Evolution of ultrapotassic volcanism on the Kaapvaal craton: deepening the orangeite versus lamproite debate

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Abstract: Orangeites are a significant source of diamonds, yet ambiguity surrounds their status among groups of mantle-derived potassic rocks. This study reports mineralogical and geochemical data for a c. 140 Ma orangeite dyke swarm that intersects the Bushveld Complex on the Kaapvaal craton in South Africa. The dykes comprise distinctive petrographic varieties that are linked principally by olivine fractionation, with the most evolved members containing minor amounts of primary carbonate, sanidine and andradite garnet in the groundmass. Although abundant groundmass phlogopite and clinopyroxene have compositions that are similar to those of cratonic lamproites, these phases show notable Ti-depletion, which we consider a hallmark feature of type orangeites from the Kaapvaal craton. Ti-depletion is also characteristic of bulk rock compositions and is associated with strongly depleted Th–U–Nb–Ta contents at high Cs–Rb–Ba–K concentrations. The resultant high large ion lithophile element/high field strength element ratios of orangeites suggest that mantle source enrichment occurred by metasomatic processes in the proximity of ancient subduction zones. The Bushveld-intersecting orangeite dykes have strongly enriched Sr–Nd–Hf isotopic compositions (initial $\varepsilon^{140}$Sm/Sm = 0.7070–0.7074; $\varepsilon$Nd = −10.6 to −5.8; $\varepsilon$Hf = −14.4 to −2.5), similar to those of other orangeites from across South Africa. Combined with the strong Ti–Nb–Ta depletion, this ubiquitous isotopic feature points to the involvement of ancient metasomatized mantle lithosphere in the origin of Kaapvaal craton orangeites, where K-rich metasomes imparted a ‘fossil’ subduction geochemical signature. Previous geochronology studies identified ancient K-enrichment events within the Kaapvaal cratonic mantle lithosphere, possibly associated with collisional tectonics during the 1.2–1.1 Ga Namaqua–Natal orogeny of the Rodinia supercontinent cycle. It therefore seems permissible that the cratonic mantle root was preconditioned for ultrapotassic magma production by tectonomagmatic events that occurred along convergent plate margins during the Proterozoic. However, reactivation of the K-rich metasomes had to await establishment of an extensional tectonic regime, such as that during the Mesozoic breakup of Gondwana, which was accompanied by widespread (1000 × 750 km) small-volume orangeite volcanism between 200 and 110 Ma. Although similarities exist between orangeites and lamproites, these and other potassic rocks are sufficiently distinct in their compositions such that different magma formation processes must be considered. In addition to new investigations of the geodynamic triggers of K-rich ultramafic magmatism, future research should more stringently evaluate the relative roles of redox effects and volatile components such as H$_2$O–CO$_2$–F in the petrogenesis of these potentially diamondiferous alkaline rocks.

Supplementary material: Mineral and bulk rock compositions including Sr–Nd–Hf isotope ratios are available at https://doi.org/10.1144/SP513-2021-84

The Kaapvaal craton in southern Africa is host to numerous occurrences of kimberlites, which traditionally have been divided into two groups based on petrographic characteristics and compositions: the so-called (1) archetypal Group 1 kimberlites and (2) highly micaceous Group 2 kimberlites, also known as ‘orangeites’ (Wagner 1914; Smith et al. 1985; Skinner 1989; Mitchell 1995). Group 1 kimberlite magmatism of the Kaapvaal craton and surrounding mobile belts has occurred repeatedly since at least 1800 Ma, with prominent eruption episodes at around 1150, 520, 240 and 90 Ma (Jelsma et al. 2009; Griffin et al. 2014; Tappe et al. 2018). Archetypal Group 1 kimberlites are both petrographically and geochemically relatively homogeneous, and their moderately depleted to slightly enriched
Sr–Nd–Hf isotopic compositions suggest an origin from the convecting upper mantle (Smith 1983; Nowell et al. 2004; Tappe et al. 2020a, b). A convecting upper mantle origin is also consistent with the lack of anomalous $^{182}$W abundances in these CO$_2$-H$_2$O-rich ultramafic magmas (Tappe et al. 2020b), which rules out significant melt contributions from lower mantle plumes to kimberlite magmatism, in contrast to many oceanic island basalt suites (Rizo et al. 2019; Mundl-Petermeier et al. 2020). The overwhelming majority of superlative diamond deposits are hosted by Group 1 kimberlites, not only across southern Africa but also on other cratons worldwide (Janse and Sheahan 1995; de Wit et al. 2016).

