Modeling of molten carbonates based on classical molecular dynamics simulations

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Because of their remarkable physicochemical properties carbonate melts receive an increasing interest in both fundamental and applied fields. To have a clear picture of these properties over a large range of thermodynamic conditions is important for developing industrial devices (e.g. fuel cell technology, carbon capture and storage) and for providing a better understanding of a number of geochemical processes (role of molten carbonates in the geodynamics of the Earth's mantle, partitionning of trace elements and volatiles between silicates and carbonates).

To model molten carbonates by atomistic simulations, we have developed an optimized classical force field in using electronic structure calculations (based on the density functional theory) as a benchmark [1]. In implementing this force field into a molecular dynamics simulation code, we have evaluated the thermodynamics (equation of state, surface tension), the liquid structure and the transport properties (viscosity, electrical conductivity and diffusion coefficients) of a set of molten carbonates (Li_2CO_3, Na_2CO_3, K_2CO_3, $MgCO_3$, $CaCO_3$ and many of their mixtures) from their melting point to the thermodynamic conditions of the Earth's upper mantle. Our results are in very good agreement with the data available in the litterature [2, 3, 4]. To our knowledge a molecular model for molten carbonates covering such a large domain of thermodynamic conditions, chemical compositions and physical properties has never been published yet.

Based on this model we will discuss the rheological properties of a carbonate magma (from Ol Doinyo Lengaï, Tanzania). We will also present some results on the solubilty of volatiles (noble gases and CO_2) in carbonate melts that is so far poorly known [5].
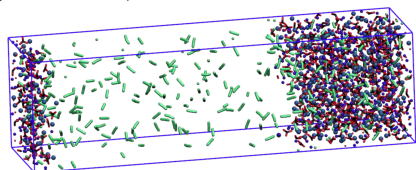
Snapshot of a CO_2 phase in contact with a carbonate melt.

[1] Vuilleumier et al. (2014), *Geochimica et Cosmochimica Acta* 141, 547–566. [2] Ward & Janz (1965), *Electrochimica Acta* 10, 849-857. [3] Gaillard et al (2008), *Science* 322, 1363-1365. [4] Kono et al (2014), *Nat Commun* 5, 1-8. [5] Wuu-Liang & Wyllie (1976), *Geochimica et Cosmochimica Acta* 40, 129-132.