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## KIMBERLITE ASCENT BY ASSIMILATION-FUELED BUOYANCY

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### INTRODUCTION

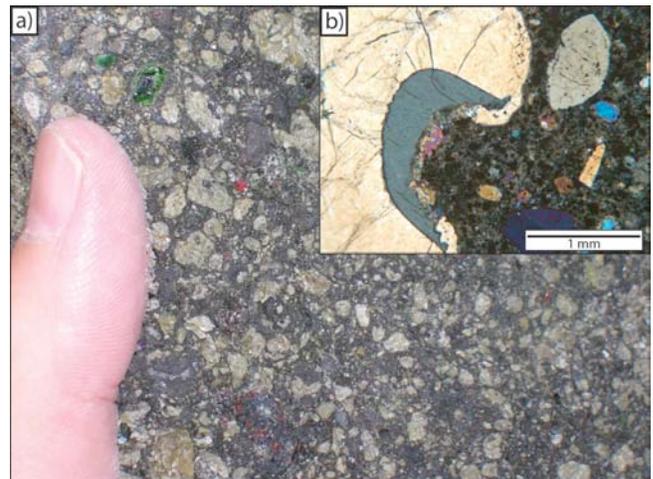
Kimberlite deposits commonly contain in excess of 25 vol% solids in the form of mantle-derived megacrysts and xenoliths implying that the original magma was enriched in solids and dense. Numerous studies of this mantle-derived cargo, including diamond, suggest that the parental magmas derive from depths > 200 km and that they are transported "rapidly" to the Earth's surface. The conundrum has been to provide the requisite buoyancy to these crystal-rich, dense magmas in order to support the implied high ascent rates. The exsolution of dissolved volatiles (CO<sub>2</sub> & H<sub>2</sub>O) is usually seen as playing a critical role in providing the requisite buoyancy, however, the source of the volatiles and the physical-chemical process behind their liberation has not been identified.

Here we present a long-sought-for mechanism for the ascent of kimberlite. Our model provides a simple but unique explanation for the rapid ascent of these deep-seated, high-density, seemingly solids-choked, enigmatic magmas. We use high-temperature analogue experiments to demonstrate the viability of the process and its relevance to kimberlite ascent. We conclude with the broader implications of our model for kimberlite ascent and new insights it provides.

### GEOLOGICAL OBSERVATIONS

One of the great impediments to the ascent of kimberlite magma is the fact that the primary compositions of kimberlite melts, especially their volatile contents, are unknown (Mitchell, 1986; Mitchell, 2008; Sparks et al., 2009). The composition of the melt remains elusive because: i) kimberlite rocks and deposits contain abundant (>30%) mantle and crustal material, ii) quenched kimberlite melt (i.e. glass) is unobserved; and iii) kimberlites are highly susceptible to alteration due to their ultrabasic composition (Mitchell, 1986) and age (Nixon, 1995). Many strategies have been used to estimate kimberlite melt compositions (cf. Sparks et al., 2009) and there is some agreement that they are SiO<sub>2</sub>-poor (<20-35 %), MgO-rich (15-25%), and that the volatile contents are high and dominated by CO<sub>2</sub> and H<sub>2</sub>O (e.g. Mitchell, 1986; Price et al. 2000; Kjarsgaard et al., 2009; Sparks et al., 2009). However, the primary composition of kimberlite melts,

including volatile contents, remains unmeasured or observed. Physical properties estimated for melts within this construct of kimberlite melt compositions include densities similar to basaltic to komatiitic melts (2800-2900 kg m<sup>-3</sup>) and low viscosities (10<sup>-2</sup> – 10<sup>2</sup> Pa s) (e.g. Kurszlauskis et al., 1998; Sparks et al., 2006; Sparks et al., 2009).



**Figure 1.** (a) Polished slab of Diavik kimberlite showing abundance of mantle-derived xenocrysts of ol, cpx, gar and ilm (courtesy of Stephen Moss). (b) Kimberlite containing fragment of disaggregated peridotite comprising grain of partially dissolved Opx within larger Ol grain (courtesy of C. Brett).