In contrast, Group 2 kimberlite or orangeite magmatism on the Kaapvaal craton and adjoining mobile belts appears to be temporally restricted to the Mesozoic between 200 and 110 Ma (Phillips et al. 1998; Griffin et al. 2014; Tappe et al. 2018), and its diamond potential is typically lower compared with that of Group 1 kimberlites (Field et al. 2008). Orangeites are a petrographically and geochemically heterogeneous group of potassic ultramafic rocks that can be reminiscent of cratonic lamproites or certain types of lamprophyres. Besides olivine-rich primitive varieties, there are several occurrences of more evolved olivine-poor orangeites that may contain groundmass sanidine or leucite (Mitchell 1995; Howarth et al. 2011). Extremely high large ion lithophile element (LILE) contents and the strongly enriched Sr–Nd–Hf isotopic compositions of orangeites are in stark contrast to archetypal kimberlites, and point to melt derivation from ancient K-metasomatized lithospheric mantle sources (Fraser et al. 1985; Nowell et al. 2004; Becker and le Roex 2006; Coe et al. 2008).

Compared with archetypal kimberlites, the mineralogy and geochemistry of type orangeite from the Kaapvaal craton are surprisingly poorly studied. For example, several decades of extensive platinum mining on the western lobe of the c. 2056 Ma Bushveld Complex exposed numerous highly micaceous ultramafic dykes that cut across the central Kaapvaal craton (Fig. 1), but there is a dearth of petrological information about these dykes in the public domain. They have been loosely referred to as ‘micaceous’ or ‘lamprophryic’ (Leeb-Du Toit 1986; Hughes et al. 2016; Daya 2019; Lerobane 2020), and most investigations have focussed on their geotechnical properties because these dykes are associated with life-threatening gas outbursts and tunnel failures in underground platinum mining operations (Daya et al. 2018; Moate 2019). Geochronology databases list the emplacement age of these diamond-poor to barren potassic ultramafic dykes on the western lobe of the Bushveld Complex at c. 140 Ma (Jelsma et al. 2009; Tappe et al. 2018). This suggests a genetic link to the c. 145 Ma diamondiferous orangeite dyke swarm near Swartruggens (Mitchell 1995), which is located c. 60 km west of the Bushveld Complex and is host to Helam Diamond Mine (Gurney and Kirkley 1996; McKenna et al. 2004; Field et al. 2008; Fig. 1).

In this contribution, we present a detailed account on the mineralogy and geochemistry, including Sr–Nd–Hf isotope data, of the potassic ultramafic dykes from the western Bushveld Complex, sampled at the Impala and Sibanye–Stillwater platinum mine leases near Rustenburg in 2019 (Figs 1 & 2). Our data show that these dyke occurrences represent typical Group 2 kimberlites or orangeites, many of which are compositionally evolved, suggesting petrographic resemblance to ultramafic lamprophyres. Although olivine fractionation was the key driver in the evolution of the orangeite magmatic system on the central Kaapvaal craton, it remains unclear why this process was apparently much more efficient compared with orangeite magmatism on the western Kaapvaal craton (Mitchell 1995). The case of the mostly barren Mesozoic orangeite dykes transecting the western Bushveld Complex provides a tangible example of how the diamond potential of potassic ultramafic magmas can rapidly diminish upon ascent – opening further new research avenues into this commercially relevant topic.

