 8% on pressure cycling that is small relative to the 30 to 60% changes in shear and longitudinal wave velocities over this pressure range. Together with observed changes in refractive index and previous results from Raman spectroscopy, these measurements reveal a continuous and reversible change in atomic packing induced by pressure. Unlike many other silicate glasses, amorphous CaSiO₃ exhibits highly reproducible properties, behaving smoothly and reversibly under pressure cycling and possessing similar structure and elasticity regardless of synthesis paths for the starting material, which suggests that the amorphous solid may mimic the liquid over the pressure range investigated.

Keywords: High pressure, glass, silicate, Brillouin

INTRODUCTION

Knowledge of the density and elasticity of silicate melts at high pressures is important for understanding the deep Earth because buoyancy drives melt migration, which in turn plays a key role in the thermal-chemical evolution of our planet. Laboratory measurements of melt properties at simultaneously high temperatures and pressures are challenging, however (Sanloup et al. 2013; Ahart et al. 2014; Andrault et al. 2020). We can instead examine silicate glasses at room temperature, which provide frozen snapshots of silicate melt structure under compression and can be studied using a wide range of probes up to high pressures (Murakami and Bass 2010, 2011; Petitgirard et al. 2017; Kono et al. 2018, Mysen and Richet 2019). The primary concern with this approach is in establishing the degree to which the kinetically frozen structure of the glass at 300 K mimics the atomic-packing configuration of the melt, as reflected in such bulk properties as density and elasticity. Reversibility upon compression and decompression is a prerequisite for inferring that relevant degrees of freedom remain unhindered in the glass at 300 K. For example, it is known that some properties of SiO₂ and MgSiO₃ glasses show evidence of kinetic hindrance (i.e., lack of reversibility upon compression and decompression) and are therefore not entirely representative of the properties of the corresponding melts (Grimsditch 1984; Sanchez-Valle and Bass 2010). In contrast, minimal hysteresis has been observed in Raman spectroscopy, IR absorption spectroscopy, X-ray diffraction, and X-ray emission spectroscopy of amorphous CaSiO₃, on compression and decompression (Kubicki et al. 1992; Serghiou et al. 2000; Shimoda et al. 2005; Shim and Catalli 2009). This reversibility suggests that CaSiO₃ glass may reflect the physical properties (e.g., density, elasticity, structure) of the melt over a wide range of pressure.

To further compare amorphous CaSiO₃ samples that have experienced different pressure-temperature histories, we characterize the elasticity and refractive index of amorphous CaSiO₃ at ambient temperature to ~40 GPa using two different types of samples: one formed by solid-state pressure-induced amorphization, and the other formed by thermal quenching from the melt at ambient pressure. We also make measurements during multiple cycles of compression and decompression. The apparent redundancy in measurements is by design; we generate high-pressure amorphous CaSiO₃ from multiple starting materials and use multiple compression cycles as a way to check both the reproducibility and reversibility of properties. To be clear, we refer to a sample lacking long-range crystalline order as being amorphous and reserve the term glass for the thermally quenched melt.

EXPERIMENTAL METHODS

Melt-quenched material from two different sources gave indistinguishable results and will therefore be described as the “glass” in all figures and subsequent text: the glass used by Richet et al. (1993) and the glass used by Kubicki et al. (1992). The glasses were polished on two sides to ~20 μm thickness, placed in the sample chambers of diamond-anvil cells with 350 μm culets and rhenium gaskets, and surrounded by an argon pressure medium.

Natural wollastonite crystals from Riverside, California, with about a millimeter in grain size, were ground into a fine powder (<3 μm grain size) with a mortar and pestle. The powder was loaded into the 140 μm diameter hole of a rhenium gasket pre-indented to 30 μm thickness. The culet diameter of the diamond anvil was 350 μm.

The crystalline powder was pressure-amorphized and studied by the following procedure. The powder sample was pressure-cycled twice to ~40 GPa with no pressure medium, loaded into a diamond cell with a methanol-ethanol-water pressure medium (mass ratio 16:3:1), compressed to 42 GPa, and decompressed