Mantle-derived xenoliths transported by kimberlite constrain the depths of formation for these magmas (Boyd and Nixon, 1973; Kopylova et al., 1999) to the base of the cratonic mantle lithosphere (CML) or deeper. Mechanical disaggregation of the xenoliths produces the large volume of xenocrysts characteristic of most kimberlites (Fig. 1). The xenocryst suite is dominated by olivine (>25 vol. %) and accessory amounts of clinopyroxene, garnet, ilmenite and, in rare cases, diamond. Notably orthopyroxene (Opx), which comprises ~15-27 % of the CML (Boyd, 1989; McDonough, 1990; Kopylova and Russell, 2000; Patterson et al., 2009), and is the most silicic mineral present, is rare to absent in kimberlite rocks. Where observed, Opx is clearly in severe disequilibrium (Fig. 1b).



Olivine xenocrysts record an early stage of grain size reduction and rounding; however, olivine overgrowths and phenocrysts indicate late-stage equilibrium with the melt and crystallization (e.g. Mitchell, 1986; Dawson, 1994; Brett et al., 2009). It has been postulated that crystallization of olivine may contribute ~25 vol. % to the total (~50%) crystal load of the magma further increasing the density of the ascending magma (Mitchell, 2008). The range of crystal contents for kimberlite, suggest bulk densities of the magma (melt plus “cargo”) in the range of 3000-3100 kg m<sup>-3</sup>.

Kimberlite magmas are reputed to ascend from the base of the mantle lithosphere (100-120 km) to the Earth's surface substantially faster (e.g. 4-20 m/s; Anderson, 1979; Canil and Fedortchouk, 1999; Sparks et al., 2006; Wilson and Head, 2007) than other xenolith-bearing magmas (0.001 to 0.15 m/s; Sparks et al., 1977). There remains the mechanistic challenge of generating the high ascent rates ascribed to these dense, crystal-rich magmas. The componentry of most kimberlite suggests aggregate magma densities  $\geq 3$  g/cc which would make them 5-10% less dense than the surrounding mantle lithosphere. However, in order to entrain and carry mantle material sampled at 120 km depth to the Earth's surface, we also need sufficient buoyancy to provide magma rise velocities that fully compensate for the settling velocities of the mantle cargo ( $\leq 1$  m s<sup>-1</sup>; Sparks et al., 2006).

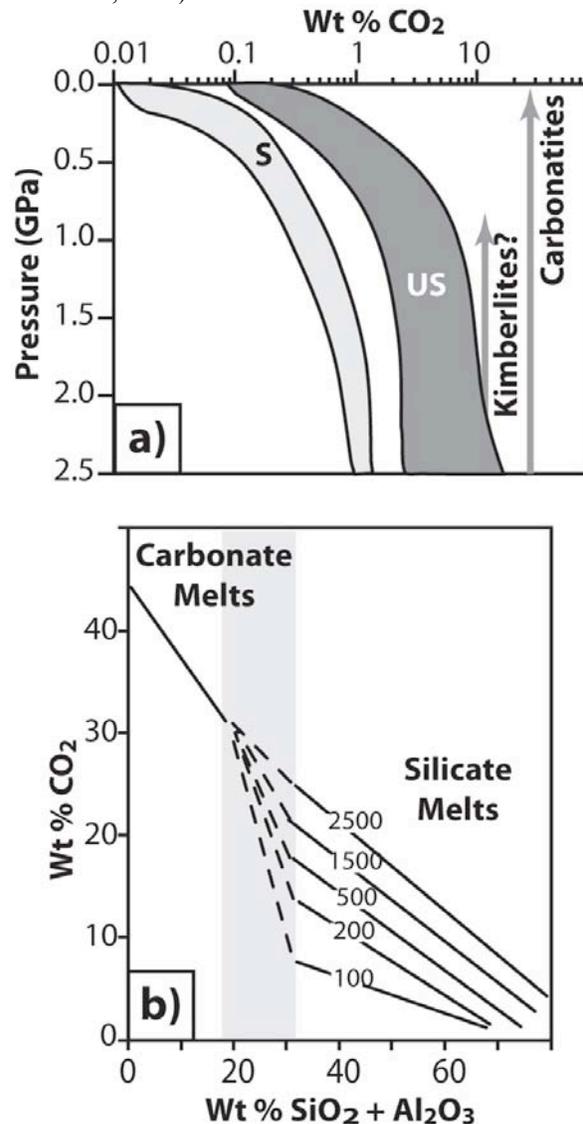
The buoyant and rapid ascent of kimberlite from depths > 200 km can be greatly facilitated by the presence of a low density fluid phase ( $\rho_{\text{CO}_2} \sim 1200$  kg m<sup>-3</sup>;  $P \sim 2$  GPa). Deep-seated exsolution of volatiles would ensure sufficient magma-mantle density contrast to provide buoyancy for rapid ascent from ~ 200km. In fact, the exsolution of dissolved volatiles (CO<sub>2</sub> & H<sub>2</sub>O) is commonly seen as playing a critical role in providing buoyancy, however, the *source* of the volatiles and the physical-chemical process behind their liberation is rarely specified (e.g. Anderson, 1979; Wilson and Head, 2007).

