Olivine in Kimberlites: Magma evolution from deep mantle to eruption

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ABSTRACT

Elucidating the origin, composition and physical properties of primary kimberlite melts is crucial to our understanding of their source, petrogenesis, ascent mechanisms and ultimately the origin of diamonds. Recently, there has been a growing interest in the study of olivine, which is one of the most abundant minerals in kimberlites with xenocrystic, metamorphic (mantle) and magmatic origins. Olivine is one of the earliest minerals to crystallise in kimberlite magmas, and presence of ubiquitous zoning (e.g., cores, internal zones, transitional zones, rims, rinds, outmost rinds) and different generations (i.e. primary, pseudosecondary and secondary) crystal/melt/fluid inclusions in euhedral olivine grains has been shown to provide fundamental insights into the composition and evolution of kimberlite melts. In this contribution, we review and evaluate:

i) The widely accepted notion that kimberlite olivine has two distinct origins – xenocrystic and magmatic. We present detailed electron microprobe X-ray element maps of well-preserved and zoned euhedral olivine microcrysts from the Koala and Mark (Lac de Gras, Canada) and Udachnaya-East (Siberia, Russia) kimberlites to show that the cores of olivine occasionally adopt euhedral shapes, which is commonly defined by the distribution of Ni. We present a scenario in which mantle olivine was recrystallised by the early (or proto-) kimberlite melt/fluid infiltrating through the lithospheric mantle to form euhedral ‘pyrocrysts’ (i.e. olivine that formed via re-crystallisation in the mantle olivine in the presence of a melt), which in turn become cores for the subsequent crystallisation of magmatic olivine during kimberlite magma ascent and emplacement.

ii) The evolution of ideas using different geochemical, petrological, experimental and melt inclusion approaches to constrain the composition of the primary/parental kimberlite melt. Based on our assessment of available data, in particular using melt inclusions, we propose that kimberlites originated from melts that were initially Si-poor, and Na-K-F-Cl-P-S-bearing and Ca-Mg-carbonate-rich. With this model composition for the primary/parental kimberlite melt considered, we emphasise the implications for the evolution of olivine and its role in the kimberlite petrogenesis. Furthermore, we present a comprehensive model outlining the key stages involved in the petrogenesis of kimberlites, ranging from the generation of the proto
kimberlite melt in the mantle, its interaction with mantle silicates during ascent, the role of liquid immiscibility in driving magma differentiation and CO$_2$ degassing and its emplacement and modification in the crust. Finally, we discuss prospective directions that may further guide the future of kimberlite petrological research.

**Keywords:** kimberlite; melt inclusion; olivine; pyrocryst; xenocryst

**INTRODUCTION**

Kimberlites are silica-poor, MgO-rich and volatile-rich (CO$_2$ ± H$_2$O) igneous rocks which originate from the deeply-derived magmas (>150 – 200 km; Pearson et al., 2014) that ascend rapidly to the surface (Mitchell, 1986; Sparks, 2013; Mitchell et al., 2019). Despite their rarity, kimberlites have captivated at least a segment of the geoscience community for several decades due to their deep mantle origin, enrichments in incompatible trace elements and the presence of abundant mantle and crustal xenoliths and xenocrysts, especially diamonds. Thus, the study of kimberlite rocks and their entrained mantle cargo can provide some of the most direct and unparalleled insights into the composition of and the processes operating within the subcontinental lithospheric mantle (SCLM) and asthenosphere.

The geochemical and mineralogical (e.g., xenocrystic, magmatic and alteration; Dawson, 1980; Mitchell, 1986; Skinner, 1989; Mitchell et al., 2019) diversity of kimberlite rocks, along with their wide variability in groundmass textures (Scott Smith et al., 2013; Sparks, 2013), has severely inhibited our ability to accurately constrain the composition of their primary (or proto-) melt in the mantle. It has been long established that “there are no hypabyssal kimberlites which are representatives of the initial primary magma; all kimberlites are hybrid and contaminated magmas” (Mitchell, 2008). This is because, once formed, the magmas that give rise to kimberlites undergo significant modification during their ascent to the surface due to: i) the entrainment and/or assimilation of mantle and crustal material, ii) exsolution of volatiles (CO$_2$, H$_2$O), and iii) differential movement of discrete components of the kimberlite magmatic system. Moreover, following their emplacement in the crust, kimberlites are almost always inevitably modified further by pervasive subaerial weathering and the interaction with externally-derived fluids, which results in the overprinting of their primary mineralogy and groundmass textures (Sparks et al., 2006; Stripp et al., 2006; Giuliani et al., 2017).
To date, identification and quantification of these processes and their effects on kimberlite melt modification remains one of the greatest challenges in kimberlite petrology (Kamenetsky et al., 2014a; Soltys et al., 2018b; Tappe et al., 2018; Mitchell et al., 2019; Giuliani et al., 2020).

Recently, our understanding of the composition and evolution of primary/parental kimberlite melts has been greatly advanced through the study of olivine. Olivine is one of the most abundant minerals in kimberlites, typically ranging from 25 – 60 %, or significantly lower in aphanitic varieties (Dawson, 1980; Clement et al., 1984; Giuliani, 2018; Mitchell et al., 2019), and is one of the earliest liquidus phases to form (Mitchell, 1986; Fedortchouk and Canil, 2004; Kamenetsky et al., 2008). Furthermore, olivine, in particular euhedral microcryts, exhibit complex compositional zoning (e.g., Kamenetsky et al., 2008; Bussweiler et al., 2015; Sobolev et al., 2015; Howarth and Taylor, 2016; Giuliani, 2018; Lim et al., 2018; Soltys et al., 2018a; Howarth and Gross, 2019; Abersteiner et al., 2020b; Dalton et al., 2020; Soltys et al., 2020; Shaikh et al., 2021; Tovey et al., 2021) and may contain multiple generations of crystal and melt/fluid inclusions (Kamenetsky et al., 2004, 2009b; Golovin et al., 2007; Abersteiner et al., 2018a), thereby providing detailed insights into the composition and evolution of the parental kimberlite melt at different stages of its evolution.

Kimberlite olivine is considered to have both xenocrystic and magmatic origins (Kamenetsky et al., 2008; Brett et al., 2009; Arndt et al., 2010; Giuliani, 2018; Lim et al., 2018; Shaikh et al., 2021), however, uncertainties remain regarding the exact proportions of xenocrystic olivine relative to magmatic olivine in kimberlite rocks, where some estimates invoke anywhere between 5% and 55% of olivine could be magmatic (Mitchell, 2008; Brett et al., 2009; Bussweiler et al., 2015; Soltys et al., 2018b). In contrast, some authors have argued for a dominantly cognate (i.e. magmatic) origin (e.g., Moore, 1988, 2012, 2017; Moore et al., 2020, 2021). Further complications in kimberlite olivine studies are exacerbated by the mechanical abrasion of grains (e.g., Jones et al., 2014) and their susceptibility to alteration (i.e. serpentinisation), which results in the poor preservation of zoning and inclusions in olivine in the majority of kimberlite localities.

In this study, we revisit the prevailing notion that kimberlite olivine cores are xenocrystic, originating from disaggregated mantle-derived lithologies, such as granular peridotites, and/or metasomatic mantle lithologies, such as megacrysts and sheared peridotites (e.g., Kamenetsky et al., 2008; Nielsen and Sand, 2008; Brett et al., 2009; Bussweiler et al., 2015; Sobolev et al., 2015; Howarth and Taylor, 2016; Moore and
Costin, 2016; Giuliani, 2018; Dongre and Tappe, 2019; Soltys et al., 2020). We present new data on well-preserved and zoned euhedral olivine grains from hypabyssal (i.e. coherent) units of the Koala and Mark kimberlites (Slave Craton, Canada) and the Udachnaya-East kimberlite (Siberian Craton, Russia). High-resolution electron microprobe X-ray maps obtained by wavelength-dispersive spectroscopy (WDS) were employed to illustrate the complex zoning patterns and distribution of major and minor elements in olivine. These results, together with previously obtained data on olivine-hosted crystal and melt/fluid inclusions (i.e. primary, pseudosecondary and secondary), are used to present new insights on the potential origin(s) and evolution of olivine in kimberlites.

Following this, we review and evaluate current data and interpretations on the origin and composition of the primary/parental kimberlite melt. We emphasise the importance of melt inclusion studies in providing the most direct evidence regarding the chemical and physical properties of parental kimberlite melts. Here, we present a comprehensive model and criteria for deducing the composition of primary/parental kimberlite melts and their subsequent evolution through the mantle and crust. With this assessment of the primary/parental kimberlite melt composition, we highlight potential avenues for future kimberlite research so that we may further decipher the origin of these rare, but fascinating rocks.

**Olivine definitions**

Some of the earliest studies of olivine in kimberlites date back to Williams (1932), who identified at least three types of olivine in kimberlites, linking some to xenoliths and others to ‘phenocrysts’ and the groundmass. It was not until the later half of the 20th century that more comprehensive petrographic and geochemical studies of kimberlite olivine were conducted, where the terms macrocrysts (i.e. grains typically larger than 0.5 – 1 mm and rounded-to-anhedral in shape) and phenocryst (i.e. grains typically smaller than 1 mm and subhedral-to-euhedral in shape) to describe the size and shape of individual grains (Boyd and Clement, 1977; Clement, 1982; Mitchell, 1973; Nixon and Boyd, 1973; Skinner and Clement, 1979). This classification was later revised by Kamenetsky et al. (2008) who recognised the compositional similarity between the cores of larger, round-to-angular olivine (olivine-I) and smaller, euhedral olivine (olivine-II), thereby prompting further investigations into the xenocrystic and magmatic origins of kimberlite olivine.
(e.g., Brett et al., 2009; Arndt et al., 2010). Although the terms ‘macrocryl’ and ‘(micro-)phenocrysts’ are often still retained in the literature, these terms no longer carry any genetic connotations (see Giuliani (2018) and references therein). In this study, we retain the term macrocryst and use the term ‘microcryst’ in place of (micro-)phenocryst. In addition to olivine macrocrysts and microcrysts, kimberlites also contain a subordinate population of larger megacrysts (i.e. grains generally larger than 1 cm; Gurney et al., 1979; Harte and Gurney, 1981) and subrounded dunitic nodules (Arndt et al., 2010), which are composed of deformed anhedral grains and recrystallised olivine grains, or ‘neoblasts’ (Shaikh et al., 2021).

Since the 2000s, more advanced and detailed petrographic and geochemical studies of kimberlite olivine have documented the presence of multiple distinct compositional zones, which are defined largely by their forsterite content \(\text{Mg} / (\text{Mg} + \text{Fe}) \times 100\) and NiO, CaO and MnO concentrations, as well as the presence of inclusions (Fig. 2; Giuliani, 2018; Lim et al., 2018; Soltys et al., 2018a, 2020; Howarth and Gross, 2019; Abersteiner et al., 2020b; Dalton et al., 2020). To avoid ambiguities in nomenclature, we present the olivine zoning terminology presented in these aforementioned studies (see also Figure 2): i) Core: the central and compositionally distinct zone common in all olivine grains. ii) Internal zone: overgrowth occurring between the core and rim. iii) Rim: overgrowth surrounding cores or internal zones. iv) Transitional zone: a zone that is diffuse and compositionally intermediate between cores and rims, or cores and internal zones. v) Rind: an overgrowth, usually marked by high-Mg, that typically surrounds rims. vi) Outmost rind: a rare overgrowth that is epitaxial to rinds, and vii) High-Mg olivine: forms along healed fractures cross cutting olivine grains.

GEOLOGY AND PETROGRAPHY

Koala and Mark Kimberlite

The diamondiferous Koala and Mark kimberlites are located in the Ekati cluster of the Lac de Gras Kimberlite field (Slave Craton, Canada; Fig. 1) and were dated to be ~53 Ma and 47.5 Ma old, respectively (Davis and Kjarsgaard, 1997; Creaser et al., 2004).

Detailed descriptions of the local country rock geology and pipe stratigraphy of the Koala kimberlite, the reader is referred to Nowicki et al. (2004) and Porritt and Cas (2011). Furthermore, descriptions of the petrography and mineral assemblages of the sample used in this study can be found in Kamenetsky et al.
The sample exhibits macrocrystic texture, which is defined by abundant (>50 %) olivine, which occurs as large (1 – 10 mm in size) anhedral-to-rounded macrocrysts and as smaller (<1 mm) subhedral-to-euhedral microcrysts. Olivine exhibits minor replacement by serpentine along the grain margins and internal fractures. A large proportion of olivine microcrysts exhibit zoning and commonly contain crystal and melt/fluid inclusions (see also Kamenetsky et al., 2013 and Lim et al., 2018).

Descriptions of the local country rock geology of the Mark kimberlite can be found in Davis and Kjarsgaard (1997). Furthermore, detailed descriptions of the petrography and mineral assemblages of the studied sample of the Mark kimberlite can be found in Abersteiner et al., (2020a, b). The sample is macrocrystic in texture and is defined by abundant (>55 %) anhedral-to-round olivine macrocrysts/megacrysts (0.2 – 1.5 cm in size) and abundant smaller (50 µm – 2 mm) subhedral-to-euhedral olivine microcrysts. In addition, some olivine grains exhibit embayments filled by interlocking mosaic clusters of subhedral-to-euhedral shaped olivine grains (or ‘neoblasts’) ranging from 10 to 200 µm in size (see Abersteiner et al., 2020b). Olivine grains are commonly zoned and contain abundant crystal and melt/fluid inclusions and exhibits excellent preservation (i.e. negligible or very low-degrees of replacement by serpentine along grain boundaries and internal fractures).