Kaapvaal craton and Bushveld Complex: an overview

The Kaapvaal craton in southern Africa presents one of the best preserved remnants of an Archean continent (de Wit et al. 1992; Kröner et al. 2019). Its rock record dates back to c. 3.66 Ga and vestiges of Hadean proto-crust have recently been identified in detrital zircon populations from the Early Archean Barberton Supergroup (Byerly et al. 2018). On the basis of its Archean geological evolution, the Kaapvaal craton has traditionally been divided into four geographical domains (i.e. eastern, northern, western, central) comprising typical granite–greenstone rock assemblages (Poujol et al. 2003), which amalgamated into a larger proto-continent during the Late Archean (Shirey et al. 2002; Smart et al. 2016). The central and eastern domains form the Witwatersrand block, which grew by magmatic and tectonic accretion between 3.6 and 3 Ga, as opposed to the younger Kimberley block (3.0–2.7 Ga) that comprises large parts of the western Kaapvaal craton (Schmitz et al. 2004). Late Archean cratonization of the Kaapvaal lithosphere was complete by 2.7 Ga, accompanied by the development of large cratonic basins (e.g. the ‘Supergroups’ filling the Pongola, Witwatersrand and Ventersdorp basins), which were to some extent rift-related
The Kaapvaal craton was mostly submerged below sea-level between 2.65 and 2 Ga when deposition of the 10 km thick Transvaal Supergroup occurred. At c. 2056 Ma, the Transvaal basinal rock succession was intruded by the giant Bushveld Complex on the central Kaapvaal craton (Zeh et al. 2015, 2019).

The 400 × 300 × 9 km saucer-shaped Bushveld Complex presents the largest known continental layered mafic intrusion, but it also includes significant volumes of felsic rocks such as the vast Lebowa granite suite and Rooiberg Group (Cawthorn et al. 2006; Fig. 1). The Rustenburg Layered Suite, up to 9 km thick, comprises various types of mafic–ultramafic cumulate rocks, with numerous relatively thin mineralized layers that form chromite and platinum-group-mineral-enriched zones commonly referred to as ‘reefs’, exploited on the three main lobes of the Bushveld Complex (i.e. western, eastern and northern lobes; Fig. 1). The pyroxenitic–anorthositic Merensky Reef and underlying UG2 chromitite reef represent world’s largest proven ore reserves of Pt–Pd–Rh and Cr, currently extracted in numerous

![Geological overview map of the c. 2056 Ma Bushveld Complex and surrounding areas on the central Kaapvaal craton in South Africa (modified from Zeh et al. 2019). The inset shows the location of the Kaapvaal craton in southern Africa. The c. 140 Ma orangeite dyke swarm on the western lobe of the Bushveld Complex is mainly exposed in the underground mines of the Impala Platinum Holdings Limited and Sibanye–Stillwater Limited leases near Rustenburg. A c. 145 Ma orangeite dyke system is mined for diamonds near the town of Swartruggens some 60 km west of the Bushveld Complex. TML, Thabazimbi–Murchison Lineament. Primary diamond deposits at: (1) ‘Swartruggens/ Helam’ in a c. 145 Ma orangeite dyke system; (2) ‘Klip springs–Marsfontein’ in a c. 155 Ma orangeite dyke–blow system; (3) ‘Palmietgat’ in c. 160 Ma kimberlite pipes; (4) ‘Palmietfontein in a c. 75 Ma kimberlite pipe; and (5) ‘Premier/Cullinan’ in a c. 1150 Ma kimberlite pipe (the ages are after Griffin et al. 2014; Tappe et al. 2018; Smart et al. 2021).](image-url)
underground mining operations between 300 and 1000 m depth below the surface. On the western lobe of the Bushveld Complex, these mining activities exposed a diamond-poor to barren swarm of thin potassic ultramafic dykes of Mesozoic age that forms the subject of this study (Fig. 2).

Methodology

Acquisition and preparation of samples

Fifteen discrete 0.1–1 m wide potassic ultramafic dykes were sampled on the western lobe of the Bushveld Complex near Rustenburg during the course of 2019 (Figs 1 & 2). The samples are derived from the underground mining operations and exploration drilling activities of Impala Platinum (Shaft-20 and surrounding areas, n = 11) and Sibanye-Stillwater (formerly Lonmin Platinum Limited, Safiy and Karee-3 shafts, n = 4). Underground sampling of the dykes occurred in production tunnels at variable depth levels in footwall drives to the Merensky and UG2 ore reefs, c. 800–1000 m below the surface (Fig. 2).

The freshest samples were cut into thin slabs with a rock saw and small blocks were used for the preparation of polished petrographic thin sections at the University of Johannesburg, South Africa. The rock slabs were washed under running water and only materials visibly free from crust-derived rock fragments were processed further. The rock slabs (c. 250 g per sample) were then wrapped in thick plastic sleeves and crushed with a hammer into <8 mm large chips. Approximately 100 g of rock chips per sample were then processed in an agate mill at the University of Johannesburg to obtain analytical grade powder.