#### A MODEL FOR KIMBERLITE ASCENT

Our model builds on the recent work of Brooker et al. (in press) and Brooker and Kjarsgaard (2010) who provide new experimental data on the solubility of volatiles (i.e. CO<sub>2</sub>) in melts across the silicate-carbonate transition (Fig. 2). These experiments elucidate the pressure and compositional controls on CO<sub>2</sub> solubility in silicic (e.g., rhyolite to basalt to melililite) to carbonate melts (Fig. 2).

Our model is inspired by three main observations from these and other studies. Firstly, CO<sub>2</sub> solubility (+H<sub>2</sub>O) in silicic melts increases with pressure but, importantly, in magmas with SiO<sub>2</sub>+Al<sub>2</sub>O<sub>3</sub> > 35 wt.% is limited to ~10-15%. This precludes extraordinary amounts of volatile being

sequestered in kimberlitic melts (Sparks et al., 2007; Wilson and Head, 2007).

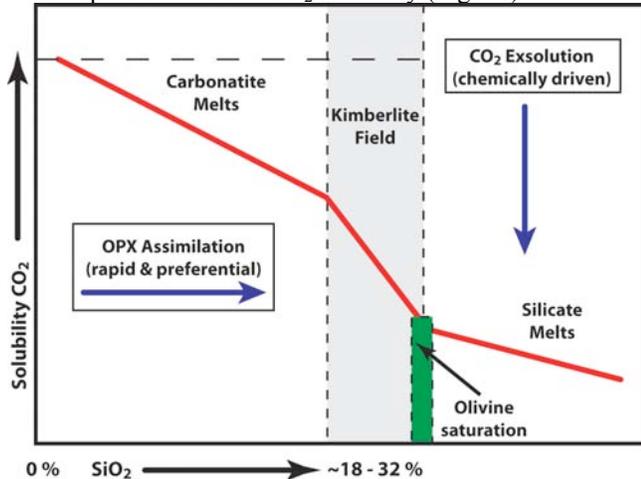


**Figure 2.** Solubility of CO<sub>2</sub> in silicic to carbonate melts (after Brooker et al. 2011). (a) Compilation of CO<sub>2</sub> solubility limits for silica-saturated (S) and undersaturated (US) melts and hypothetical solubilities of carbonatite and kimberlite melt. (b) Pressure and compositional dependence of CO<sub>2</sub> solubility across the carbonate-silicate transition. Solid lines illustrate limited effects of pressure (MPa) on CO<sub>2</sub> solubility vs. the effects of composition.