Udachnaya-East Kimberlite

The diamondiferous Udachnaya-East kimberlite is located in the Siberian Craton (Russia; Fig. 1) and was dated to be ~365 Ma (Kinny et al., 1997; Kamenetsky et al., 2009c). For detailed descriptions of the pipe structure, stratigraphy and country rocks, the reader is referred to Marshintsev (1986), Kharkiv et al. (1991), Zinchuk et al. (1993) and Abersteiner et al. (2018b).

The studied samples (see also Kamenetsky et al., 2008) of the Udachnaya-East kimberlite were derived from a petrographically and geochemically unique horizon (410 – 500 m) depth that is characterised by exceptionally well-preserved serpentine-free kimberlite (i.e. ‘serpentine-free Udachnaya-East’ - SFUE; Abersteiner et al., 2018b; Kamenetsky et al., 2014a; Kopylova et al., 2016 – see also Sharygin et al., 2003; Kamenetsky et al., 2008, 2012 for detailed descriptions on kimberlite petrography and mineral assemblages).

The samples used in this study are fragments of coherent kimberlite rock, which contains abundant fresh
olivine (up to 60 vol.%). Olivine occurs as larger anhedral-to-round macrocrysts (>0.5mm) and as smaller (<250 µm) euhedral-shaped microcrysts. Rare olivine nodules (several mm in size) and fragments of olivine megacrysts up to 1.5 cm have also been documented (Abersteiner et al., 2019b).

METHODS

Detailed descriptions for sample preparation and analyses for each kimberlite locality can be found in: i) Koala – Kamenetsky et al. (2013), ii) Mark (Abersteiner et al., 2020a, b), iii) Udachnaya-East (Kamenetsky et al., 2008; Abersteiner et al., 2018b). Electron microscope X-ray element mapping of olivine in the Koala, Mark and Udachnaya-East kimberlite (Figs. 5 – 7) were performed at the Central Science Laboratory (CSL), University of Tasmania (see Abersteiner et al., 2020b for methodology). Other olivine EMP X-ray element maps (Fig. 8) from the Udachnaya-East kimberlite were undertaken at the Max Planck Institute for Chemistry (Mainz, Germany; see Kamenetsky et al. (2008) for methodology).

OLIVINE CHEMISTRY

Detailed X-ray element maps of euhedral olivine microcrysts and olivine nodules showing the distribution of Mg, Fe, Ca, Al, Ni, Mn and Cr were produced using EMP-WDS (see Methods). The olivine grains examined were usually exposed at a mid-planar level parallel to the c-axis. The key advantage of X-ray maps is that they provide a more complete two-dimensional cross-sectional view of element distributions in olivine grains than single point or line analyses. Compositional zoning in olivine is marked by variations in forsterite content, which is reflected in BSE images by lighter (Fe-rich) and darker areas (Mg-rich; Fig. 2). The compositions of individual zones within X-ray element maps of olivine from the Koala, Mark and Udachnaya-East kimberlites were extracted and are presented along with previous geochemical data of olivine from the Koala (Kamenetsky et al., 2013; Lim et al., 2018), Mark (Abersteiner et al., 2020b) and Udachnaya-East (Kamenetsky et al., 2008) kimberlites (Fig. 3; Supplementary Tables S1 – 4).

Koala

Four EMP-WDS X-ray element maps of euhedral olivine microcrysts (250 – 400 µm in size) from grain separates were selected from the Koala kimberlite (Fig. 4). Olivine cores are variable in composition in terms
of Fo-content (89.9 – 93.1 mol.%; Fig. 3; Supplementary Table S1, 4) and exhibit higher NiO and Cr₂O₃, and lower CaO and MnO relative to the exterior zones (Fig. 3; Supplementary Table S1, 4). A striking feature of olivine grains is that there may be as many as five distinct cores of variable size and shape within an individual olivine grain (Fig. 4b – d). In one grain (Fig. 4c), there appears to be both a Mg-rich and Fe-rich core within the same olivine grain. The distribution of Ni commonly defines the shape of the core, as it shows the strongest compositional contrast (i.e. higher concentrations) relative to the exterior zones (e.g., internal zones, rims and rinds). The distribution of Ni also shows that the core(s) of olivine range in shape from subhedral to well-developed euhedral (Fig. 4). Moreover, the distribution of Ni is variable within olivine cores, where it is sometimes relatively homogeneous (Fig. 4a, b), but in other cases it is heavily concentrated along the core margins leaving the interior relatively more ‘depleted’ in Ni (Fig. 4c, d).

Olivine cores are surrounded by a rim, which is defined by a narrower range in Fo-content (90.8 – 92.2 mol.%), and lower NiO and higher MnO and CaO relative to the core (Supplementary Table S1; Fig. 3). Olivine rims usually adopt a well-developed euhedral shape and ‘fuse’ together the multiple olivine cores into a single grain (Fig. 4).

Rinds form the outmost zone of olivine grains and are characterized by high Fo-content (92.1 – 95.0 mol.%), very low NiO (<0.17 wt.%), and higher CaO (up to 0.56 wt.%) and MnO (up to 0.32 wt.%) relative to the cores and rims (Supplementary Table S1, 4; Fig. 3). In some cases, these rinds exhibit feint oscillatory zoning patterns that are on the scale of only a few microns thick, which is reflected by minor variations in Fo-content, CaO, NiO and MnO (Fig. 4).

Mark

Three EMP-WDS X-ray element maps of subhedral-to-euhedral olivine microcrysts (200 – 500 µm in size) in situ in the groundmass were selected from the Mark kimberlite (Fig. 5). The sizes and shapes of cores are highly variable (euhedral – Fig. 5a; angular-to-rounded fragments – Fig. 5b, c) and are best illustrated by Ni-distribution, as the compositional change to internal zones are not easily identifiable in BSE images. These cores are characterized by variable Fo-content (~88.1 – 93.6 mol.%), high NiO (0.23 – 0.40 wt.%), marginally elevated Cr₂O₃, and low MnO (<0.17 wt.%) and CaO (<0.05 wt.%; Supplementary Table S2, 4;
Fig. 3). In one case (Fig. 5a), Ni-distribution shows the outline of a euhedral core with an irregularly-shaped inner zone characterized by low-Ni interior zone. Similar to analyses of olivine from the Koala samples, all elements appear relatively homogeneously distributed throughout the core, except for Ni, which in some cases is most heavily concentrated along the core boundary (Fig. 5a, c).

Olivine cores are sometimes surrounded by a rim (Fig. 5b; see also Abersteiner et al., 2020b), which is defined by narrower range in Fo-content (91.4 – 92.1 mol.%), as well as lower NiO and only marginally higher MnO and CaO relative to the core (Fig. 3; Supplementary Table S2, 4). Some olivine grains occasionally exhibit diffuse transitional zones (few tens of microns) and/or discrete internal zones between the core and rim (Fig. 5), which have compositions intermediate between the core and rim.

Olivine rims and/or cores are commonly overgrown by a rind of extremely Mg-rich olivine (Fo: 94.0 – 98.1 mol.%) that is defined by very low NiO (<0.14 wt.%) and appreciably higher MnO (up to 0.39 wt.%) and CaO (up to 1.73 wt.%; Supplementary Table S2, 4; Fig. 3) and extremely dense clusters of melt/fluid inclusions (see Abersteiner et al., 2020b). These melt/fluid inclusions in olivine rinds are usually marked by anomalously high Ca and Al (e.g., daughter minerals of carbonates, apatite, phlogopite) in X-ray element maps (Fig. 6).

Olivine rinds (and sometimes rims) are surrounded by an outmost rind, which has intermediate compositions between the rims and rind (Fo: 92.7 – 95.4 mol.%; Figs. 3, 5; Supplementary Table S2, 4), and is devoid of any inclusions.

Udachnaya-East

Four (at UTas; see methods) and three (at Mainz; see methods) EMP-WDS X-ray element maps of euhedral-to-subhedral olivine microcrysts (300 – 500 µm in size) from grain separates were selected from the Udachnaya-East kimberlite (Figs. 6, 7). The cores of olivine microcrysts show wide variability in Fo-content (85.5 – 94.4 mol.%) and exhibit high NiO (up to 0.46 wt.%) and low CaO (<0.16 wt.%) and MnO (<0.22 wt.%) contents relative to surrounding zones (Fig. 3; Supplementary Table S3, 4). In some cases, up to four distinct cores of variable composition (both containing Fe-rich and Mg-rich types) were distinguished (Fig. 6b, d). The Fe-rich cores in Figure 6b are an exception, where the Ni is depleted relative to the surrounding
internal zone. The distribution of Cr$_2$O$_3$ in olivine cores shows that it is marginally more depleted in some cores (e.g., Fig. 6a, b, d) relative to exterior zones, and in other cores (e.g., the Mg-rich core in Fig. 6c) is more enriched in Cr relative to the exterior zones. Similar to in the Koala and Mark kimberlites, olivine cores from Udachnaya-East are clearly identifiable by the distribution of Ni. The distribution of Ni is usually uniform along the core boundary, but in some cases (Figs. 6a, c) forms a diffuse boundary. EMP X-ray element maps also showed that Ni distribution can be unperturbed by the transition between different zones (Figs. 6d, 7b, c), where there are significant changes in Fo-content. Here, this Ni-enriched zone is homogeneously distributed and extends beyond the Fo-defined core boundary and adopts its own a euhedral shape independent of Fe-Mg.

Olivine cores are surrounded by an internal zone, which is characterized by generally higher FeO and lower NiO. Noteworthy is that Ni appears slightly more heavily concentrated along the peripheries of internal zones (e.g., Fig. 6a, c). In some cases, the concentrations of MnO may be unperturbed by the transition from core to internal zone (e.g., Fig. 6a, c). In cases where there are multiple olivine cores (Fig. 6b, d), this internal zone fuses these cores into a single grain. This internal zone usually adopts a very well-developed euhedral shape.

Internal zones in olivine microcrysts are surrounded by a rim, which is defined by a constant and narrow range in Fo-content (88.5 – 89.6 mol.%), and notably lower NiO (<0.31 wt.%), and marginally higher MnO (up to 0.19 wt.%) and CaO (up to 0.15 wt.%; Fig. 3; Supplementary Table S3, 4) compared to the cores and internal zones.

**Olivine nodules**

The Mark (Fig. 8a – c, e, f) and Udachnaya-East (Fig. 8d) kimberlites are characterized by the presence of a very small population (i.e. <1%) of multigranular olivine ‘nodules’ (see definitions by Arndt et al., 2010 and Shaikh et al., 2021), which are large (100s of μm in size and even >1 cm) olivine grains that are partially (e.g., Fig. 8a, b, f) to completely (e.g., Fig. 8c – e) composed of interlocking smaller (<10 μm to 100s of μm in size) asymmetric olivine grains, which have been referred to as ‘tablets’ or ‘neoblasts’ (e.g., Arndt et al., 2010; Shaikh et al., 2021). In olivine nodules that are partially composed of neoblasts, individual neoblasts
are randomly distributed and orientated throughout the nodule (e.g., Fig. 8a, b, f) or form tightly packed and interlocking clusters.

Two EMP-WDS X-ray element maps of in-situ olivine nodules were selected from the Mark kimberlite. One of the olivine nodules (Fig. 8e) is completely composed of olivine neoblasts, whereas the second one (Fig. 8f) is only partially composed of olivine neoblasts. Both X-ray element maps revealed olivine neoblasts to be compositionally homogenous and identical to each other (i.e. Fig. 8f).

INCLUSIONS IN OLIVINE

Since the early 2000s, there has been an increasing interest in the study of crystal and melt/fluid (i.e. primary, secondary and pseudosecondary) inclusions in euhedral olivine grains as a method of deciphering the composition and evolution of the primary/parental kimberlite melt. These studies have been restricted to kimberlite localities where olivine is well-preserved, and includes studies of the Koala (Kamenetsky et al., 2013), Mark (Abersteiner et al., 2020b) and Udachnaya-East (Kamenetsky et al., 2004, 2007a, 2008, 2009a, 2012, 2014a; Golovin et al., 2007; Mernagh et al., 2011; Abersteiner et al., 2018b) kimberlites (Table 1). Primary melt/fluid inclusions and crystal inclusions are entrapped by the host olivine crystal from the medium in which it forms, thereby providing 'snapshots' of the parental melt at a particular stage of its evolution. Secondary melt/fluid inclusions are formed if the host crystal is fractured, whereby the surrounding melt/fluid can enter and become entrapped as the fracture heals. Pseudosecondary inclusions are broadly intermediate between primary and secondary inclusions. These are formed if the host crystal becomes fractured during growth, thus appearing petrographically similar to secondary inclusions (Roedder, 1984). Secondary inclusion trails cross-cutting olivine usually intersect the boundary of olivine grains in contact with the groundmass, whereas pseudosecondary melt inclusion trails terminate at the olivine rind or rind interface (Figs. 2, 9). After their entrapment, melt/fluid inclusions are potentially shielded from external modification and can thus provide insights into magma differentiation prior to eruption, late-stage crystallization and post-magmatic alteration (e.g., Lowenstern, 2003). In this section, we present an overview of the different types and chemistry of inclusions hosted in olivine from the Koala, Mark and Udachnaya-East kimberlites.
Crystal Inclusions

Two distinct types of inclusions crystal (i.e. monomineralic) inclusions occur in kimberlite olivine macrocrysts and microcrysts: i) minerals typical of the lithospheric mantle, and ii) minerals typical of the kimberlite groundmass (Table 1).

