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Effects of temperature, pressure and chemical compositions on the electrical conductivity of carbonated melts and its relationship with viscosity

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ABSTRACT

Carbonated melts constitute a key medium in the global deep carbon cycle: their impact on the geochemical signature of deep rocks is well studied because of their role as metasomatic agents in the deep mantle. However, their physical properties and in particular their electrical conductivity at high temperature and high pressure remain poorly constrained. In this study, we investigated the effect of chemical composition on the electrical conductivity of carbonated melts. We characterized this effect for various temperatures (1000–1700 °C) and pressures (1 to 4 GPa). Measurements show a very high electrical conductivity ($>100\ S\cdot m^{-1}$) with weak temperature, pressure and chemical composition dependence. Carbonated melts are five orders of magnitude more conductive than mantle olivine, and up to two orders of magnitude more conductive than basalts at similar T and P. The electrical conductivity of molten carbonates follows an Arrhenius law and the different parameters were determined. A common activation volume was defined with $\Delta V = 0.275$ J/bar. As a result, we are able to calculate the electrical conductivity for larger temperature and pressure ranges for the melt compositions considered here. By combining the Nernst–Einstein and Eyring equations, a remarkably simple correlation was established between electrical conductivity and viscosity. The viscosity of carbonated melts, which is a key parameter defining the rate of metasomatic fluids flowing in the earth's mantle, can therefore be calculated as a function of pressure and temperature.

We used these new data to interpret the high electrical conductivity recently observed in the mantle under the Brazilian craton. The anomalously elevated conductivity most likely images the process of lithospheric rejuvenation involving 0.03 to 0.2% of carbonated melt.

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1. Introduction

Melting in the Earth's mantle due to CO_2 has long been proved by geochemical observations (Yaxley et al., 1991; Rudnick et al., 1993; Walter et al., 2008; Collerson et al., 2010; Simonetti and Neal, 2010), and experimental petrology (Wyllie and Huang, 1975; Falloon and Green, 1990; Presnall and Gudfinnsson, 2005; Dasgupta and Hirschmann, 2006; Ghosh et al., 2014) on peridotitic and eclogitic rocks (Hammouda, 2003; Dasgupta et al., 2004). All these studies reported that carbon dioxide can strongly reduce mantle solidus temperature even at the very low concentration levels expected in the mantle. Carbonate melts are therefore expected in most of the P–T domains of the upper mantle, being merely confined by redox boundaries marking the diamond stability field (Rohrbach and Schmidt, 2011; Stagno et al., 2013).

Their physical properties are much less known (Jones et al., 2013), in particular their electrical conductivity. Existing data were obtained at low pressure (Gaillard et al., 2008; Kojima et al., 2008) on alkali-rich carbonate melts or at high pressure (Sifré et al., 2014) and on alkali-free carbonate melts with poorly defined temperature and pressure dependence (Yoshino et al., 2010, 2012). Gaillard et al. (2008) showed that carbonated melts are five orders of magnitude more conductive than mantle olivine, and up to three orders of magnitude more conductive than basalt at atmospheric pressure. This observation was corroborated by Sifré et al. (2014) at 3 GPa. However, in the absence of systematic data on the effect of chemical composition, pressure and temperature on the conductivity of carbonated melts, it is difficult to quantitatively interpret mantle electrical properties.

In this study, we report measurements of electrical conductivities of dry and hydrous carbonated melts at HT (>1000 °C) and HP (1 to 4 GPa) in a piston cylinder. We describe the effect of chemical composition, temperature and pressure on the electrical conductivity. We propose a calculation to predict the effect of T and P by using the Arrhenius

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parameters determined in this study. These new data provide a link between electrical conductivity and viscosity of carbonatic melts, with a remarkably simple equation. Finally, laboratory-based values and geophysical measurements are confronted allowing quantitative estimations of melt fraction below the Brazilian craton.

2. Material and methods

2.1. Starting material

Electrical conductivity measurements were performed on six different mixtures (Tables 1 and 2): 5 dry carbonated melts (CO_2 ranging from 38 to 49 wt.%) and a hydrous carbonated melt ($CO_2 = 28$ wt.%; $H_2O = 8$ wt.%), the latter corresponding to fluid inclusion compositions found in diamonds (Klein-Bendavid et al., 2009). To obtain these mixtures, starting materials were natural calcite ($CaCO_3$), natural dolomite ($MgCa(CO_3)_2$), natural magnesite ($MgCO_3$), potassium carbonate (K_2CO_3), sodium carbonate (Na_2CO_3), silicon dioxide (amorphous SiO_2), halite (NaCl) and brucite ($Mg(OH)_2$). The starting materials were stored in an electrical oven at 120 °C before use. Potassium and sodium carbonates were however previously dried at 250 °C to ensure that they were fully dehydrated. Samples C and MC were cored from natural rocks (Carrara Marble and natural dolomite, Table 1) whereas the other mixtures were weighed in suitable amounts for the desired compositions (MK, CK, MN and MCKNw samples) and then cold pressed into pellets. For samples MK, CK and MN, a one to one molar ratio was respected, that is $MgCO_3/K_2CO_3 = 1$ for MK, $CaCO_3/K_2CO_3 = 1$ for CK and $MgCO_3/Na_2CO_3 = 1$ for MN. Concerning composition MCKNw, a molar ratio of $CO_2/(CO_2 + H_2O) = 0.65$ was considered for the volatile content.