Secondly, carbonate melts have substantially higher CO<sub>2</sub> solubilities that are essentially independent of pressure (Fig. 2b). For example, CO<sub>2</sub> solubility in carbonate melts can be >40% and is limited only by melt stoichiometry (Hrma, 1985; Dobson et al., 1996; Dalton and Presnall, 1998b, 1998a). Recent work by Keppler (2003) has shown that H<sub>2</sub>O solubility in carbonate melts is also substantially higher than in silicic melts. Carbonate melts can dissolve up to 10% H<sub>2</sub>O at 0.1 GPa without compromising the solubility



of CO<sub>2</sub>. Lastly, the transition from carbonatitic to silicic (i.e. kimberlitic) melt compositions is accompanied by a catastrophic decrease in CO<sub>2</sub> solubility (Fig. 2b).

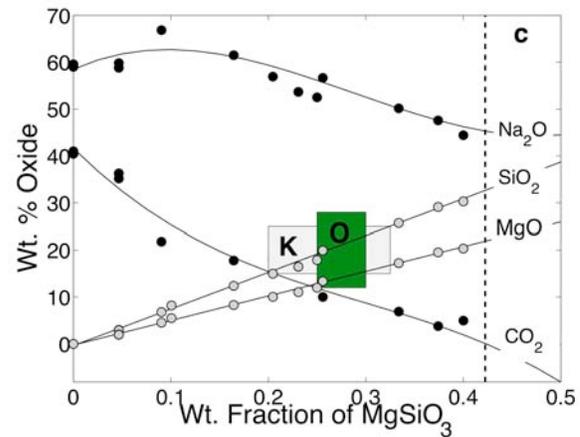


**Figure 3.** Schematic model for assimilation-induced vesiculation of carbonatite. Opx assimilation drives melts to more silicic compositions causing decrease in CO<sub>2</sub> solubility and exsolution of a fluid phase. Continued assimilation causes continued exsolution and results in a kimberlitic melt and, ultimately, olivine saturation.

We suggest that the parental melts to kimberlite may be carbonatitic or near-carbonatitic in composition. The carbonatitic melts begin with near stoichiometric CO<sub>2</sub> contents ( $\geq 40$  wt.%) and are buoyant relative to the mantle ( $\sim 2.8$  vs  $\sim 3.3$  g cm<sup>-3</sup>) at 1-3 GPa (Dobson et al., 1996). Carbonatitic melts entering the cratonic mantle lithosphere initiate the mechanical incorporation and disaggregation of xenoliths (e.g. Lensky et al., 2006) producing the suite of mantle-derived xenocrysts and ensuring that newly liberated minerals are continuously fed into the melt. At these pressures (< 2.5 GPa) and temperatures (>1250 °C), all mantle minerals are out of equilibrium and reacting with the low a<sub>SiO<sub>2</sub></sub> carbonatitic melt. Orthopyroxene, as the most silica-saturated phase, has the highest affinity for dissolution and is assimilated rapidly and preferentially over other phases (Nicholls et al., 1971; Mitchell, 1973; Edwards and Russell, 1998; Shaw, 1999; Mitchell, 2008; Luth, 2009). This process of assimilation drives the melt to more silicic compositions turning the system "kimberlitic". The key to successful ascent of kimberlite is the effect of this assimilation on volatile solubility (Fig. 3). The increased SiO<sub>2</sub> content causes a decrease in CO<sub>2</sub> solubility and immediate exsolution of a fluid phase deep within the CML. The process is gradual until Opx assimilation produces a "kimberlitic" melt composition in excess of ~18 wt.% SiO<sub>2</sub> and causes a catastrophic drop in CO<sub>2</sub> solubility. This results in massive exsolution of a volatile phase that reduces magma density, increases buoyancy, and supports rapid and accelerating ascent.

### High-Temperature Experiments

We present a set of high-temperature (high-T) melting experiments as a test of the inductive ideas advanced above. They are designed to capture these basic concepts and, although not intended to be identical to the natural process, illustrate how fast and efficiently the process is likely to operate during magma transport.



**Figure 4.** Compositions of melts from high-T analogue melting experiments vs. Opx content. Shaded field (K) marks range of SiO<sub>2</sub> contents for putative kimberlite and hypothetical field where melt might support olivine saturation and crystallization.

