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Viscosity of carbonate-rich melts under different oxygen fugacity conditions

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Viscosity is a fundamental property of many materials and its changes affects the fluid dynamics of natural system as well as industrial processes.

The mobility of carbonatitic melts, which are carbonate-rich and very fluid melts, has attracted renewed interest in both earth science and industry. In fact, these melts are considered the main transport agent of carbon from the mantle to the crust and may be intimately linked to the generation of kimberlites.

At the same time lithium, potassium and sodium carbonate are used as electrolytes in molten carbonate fuel cells which operate at high temperatures ($\sim$650°C) for the production of electricity without CO_2 emissions.

Accurate measurement of the transport property (i.e. viscosity) of carbonatitic melts is a priority in order to understand the carbonatite mobility and reaction rates. Additionally, obtaining accurate viscosity measurements of such low viscosity melts is however an experimental challenge due to volatility, very low torques and chemical melt instability in the viscometer.

To overcome these limitations we have customized a Modular Compact Rheometer (MCR 502 from Anton Paar) ad hoc equipped with 2 narrow gap concentric-cylinder geometries of steel and Pt-Au. The rheometer is characterized by an air-bearing-supported synchronous motor with torque ranging between 0.01 μNm and 230 mNm (resolution of 0.1 nNm), achieving very low viscosity measurements in the order of mPa s, temperatures up to 1000°C and shear rates ranging between 1 and 100 sec^{-1}. These experimental conditions well match the temperature-viscosity-shear rate window relevant for carbonate melts.

Here we present the calibration of the rheometer and the results of a rheological characterization study on a series of very low viscous synthetic and natural carbonatitic melts at different oxygen fugacity (air and CO_2 saturated atmosphere).

Viscosity measurements on carbonate melts have been performed in the temperature range between $\sim$650 and 1000°C. Measured values range between $\sim$2 and 20 mPa sec. The results point out that the viscosity of synthetic samples is inversely related to the cations radius, being Li_2CO_3 melt the more viscous. Viscosity measurements on natural samples (carbonatitic lava from Lengai volcano, Tanzania), reveal a higher viscosity ($\sim$1000 mPa s) and a dramatic higher activation energy than the synthetic samples.

Our results have been compared with literature data in order to determine the effect of chemical composition and oxygen fugacity conditions on the liquid viscosity of carbonatitic melts.