



## CO<sub>2</sub> solubility in kimberlite melts



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### ABSTRACT

Carbon dioxide is the most abundant volatile in kimberlite melts and its solubility exerts a prime influence on the melt structure, buoyancy, transport rate and hence eruption dynamics. The actual primary composition of kimberlite magma is the matter of some debate but the solubility of CO<sub>2</sub> in kimberlitic melts is also poorly constrained due to difficulties in quenching these compositions to a glass that retains the equilibrium CO<sub>2</sub> content. In this study we used a range of synthetic, melt compositions with broadly kimberlitic to carbonatitic characteristics which can, under certain conditions, be quenched fast enough to produce a glass. These materials are used to determine the CO<sub>2</sub> solubility as a function of chemical composition and pressure (0.05–1.5 GPa). Our results suggest that the solubility of CO<sub>2</sub> decreases steadily with increasing amount of network forming cations from ~30 wt.% CO<sub>2</sub> at 12 wt.% SiO<sub>2</sub> down to ~3 wt.% CO<sub>2</sub> at 40 wt.% SiO<sub>2</sub>. For low silica melts, CO<sub>2</sub> solubility correlates non-linearly with pressure showing a sudden increase from 0.1 to 100 MPa and a smooth increase for pressure > 100 MPa. This peculiar pressure–solubility relationship in low silica melts implies that CO<sub>2</sub> degassing must mostly occur within the last 3 km of ascent to the surface having potential links with the highly explosive nature of kimberlite magmas and some of the geo-morphological features of their root zone. We present an empirical CO<sub>2</sub> solubility model covering a large range of melt composition from 11 to 55 wt.% SiO<sub>2</sub> spanning the transition from carbonatitic to kimberlitic at pressures from 1500 to 50 MPa.

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### 1. Introduction

Experimental investigation of the solubility of CO<sub>2</sub> in kimberlite melt has been limited due to the difficulty in quenching such melt fast enough to form a pure glass (e.g. Brey and Ryabchikov, 1994). Previous studies have therefore estimated the CO<sub>2</sub> solubility based on: (i) extrapolation from trend defined in silicate melts (Brooker et al., 2001) (ii) solubility of melt in partially to highly crystallised experiments (Brooker et al., 2011) (iii) solubility at 0.1 MPa of simple, four components, synthetic compositions considered as analogue to the natural system (Russell et al., 2012) and (iv) molecular dynamics simulation studies (Guillot and Sator, 2011). While these studies all suggest that the solubility of CO<sub>2</sub> decreases as the melt becomes enriched in silica, the effect of pressure has remained speculative. Our current knowledge from experimental studies on silicate melt suggests that CO<sub>2</sub> solubility increases with pressure (Blank and Brooker, 1994; Morizet et al., 2002, 2014; Iacono-Marziano et al., 2012) while in pure carbonate melts, CO<sub>2</sub> solubility is more related to satisfying stoichiometric requirements and therefore less dependent on pressure as

exemplified by Oldoinyo Lengai's alkali carbonatite lava (Kervyn et al., 2008). However, experiments from Wyllie (1989) and Brooker and Kjarsgaard (2011) suggest that there is some 'excess' CO<sub>2</sub> solubility in pure CaCO<sub>3</sub> melt, apparently causing a decrease in liquidus temperature.

The composition of primary kimberlite melts in term of volatile and major element chemistry is poorly constrained. Not only are natural kimberlite laden with xenolith and xenocryst (Mitchell, 2008), but the strong alteration by external fluids they typically endured, in particular serpentinization, strongly affects the volatile and major element composition (Sparks et al., 2009; Brooker et al., 2011). Latest attempt to back-calculate the kimberlite melt chemistry by removal of the serpentinization process points towards an original melt being richer in CO<sub>2</sub> and CaO and poorer in SiO<sub>2</sub>, water and MgO than calculated from "uncontaminated" kimberlites (Sparks et al., 2009; Brooker et al., 2011). This findings are corroborated with the fact that the Mg number in kimberlite is too high for equilibrium with a mantle sources (Price et al., 2000; Kopylova et al., 2007) and that liquidus temperature for "uncontaminated" kimberlite composition of 1400 °C (Brooker et al., 2011) are higher than predicted from mineral thermometry (Fedortchouk et al., 2002). We therefore chose here to follow from the work of Brooker et al. (2011) who argued that kimberlite melts are originally transitional, that is, they have a composition intermediate between a

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carbonate and a silicate liquid, and investigated several melt compositions with SiO<sub>2</sub> contents between 11 and 32 wt.%.