2.2. High-pressure conductivity measurements

Experiments were performed in ½-inch piston cylinders (graphite–Pyrex–talc assemblages) at pressures ranging from 1 to 4 GPa. These experiments were connected to a 1260 Solartron Impedance/Gain Phase Analyzer for the electrical conductivity measurements. Temperature was measured with a B-type thermocouple localized on top of the sample (Figs. 1 and S1). The oxygen fugacity (fO_2) was not controlled during the measurements but the presence of the graphite furnace and molten carbonate samples should imply a fO_2 close to FMQ-2 (Stagno et al., 2013).

A protocol used for measurements has been specifically developed for electrical conductivity measurements on highly conductive molten materials (Fig. 1). A pseudo 4-wire configuration was designed, which removed the electrical contribution of the electrical cell itself. A blank test is presented in Fig. 2, the measured resistance consisting of the blank assemblage (i.e. the metal plug (Fig. 1), the Pt foil (25 μm in thickness and 15 mm length) and a Ni sample). Fig. 2 shows that the electrical conductivity of the electrical cell itself is two orders of magnitude lower than the carbonated melt, confirming the use of a 4-wire configuration. Such a configuration previously adapted at 1 atm (Gaillard, 2004; Gaillard et al., 2008) and 3 GPa (Sifré et al., 2014) was judged necessary for our measurements at high pressure.

An inner Pt electrode (1 mm diameter) was placed in the centre of the cold-pressed pellets or the centre of the natural cores (5 mm outer diameter). A Pt foil was positioned around the sample and used as the second outer electrode. The sample (approx. 2 mm length) was surrounded at the top and at the bottom by two MgO plugs (Fig. 1) that were previously annealed at 1000 °C/1 atm for 2 h. The length of each MgO plug was adjusted in order to position the sample at the centre of the furnace. The entire electrical cell was isolated from the graphite furnace by an alumina jacket (Fig. 1). The sample impedance was therefore measured between the two electrodes in a co-axial geometry (Gaillard, 2004; Hashim et al., 2013). Connection between the inner electrode and the impedance spectrometer was achieved by means of the thermocouple wires (Hashim et al., 2013). The outer electrode was connected to a nickel cylinder (metal plug in Fig. 1), the latter located 5 mm above the sample, which was mounted in series with two additional wires (B-type thermocouples), as shown in Fig. 1.

Impedance spectra were collected during heating and cooling cycles at different temperature plateaux in the frequency range of 1 Hz to 1 MHz (data collection < 2 h, Fig. S2). In the case of the hydrated sample (MCKNw), the heating cycle was slightly different: the temperature was rapidly (less than 2 min) increased to the maximum temperature (Table 3) in order to trap the volatiles (released by the brucite compound) into the molten sample. Electrical conductivity measurements were therefore conducted during the cooling cycle and the second heating cycle (except for the run at 4 GPa where no measurement was made during the second heating cycle; Table 3).

For temperatures lower than the melting temperature, T_m, the collected spectra showed impedance arcs, whereas for temperatures $> T_m$ the produced spectra exhibited vertical lines. These low- and high-temperature spectra correspond to capacitance-dominated and inductance-dominated signals, where the intercept of each spectrum with the X-axis yielded the resistance of the studied sample.

Validation of each electrical value was achieved by reproducing the measurements during both heating and cooling cycles. Any measurements failing in satisfying this requirement were discarded.

The electrical conductivity of the different samples was calculated from the measured resistance, R (in Ω) (Gaillard, 2004; Hashim et al., 2013), using:

$$\sigma = (\ln(r_{out}/r_{in}))/(2\pi h R) \qquad (1)$$

Table 1
Chemical compositions of the starting materials (wt.%).

Sample	Dry					Hydrous
	C	MC	MK	CK	MN	MCNKw
SiO_2	0.04 (0.04)	0.13 (0.20)	0.01	0.02	0.01	5.17
TiO_2	–	–	0.02	0.00	0.02	0.02
Al_2O_3	0.02 (0.02)	0.06 (0.05)	0.02	0.01	0.02	0.01
FeO	>0.01 (0.01)	0.13 (0.10)	0.03	0.00	0.03	0.02
MgO	0.64 (0.02)	18.22 (0.27)	18.12	0.27	21.19	21.21
CaO	54.69 (0.35)	31.98 (0.85)	0.01	22.84	0.01	17.11
Na_2O	0.01 (>0.01)	–	0.01	0.00	32.58	4.04
K_2O	0.01 (0.15)	–	42.34	39.30	0.01	14.57
MnO	0.01 (0.02)	0.07 (0.09)	0.01	0.00	0.02	0.01
P_2O_5	0.04 (0.05)	0.02 (0.06)	0.02	0.02	0.03	0.30
NaCl	–	–	–	–	–	2.07
Total	55.46 (0.40)	50.61 (0.50)	60.59	62.45	53.92	64.52
CO_2	44.54	48.81	39.41	37.55	46.08	28.03
H_2O	–	–	–	–	–	7.45

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