The first type of crystal inclusions are extremely rare (< ~1% of olivine grains; e.g., Kamenetsky et al., 2009a) and consist mostly of clinopyroxene (Cr-diopside to Cr-omphacite) and orthopyroxene (enstatite), along with subordinate Cr-pyrope and rare picroilmenite, low-Ti Cr-spinel and subhedral olivine (e.g., Fig. 9a – c; Kamenetsky et al., 2008, 2009a; Sobolev et al., 2015; Moore and Costin, 2016; Giuliani et al., 2017; Abersteiner et al., 2020b; Shaikh et al., 2021). In addition, olivine cores are sometimes associated with randomly distributed globules of Fe-Ni-Cu sulphides, which may represent monosulphide solid solutions (MSS) and/or intermediate sulphide solid solutions (ISS). Silicate and oxide crystal inclusions are typically round-to-elongate in shape and variable in size (up to 500 µm), and were noted to be generally restricted to the Mg-rich olivine cores (Soltys et al., 2020). Kamenetsky et al. (2009a) noted that in some olivine grains from the Udachnaya-East kimberlite, orthopyroxene inclusions may also occur along the diffusive boundary between the core and rim (or, using the revised definitions – the transitional zone) or even within the rim (see Figure 7 of Kamenetsky et al., 2009a). Furthermore, some of these orthopyroxene inclusions were shown to associate with other mineral, melt and fluid phases (Kamenetsky et al., 2009a), where upon heating, recrystallised to form complex assemblages of orthopyroxene rimmed by clinopyroxene, along with silicate glass and CO₂-rich bubbles.

Cr-diopside inclusions in olivine cores show compositional heterogeneity from inclusion to inclusion and exhibit a general overlap in terms of major and trace element chemistry with Cr-diopside megacrysts and Cr-diopside in granular peridotite xenoliths (Kamenetsky et al., 2009a; Sobolev et al., 2015; Soltys et al., 2020). Kamenetsky et al. (2009a) noted that in olivine from the Udachnaya-East kimberlite, low-Mg# Cr-diopside inclusions contain appreciably higher concentrations of CaO and lower TiO₂ than Cr-diopside from mantle xenoliths. In some cases, individual Cr-diopside inclusions in olivine exhibit compositional heterogeneity, where the Mg#, CaO increase and Na₂O, Cr₂O₃ and Al₂O₃ decrease from core to rim (Kamenetsky et al.,
In contrast, analytical traverses of clinopyroxene inclusions in olivine from the Colossus kimberlite revealed no such compositional zoning from core to rim (Moore and Costin, 2016). In fractured or partially altered (i.e. serpentinised) olivine grains, the margins of pyroxene inclusions in contact with the host kimberlite groundmass are partially-to-completely resorbed, where they are replaced by secondary assemblages of MUM spinel, monticellite or phlogopite (Kamenetsky et al., 2009a; Russell et al., 2012; Soltys et al., 2020). Furthermore, some pyroxene inclusions in olivine from the Udachnaya-East kimberlite contain multiphase inclusions of alkali-bearing carbonates and chlorides that cluster along the margins of the pyroxene host, or align along cleavage planes (Kamenetsky et al., 2009a). Cr-pyropes inclusions (e.g., Fig. 9c) in olivine do not show any pronounced compositional zoning but are commonly altered along the grain peripheries and internal fractures to kelyphitic assemblages.

The second type of crystal inclusions in olivine are confined to the rims, rinds or internal zones surrounding the core (Fig. 2, 9d – n). The most common minerals found in olivine rims are euhedral-shaped Cr-spinel (Fedortchouk and Canil, 2004; Kamenetsky et al., 2008; Soltys et al., 2018a; Abersteiner et al., 2020b), which sometimes show minor compositional zoning towards titanian magnesian aluminous chromite (TIMAC) or magnesian ulvöspinel-magnetite (MUM) compositions (i.e. typical of the magmatic spinel trend; Roeder and Schulze (2008). In addition, euhedral grains of TIMAC/MUM spinel, perovskite, phlogopite, ilmenite and rutile are also common. These crystal inclusions in olivine rims are compositionally analogous to their magmatic mineral counterparts in the host kimberlite groundmass (e.g., Koala – Kamenetsky et al., 2013; Mark – Abersteiner et al., 2020; Udachnaya-East – Kamenetsky et al., 2008; Abersteiner et al., 2018b). The Mg-rich rinds in olivine from the Mark kimberlite host abundant crystal inclusions of minerals that are typical of the host kimberlite groundmass, such as spinel (MUM, pleonaste, Mg-magnetite), monticellite, perovskite, periclase, apatite and phlogopite (see Abersteiner et al., 2020b; e.g., Fig. 9d).

**Melt/Fluid Inclusions**

Fluid-dominated inclusions in kimberlite olivine are ubiquitous and typically occur along healed fractures crosscutting olivine grains and are thus interpreted to be secondary or pseudosecondary in origin. Fluid
inclusions are typically small (<5 – 10 µm in size), round in shape and consist of variably dense CO₂ bubbles that sometimes show evidence of decrepitation (e.g., Sobolev et al., 1989; Kamenetsky et al., 2004, 2008; Golovin et al., 2007; Brett et al., 2009; Abersteiner et al., 2020b).

Melt inclusions are common in kimberlite olivine, where they mostly occur as secondary or pseudosecondary trails aligned along healed fractures cross cutting olivine (Figs. 2, 9; Table 1). Secondary and pseudosecondary melt inclusions are typically interconnected by thin channels, indicating that they were potentially modified by ‘necking down’ (see Roedder, 1984). Melt inclusions are sometimes surrounded by patchy and uneven zones that are very Fo-rich (up to 98.9 mol.%; or ‘Mg-rich zones’), along with high CaO, MnO and low NiO contents (e.g., Abersteiner et al., 2018b; Soltys et al., 2020). Primary melt inclusions have only been confidently identified in the rims of olivine from the Bultfontein kimberlite (Giuliani et al., 2017), Koala kimberlite (Kamenetsky et al., 2013) and in Mg-rich olivine rinds in the Mark kimberlite (Abersteiner et al., 2020b).

In general, melt inclusions in olivine are extremely variable in size (up to 50 – 150 µm in size) and shape (e.g., round, irregular or elongate), where they may consist of both fluid (e.g., CO₂) and daughter mineral assemblages. Combined SEM-EDS and Raman analyses demonstrated that regardless of kimberlite locality or generation (i.e. primary, pseudosecondary or secondary), olivine-hosted melt inclusions are dominated by a diverse array of Ca-Mg, alkali (Na, K) and alkali-earth (Ba, Sr) bearing carbonates. In addition, alkali-bearing chlorides, phosphates (e.g., apatite) and sulphates are also common, but are generally subordinate to carbonates. Other common daughter minerals include oxides (e.g., various types of Fe-Mg-Cr-Ti-Al-bearing spinel, perovskite, periclase), Fe-Ni-Cu (±K-Cl) sulphides (e.g., djerfisherite) and subordinate silicates (e.g., monticellite, humite/clinohumite, (tetraferri)phlogopite (Fig. 9; Golovin et al., 2007; Mernagh et al., 2011; Giuliani et al., 2017; Abersteiner et al., 2018a, 2018b, 2020b; Kamenetsky et al., 2004, 2009b, 2012, 2013).

Heating experiments (1 atm) of pseudosecondary/secondary melt/fluid inclusions in olivine from the Udachnaya-East and Koala kimberlites showed that homogenization is typically achieved between 660 – 800 °C (Fig. 10; Kamenetsky et al., 2004, 2009b, 2013; Golovin et al., 2007). Despite the variability of inclusion sizes and host grains, similar behavior was recorded in these melt/fluid inclusions during heating and cooling
(i.e. melting, immiscibility, homogenization and re-crystallization as heterogeneous mineral assemblages).

Similar observations were made during identical thermometric analyses of olivine-hosted melt/fluid inclusions in the Aaron, Jericho and Leslie kimberlites (Lac de Gras, Canada) and Majuagaa kimberlite dyke (Greenland; Kamenetsky et al., 2009b). No quenched or crystallized silicate melts were identified in these melt/fluid inclusions from kimberlite olivine, with the exception of the rare case of heated orthopyroxene inclusions associated with glass in the Udachnaya-East kimberlite (Kamenetsky et al., 2009b) and reports of quenched glass-rich polynmineratic inclusions in olivine megacrysts from the Monastery kimberlite (South Africa; Howarth and Büttner, 2020).

DISCUSSION

The ubiquity of olivine in kimberlites, along with its extensive crystallization history recorded by chemical zoning and inclusions, renders it an invaluable petrological tool for examining the origin, composition and evolution of kimberlite magmas. In the following sections, we first review the established ideas on the origin(s) of kimberlite olivine and then discuss the implications of newly obtained data from EMP-WDS element maps, along with data on crystal and melt/fluid inclusions, from the Koala, Mark and Udachnaya-East kimberlites. Following this, we evaluate the potential origin(s) of olivine, with a special emphasis on the origin of cores, and ultimately its link to the evolution of the host kimberlite magma. To illustrate our ideas in the sections presented below, we present a schematic diagram depicting the evolution of olivine and the host kimberlite magma, ranging from its ascent through the mantle to emplacement in the crust (Fig. 12).

Origin of olivine in kimberlites: An overview

It has been long established that the cores of kimberlite olivine, regardless of morphological type (i.e. macrocryt, microcryt), are compositionally homogeneous, but exhibit widespread intergranular variability (Mitchell, 1973; Nixon and Boyd, 1973; Kamenetsky et al., 2008; Brett et al., 2009; Arndt et al., 2010; Bussweiler et al., 2015; Howarth and Taylor, 2016; Giuliani, 2018; Lim et al., 2018; Abersteiner et al., 2020b; Shaikh et al., 2021). Recently, extensive geochemical datasets on olivine core compositions have subdivided them into three broad, but overlapping compositional categories, which is based on Mg#, NiO, MnO and CaO contents: i) ‘Mg-rich’ olivine cores (Mg#~89 – 94; CaO <0.05 wt.%), which are generally the
most dominant compositional type. Subordinate populations of: ii) ‘Mg-Ca-rich’ (Mg# >89; CaO >0.05 wt.%) cores, and iii) ‘Fe-rich’ cores (Mg# <89 and as low as Mg# ~78; Kamenetsky et al., 2008; Howarth and Taylor, 2016; Giuliani, 2018; Lim et al., 2018; Soltys et al., 2020). Superseding earlier studies that suggested groundmass olivine was largely of magmatic origin based on its ‘phenocrystic’ shape (e.g., Boyd and Clement, 1977; Mitchell, 1986; Moore, 1988), a strong contingent of geochemical studies have assigned a xenocrystic origin for the cores of both olivine macrocrysts and microcrysts (Kamenetsky et al., 2008; Brett et al., 2009; Arndt et al., 2010; Pilbeam et al., 2013; Bussweiler et al., 2015; Giuliani, 2018; Dongre and Tappe, 2019; Soltys et al., 2020).

One of the main lines of evidence presented for the xenocrystic origin for olivine cores is their close compositional overlap with entrained peridotitic mantle xenoliths, which indicates that olivine cores originated from disaggregated mantle lithologies (see Fig. 11). The fragmented-to-rounded and/or embayed shapes exhibited by many olivine cores are also viewed as evidence of their disequilibrium within the host kimberlite magma, where they were subject to partial chemical resorption (Kamenetsky et al., 2008; Brett et al., 2015; Giuliani, 2018), likely in combination with mechanical abrasion during magmatic ascent (Jones et al., 2014). These xenocrystic olivine cores are considered to be nucleation points for the subsequent crystallisation of magmatic olivine (e.g., internal zones, rims, rinds, outmost rinds), which precipitated directly from the host kimberlite melt under varying conditions of increasing oxygen fugacity and evolving melt composition (Kamenetsky et al., 2008; Brett et al., 2009; Arndt et al., 2010; Pilbeam et al., 2013; Bussweiler et al., 2015; Cordier et al., 2015; Sobolev et al., 2015; Howarth and Taylor, 2016; Giuliani, 2018; Lim et al., 2018; Abersteiner et al., 2020b; Soltys et al., 2020). Recently, Giuliani et al. (2020) demonstrated a strong linear trend correlating the Mg# of olivine cores and rims, and the relative abundance of kimberlite-metasomatised cores from kimberlites worldwide, indicating that kimberlite melts were broadly similar prior to lithospheric mantle assimilation.

The intragranular compositional diversity of olivine cores (e.g., Mg-rich, Ca-Mg-rich, Fe-rich cores) within different kimberlite localities, even within individual rock samples, suggests that these olivine cores originated from multiple sources, including: i) granular peridotite xenoliths derived from the cratonic mantle (e.g., Mg-rich olivine cores), and ii) mantle lithologies linked to precursor (or proto-) kimberlite melt-related
metasomatism, such as megacrysts and dunites (e.g., Fe-rich olivine cores) and sheared peridotites (e.g., Mg-Ca-rich olivine cores; e.g., Moore and Belousova, 2005; Kamenetsky et al., 2008; Nielsen and Sand, 2008; Brett et al., 2009; Bussweiler et al., 2015; Sobolev et al., 2015; Howarth and Taylor, 2016; Moore and Costin, 2016; Giuliani, 2018; Howarth and Giuliani, 2020; Soltys et al., 2020) or possibly from earlier crystallised products of ‘failed’ kimberlite intrusions in the mantle (e.g., polymict breccias; Giuliani et al., 2014b). Moreover, olivine cores revealed δ18O compositions typical of the Earth’s mantle (Giuliani et al., 2019).

Additional data supporting a xenocrystic, mantle-derived origin for olivine cores in kimberlites is the presence of inclusions of high-pressure lithospheric mantle minerals (e.g., pyroxene, garnet; Kamenetsky et al., 2008, 2009a; Arndt et al., 2010; Sobolev et al., 2015; Soltys et al., 2020; Shaikh et al., 2021), which exhibit compositional overlap with their mineral counterparts in granular peridotites (Moore and Lock, 2001; Gibson et al., 2008). Olivine cores containing inclusions of clinopyroxene and/or garnet have been ascribed a lithospheric mantle origin, where they underwent metasomatism by the host kimberlite melt prior to magmatic ascent (Sobolev et al., 2015), or were derived from the Cr-rich megacryst suite (Moore and Costin, 2016). Furthermore, deformation features in kimberlite olivine cores, such as undulose extinction, kink banding and mosaic recrystallisation predating the formation of magmatic overgrowths (e.g., rims) are commonly presented as evidence of solid-state deformation in the mantle (Skinner, 1989; Arndt et al., 2010; Brett et al., 2015; Cordier et al., 2016).