The experiments involve melting (T~1000-1100 °C) mixtures of Na<sub>2</sub>CO<sub>3</sub> (T<sub>m</sub> = 849 °C) and natural powdered orthopyroxene (Opx, MgSiO<sub>3</sub>) (Fig. 4) and follow the work of Hirma (1985). Two types of experiments were conducted. Transient experiments involved lowering samples (Na<sub>2</sub>CO<sub>3</sub> + Opx) from a balance into a preheated furnace. In these experiments the Na<sub>2</sub>CO<sub>3</sub> powder melts reacts with the admixed powdered Opx to cause strong and immediate effervescence of CO<sub>2</sub>. The melt assimilates the Opx (< 25 mins) creating a more silicic melt oversaturated in dissolved CO<sub>2</sub> and, thus, the melt exsolves (effervesces) the dissolved CO<sub>2</sub> (i.e., decarbonation) causing a weight loss. This reaction continues until the solubility limit of the new more silicic melt composition is reached. The experiment records the change in mass as a function of time.

We used a series of static experiments (Fig. 4) to create an assimilation-induced decarbonation phase diagram for Na<sub>2</sub>CO<sub>3</sub>-Opx melts (Fig. 4) and record the extent of decarbonation (i.e. Δwt. %) driven by the reduction in CO<sub>2</sub> solubility as the melt becomes ever more SiO<sub>2</sub> rich (i.e. Opx content). The SiO<sub>2</sub> and MgO contents of the hybrid melts are greater than the bulk compositions due to enrichment by CO<sub>2</sub> loss. Na<sub>2</sub>O shows an initial increase and then decreases due to the combined effects of dilution (addition of Opx) and enrichment (decarbonation). CO<sub>2</sub> decreases dramatically. The addition of ~42 wt. % MgSiO<sub>3</sub> produces SiO<sub>2</sub>-rich (~32.7 wt. %) melts in which all CO<sub>2</sub> is lost due



to decarbonation. Lastly, we show fields for putative kimberlite based on SiO<sub>2</sub> contents and where the assimilation process might produce an olivine-saturated melt (Fig. 4). The maximum amount of CO<sub>2</sub> that could be released from the magma during assimilation is ~ 25 wt. %.

### Consequences & Implications

Our model requires that kimberlite magmas originate as primary mantle-derived carbonatitic melts. Indeed, recent experimental work shows that the solidus melts of natural and synthetic carbonated (~0.15%) peridotite at > 2.5 GPa will always be CO<sub>2</sub> rich and SiO<sub>2</sub> poor as long as carbonate is stable in the mantle assemblage (Falloon and Green, 1989; Dalton and Wood, 1993). These melts contain in excess of 40% dissolved CO<sub>2</sub> and we suggest that the asthenospheric production of such melts marks the onset of kimberlite ascent. Our model supports geochemical-based models that describe the diversity of kimberlite bulk rock compositions as resulting from mechanical mixing of mantle olivine (70-80%) and a carbonate-rich melt (Canil and Bellis, 2008) combined with assimilation of mantle orthopyroxene (20-30%) (e.g. Patterson et al., 2009).

The assimilation of orthopyroxene is key to this process because it dissolves on timescales that compete with kimberlite ascent rates (Nicholls et al., 1971; Edwards and Russell, 1998; Shaw, 1999; Luth, 2009). The rapid and efficient dissolution of Opx, relative to the other mantle macrocrysts, explains the lack of xenocrystic Opx in kimberlite (Mitchell, 2008). However, the main mechanistic consequence of Opx assimilation is to decrease the CO<sub>2</sub> solubility of the melt thereby driving deep-seated exsolution and effervescence of a CO<sub>2</sub>-rich volatile phase providing buoyancy. Ultimately, the Opx assimilation process also allows for late stage crystallization of olivine and can explain the pervasive olivine overgrowths observed on rounded olivine xenocrysts and the pervasive microphenocrysts of olivine in the groundmass (Mitchell, 1986; Mitchell, 2008; Brett et al., 2009). This crystallization event would logically coincide with the later stages of kimberlite ascent and emplacement and clearly follows on from an earlier and more extensive period of olivine disequilibrium. Our model for kimberlite ascent is unique in that it begins with a melt that contains stoichiometric amounts of dissolved CO<sub>2</sub>. The dissolved CO<sub>2</sub> is stable in the melt across a wide range of P-T conditions and, thus, exsolution of the fluid phase is not driven by depressurization as it is in most magma ascent models (e.g. Spera, 1984). Rather the exsolution of the CO<sub>2</sub> fluid phase is driven only by an inevitable chemical reaction (i.e. assimilation) that changes the melt composition, thereby creating a new melt whose CO<sub>2</sub> solubility is exceeded. Consequently, the production of the fluid phase does not depend on depressurization as the magma transits through the lithosphere but does depend on

