



Effect of pressure on the short-range structure and speciation of carbon in alkali silicate and aluminosilicate glasses and melts at high pressure up to 8 GPa: ^{13}C , ^{27}Al , ^{17}O and ^{29}Si solid-state NMR study

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Abstract

Despite the pioneering efforts to explore the nature of carbon in carbon-bearing silicate melts under compression, *experimental* data for the speciation and the solubility of carbon in silicate melts above 4 GPa have not been reported. Here, we explore the speciation of carbon and pressure-induced changes in network structures of carbon-bearing silicate ($\text{Na}_2\text{O}\text{-}3\text{SiO}_2$, NS3) and sodium aluminosilicate ($\text{NaAlSi}_3\text{O}_8$, albite) glasses quenched from melts at high pressure up to 8 GPa using multi-nuclear solid-state NMR. The ^{27}Al triple quantum (3Q) MAS NMR spectra for carbon-bearing albite melts revealed the pressure-induced increase in the topological disorder around 4 coordinated Al ($^{[4]}\text{Al}$) without forming $^{[5,6]}\text{Al}$. These structural changes are similar to those in volatile-free albite melts at high pressure, indicating that the addition of CO_2 in silicate melts may not induce any additional increase in the topological disorder around Al at high pressure. ^{13}C MAS NMR spectra for carbon-bearing albite melts show multiple carbonate species, including $^{[4]}\text{Si}(\text{CO}_3)^{[4]}\text{Si}$, $^{[4]}\text{Si}(\text{CO}_3)^{[4]}\text{Al}$, $^{[4]}\text{Al}(\text{CO}_3)^{[4]}\text{Al}$, and free CO_3^{2-} . The fraction of $^{[4]}\text{Si}(\text{CO}_3)^{[4]}\text{Al}$ increases with increasing pressure, while those of other bridging carbonate species decrease, indicating that the addition of CO_2 may enhance mixing of Si and Al at high pressure. A noticeable change is not observed for ^{29}Si NMR spectra for the carbon-bearing albite glasses with varying pressure at 1.5–6 GPa. These NMR results confirm that the densification mechanisms established for fluid-free, polymerized aluminosilicate melts can be applied to the carbon-bearing albite melts at high pressure.

In contrast, the ^{29}Si MAS NMR spectra for partially depolymerized, carbon-bearing NS3 glasses show that the fraction of $^{[5,6]}\text{Si}$ increases with increasing pressure at the expense of Q^3 species ($^{[4]}\text{Si}$ species with one non-bridging oxygen as the nearest neighbor). The pressure-induced increase in topological disorder around Si is evident from an increase in peak width of $^{[4]}\text{Si}$ with pressure. ^{17}O NMR spectrum shows that the fraction of $\text{Na}\cdots\text{O}\text{---}^{[5]}\text{Si}$ in carbon-bearing NS3 glasses is less than that of carbon-free NS3 glasses at 6 GPa potentially due to the formation of bridging carbonate species. While its presence is not evident from the ^{17}O NMR spectrum primarily due to low carbon concentration, ^{13}C MAS NMR results imply the formation of bridging carbonates, $^{[4]}\text{Si}(\text{CO}_3)^{[4]}\text{Si}$, above 6 GPa. The spin-lattice relaxation time (T_1) of CO_2 in albite melts increases with increasing pressure from 42 s (at 1.5 GPa) to 149 s (at 6 GPa). Taking the pressure-induced change in T_1 of carbon species into consideration, total carbon content in carbon-bearing albite melts increases with pressure from ~ 1 wt% at 1.5 GPa to ~ 4.1 wt

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% at 6 GPa. The results also reveal a noticeable drop in the peak intensity of free carbonates in carbon-bearing NS3 melts at 6 GPa, implying a potential non-linear change in the carbon solubility with pressure. The current results of carbon speciation in the silicate melts above 4 GPa provide an improved link among the atomic configurations around carbon species, their carbon contents, and isotope composition of carbon-bearing melts in the upper mantle.

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1. INTRODUCTION

CO₂ is among the most abundant volatile components in Earth's interior, and understanding the nature of carbon (e.g., speciation, solubility) in silicate melts at high pressure is essential to unraveling the property changes in carbon-bearing silicate melts in the mantle. The presence of CO₂ molecule and/or other carbon-bearing species in silicate melts changes the stability field of silicate crystals (Mysen and Richet, 2005), which controls the composition of partial melts generated by decompressional melting beneath the mid-ocean ridge (e.g., Egglar, 1976; Wallace, 2005; Dasgupta and Hirschmann, 2006; Dasgupta et al., 2013; Evans et al., 1999). The addition of CO₂ with H₂O in peridotitic melts is expected to decrease the solidus, which may have contributed to the presence of a low-velocity zone in the asthenosphere (e.g., Egglar, 1976; Lee, 2010; Dasgupta et al., 2004; Evans et al., 1999; Gu et al., 2005; Massuyeau et al., 2015; Song et al., 2004). The dissolved CO₂ in silicate melts affects the viscosity of silicate melts: the viscosity of KAlSi₃O₈ melts shows non-linear behavior as the viscosity decreases with increasing CO₂ content up to 5 wt% and then increases with more CO₂ in the melts, perhaps because of the complex role of CO₂ in silicate melts (White and Montana, 1990). In contrast, the viscosity of NaAlSi₃O₈ (albite) melts continuously decreases with increasing CO₂ contents in the silicate melts (Brearley and Montana, 1989).