In stark contrast to the prevailing notion advocating a xenocrystic origin for kimberlite olivine cores, a series of studies (Moore, 1988, 2012, 2017; Moore et al., 2020, 2021) have proposed that the majority of cores are cognate, where they crystallised over a wide P-T interval ranging from the mantle source to the surface. This series of publications highlighted some inconsistencies in the classification of xenocrystic olivine cores in kimberlites, such as: i) the compositional overlap between kimberlite olivine cores and olivine of mantle peridotite origin does not necessarily imply a mutual origin, as olivine crystallising from magmas in equilibrium with the mantle olivine would be expected to have a similar composition (Moore, 2017; Moore et al., 2021), ii) there is sometimes a disparity between kimberlite olivine core compositions and associated mantle peridotites within the same kimberlite locality (Moore et al., 2021), iii) a magma crystallising olivine
in equilibrium with mantle peridotite would produce olivine with overlapping Mg# and Ni-concentrations and that the sharp compositional changes observed at the core-rim interface of olivine grains could be explained in terms of abrupt increases in Ni partition coefficient (olivine-liquid) due to decreasing temperature; iv) deformation textures commonly observed in olivine cores cannot be a reliable indicator of a mantle origin as routinely assumed, but may also develop at shallower, crustal depth (Moore et al., 2020). Thus, the complexity of kimberlite olivine cores and interpretation of their origin is still outstanding.

**A new perspective on the origin of olivine cores**

As outlined in the previous section, kimberlite olivine cores have been broadly broken down into two distinct origins, where they are considered to derive from disaggregated mantle lithologies and/or form as cognate phenocrysts. In the following sections, we discuss new evidence to support links between the early (or ‘proto-’) kimberlite melt and the crystallization of euhedral olivine grains, including their cores. The combined use of WDS EMP X-ray element maps, along with systematic documentation of the different types of inclusions, in euhedral kimberlite olivine grains is used to present an additional view for their origin.

**Euhedral Olivine Cores**

Data on euhedral olivine microcryst cores from the Koala, Mark and Udachnaya-East (e.g., Figs. 4 – 7) kimberlites show that their compositions broadly overlap with: i) Mg-rich olivine, and ii) Fe-rich olivine (i.e. Mg# <89; e.g., Fig. 11). Based on the compositions of these olivine cores and their geochemical overlap with mantle lithologies, it would be tempting to assign a xenocrystic origin to them based purely on these criteria. However, we note that olivine microcryst cores are sometimes defined by subhedral-to-euhedral shapes, which are best defined by Ni distribution (Figs. 4 – 7). Interestingly, previous studies suggest that euhedral-shaped olivine cores are a relatively rare feature in kimberlites (e.g., Moore, 2012, Howarth and Gross, 2019; Moore et al., 2021). For example, Soltys et al. (2020) documented that <0.1% of grains had euhedral-shaped cores in their study of the Bultfontein kimberlite (South Africa). This apparent rarity of euhedral-shaped olivine cores could be attributed to the difficulty in their identification through conventional techniques (e.g., contrast differences in BSE imaging and the two-dimensional cross sections produced by analytical traverses).
The significance of these euhedral-shaped cores in olivine microcrysts is that they do not appear to be consistent with a common disaggregated mantle xenolith (e.g., granular peridotite) source, where olivine grains in mantle xenoliths typically display a wide variety of shapes and textures (e.g., deformed and undeformed, mosaic, porphyroclastic) and grain habits (e.g., anhedral; Harte, 1977; Agashev et al., 2013; Doucet et al., 2013; Soltys et al., 2016; Kargin et al., 2017b; Golovin et al., 2019). Furthermore, the cores of olivine microcrysts in this study (Figs. 4 – 7) appear more symmetrical than the polyangular-to-angular core shapes shown by EMP X-ray element maps for some olivine microcryst cores from the Benfontein kimberlite sills (Howarth and Gross, 2019). The closest analogies that could be related to these euhedral-shaped olivine cores could be: i) disaggregated olivine neoblasts from sheared peridotite xenoliths (e.g., Tappe et al., 2021; Drury and Van Roermund, 1989), which are composed of fine-grained interlocking clusters of small (~<100 µm – 400 µm) and undeformed euhedral olivine ‘neoblasts’. ii) Multigranular dunitic olivine ‘nodules’ (Arndt et al., 2010), which are relatively rare but widespread features in kimberlites (e.g., Kangamiut, Greenland; Diavik, Canada; Igwisi Hills, Tanzania; Brett et al., 2009; Arndt et al., 2010; Cordier et al., 2015; Shaikh et al., 2021), aillikites (Rooney et al., 2020) and orangeites (Howarth and Nembambula, 2021), as well as Mark (Abersteiner et al., 2020b) and Udachnaya-East (Fig. 8). Dunitic nodules are partially-to-completely composed of asymmetric equant olivine ‘tablets’ or ‘neoblasts’ that exhibit variable shapes, ranging from sub- to euhedral (Arndt et al., 2010; Shaikh et al., 2021). Although there are no robust estimates on the abundance of these dunitic nodules in kimberlites, our assessment of the Koala, Mark and Udachnaya-East kimberlites suggest that they comprise only a very small fraction of the olivine population (e.g., ≤ 1 vol.%). Based on the euhedral shapes of some individual neoblasts in dunitic nodules (Fig. 8), the disaggregation of these nodules may be a potential source for the euhedral cores in some olivine grains, where the complete disaggregation of a single nodule could disperse tens or even hundreds of olivine neoblasts into the kimberlite magma. X-ray element mapping of dunitic nodules from the Mark kimberlite (Fig. 9) showed that individual neoblasts are compositionally homogenous and do not exhibit any zoning patterns reminiscent to what we observed in olivine microcryst cores.

Decoupling between Fe-Mg and Ni in Olivine Cores
Variations in Fo-content in olivine usually show a close and systematic coupling with Ca, Mn and Cr (Figs. 4 – 6), whereas the relationship between Fo and Ni-content between individual olivine grains is more complex. For example, the distribution of Ni in olivine can display either close coupling with Fo-content (i.e. variations in Fo correspond with changes in Ni-content; e.g., Fig. 4b, d, 5b; 6a, b – see Fe-rich cores, c) or decoupling from Fo-content (i.e., Ni distribution is independent from changes in Fo; Fig. 4a, c – see Fe-rich core, 6a, c, 7b – Mg-rich core, d; 7). Although this decoupling relationship between Fo and Ni distribution has been documented in numerous studies of zoning in kimberlite olivine (see Nixon and Boyd, 1973; Kamenetsky et al., 2008; Bussweiler et al., 2015; Cordier et al., 2015, 2017; Sobolev et al., 2015; Giuliani and Foley, 2016; Howarth et al., 2016, 2019; Moore, 2017; Soltys et al., 2020), X-ray element maps obtained in this study reveal this relationship between Fo and Ni may be more complex than previously envisaged.

This decoupling is best illustrated where there are systematic changes in Fo-content, along with Ca and Mn (e.g., internal zones, rims), whereas the distribution of Ni content appears almost completely unperturbed (e.g., Figs. 4a, c, 5a, c, 6b, d, 7). In several cases, Ni-distribution exhibits a euhedral (i.e. orthorhombic) shape that extends beyond the “Fo-defined” core boundary into outer zones, such as the transitional zone, internal zone or rim. It is noteworthy that the boundaries of Ni distribution in the core are sometimes ‘invisible’ in BSE imaging. In addition, the distribution of Ni throughout olivine cores can be extremely heterogeneous between individual olivine grains. For example, Ni distribution may be uniform throughout the cores of some euhedral olivine grains (Fig. 4a, 5b, 6a, b, 7), whereas in other cores it may be extremely heterogeneous, where it often concentrates along the peripheries of the core, leaving the interior of the core relatively ‘depleted’ in Ni (Fig. 4b, c, d, 5a, c). Consequently, this complex and seemingly capricious behaviour of Ni distribution within each individual olivine grain adds yet another caveat when interpreting the crystallisation of olivine in kimberlites.

The diversity of Ni distribution patterns in euhedral olivine grains in our three studied kimberlite localities suggests that olivine experienced complex crystallisation histories, likely coupled with varying degrees of re-equilibration. For example, olivine cores where Fo, Ca, Mn and Ni distribution is homogenous (Figs. 5b, 6b) may indicate limited diffusion (or re-equilibration) of elements occurred, whereas cores with gradational changes (e.g., diffuse boundaries or higher concentrations of Ni along the core boundaries; Figs. 4b, c, d, 5a,
c, 6a, c, d) may represent zones where protracted diffusion occurred, indicating that there was partial re-equilibration between the olivine core and the host kimberlite melt (e.g., Howarth and Gross, 2019). This compositional ‘accumulation’ of Ni along zone boundaries, in particular in the olivine core, could be attributed to the stalling of Ni-diffusion from the olivine core into the host kimberlite melt.

Experimental studies of trace elements in mantle-derived peridotitic olivine demonstrated that the concentrations of elements, such as Mg, Fe and Mn, can be homogeneously distributed in olivine, whereas the behaviour of Ni and Cr show decoupling from other elements and exhibit significant heterogeneity across olivine grains (Jackson and Gibson, 2018). Furthermore, the diffusion of Ni can be strongly influenced by the silica activity of the host melt, where Ni diffusion is much slower in Si-poor melts, such as those that give rise to kimberlites (Zhukova et al., 2014; Jackson and Gibson, 2018). Noteworthy is that there have been no detailed parameters for the diffusion rates of Ni in the carbonate-rich melts. Interestingly, no compositional accumulations of other major and minor elements (e.g., Fe, Mg, Ca, Mn or P) were observed along the boundaries of olivine cores (Figs. 4 – 7; see also Sobolev et al., 2015; Howarth and Gross, 2019). This may reflect the faster diffusion rates of these elements (i.e. carbonate compatible elements) into the kimberlite melt, whereas the diffusion of Ni in olivine was significantly slower as it is incompatible in carbonate-rich melts.

Olivine pyrocrysts: An alternative origin for olivine cores in kimberlites

In order to explain the origin of euhedral-shaped cores in olivine microcrysts, we propose a scenario whereby the euhedral cores in olivine grains formed via re-crystallisation of mantle olivine, likely in the presence of a melt and/or fluid. In our proposed model, we adopt the term ‘pyrocryst’ to describe these euhedral mantle-like olivine cores. Although this term was previously applied by Scott-Smith et al. (2013) in kimberlite literature to describe a “crystal pyroclast completely separated during pyroclastic emplacement processes from the original host kimberlite melt before solidification”, we consider a more fitting definition to be by Herzberg et al. (2015) to describe olivine grains that were “accidental crystals that were plucked from the residual peridotite melting” and formed via the re-crystallisation of mantle olivine in the presence of a melt. An example that we consider illustrates this process is the recrystallisation of mantle olivine (as well as other
phases, such as clinopyroxene and Cr-spinel) from a peridotitic xenolith (see Supplementary Figure S1) hosted in basalt is from the Tolbachik volcano (Kamchatka, Russia). Here, xenolith-forming olivine, clinopyroxene and Cr-spinel show evidence of partial re-crystallisation, where they exhibit subhedral-to-euhedral morphologies. This process of recrystallisation probably occurred following the xenolith’s entrainment in the host basalt magma and was facilitated by the infiltration of the host basaltic melt, which crystallised as glass within xenolith-forming mineral interstices. The varying degrees of core-to-rim zoning in olivine grains from this xenolith (Supplementary Figure. S1b) is dependent on the size of the grains, where smaller grains appear compositionally homogenous compared to the more strongly zoned larger grains.

In the case of kimberlites, this melt-facilitated recrystallisation process could be analogous to the formation of multigranular olivine (or dunitic) nodules (i.e., Arndt et al., 2010; Cordier et al., 2015; Shaikh et al., 2021; Fig. 8). Although the origin of these dunitic olivine nodules in kimberlites is still debated, their formation has been linked to: i) ‘defertilization’, which involves the removal of pyroxenes and garnet from peridotite due to reactions with the proto-kimberlite melt in the mantle lithosphere (Arndt et al., 2010; Cordier et al., 2015); ii) coarse-grained peridotites and olivine megacrysts (Giuliani and Foley, 2016; Moore, 2017); iii) melt/fluid assisted recrystallisation of olivine in magmatic conduits in the mid-lithospheric to lower cratonic mantle (Shaikh et al., 2021). Regardless, the involvement of a precursor (or proto-) kimberlite melt is inferred to be a prerequisite in the formation of these dunitic nodules. In addition, similar processes have been suggested for the formation of euhedral and undeformed olivine neoblasts in sheared peridotite xenoliths, where their origin is linked to fluid-assisted static recrystallisation (Drury and Van Roermund, 1989). In contrast to the widely spaced olivine grains and melt-filled interstices, along with the varying degrees of zoning within individual olivine grains (Supplementary Figure S1) in the Tolbachik xenolith example, we attribute the highly compact and interlocking nature of olivine neoblasts in dunitic nodules (Fig. 8) and sheared peridotites to a high-pressure origin, whereby complete re-equilibration of individual neoblasts was achieved, and interstitial melt was essentially ‘squeezed out’.

