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CO_2 speciation and transport properties of CO_2-bearing silicate melts.

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There are growing evidences of the existence of CO_2-rich magmas in the upper mantle (e.g. Dasgupta and Hirschmann, 2006; Zeng et al., 2010; Helo et al., 2011). So the role of carbon-rich melts at depth is now becoming a credible scenario to explain the extraction of CO_2 from the source region to the surface. During the last three decades many studies have been devoted to measure the solubility of CO_2 in silicate melts of various composition. But due to experimental difficulties these studies were generally restricted to low and moderate pressures (below ~20 kbar). IR spectroscopy has emphasized the importance of CO_2 speciation which may exist either as molecular CO_2 or as carbonate ion (CO_3^{2-}), the molecular form being favored in polymerized (silicic) melts while the carbonate ion is dominant in depolymerized (basic and ultrabasic) melts. However it has been suggested recently (e.g. Morizet et al., 2007, Spickenbom et al., 2010) that the CO_2 speciation observed in quenched glasses by IR spectroscopy may not be representative of that in silicate melts equilibrated at high temperature: in particular, the abundance of molecular CO_2 could be underestimated in the liquid at magmatic temperatures

Recently, in introducing an empirical force field to describe the chemical reactivity of CO_2 in silicate liquids ($CO_2+(O^{2-})_{melt}\leftrightarrow CO_3^{2-}$), it has been possible to evaluate by classical molecular dynamics (MD) calculation the solubility and speciation of CO_2 in silicate melts of various composition (Guillot and Sator, 2011). The conclusions of these MD calculations are twofold, (i) The solubility of CO_2 increases markedly with the pressure and reaches value as high as ~20 wt% in basaltic melts at 8 GPa, and (ii) The proportion of molecular CO_2 is found to be significant in CO_2-saturated basic and ultrabasic melts at superliquidus temperature and high pressures, a result at variance with post mortem analysis of basaltic glasses where only carbonate ions are detected. So, to check the consistency of the above MD calculations we have performed *ab initio* molecular dynamics simulations (AIMD) of CO_2-saturated basaltic and kimberlitic melts. The results of these electronic structure calculations are presented here. They support the aforementioned conclusions reached with classical MD calculations. The effect of CO_2 on viscosity and electrical conductivity of basaltic and kimberlitic melts has also been evaluated by the two methods (classical and *ab initio*). It will be discussed how our simulation data may help to better understand the migration and emplacement of CO_2-rich magmas.

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