Electron microprobe analysis

Quantitative major and minor element analysis of rock-forming minerals was conducted on representative thin sections employing a Cameca SX100 electron probe micro-analyser at the University of Johannesburg. The polished thin sections were covered with c. 25 nm thick carbon layers under vacuum in a Quorum Q300T ES coater. The electron microprobe instrument was calibrated using a variety of natural standards: jadeite (Na), olivine (Mg), almandine (Al), diopside (Si), orthoclase (K), wollastonite (Ca), rhodonite (Mn), hematite (Fe), barite (Ba), fluorite (F) and halite (Cl). Synthetic pure metal oxides were used as calibration standards for Ti, Cr and Ni. Chemical elements of interest were measured on their respective X-ray Kα lines, and matrix corrections were performed with the ‘X-PHI’ method, which is a ϕ(ρz) offline correction routine. The lower limit of detection for the quantified major and minor element abundances is c. 0.05 wt%. The diverse mineralogy of the potassic ultramafic dykes required different electron
probe micro-analyser setups for ‘dry’ phases (e.g. olivine, diopside, spinel-group minerals) and ‘volatile-bearing’ phases (e.g. phlogopite). Therefore, the electron beam parameters and element signal counting times varied between setups. For example, the electron beam acceleration voltage was adjusted to 15 kV for phlogopite analyses, whereas 20 kV was applied during analysis of the ‘dry’ phases. The electron beam current was adjusted to 20 nA for both analytical routines. Peak intensity counting times varied between 10 and 60 s depending on the chemical element of interest. The electron beam diameter for spot analysis varied between setups: i.e. 1 µm for ‘dry’ and 2 µm for ‘volatile-bearing’ phases. The in-house almandine-rich garnet (Alma-PH1) and diopside-rich clinopyroxene (Diop-PH1) standards were repeatedly analysed during this study to monitor data accuracy and precision (Ngwenya and Tappe 2021).

**X-ray fluorescence analysis and \( \text{CO}_2 \) determination**

Bulk rock major and minor element concentrations were determined with a PANalytical MagiX PRO X-ray fluorescence spectrometer (XRF) at the University of Johannesburg. Sample powders were dried in an oven at 105°C prior to fusion into glass discs with a Li\(_2\)B\(_4\)O\(_7\)-LiBO\(_2\) flux. The XRF method detection limit is c. 0.05 wt% for all chemical elements reported in Table 1. Instrument calibration was conducted with mixtures of pure metal oxides, and the accuracy of the XRF method was monitored by analysis of certified reference materials (BE-N, JSy-1, SARM-2, SARM-16), as well as analysis of carbonatite ST199 and kimberlite (BE-N, JSy-1, SARM-2, SARM-16). Neodymium was subsequently isolated from the REE fraction using LN Spec resin (50–100 mesh). Neodymium was then added to each beaker, and the previous heating step was repeated twice. Trace element analysis was performed with a Perkin Elmer ELAN DRC-e Q-ICP-MS instrument. Internal standards were Rh, Re, In and Bi, and ICP-MS calibration standards were made from certified solutions (Wilson 2012). During this study, replicate analyses of the USGS basaltic reference material BHVO-2 (Jochum et al. 2016) and the Mintek kimberlite reference material SARM-39 (Roy et al. 2007; Tappe et al. 2020a) yielded trace element concentrations within 10% of the recommended values, excluding Gd, Tm, Nb and Ta (within 20%).

**\( \text{Sr–Nd–Hf} \) isotope ratio measurements**

Purification of bulk rock Sr, Nd and Hf cuts for isotope ratio measurements was achieved through ion exchange chromatography at the University of Münster, and detailed descriptions of the clean-laboratory and mass spectrometry methods are provided in Tappe et al. (2020a, b). Approximately 100 mg of each sample powder were dissolved in HF-HNO\(_3\) at 180°C (in Savillex beakers) and re-dissolved twice in 6 M HCl until clear solutions were obtained. Final sample digests were split into two solution aliquots for further processing for \( \text{Sr–Nd} \) and \( \text{Hf} \) isotope analysis, respectively. Strontium and the REE fractions were collected using conventional cation exchange chromatography (AG50W-X8, 200–400 mesh). Neodymium was subsequently isolated from the REE fraction using LN Spec resin (50–100 mesh). Hafnium was isolated in a separate procedure that also uses LN Spec resin. The total procedural blanks are negligible at <50 pg Sr, <30 pg Nd and <10 pg Hf (Bast et al. 2015; Tappe et al. 2020a).