In our view, the ascent of the proto-kimberlitic melt into the lithospheric peridotitic mantle may have initiated this process of pyrocryst formation, similar to or even the same as the process described for dunitic olivine nodule formation, whereby pre-existing mantle olivine underwent re-crystallisation to form
subhedral-to-euhedral morphologies. Based on the compositional diversity of kimberlite olivine cores, including pyrocrysts, it is likely that olivine from various mantle lithologies was sampled prior to this recrystallisation (i.e., pyrocryst formation) event. An alternative scenario could be that these subhedral-to-euhedral pyrocrystic olivine cores may represent neoblasts derived from disaggregated dunitic olivine nodules. The dislocation or disaggregation of individual subhedral-to-euhedral olivine neoblasts from these dunitic nodules during kimberlite magma ascent and emplacement may have produced ‘seed’ cores for the subsequent crystallisation of magmatic olivine overgrowth. Furthermore, the dislocation of olivine neoblasts from dunitic nodules during the latter stages of kimberlite magma ascent could in part explain the excellent preservation of their euhedral shapes, as opposed to many mantle-derived xenocrysts, which commonly show evidence of mechanical and chemical abrasion (see Giuliani, 2018 and references therein).

Although our study demonstrates the presence of kimberlite olivine grains with well-developed euhedral cores, those examined represent only a carefully selected subset of the olivine population that could be assigned a pyrocrystic origin. Therefore, we cannot place any quantitative estimates on their potential abundances within their respective host kimberlites. In addition to these euhedral-shaped olivine cores, the origin of olivine cores with more fragmented or irregular-to-rounded shapes (e.g., Figs. 4c, 5, 6c) are more ambiguous, as they could represent either xenocrysts or even pyrocryst cores that underwent different evolutionary paths, associated with differing degrees of growth, dissolution, recrystallisation, diffusion and/or abrasion during magmatic ascent (e.g., Kamenetsky et al., 2008; Brett et al., 2015; Giuliani, 2018).

**Inclusions in olivine: Insights into composition and evolution of kimberlite melts**

To our knowledge, no primary melt inclusions have been reported in kimberlite olivine cores, however, cores are commonly pervaded by abundant trails of secondary and/or pseudosecondary melt inclusion trails (Fig. 9; Kamenetsky et al., 2004, 2008, 2012; Golovin et al., 2007; Abersteiner et al., 2020b). These secondary/pseudosecondary melt inclusions in kimberlite olivine, regardless of the host locality, are consistently shown to be dominated by alkali-bearing (Na, K) carbonates, chlorides and phosphate daughter minerals, along with oxides, sulphides/sulphates and subordinate H₂O-bearing and silicate minerals (Table 1; Kamenetsky et al., 2004, 2008, 2009b, 2012; Golovin et al., 2007; Mernagh et al., 2011; Abersteiner et al.,
The intersection between these secondary/pseudosecondary melt inclusion trails with crystal inclusions (e.g., pyroxene, garnet) in olivine cores are commonly marked by disequilibria features in the crystal inclusion, such as core-to-rim zoning in Cr-diopside and kelyphite rims around Cr-pyrope (Kamenetsky et al., 2008; Sobolev et al., 2015; Moore and Costin, 2016; Abersteiner, 2021; Fig. 9a–c). This indicates that the infiltrating kimberlite melt was highly reactive with these lithospheric mantle mineral inclusions. However, due to the secondary and/or pseudosecondary origin of these melt inclusions in olivine cores, constraints on the timing of kimberlite melt entrapment is uncertain (i.e. whether they represent early proto-kimberlitic melt infiltration at mantle depths or more evolved melts sampled during ascent and/or emplacement).

The Mark kimberlite is an extremely rare case demonstrating the entrapment of abundant primary melt inclusions within the Mg-rich rinds around olivine (Abersteiner et al., 2020b). Based on fluid densities obtained from decrepitated melt-fluid inclusions in olivine rinds from the Mark kimberlite, these inclusions were estimated to have been entrapped at upper crustal levels of at least ~6 – 15 km. Similar to secondary/pseudosecondary melt inclusions in olivine, primary melt inclusions in olivine rinds were also shown to be largely dominated by identical daughter mineral assemblages (i.e. alkali-carbonate dominated and silica-poor). In addition to studies of kimberlite olivine-hosted inclusions, similar daughter mineral assemblages were documented in polymineralic inclusions in kimberlite-hosted megacrysts (e.g., clinopyroxene, olivine, zircon, ilmenite; Kamenetsky et al., 2014b; Abersteiner et al., 2019b), as well in secondary melt inclusion trails in olivine from sheared peridotite xenoliths (Golovin et al., 2017, 2018 2019; Sharygin et al., 2021), which are interpreted to represent some of the deepest derived mantle rocks that originate from localized deformation zones across the expanse of the lithosphere-asthenosphere transition zone (Tappe et al., 2021). These melt inclusions were interpreted to form due to the early entrapment of kimberlite melt following in-situ fracturing of the host mineral either in the mantle or during transport in the kimberlite magma to the surface (Sharygin et al., 2021).

Analogous studies were undertaken on primary melt inclusions hosted in magmatic kimberlite groundmass minerals (e.g., Cr-spinel, monticellite, apatite, perovskite; Kamenetsky et al., 2013; Abersteiner et al., 2017, 2018a, c, 2019a; 2020a; Giuliani et al., 2017), where they produced similar results, showing that these
magmatic host minerals entrapped a broadly similar, but often more evolved (i.e. more enriched in carbonates, alkalis and chlorine; see Abersteiner et al., 2017) daughter mineral assemblages to olivine-hosted melt inclusions.

In summary, melt inclusions in kimberlite olivine present some of the earliest and most direct insights into the composition of the parental kimberlite melt, as well as insights into its subsequent geochemical evolution. Kimberlite olivine-hosted melt inclusions broadly demonstrate that parental kimberlite melt was silica-poor and Ca-Mg carbonate-rich, with variable but high concentrations of alkalis (Na, K), halogens (F, Cl), P and S.

PERSPECTIVES ON THE COMPOSITION OF KIMBERLITE MELTS

During the last 20 years, our understanding of the composition of kimberlite melts and magma petrogenesis has greatly advanced through the study of kimberlite olivine morphology, chemistry and zoning, and entrapped inclusions. These advancements follow a general hiatus in kimberlite petrological and volcanological research during the late 1980’s and throughout the 90’s, which was punctuated by the seminal works of (Mitchell, 1986) and (Mitchell, 1995). The sparked revival of interest in kimberlite research was catalyzed by the discovery of the late-Cretaceous to Eocene age Lac de Gras kimberlites (Pell, 1997; Graham et al, 1999) in Canada during the 1990s. The Lac de Gras kimberlites have been well revealed by mining and, in some cases, contain well-preserved olivine and groundmass mineralogy, features that are rarely found in many other kimberlite provinces around the world.

Although significant strides to develop our understanding of kimberlite petrogenesis have been undertaken, the story linking the origin of olivine to the early stages of kimberlite melt generation through to emplacement remains incomplete. In the following sections, we present and discuss our current and previously discussed views on the composition and evolution of kimberlite magmas.

Geochemical approaches for constraining kimberlite melt compositions

Perhaps one of the greatest challenges in study of kimberlite petrogenesis is our evaluation of the origin and composition of their primary or proto-kimberlitic (i.e. melt that formed in the mantle at which it segregated
from the source region and is no longer in equilibrium with it) and *parental* (i.e. melt that evolved by fractionation to eventually crystallised the kimberlite rock) melt. Resolving this is a crucial step that underpins our ability to construct a holistic model of kimberlite petrogenesis. The ambiguities surrounding the nature of the primary kimberlite melt stems from the inherent complexity of its evolution (i.e. the melts that give rise to kimberlites could be considered a continuously changing entity as they undergo extensive modification) and the hybrid mineralogy of kimberlite rocks (Giuliani et al., 2019; Mitchell et al., 2019). Consequently, kimberlite rocks, even “aphanitic” varieties, cannot be considered pristine analogues of their primary melts because of these complexities and has thus hampered the traditional approach of using whole-rock chemistry to characterise the primary/parental kimberlite melt composition. Nevertheless, this approach is still in use and describes the parental kimberlite melt to be broadly ultramafic (~20 – 31 wt.% MgO), silica-poor (~21 – 32 wt.% SiO₂) and contain widely varying volatile (H₂O ~6 – 9.4 wt.% and CO₂ ~5 – 17 wt.%) abundances (see Price et al., 2000; le Roex et al., 2003; Becker and le Roex, 2006; Kopylova et al., 2007; Kjarsgaard et al., 2009). Refinements to this whole-rock approach have attempted to more accurately discriminate and quantify processes of kimberlite melt contamination, volatile exsolution and post-magmatic alteration, where they have suggested a parental kimberlite melt composition that is ‘transitional silicate-carbonate’ (see Nielsen and Sand, 2008; Arndt et al., 2010; Pilbeam et al., 2013) with even lower concentrations of SiO₂, MgO and H₂O, and higher CaO and CO₂ (up to 10 – 20 wt.%) and alkalis than previous estimates (e.g., Soltys et al., 2018b; Dongre and Tappe, 2019). Even if these compositions are ultimately accepted, there are still contradictory issues about them, as these parental melt compositions:

i) Are considered to be too Mg-rich to have been in equilibrium with mantle source rocks (Kopylova et al., 2007). Furthermore, the kimberlite whole-rock compositions are evidently low in “basaltic” components (i.e. low Al₂O₃ and Na₂O and high CaO/Al₂O₃) and are undersaturated in pyroxenes and feldspathoids, which are common amongst other silicate melts.

ii) Require high degrees of partial melting (>30 – 40%; see Kamenetsky et al., 2012) of a peridotitic mantle source. This is inconsistent with the enriched trace element characteristics of kimberlites, which likely indicates very low-degree partial melting of a volatile fluxed (e.g., CO₂, H₂O) and garnet-bearing mantle source in an incipient melting regime (see Tappe et al., 2018).
Furthermore, the high temperatures required for producing ultramafic melts is inconsistent with the preservation of diamonds in kimberlites (Fedortchouk et al., 2005) and absence of contact metamorphism in surrounding wall-rocks and entrained organic material (e.g., Mitchell, 1986; Kamenetsky et al., 2012), indicating relatively low temperatures of emplacement ~<700°C.

iii) Are unable to accommodate measured whole-rock volatile (H_2O, CO_2) concentrations, especially at low pressure (Sparks et al., 2009; Brooker et al., 2011; Moussallam et al., 2014, 2016).

iv) Do not explain the high buoyancy and ascent rates of kimberlite magmas, as olivine-laden Mg-rich magmas are dense (kimberlite rock density is 3.0 – 3.1 g cm^-3; Russell et al., 2012).

v) Are unable to satisfactorily explain the low concentrations of Na (typically <~0.1 wt.%) in kimberlite rocks, as primary kimberlite melts produced with such low Na would not be in equilibrium with mantle clinopyroxene (Kamenetsky et al., 2014a; Stamm and Schmidt, 2017).

In recognition of the inherent complexities during kimberlite magma petrogenesis and emplacement, and post-magmatic alteration, a series of studies on the Udachnaya-East kimberlite provided new insights into the origin, composition and physical parameters (e.g., viscosity, temperature) of parental kimberlite melts (Kamenetsky et al., 2004, 2007b, 2008, 2009a, b, c; Golovin et al., 2017, 2018, 2019; Abersteiner et al., 2018b). These studies were centred on a petrologically unique SFUE horizon within the Udachnaya-East pipe that contains, in addition to minerals typical of the kimberlite groundmass worldwide (e.g., olivine, spinel, perovskite, monticellite, phlogopite, apatite), abundant Na-K-Cl-S-rich minerals (carbonate, chloride, sulphate/sulphide, sodalite; Shirygin et al., 2003; Kamenetsky et al., 2004, 2012; Kopylova et al., 2016).

This led to the notion that this part of the Udachnaya-East kimberlite is “pristine” (i.e. unaffected by secondary alteration), where, based on its combined geochemistry, mineralogy, radiogenic (Sr-Nd-Pb; Maas et al., 2005; Kamenetsky et al., 2009; Kitayama et al., 2021) and stable (C-O-S; Kamenetsky et al., 2007b, 2012; d’Eyrames et al., 2017; Kitayama et al., 2017) isotope characteristics, “originated in the mantle as a chloride-carbonate liquid, devoid of ‘ultramafic’ or ‘basaltic’ aluminosilicate components, but became olivine-laden and olivine-saturated by scavenging olivine crystals from the pathway rocks and dissolving them en route to the surface” (Kamenetsky et al., 2008). However, a drawback of using the Udachnaya-East as a standard composition for the parental kimberlite melt is that it is petrologically unique amongst
kimberlites worldwide, as no other kimberlites contain abundant alkali-C1-S-rich minerals in the groundmass.

**Experimental and petrological approaches for constraining the composition of kimberlite melts**

In order to elucidate the nature of the primary/parental kimberlite melt, various indirect approaches have been employed. These have largely been based on: i) experimenting with putative melt compositions, and ii) petrological modelling of mineralogical and geochemical (e.g., based on trace element composition of equilibrium melts) modification of xenoliths and xenocrysts entrained by kimberlite magmas during the ascent through the SCLM to the surface.

One such experimental method attempts to explain the inferred buoyant and rapid ascent of kimberlite magmas (e.g., Canil and Fedortchouk, 2006; Sparks et al., 2006; Sparks, 2013; Russell et al., 2019), which is commonly attributed to continuous exsolution of CO$_2$ (Wilson et al., 2007; Russell et al., 2012). Both atmospheric pressure (Russell et al., 2012) and high pressure (Kamenetsky and Yaxley, 2015) experiments have suggested that an initially alkali-bearing (or dolomitic; see Wallace and Green, 1988) carbonatitic melt could be a suitable candidate for the primary kimberlite melt, as it could effectively react with mantle silicates (e.g., orthopyroxene), exsolving CO$_2$ and thereby driving magmatic ascent. However, these melt compositions have been deduced through *a priori* rationale rather than demonstrated empirically.