In this contribution, we present results on the first experimental kimberlitic glasses synthesised using an internally heated pressure vessel (IHPV) equipped with an optimised quench configuration. We produced a series of melt compositions across the “transitional” melt field (SiO<sub>2</sub> ~ 18 to ~32 wt.%) to assess the effect of silica activity on CO<sub>2</sub> solubility. The effect of pressure was then investigated by equilibrating melts of similar composition at different pressures (50 to 1500 MPa).

## 2. Experimental methodology

### 2.1. Starting material

Starting materials were produced by mixing powders from a natural lamproite from Torre Alfina, Italy (Table 1, see Peccerillo et al., 1988) with various amounts of synthetic powders of pure oxides and natural dolomite. In order to ensure homogeneity and remove the volatiles present in the Torre Alfina rock, it was fused twice in air at 1400 °C and quenched to glass. The composition of all mixtures used as starting materials of all experiments is reported in Table 1. The melt compositions we have synthesised here are very Ca-rich when compared to the more Mg-rich typical “contamination free” kimberlite (Mitchell, 1986) but later compare our findings to experiments with more Mg-rich compositions. Therefore, an underlying supposition of our work is that Mg and Ca cations behave in a similar way with respect to CO<sub>2</sub> solubility in the melt (see Brey et al., 1991; Brey and Ryabchikov, 1994 for an investigation of the effect of Mg–Ca substitution on CO<sub>2</sub> solubility in kimberlite melts and see discussion in Section 5). The Al content of the composition presented here might also be considered slightly high by some authors while the Fe content would be considered low. Notwithstanding these differences, the compositions we explored here provide a close analogue to kimberlite melts (in terms of NBO/T for instance) for which the true composition remains unknown and debated (see review by Sparks, 2013).

### 2.2. High pressure experiments; internally heated pressure vessels and piston cylinder

To investigate the solubility of CO<sub>2</sub> in transitional melts at crustal pressure, we performed a series of solubility experiments in the pressure range from 50 to 350 MPa at relatively constant temperature (1225 to 1270 °C) in relatively dry and oxidized conditions (log<sub>f</sub>O<sub>2</sub> = FMQ + 3). We used internally heated pressure vessels at the ISTO-CNRS laboratory in Orléans, which can reach pressures of up to 400 MPa (± 3 MPa) under controlled temperature up to 1300 °C (± 2 °C). The vessel was pressurised using argon gas as the pressure medium. A two-Mo-winding vertical furnace was used, creating a

5 cm isothermal (± 5 °C) “hot-spot” zone and controlled by two S-type thermocouples located on both sides of the sample.