Whereas the focus of the study is to reveal the effect of pressure on the speciation of carbon in silicate melts, carbon speciation has strong implication for carbon solubility in melts. The solubility of CO₂ in the silicate melts up to ~3.5 GPa has been experimentally explored. Here, we briefly summarize the solubility of carbon in silicate melts below 3.5 GPa. The total solubility of carbon in basaltic melts linearly increases up to 0.5 wt% with increasing pressure up to 0.2 GPa (Parfitt and Wilson, 2008). The solubility of carbon in the silicate melts tends to increase with increasing pressure, the ratio of non-bridging oxygen over tetrahedron (NBO/T), and the fraction of Mg and Ca, but it decreases with increasing temperature, the SiO₂ content, and the Na content (e.g., Egglar and Rosenhauer, 1978; Egglar and Kadik, 1979; Fine and Stolper, 1985; Kohn et al., 1991; Morizet et al., 2010; Mysen et al., 1975; Nowak et al., 2004). For instance, the total solubility of carbon species in synthesized NaAlSi₄O₁₀ melts increases up to 1.0 wt% at 2.5 GPa (Fine and Stolper, 1985), while the solubility in albite melts increases up to 2.5 wt% at 3.5 GPa (Brooker et al., 1999). In the natural olivine nephelinite-composition melts, the solubility ranges up to

17.7 wt% with increasing pressure up to 3 GPa (Egglar and Kadik, 1979; Kohn et al., 1991; Mysen et al., 1975).

The mechanisms for dissolving CO₂ and other carbon species (e.g., CO₃²⁻, CO, and CH₄) into silicate melt are relatively well understood in the pressure range up to 3.5 GPa (Blank and Brooker, 1994; Behrens et al., 2004; Brooker et al., 1999; Kohn et al., 1991; Morizet et al., 2002, 2014a, 2015; Tossell, 1995; Mysen et al., 1976, 2009, 2011; Pan et al., 1991). FTIR and Raman spectroscopies are extensively used to identify the speciation and exact structural information of carbon in silicate glasses and melts (Rosso and Bodnar, 1995; Burke, 2001; Ni and Keppler, 2013; Brooker et al., 1999; Hacura et al., 1990; Morizet et al., 2009; Moussallam et al., 2016; Mysen et al., 2009; Pawley et al., 1992; Seitz et al., 1996). Those studies found diverse molecular species (superscript mol), such as CO₂^{mol} and CO^{mol}, and other structurally bound species, such as bridging carbonate species and dissolved CO₂ (a molecule weakly bound to the bridging oxygen in the silicate network). In addition, the information from these vibrational spectroscopies can be useful in inferring the pressure conditions and slight changes in the topology of carbon species with increasing pressure and temperature. For instance, the peak splitting in the main CO₂ Raman peak (at 1337 cm⁻¹, i.e., the Fermi resonance) in fluid inclusions provides the internal pressure of the fluid inclusion (Rosso and Bodnar, 1995; Bakker, 2003; Bodnar, 2003; Seitz et al., 1996), and the gap between the two split carbonate peaks (at ~1500 cm⁻¹) in the FTIR spectra can give insights into the distortion of carbonate species in the silicate glasses (Brooker et al., 1999; Morizet et al., 2002).

NMR spectroscopy is complementary to the vibrational spectroscopy. It provides direct, element-specific, and quantitative information of atomic structures around nuclides of interest in silicate melts and glasses (e.g., Kirkpatrick and Brow, 1995; Stebbins, 1995b; Lee and Stebbins, 2003; Park and Lee, 2012; Kohn et al., 1991; Lee et al., 2001; Malfait et al., 2012; Papenguth et al., 1989; Phillips et al., 2000; Stebbins and Xue, 2014; Xue et al., 1989). ¹³C NMR in particular has been effective in identifying carbon species in silicate melts and glasses (Herzfeld and Berger, 1980; Cody et al., 2011; Kohn et al., 1991; Morizet et al., 2014b, 2015; Mysen, 2012). Previous studies on ¹³C magic angle spinning (MAS) NMR have identified CO₂ (at ~125 ppm), CO₃²⁻ (at ~160–170 ppm), and CO species (~184 ppm) in silicate glasses (Tossell, 1995; Mysen, 2013; Brooker et al., 1999; Feng et al., 2006; Kim et al., 2016; Kohn et al., 1991). The bridging carbonate ions that are linked to two framework cations, such as [⁴Si(CO₃)⁴Si, [⁴Si(CO₃)⁴Al, or [⁴Al(CO₃)⁴Al], and the free carbonate

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