Experimental studies have similarly provided constraints to model the reactions and behavior of putative silica-poor and alkali-carbonate-rich kimberlite melt compositions under source P-T conditions and mantle lithologies (e.g., Stone and Luth, 2016; Sharygin et al., 2017; Stamm and Schmidt, 2017; Sokol and Kruk, 2021), however, experiments alone do not provide direct evidence for the existence of such a melt composition (see also Foley et al., 2019).

The petrological approach examines metasomatised xenolith and xenocryst suite minerals, which were entrained by kimberlite magmas during their ascent through the SCLM to the surface, and includes:

i) PIC (phlogopite-ilmenite-clinopyroxene) rocks, lherzolites, wehrlites (Fitzpayne et al., 2018a, b, 2019; 2020) and polymict breccias (e.g., polymict breccias; Giuliani et al., 2014b), which are
considered to have formed due to intense metasomatism of mantle peridotites by earlier pulses of
‘failed’ kimberlite magmas (i.e. stalled at mantle depths).

ii) Entrained mantle-derived peridotite and eclogite xenoliths and xenocrysts, which show variable
petrographic and geochemical evidence of metasomatism by melts presumed to be related to the
host kimberlite melt at different stages of its evolution (e.g., Spetsius and Taylor, 2002; Misra et
al., 2004; Giuliani et al., 2012, 2013; Bussweiler et al., 2016; Giuliani and Foley, 2016; Kiseeva
et al., 2016; Soltys et al., 2016; Fitzpayne et al., 2018b; Lim et al., 2018; Smart et al., 2021).

iii) Large (cm to >20cm in size) megacryst suite minerals (e.g., olivine, orthopyroxene,
clinopyroxene, phlogopite, garnet, ilmenite, zircon; e.g., Gurney et al., 1979), which are
interpreted to have formed due to have crystallised from from proto-kimberlitic melts prior to, or
contemporaneous with kimberlite magmatism (e.g., Bell and Moore, 2004; Hops et al., 1992;
Kostrovitsky et al., 2004; Howarth et al., 2019; Nkere et al., 2021), or formed as a reaction
product of multi-stage metsomatism between proto-kimberlite melts and the lithospheric mantle
(Kopylova et al., 2009; Kamenetsky et al., 2014a; Kargin et al., 2017a; Bussweiler et al., 2018;
Abersteiner et al., 2019b; Cone and Kopylova, 2021; Kargin, 2021; Tappe et al., 2021).

iv) Pyrocryst olivine cores (this study) and dunitic multi-granular nodules (Arndt et al., 2010;
Shaikh et al., 2021), which have been linked to the proto-kimberlite melt interacting with mantle
peridotite.

In summary, these aforementioned petrological studies of mantle-derived xenogenic cargo in kimberlites
highlight the volatility, mobility and reactivity of early (proto) kimberlite melts as a powerful metasomatic
agent in the mantle and crust. However, the composition of these inferred kimberlite melts is generally
poorly defined with widely varying inferences on their SiO$_2$, alkalis and volatiles (H$_2$O and CO$_2$) contents
(e.g., Giuliani et al., 2012, 2013; Kamenetsky et al., 2014a; Kargin et al., 2017; Abersteiner et al., 2019b;
Howarth et al., 2019; Cone and Kopylova, 2021; Kargin, 2021).

Melt inclusion approach for constraining the composition of kimberlite melts
In order to circumvent the inherent complexities, especially post-magmatic alteration, associated with kimberlite rock petrogenesis, the study of melt inclusions is an additional means of analysing the composition of the parental kimberlite melt at different stages of its magmatic evolution. It has been shown that regardless of kimberlite locality (e.g., southern Africa, Siberia, Canada, Finland) or origin (i.e. primary vs. secondary) of the melt inclusions in kimberlite minerals, the melt entrapped by these inclusions is generally devoid of “ultramafic” and “basaltic” aluminosilicate components, H₂O-poor, Na-K-Cl-S-bearing and Ca-Mg-carbonate-rich. These melt inclusion compositions have been argued to be representative of the parental kimberlite melt and even provide insights into the composition of the primary melt (Kamenetsky et al., 2009b, 2012; Golovin et al., 2019). In addition, fibrous diamonds entrained by kimberlite magmas have been shown to contain four broad compositional endmembers: high-Mg and low-Mg carbonatitic, silicic and saline high-density fluids (HDFs; e.g., Navon et al., 1988; Weiss et al., 2009, 2015 and references therein), which are all enriched in alkalis. Although there is no direct evidence linking diamond and kimberlite formation to the same parental melt/fluid, there are numerous similarities (e.g., Ca, K, Na contents) between saline HDFs in diamonds and those in kimberlite mineral-hosted melt inclusions, suggesting similar enrichments in alkalis in their mantle source.

There is an evident mismatch between the daughter mineralogy of melt inclusions and that of the host kimberlite groundmass, where phases common in kimberlite-hosted minerals, such as alkali-carbonates and chlorides, are notably rare or absent from the groundmass of kimberlites. The serpentine-free units of the Udachnaya-East kimberlite provides one such example that reconciles this paradox, as there is a strong correlation between the mineralogy of melt inclusions in xenocrystic and magmatic kimberlite minerals and that of the groundmass (Abersteiner et al., 2018b, c, 2021; Kamenetsky et al., 2004, 2008, 2009a, b; 2012, 2014a; Golovin et al., 2007, 2017, 2018, 2019). This has led to the notion that melt inclusions in kimberlite minerals may indeed be representative of the parental medium.

What was the composition of the primary kimberlite melt?

Although we cannot provide a quantitative reconstruction of the primary kimberlite melt composition in this study, we emphasise the impact of numerous geochemical, experimental and melt inclusion studies which
have progressively narrowed down the potential range of compositions for the primary and parental Kimberlite melt compositions. These studies have steadily shied away from the early notion that the parental Kimberlite melt was H₂O-rich ultramafic and silicate-poor in composition, and instead shifted towards a (and in our preferred melt composition model) potentially Si-poor Ca-Mg-carbonate-rich (or even carbonatitic) melt, which contained appreciably higher concentrations of alkalis (e.g., Na, K), halogens (F, Cl), phosphorus and sulphur, than what is typically measured in the Kimberlite whole-rock. In contrast to a silica-poor ultramafic primary melt composition, an initially carbonate-rich melt can more adequately explain numerous chemical and physical properties of Kimberlite magmas, including:

i) Enrichment in incompatible elements, which is the result of melting of low-degrees of partial melting.

ii) Metasomatism of mantle lithologies, which is commonly associated with the carbonate-rich (proto-) Kimberlite melt. Moreover, an initially carbonate-rich Kimberlite melt supports theoretical, experimental and petrographic studies that show mantle orthopyroxene was unstable and dissolved into the Kimberlite melt during its ascent through the mantle.

iii) Low temperatures of Kimberlite magma emplacement (as evidenced by the lack of thermal contact metamorphism of surrounding wall-rocks). Heating experiments of melt inclusions in Kimberlite olivine show that homogenisation can occur as low down as ~660 – 800°C (Figure 10; Golovin et al., 2007; Kamenetsky et al., 2004, 2009c, 2013, 2014a).

iv) Low melt viscosity and high buoyancy of Kimberlite melts (Kamenetsky et al., 2007a).

v) Liquidus assemblage minerals (e.g., perovskite, phlogopite, apatite, carbonate) are consistent with crystallisation from an alkali-bearing carbonate melt.

vi) Primitive Kimberlites and carbonatites occurring together in space and time (e.g., Kimberlite dykes and carbonatite sheets occurring at Tikusaaq, West Greenland - Tappe et al., 2017; and Premier Kimberlite pipe and carbonatite dykes, South Africa; Tappe et al., 2020)

In summary, a complete understanding of the primary Kimberlite melt remains far from fully resolved, and that significant strides still need to be undertaken to more accurately disentangle the complex interplay of
contamination processes that modify kimberlite melts during their ascent to the surface and after emplacement.

KIMBERLITE PETROGENESIS: PROSPECTIVE RESEARCH

Based on progress in our understanding of the primary/parental kimberlite melt composition, we may suggest pathways for future studies that may clarify the following points:

1) Geochemical trends in mantle minerals
   - So far, we have identified three kimberlite localities (Koala, Mark and Udachnaya-East) that contain olivine pyrocryst cores. We speculate these pyrocryst olivine cores may be a common occurrence in kimberlites worldwide and advocate further examination of olivine grains using EMP X-ray elemental mapping (including phosphorus), possibly in conjunction with laser ablation inductively coupled plasma mass spectrometry (LA-ICPMS), as an additional technique for their identification and geochemical fingerprinting.
   - Microanalyses of reaction textures xenocrystic minerals in kimberlites (e.g., kelyphite rims around xenocrystic garnet, replacement of xenocrystic orthopyroxene by clinopyroxene, disequilibrium features in olivine) as to deduce the composition of the reacting host melt.
   - Experiments to: i) improve characterisation of element (e.g., Fe, Mg, Ca, Mn, Ni, Cr, Al P) partitioning between olivine (e.g., Gordeychik et al., 2020), as well as other mantle-derived minerals, and carbonate-rich melts at different P-T conditions and oxygen fugacity to assist in understanding diffusion chronometry and the composition of the melt that these minerals were in equilibrium with. ii) Model the interaction between Ca-Mg-carbonate-rich melts (with varying concentrations of alkalis (Na, K) and halogens (Cl, F)) and different types of mantle lithologies, as well as individual mantle minerals (e.g., olivine, pyroxenes, garnet), at differing P-T conditions. Experiments should take into account compositional parameters obtained from melt inclusion constraints from kimberlite minerals and kimberlite-hosted xenocrysts.

2) Melt inclusions as a proxy for reconstructing primary/parental kimberlite melt composition
- As stated by Pearson et al., (2019), “no single kimberlite sample is likely to represent a primitive melt composition because of its hybrid nature”. We suggest additional studies of melt inclusions entrapped in deep mantle-derived xenocrysts and xenoliths from more kimberlites, including areas previously unstudied in detail (e.g., South America, Australia, Antarctica, India, China). This will assist in providing some of the earliest and most direct insights into the composition of the parental kimberlite melt (Golovin et al., 2019) and help ascertain geochemical variability in the source region underlying different cratons. Furthermore, homogenisation experiments of melt inclusions may assist in ascertaining temperatures of melting, observing immiscibility textures and producing more homogenous compositions after quenching.

- The implementation of additional analytical techniques previously under-represented in the study of melt inclusions in kimberlite minerals. This could include, but is not limited to: i) LA-ICPMS for the purpose of bulk-inclusion analyses and their trace element budget. ii) Secondary ion mass spectrometry (SIMS) for in-situ determination of trace element chemistry and/or C-O isotopes in daughter minerals in melt inclusions, especially carbonates.

3) **The role of liquid immiscibility in driving magma differentiation and volatile exsolution**

In addition to liquid immiscibility being a potential process responsible for driving kimberlite magma differentiation (i.e. promoting magma buoyancy) and CO$_2$ exsolution, we speculate that the unmixing of the kimberlite melt into silicate globules suspended within a carbonate melt matrix could assist in explaining: i) the survival of euhedral-shaped olivine pyrocrysts, and ii) the formation of transitional zones and/or unevenly zoned olivine cores. Firstly, olivine pyrocrysts suspended within silicate globules may be shielded from mechanical abrasion during magmatic ascent, as well from chemical disequilibria (i.e. dissolution) as they are suspended within silicate-carbonate melt globules. Secondly, silicate-carbonate liquid immiscibility experiments demonstrate that olivine suspended in carbonate-silicate globules exhibit transitional zones between the cores and rims that are analogous to those observed in natural samples (see Kamenetsky and Yaxley, 2015). Transitional zones in kimberlite olivine have been considered to result from metasomatism induced by kimberlite or kimberlite-related (i.e. proto) melts, which were likely linked to re-equilibration of olivine with the ascending kimberlite melt (Howarth and Taylor, 2016; Lim et al., 2018) or previous pulses
of kimberlite melt (e.g., Cordier et al., 2015; Soltys et al., 2020). We speculate that olivine grains, which exhibit unevenly zoned cores (e.g., diffusely zoned on one particular side but abruptly zoned on the other; e.g., Figs. 4c, 5a, c, 6a, d), could be explained by olivine grains adhering to the peripheries of these immiscible carbonate-silicate globules during magmatic ascent. Here, the side of the olivine grain in contact with the surrounding carbonate melt matrix may cause diffusive re-equilibration.

In order to test these aforementioned hypotheses, as well as elucidate the potential role of liquid immiscibility in kimberlite petrogenesis, we suggest the following avenues for future research:

- Petrographic studies to search for possible evidence of liquid immiscibility textures in kimberlite rocks. One such example could be magmaclasts, which is a general term to describe discrete spheroidal-to-ovoid fragments (ranging in size from 1 – 100 mm) of kimberlite material, where they contain a core fragment (e.g., mineral or xenolith) that is surrounded by concentrically aligned kimberlite material (e.g., Ferguson et al., 1973; Field and Scott Smith, 1998, 1999; Mitchell et al., 2009; Abersteiner et al., 2017). Magmaclasts have also been referred to as nucleated autoliths and globular segregations and are relatively widespread feature in hypabyssal kimberlites (e.g., Danchin et al., 1975; Mitchell, 1986; Abersteiner et al., 2017) and other types of alkaline rocks (e.g., aillikites; Tappe et al., 2006). Although magmaclasts are broadly considered to form due to fragmentation/segregation of kimberlite magmas prior to emplacement (Danchin et al., 1975; Wilson et al., 2007; Scott Smith et al., 2013), we propose an alternative hypothesis to suggest that kimberlite magmaclasts may represent relics of liquid immiscibility (see Lloyd and Stoppa, 2003) in many kimberlites worldwide. We present a few example photographs from the Legkaya, Komsomol’skaya-Magnitnaya and Leningradskaya kimberlites (Siberia, Russia; see Supplementary Figure. S2; see also Fig. 4 in Kamenetsky et al., 2013 and Fig. 5 in Soltys et al., 2015), where they contain abundant rounded globules that vary widely in size from millimetre to a few centimetres in size which are suspended in a carbonate-rich kimberlite matrix. These globules commonly contain central macrocrystic mineral (e.g., olivine, garnet) or crustal xenolith (Supplementary Figure. S2) core(s) that is surrounded by a largely silicate-carbonate kimberlite matrix. In some cases, these globules show evidence of sintering, indicating that they were in a liquid or semi-liquid state.
High P-T (i.e. mantle conditions) experimental studies using our proposed primary/parental kimberlite melt composition (i.e. Ca-Mg-carbonate-rich with the addition of varying concentrations of alkalis (Na, K) + halogens (Cl, F)) should aim to model its interaction with different lithospheric mantle lithologies in order to constrain the behaviour of liquid immiscibility and the extent that CO₂ is exsolved from the melt due to compositional changes (i.e. increasing silica activity) and changes in pressure.