access to and assimilation of Opx (see below). Although the initial fluid production is not driven by  $\Delta P$ , the buoyancy of the fluid saturated magma is greatly enhanced by  $\Delta P$  as depressurization will cause volumetric expansion of the fluids. This secondary process would allow for continuous acceleration of the magma, entrainment of greater quantities of mantle material, and decoupling of a CO<sub>2</sub>-rich fluid phase from the ascending magma.

Our model provides an explanation for the linkage between kimberlites and cratons; it is clear that, if the parental magmas are carbonatitic, that a ready source of highly reactive orthopyroxene is needed to create a more silica-rich melt (i.e. kimberlite) and to initiate deep seated vesiculation to provide buoyancy to transport the evolving magma (melt + mantle cargo). The cratonic mantle lithosphere has two attributes that support this transformation (carbonatite to kimberlite): i) firstly it is enriched in Opx (15-30%) relative to other mantle lithosphere (Boyd, 1989; McDonough, 1990; Kopylova and Russell, 2000); and ii) secondly it is thick (90-70 km) and, thus, provides ample opportunity for continuous entrainment of Opx enriched mantle material. As long as the evolving carbonatitic melt is exposed to new Opx, the assimilation-induced exsolution of CO<sub>2</sub> will continue and buoyancy is maintained or increased. Therefore, we might expect the diversity of kimberlite at the Earth's surface to actually be reflective of the underlying mantle lithosphere rather than the source region to the melt (Francis and Patterson, 2009; Tappe et al., 2011).

The corollary is that if Opx were absent this process would not be viable because the dissolution rates for other mantle minerals (which could also promote an assimilation-driven reduction in CO<sub>2</sub> solubility and, hence, fluid exsolution) are too low to keep pace with ascent rate. In addition, the enthalpy of orthopyroxene dissolution is less than the heats of crystallization for olivine which means that Opx assimilation is energetically supported by smaller quantities of olivine crystallization and little cooling (Brett et al., 2009). In parts of the planet where carbonatitic magmas do not transit the cratonic mantle lithosphere there are only two possible outcomes. The carbonatitic melt successfully travels through a relatively thin lithosphere to be emplaced or erupted as carbonatite (e.g. Woolley and Church, 2005), or the carbonatite melt incorporates Opx-poor mantle and crystallizes at depth because it fails to assimilate mantle silicate phases efficiently enough to promote a deep-seated fluid phase and the requisite buoyancy.

### CONCLUSIONS

We suggest that all kimberlites start life as carbonatitic melts produced through partial melting of carbonated peridotite in the subcratonic mantle. The primary carbonatitic melts contain near stoichiometric CO<sub>2</sub> contents



(~40 wt. %). As the melts coalesce and ascend they begin to mechanically sample the mantle lithosphere incorporating peridotite xenoliths. These xenoliths disaggregate and the low  $a_{\text{SiO}_2}$  of the carbonatitic melt ensures that Opx dissolves rapidly and preferentially relative to the other silicate mantle minerals. This increases the silica content of the melt and triggers immediate exsolution of  $\text{CO}_2$ , reducing the buoyancy of the magma and facilitating rapid ascent. The faster the magma rises the more mantle material will be entrained and the more Opx will be dissolved. Providing Opx is available, this mechanism enables the continuous and accelerating ascent of the magma and the evolution of the melt from carbonatitic to kimberlitic compositions.

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