Experimental charges consisted of anhydrous sample powder (30 to 150 mg) loaded in gold–palladium (Au<sub>80</sub>Pd<sub>20</sub>) or platinum capsules (2 to 4 cm in length, 2.5 mm inner diameter and 2.9 mm outer diameter). The capsules were welded shut. Although the initial powder mixtures were stored at 120 °C, the strongly hygroscopic character of some oxides (in particular MgO, NaO and K<sub>2</sub>O) absorbing atmospheric H<sub>2</sub>O within the 15 min necessary to load the capsules, made it impossible to obtain fully water-free experimental charges, hence lowering slightly the XCO<sub>2</sub> in the fluid in equilibrium with the melt at P & T. For each experiment, one to three capsules were hanged by a thin Pt wire. The temperature gradient along the “hot-spot” zone, where the capsules were located, was maintained at about 10 °C; the hot zone was always on the top end of the sample. This gradient, although inducing an uncertainty in the run temperature, was part of the quench optimisation strategy as it imposed a thermal profile in the furnace that ensures a strong thermal contrast over a small vertical length located directly below the hanged capsule. Rapid quenching was achieved by passing an electrical current to the holding Pt wire so that the sample dropped into the cold, bottom part of the furnace. The cooling rate was estimated at >100–200 °C s<sup>-1</sup> although no precise estimate could be obtained. After each experiment, capsules were weighed (i) before and (ii) after, opening in order to (i) verify that no leakage had occurred, and (ii) determine the amount of gas not incorporated in the glass. Capsules were then opened and a fragment of the charge was embedded in an epoxy resin and polished for SEM and microprobe analyses. We conducted a total of 18 solubility experiments. The strategy was first to produce a complete isobaric section at 350 MPa, for a range of melt chemistry spanning SiO<sub>2</sub> from 11 to 38 wt.% (expressed as nominal starting composition, Table 1). We then explored the effect of pressure on several compositions (between 17 and 38 wt.% SiO<sub>2</sub>; expressed as nominal starting composition) from 50 to 350 MPa at temperatures of 1225 to 1270 °C. While most experiments successfully produced a pure glass (Fig. 1 and Fig. S1), experiments performed at 50 MPa (run number: TA6\_1\_6 and TA7\_1\_3), one experiment at 100 MPa (run number: TA10\_1\_2) and the experiment with lowest SiO<sub>2</sub> (run number TA15\_1\_1) could not be quenched fast enough and quench crystals were present throughout the experimental charge (these crystals could not be analysed free of the glass component).

A single piston cylinder experiment was performed at 1.5 GPa and 1300 °C in a ½ inch piston-cylinder apparatus. Experimental charge consisted of natural anhydrous sample powder (30 mg) loaded in gold–palladium (Au<sub>80</sub>Pd<sub>20</sub>) capsules (1 cm in length, 2.5 mm inner diameter and 2.9 mm outer diameter). The capsule was introduced in a talc–pyrex–graphite furnace assembly and surrounded by MgO. A B-type thermocouple was located at ~1 mm atop of the capsule and the run temperature should be considered as a minimum value. The quench of the piston cylinder assembly was not fast enough to prevent the pervasive growth of quench crystals (Fig. 1; Fig. S1; experiment TA6\_1\_5).

## 3. Analytical techniques

### 3.1. EMPA

All experimental products were examined by optical microscope and scanning electronic microscope (SEM) to check for the presence of quench crystals (Fig. S1). Electron microprobe analyses (EMPA) were performed on a Cameca SXFive at the ISTO-CNRS laboratory in Orléans. Glasses were analysed using an accelerating voltage of 15 kV, a beam current of 6 nA and a defocused beam of 10 μm. Na and K were analysed first in order to minimise alkali loss during analysis. The shortfall in the totals for analysed glasses was broadly consistent with the CO<sub>2</sub> content estimated by other techniques, but systematically higher (Fig. S2).

**Table 1**

Starting oxide and natural rock powder mix compositions. The composition of a natural lamproite from Torre Alfina, Italy, (Peccerillo et al., 1988), fused twice at 1400 °C and used to prepare the oxide-mix compositions is reported at the top of the table.

Name	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	CO <sub>2</sub>	Total
TA	55.74	1.34	13.05	5.83	9.43	5.45	0.96	7.68	0.51	0.00	100
TA15	11.01	0.09	2.34	0.41	8.45	36.36	0.27	2.14	0.04	38.90	100
TA6	16.60	0.20	3.85	0.86	6.93	35.48	0.28	2.27	0.07	33.47	100
TA7	20.08	0.25	4.66	1.08	8.16	32.50	0.34	2.74	0.10	30.09	100
TA8	21.66	0.30	5.03	1.30	8.21	31.30	0.37	2.96	0.11	28.75	100
TA9	23.11	0.34	5.37	1.49	8.26	30.20	0.40	3.16	0.13	27.53	100
TA10	23.79	0.36	5.53	1.58	8.29	29.68	0.41	3.26	0.14	26.95	100
TA11	30.18	0.56	7.04	2.43	8.52	24.84	0.52	4.14	0.21	21.56	100
TA12	37.70	0.79	8.81	3.43	8.78	19.14	0.65	5.18	0.30	15.22	100

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