4) The modification of the kimberlite melt during emplacement and crystallisation

Upon emplacement in crust, the kimberlite magmas undergo further modification, where simultaneous and sometimes counteracting processes operate to change the melt composition, including:

i) Stratification of the kimberlite magma is inferred to occur in the propagating dyke tip during ascent (Russell et al., 2012; Brett et al., 2015). In some cases where kimberlites were emplaced intrusively, such as dyke and sill complexes (e.g., Benfontein – Dawson and Hawthorne, 1973; Abersteiner et al., 2019a; Wemindji – Zurevinski and Mitchell, 2011; Amon – Tappe et al., 2014), kimberlite rocks commonly show evidence of flow differentiation, where the carbonate-rich component of the melt separated from its entrained mantle cargo and early-forming magmatic minerals (e.g., olivine, spinel) due to density and viscosity contrasts.

ii) During the late-stages of groundmass crystallisation, the residual kimberlite melt composition becomes “re-enriched” carbonates, alkalis-halogens-P-S and other incompatible elements (e.g., Kitayama et al., 2021), which are commonly recorded by primary melt inclusions in groundmass minerals (Abersteiner et al., 2017). This late-stage residual melt is considered to bear resemblances to the initial parental (or even primary) kimberlite melt composition (Kamenetsky et al., 2008).

iii) Upon emplacement in the crust, kimberlites magma continues to undergo volatile exsolution (CO₂ ± H₂O), further affecting the composition of the residual melt and groundmass mineralogy (e.g., removal of carbonate; Sparks et al., 2006; Brooker et al., 2011; Giuliani et al., 2014a). In-situ decarbonation reactions have also been inferred to occur after emplacement of some kimberlites, where olivine was shown to have reacted with the carbonate component of the
kimberlite melt to produce monticellite, periclase and CO₂ (Abersteiner et al., 2018a). Moreover, accumulated alkalis (Na, K, Rb) and halogens (Cl) in the residual kimberlite melt/fluids could be exsolved into the surrounding wall-rock lithologies (e.g., Smith et al., 2004; Soltys et al., 2018b; Tovey et al., 2020; Hoare et al., 2021).

Kimberlite groundmass minerals, their zoning patterns and melt inclusions can provide an important record of kimberlite melt evolution during its emplacement in the crust. In order to further our understanding of the parental kimberlite melt, we suggest the study of:

- Intrusive kimberlites (e.g., sill complexes). This may provide clearer insights into the composition and differentiation of kimberlite magmas prior to eruption, especially their volatile inventory which is commonly lost during eruption.
- Further investigation into mineral zoning patterns (e.g., atoll-spinel, zoning in perovskite/phlogopite/apatite, monticellite rims around olivine, ‘necklaces’ of perovskite around olivine) and how they reflect compositional changes in the parental melt during crystallisation.
- Correlating constructed parental kimberlite melt compositions with that of the groundmass mineralogy and mineral zoning patterns (e.g., Lim et al., 2018; Giuliani et al., 2020).

5) **Post-emplacement alteration of kimberlite rocks**

Following solidification, kimberlite rocks are *almost always* affected by post-magmatic alteration caused by externally derived groundwater. Post-magmatic alteration of kimberlite rocks is also inferred to be responsible for the removal of the majority of alkali/alkali-earth/halogen/phosphorous/sulphur-bearing groundmass minerals, which are considered to crystallise as late-stage and water-soluble products from the residual kimberlite melt, such as in the Udachnaya-East kimberlite. The absence of these phases (i.e. phases analogous to the daughter mineral assemblages commonly found in melt inclusions from kimberlite minerals from localities worldwide – Table 1) in the groundmass of kimberlites is attributed to their instability during their interaction with externally derived hydrous fluids. Analyses of secondary formed minerals in kimberlites, such as serpentine and iowaite, have revealed that these minerals can host small, but appreciable concentrations of alkalis (e.g., K in serpentine; Abersteiner et al., 2020a) and Cl (e.g., iowaite; Kopylova et
al., 2016; Abersteiner et al., 2018b). We anticipate that further investigations into alteration assemblages in kimberlites may reveal how alkalis and halogens are remobilised in the groundmass during the late and/or post-magmatic stages of alteration.

CONCLUDING REMARKS

As stated by Mitchell (1986), “Are kimberlite studies in the next decade to meander, as they have done in the past, without a common direction; flounder in the abyss of increasing specialization with the inevitable concomitant isolationism; or progress by means of comprehensive co-operative studies of all facets of kimberlite petrology?”. Unfortunately, following the inception of this seminal work, the study of kimberlites endured a prolonged hiatus until the start of the 2000s. Moreover, this remark continues to remain relevant to the study of kimberlite petrology. Although the study of all facets of kimberlite geology, mineralogy, geochemistry and petrogenesis have greatly expanded, progress on all fronts continues to be hampered by the absence of a clear understanding of the primary kimberlite melt composition. Moreover, traditional geochemical approaches applied to this non-traditional rock type have produced divergent and sometimes-contradictory views on kimberlite petrogenesis that persist in the literature. Without a clear determination of the primary kimberlite melt composition, the future of kimberlite research may fall short and diverge towards ever-growing esoteric frontiers. Elucidating the primitive kimberlite melt composition remains a primary task that underpins our ability to construct a holistic understanding of kimberlite magma petrogenesis, ranging from its generation in the mantle to its emplacement in the crust. To aid in this endeavor, we reviewed and evaluated the wide array of geochemical, experimental, petrological and melt inclusion studies. We hope that this issue of Perspectives in Petrology will provide the necessary steppingstones to further embolden interest and enthusiasm in the study of kimberlites.

ACKNOWLEDGEMENTS

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REFERENCES


Table 1. Summary of the types of olivine-hosted crystal, fluid and melt inclusions in the Udachnaya-East, Mark and Koala kimberlites.

<table>
<thead>
<tr>
<th>Locality</th>
<th>Type of Inclusion</th>
<th>Location</th>
<th>Phases</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Udachnaya-East</td>
<td>Crystal</td>
<td>Core</td>
<td>Low- and High-Ca pyroxene (i.e. diopside, enstatite), garnet, picroilmenite, sulphides</td>
<td>Abersteiner et al., 2018b, Golovin et al., 2017, Golovin et al., 2007, Kamenetsky et al., 2012, Kamenetsky et al., 2004, Kamenetsky et al., 2003, Sobolev et al., 1989, Sharygin et al., 2007, Kamenetsky et al., 2008, Kamenetsky et al., 2009a, Kamenetsky et al., 2009b, Kamenetsky et al., 2014</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Rim</td>
<td>Phlogopite, rutile, Cr-Ti-Fe-spinel, magnetite, perovskite</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Secondary Fluid</td>
<td>Core, Rim</td>
<td>Core, Rim</td>
<td>CO₂</td>
<td></td>
</tr>
<tr>
<td>Secondary Melt</td>
<td>Core, Rim</td>
<td>Core, Rim</td>
<td>Na-K chlorides (halite/sylvite), Ca-Mg-bearing alkali/alkali-earth carbonates (Na-K-Ba-Sr ± S; e.g., dolomite, calcite, shortite (Na₂Ca₂(CO₃)₃)), uyerereite (Na₂Ca(CO₃)₂), northupite (Na₂Mg(CO₃)₂Cl), burbeite (Na₂CO₃(SO₄)₂), holdawaite (Mn₃(CO₃)₂(OH)₂(Cl, OH)), gregoryite ((Na₂,K₂,Ca)CO₃), natrite (Na₂CO₃), strontianite (SrCO₃), Fe-Ti-Mg-Cr-Al spinel, Na-K-sulphates (e.g., arcanite (K₂SO₄), tetraferriphlogopite (KMg₃Fe³⁺Si₃O₁₀(OH)₂), apithilite ((K,Na)₃Na(SO₄)₂), djerfisherite (K₆Na(Fe²⁺,Cu,Ni)₂₆S₂₆Cl), Fe-Ni sulphides, phlogopite, monticellite, humite-clinohumite ((Mg,Fe)₇(SiO₄)₃(F,OH)₂), perovskite, apatite</td>
<td></td>
</tr>
<tr>
<td>Mark</td>
<td>Crystal</td>
<td>Core</td>
<td>Cr-diopside, enstatite, Cr-pyrope</td>
<td>Abersteiner et al., 2020a</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Rims + some phenocryst cores</td>
<td>Cr-spinel, Ti-Mg-Al-Cr-spinel, magnesian ulvöspinel magnetite</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Rind</td>
<td>Magnesian ulvöspinel magnetite, pleonaste, Mg-magnete, monticellite, perovskite, periclase, apatite, phlogopite</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Core, Rim, Rind</td>
<td>CO₂</td>
<td></td>
</tr>
<tr>
<td>Secondary Fluid</td>
<td>Outmost Rind</td>
<td>Ca-Mg- (e.g., calcite, dolomite, Mg-calcite) and alkali (K, Na, Ba, Sr) carbonates (±F) (e.g., fairchildite ((K_2Ca(CO_3)_2)), gregoryite ((Na_2K_2CaCO_3)), zemkorite ((Na,K)_2Ca(CO_3)_2), witherite ((BaCO_3))) sylvite/halite, spinel (e.g., Mg-magnetite, magnesian ulvöspinel magnetite, pleonaste), phlogopite, monticellite, perovskite, apatite, Fe-Cu sulphides</td>
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<tr>
<td>-----------------------</td>
<td>--------------</td>
<td>---------------------------------------------------------------------------------------------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pseudosecondary Melt</td>
<td>Cores, Rims</td>
<td>Similar to primary melt inclusions + magnesite, tetraferriphlogopite, Ba- or K-sulphates (e.g., barite, arcanite), kalsilite, Na-phosphates (bradleyite ((Na_3Mg(PO_4)(CO_3))), Cu-Fe-sulphides, unidentified rare earth element (REE)-Nb-Zr phase</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Koala</td>
<td>Crystal</td>
<td>Cr-spinel</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Primary Melt</td>
<td>Core-Rim interface</td>
<td>Tetraferriphlogopite, dolomite, eitelite ((Na_2Mg(CO_3)_2)), shortite (Na_2Ca_2(CO_3)_3), nyerereite (Na_2Ca(CO_3)_2), zemkorite ((Na,K)_2Ca(CO_3)<em>2), and fairchildite ((K_2Ca(CO_3)<em>2)), bradleyite ((Na_2Mg(PO_4)(CO_3))), chlorides (halite and sylvite), sulphates (aphthitalite ((Na</em>{0.25}K</em>{0.75})_2SO_4)),</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pseudosecondary + Secondary Melt</td>
<td>Core, Rim</td>
<td>Kamenetsky et al. 2013</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Figure 1. Map of the Slave Craton (Canada) and Siberian Craton (Russia) and the approximate locations of the Koala, Mark and Udachnaya-East kimberlites.

Figure 2. Schematic diagram showing the different types of compositional zones in euhedral olivine microcrysts. (a, b) Back-scattered electron (BSE) scanning electron microscope (SEM) images of zoned olivine grains from the Mark kimberlite (Canada). The core in BSE imaging is usually irregular in shape, whereas the surrounding rims, rinds and outmost rinds typically adopt more euhedral shapes. High-Mg olivine occurs along healed trails where pseudosecondary/secondary melt/fluid inclusion trails occur.

Figure 3. Bivariate plots of Fo. (mol.%) vs. NiO, MnO and CaO for olivine cores (diamonds), rims (squares), rinds (circles), outmost rinds (OR; triangles), internal zones (Int; plus sign) and transitional zone (TZ; multiply sign) from the Koala, Mark and Udachnaya-East (UDE) kimberlites. Previous literature (Lit) data values (grey) for olivine from the Koala kimberlite are from Kamenetsky et al. (2013) and Lim et al. (2018), for the Mark kimberlite from Abersteiner et al. (2020b), and for the Udachnaya-East kimberlite from Kamenetsky et al. (2008). The grey compositional fields represent olivine compositions from granular peridotite xenoliths from kimberlite occurrences in the Slave Craton and Udachnaya-East kimberlite, respectively. The green composition field represents olivine from sheared peridotite xenoliths from the Udachnaya-East kimberlite. Full dataset in Supplementary Tables S1 – 4.

Figure 4. Back-scattered electron (BSE) scanning electron microscope (SEM) images and wavelength-dispersive spectroscopy (WDS) X-ray element maps of zoned olivine (Ol) microcrysts from the Koala kimberlite. (a – d) Olivine grains consist of a geochemically distinct core, rim and rind. (b – d) Olivine consists of multiple cores, which are best defined by high-Ni content. These cores are sometimes characterised by well-defined euhedral shapes. (b) The olivine rims encompass the cores ‘fusing’ them together into a single grain. In the case of (c) and (d), the rind encompasses multiple olivine grains with rims, ‘fusing’ these into a single grain. Olivine rims are characterised by low Ni and marginally higher Mn and Ca relative to the core. Rinds are characterised by higher Mg, Ca and Mn, and lower Ni and Cr relative to the cores and rinds. In addition, rinds (b – d) are sometimes characterised by minor oscillatory zoning (see text for detailed descriptions). The compositional range (wt.%) for each element map is: (a) 20 – 40, Fe: 0 – 15, Ca: 0 – 0.5, Al: 0 – 0.3, Ni: 0 – 0.4, Cr: 0 – 0.3. (b) Mg: 15 – 40, Fe: 0 – 10, Ca: 0 – 0.3, Al: 0 – 0.3, Ni: 0 – 0.4, Mn: 0 – 0.3, Cr: 0 – 0.2. (c) Mg: 20 – 36, Fe: 2 – 14, Ca: 0 – 0.3, Al: 0 – 0.3, Ni: 0 – 0.5, Mn: 0 – 0.5, Cr: 0 – 0.2. (D) Mg: 20 – 38, Fe: 2 – 8, Ca: 0 – 0.3, Al: 0 – 0.3, Ni: 0 – 0.45, Mn: 0 – 0.3, Cr: 0 – 0.2.
**Figure 5.** Back-scattered electron (BSE) scanning electron microscope (SEM) images and wavelength-dispersive spectroscopy (WDS) X-ray element maps of zoned olivine (Ol) grains from the Mark kimberlite. (a – c) Olivine cores are best defined by high Ni-content. In some cases, Ni cores develop a euhedral-shape core (a), or form irregular-to-angular shapes (b) or consist of potentially multiple cores (c), which are defined by the yellow dotted lines. Olivine cores are surrounded by a transitional zone (TZ), rim, intermediate zone (Int), rind and outmost rind (OR). Transitional zones occur between the core and rim and are defined by diffuse and intermediate compositions. An internal zone exists between the olivine core and rim in (b) and is defined by distinctly lower Mg than the adjacent core and rim, but with intermediate Ni-, Ca- and Mn-content. The olivine rim in (b) is characterised by higher Mg, Ca and Mn, and lower Ni relative to the core. Olivine rinds have the highest Mg-, Ca- and Mn-content, and lowest Ni-content relative to other zones/core. In addition, olivine rinds are marked by the presence of abundant melt inclusions, which are evident in Ca, Al and Cr X-ray maps. Noteworthy, a thin internal zone exists between the rim and rind in (b), which is characterised by elevated Ni- and lower Mg-content. Outmost rinds are characterised by slightly lower Mg-, Ca- and Mn-contents and marginally higher Ni-contents than the Mg-rich rinds (see text for detailed descriptions. The compositional range (wt.%) for each element map is: (a) Mg: 15 – 40, Fe: 0 – 10, Ca: 0 – 2.5, Al: 0 – 2, Ni: 0 – 0.5, Mn: 0 – 0.3, Cr: 0 – 0.2. (b) Mg: 15 – 40, Fe: 0 – 10, Ca: 0 – 2, Al: 0 – 0.2, Ni: 0 – 0.5, Mn: 0 – 0.5, Cr: 0 – 0.4. (c) Mg: 18 – 40, Fe: 0 – 10, Ca: 0 – 1, Al: 0 – 0.5, Ni: 0 – 0.5, Mn: 0 – 0.3, Cr: 0 – 0.3.

**Figure 6.** Back-scattered electron (BSE) scanning electron microscope (SEM) images and wavelength-dispersive spectroscopy (WDS) X-ray element maps of zoned olivine (Ol) grains from the Udachnaya-East kimberlite. (a, d) Olivine consists of multiple cores, which are best defined by Ni-distribution. In some cases, olivine contains multiple cores (b) of differing composition (i.e. Mg-rich and Fe-rich composition). Furthermore, in some cases olivine cores may exhibit well-developed euhedral shapes (b). Mg-rich olivine cores (a – c) are characterised by high Ni-content relative to the rim or internal zone (Int), whereas Fe-rich cores (b) exhibit lower Ni-contents than the internal zones, but may be similar to the rim. Grain (d) appears to be complex, where the Ni-rich core exhibits a diffusely zoned ‘inner core’, which is defined by a slight coupled increase in Mg and Ni content. In addition, grain (d) also contains a Ni-poor and Fe-rich ‘sub-core’ inclusion in the outer part of the core. (a – c) The internal zone surrounding cores forms a well-developed euhedral shape and typically encompasses multiple olivine cores, ‘fusing’ them together. Rims overgrow the internal zones and/or cores and adopt the same euhedral shape. Rims are defined by a consistent range of Mg-content, as well as higher Ca- and Mn-content, and lower Ni-content relative to the core or internal zone. The compositional range (wt.%) for each element map is: (a) Mg: 15 – 36, Fe: 0 – 12, Ca: 0 – 0.4, Al: 0 – 0.3, Ni: 0 – 0.38, Mn: 0 – 0.4, Cr: 0 – 0.2. (b) Mg: 20 – 35, Fe: 4 – 10, Ca: 0 – 0.3, Al: 0 – 0.3, Ni: 0 – 0.4, Mn: 0 – 0.3, Cr: 0 – 0.2. (c) Mg: 12 – 36,
Fe: 2 – 14, Ca: 0 – 0.3, Al: 0 – 0.4, Ni: 0 – 0.3, Mn: 0 – 0.5, Cr: 0 – 0.2. (D) Mg: 15 – 36, Fe: 3 – 12, Ca: 0 – 0.3, Al: 0 – 0.3, Ni: 0 – 0.3, Mn: 0 – 0.4, Cr: 0 – 0.2.

**Figure 7.** Back-scattered electron (BSE) scanning electron microscope (SEM) images and wavelength-dispersive spectroscopy (WDS) X-ray element maps of zoned olivine (Ol) microcrysts from the Udachnaya-East kimberlite showing the decoupling of Fe, Mg and Ca from Ni in the cores. Colour scale indicates increasing intensity (and thus concentration).

**Figure 8.** Optical transmitted light (a) and back-scattered electron (BSE) scanning electron microscope (SEM) images (b – d) of olivine (Ol) grains that are partially (a, d) to completely (b, c) composed of olivine tablets from the Mark (a, c, d) and Udachnaya-East (b) kimberlites. Note, olivine grains composed entirely of tablets (b, c) are termed ‘multigranular nodules’, following the definitions by Arndt et al. (2010). (C, D) Back-scattered electron (BSE) scanning electron microscope (SEM) images and wavelength-dispersive spectroscopy (WDS) X-ray element maps of zoned olivine (Ol) microcrysts from the Mark kimberlite. Note, each individual olivine tablet exhibits compositional internal homogeneity and between individual tablets. The compositional range (wt.%) for the element maps are: Mg: 0 – 70, NiO: 0 – 0.75, MnO: 0 – 0.75, CaO: 0 – 1.

**Figure 9.** Optical transmitted light (a, b, d – h) and back-scattered electron (BSE) scanning electron microscope (SEM; c, i – n) images of crystal, melt and fluid inclusions in olivine (Ol) from the Koala, Mark and Udachnaya-East kimberlites. (a – c) Rounded crystal inclusions of Cr-diopside (Cr-diop) and garnet (Grt) in olivine. Note the presence of olivine tablets and inclusions along the peripheries of the Cr-diopside inclusion in olivine in (b). (d – e) Primary melt and crystal inclusion trails in olivine rinds (Rd). Exposed secondary (S.I.; g – h; j – n) and primary (P.I.; i) multiphase melt inclusions in olivine. Note the decrepitation of the melt inclusion in (N). C: core, REE: rare earth element, dol: dolomite, ox: oxide, tet: tetraferriphlogopite, Alks: alkalis, Cb: carbonate, Mtc: monticellite, Ap: apatite, Hal: halite.

**Figure 10.** Transmitted light images showing the phase transformations during experimental heating, homogenisation and cooling in a pseudosecondary melt inclusions in an olivine grain from the Koala kimberlite (see also figure 11 of Kamenetsky et al., 2013). Note that upon cooling to 505°C, spinifex-like immiscibility occurs and upon further cooling, phase re-solidify into crystalline phases.

**Figure 11.** Bivariate plots of Mg# vs. NiO, MnO and CaO for olivine cores (red diamonds) for kimberlite localities worldwide: Koala (Kamenetsky et al., 2013; Lim et al., 2018), Mark (Abersteiner et al., 2018b), Limpeza-18, Grizzly, Tres Ranchos, Samada, New Robinson (Lim et al., 2018), Udachnaya-East (Kamenetsky et al., 2008), Bultfontein (Giuliani et al., 2017), De Beers dyke (Soltys et al., 2020), Benfontein sills (Arndt et al., 2010; Howarth and Taylor, 2016), DIavik A54N (Brett et al., 2009), Kangamiut (Arndt et al., 2010; Cordier et al., 2015), Majuagaa (Nielsen and
Sand, 2008), Collusus (Moore and Costin, 2016), Kuopio-Kaavi Finnish kimberlites (Dalton et al., 2020) and Tongo kimberlite (Howarth and Giuliani, 2020). The green shaded field shows the compositional distribution of olivine megacrysts from kimberlite localities worldwide (Nielsen and Sand, 2008; Boyd, 1974; Boyd and Nixon, 1975; Gurney et al., 1979, Hops et al., 1992; Eggler et al., 1979; Garrison and Taylor, 1980; Abersteiner et al., 2019b). The blue shaded fields show the composition of olivine from granular peridotites (Aulbach et al., 2007; Bernstein et al., 2006; Menzies et al., 2004; Sobolev et al., 2009; Rehfeldt et al., 2008; Simon et al., 2007; Hervig et al., 1980; De Hoog et al., 2010) and the orange shaded field shows the composition of olivine from sheared peridotites (Aulbach et al., 2007; Simon et al., 2007; Agashev et al., 2013; Sobolev et al., 2009; Boyd, 1974; Boyd and Nixon 1975). Full dataset in Supplementary Table S5.

**Figure 12.** Generalised schematic diagram showing the crystallisation history of euhedral olivine pyrocrysts and the petrogenesis of kimberlite magmas, ranging from the formation of the ascent of the proto-kimberlitic melt in the mantle through to the emplacement of the kimberlite magma in the crust. Note, we do not consider xenocrystic olivine cores in this diagram for simplicity. The diagram is divided into five broad stages (see text for detailed descriptions): (i) the generation of the initially Si-poor, and Na-K-F-Cl-P-S-bearing and Ca-Mg-carbonate-rich kimberlite melt in the mantle. (ii) The ascent of the proto-kimberlite melt through the lithospheric mantle, where it is contaminated due to mantle silicate assimilation (e.g., orthopyroxene resorbed). The re-crystallisation of mantle olivine leads to the formation of olivine pyrocrysts. Entrainment of mantle xenoliths (e.g., peridotite, eclogite) and xenocrysts (e.g., megacrysts, diamond). (iii) The hybrid silicate-carbonate kimberlite melt unmixes to form immiscible carbonate and silicate-carbonate fractions. (iv) Exsolution of CO$_2$ is driven by decompression and increasing silica activity (i.e. in response to mantle silicate assimilation) promotes buoyant and rapid magma ascent. Crustal xenoliths are entrained (v) Emplacement of the kimberlite magma in the upper crust. Here, the interplay of magma differentiation, fractional crystallisation, degassing and post-magmatic alteration modify the kimberlite magma/rock. Fractional crystallisation drives the residual melt towards proto-kimberlite-like alkali-carbonate enriched compositions. Post-emplacement exsolution removes volatiles (CO$_2$, H$_2$O). Alteration removes soluble components and secondary minerals form (e.g., serpentine) increasing, the H$_2$O content. Ol: olivine, Cr-Spl: Cr-spinel, Mtc: monticellite, Prv: perovskite, Opx: orthopyroxene, Cpx: clinopyroxene.
Fig. 3.
Fig. 6.
Fig. 8.
Fig. 9.
Fig. 11.
**Fig. 12.**

- **Olivine Stage 4:** Formation of rinds ± outmost rinds ± high-Mg olivine.
- **Olivine Stage 3:** Formation of rinds ± transitional zones.
- **Olivine Stage 2:** Formation of internal zones ± transitional zones.
- **Olivine Stage 1:** Formation of “pyroclastic” olivine due to partial re-crystallisation of entrained mantle olivine + entrainment of olivine from disaggregated mantle xenoliths and wall-rock.

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**Silicate-Carbonate Immiscibility followed by degassing during decompression**

**Exsolution**

Reaction with wall-rocks. Entrainment, assimilation and disaggregation of mantle xenoliths

**Post-Emplacement**

Exsolution + Alteration

↑ H₂O, H₂O, (F, Mg)

↓ CO₂, CO₂, H₂O, K, Na, P, Cl, F, S

**Crystallisation**

↑ CO₂, CO₂, K, Na, Cl, F, S

↓ Si, Mg, Al, Fe, Cr, Ti, CO₂

**Lithosphere**

- Olivine
- Olivine Pyroclyst
- Orthopyroxene
- Garnet
- Clinopyroxene
- CO₂
- Peridotite xenolith
- Eclogite xenolith
- Crustal xenolith
- Carbonate melt
- Silicate-Carbonate melt
- Volatiles-rich melt/fluid

**Peridotite**

Si-poor, Ca-Mg-carbonate-rich Na-K-Cl-F-P-S-bearing melt

**Proto Kimberlite Melt**

Partial dissolution of mantle-derived opx and replacement by cpx

**Contamination**

↑ Si, Mg (Al, Cr, Ni)

**Walk-rock/melt-rock interaction**

**Olivine Pyroclyst**

- Fe-Mg ± Ca-Mn and Ni decoupling.
- Ni-defined core.