Strontium isotope ratio measurements were performed by thermal ionization mass spectrometry on a Thermo Scientific Triton instrument, whereas the Nd and Hf isotope ratios were measured by plasma ionization with a Thermo Scientific Neptune Plus instrument. Measured \(^{87}\text{Sr}/^{86}\text{Sr}, ^{143}\text{Nd}/^{144}\text{Nd} and ^{176}\text{Hf}/^{177}\text{Hf} \) ratios were normalized using an

**Inductively coupled plasma mass spectrometry**

Sample powder digestion for trace element inductively coupled plasma mass spectrometry (ICP-MS) analysis was undertaken at the University of the Witwatersrand in Johannesburg. Approximately 50 mg of rock powder per sample were loaded into microwave Teflon vessels. Concentrated HF-HNO\(_3\) (6 ml) was added to each vessel, and powders were dissolved in a MARS microwave digester for 60 min. The sample solutions were then transferred to 15 ml Savillex beakers, which were placed on a hotplate at 60°C for 24 h and subsequently dried down at 70°C. Approximately 2 ml HNO\(_3\) were then added to each beaker, and the previous heating step was repeated twice. Trace element analysis was performed with a dual inlet. The detection limit for the \( \text{Sr–Nd–Hf} \) isotope ratio measurements was determined relative to a natural carbonate standard using a Thermo-Finnigan Delta Plus gas source mass spectrometer fitted with a dual inlet. The detection limit for the \( \text{CO}_2 \) concentration measurements was 0.01 wt% (Table 1).
Table 1. Major (wt%) and trace (ppm) element compositions, as well as Sr–Nd–Hf isotopic compositions of Mesozoic Group 2 kimberlite dykes (orangeites) that intersect the western Bushveld Complex, central Kaapvaal craton, South Africa

| Sample ID: | Impala | Evolved orangeite | Sibanye (Marikana) | Evolved orangeite |
|-----------|--------|------------------|-------------------|------------------|
|           | Orangeite | Evolved orangeite | Orangeite | Evolved orangeite | Orangeite | Evolved orangeite |
| Mining block: | IP20-1 | IP20-3 | IP20-4 | IP20-5 | IP20-7 | IP20-11 | IP20-2 | IP20-6 | IP20-8 | IP20-9 | IP20-10 | LM-4 | LM-5 | LM-6B1 | LM-6B2 |
| SiO₂ | 39.0 | 36.7 | 41.2 | 40.2 | 40.2 | 38.4 | 38.6 | 36.4 | 32.2 | 38.2 | 40.1 | 40.1 | 38.6 | 40.1 | 37.8 |
| TiO₂ | 0.91 | 0.78 | 1.24 | 1.08 | 1.09 | 0.87 | 2.93 | 2.83 | 3.04 | 1.63 | 1.31 | 1.82 | 1.82 | 1.82 | 1.78 |
| Al₂O₃ | 4.74 | 6.22 | 5.55 | 4.77 | 4.64 | 6.64 | 4.41 | 4.55 | 5.66 | 5.86 | 5.46 | 6.37 | 6.59 | 6.48 | 6.28 |
| Fe₂O₃ | 7.64 | 8.05 | 9.46 | 8.56 | 9.39 | 7.70 | 12.3 | 11.3 | 11.7 | 10.2 | 9.06 | 12.0 | 10.1 | 10.3 | 10.1 |
| MnO | 0.13 | 0.18 | 0.16 | 0.14 | 0.14 | 0.16 | 0.22 | 0.19 | 0.18 | 0.19 | 0.15 | 0.18 | 0.17 | 0.16 | 0.17 |
| MgO | 26.1 | 22.6 | 22.8 | 25.7 | 22.9 | 23.2 | 16.5 | 16.5 | 15.1 | 18.8 | 20.2 | 15.5 | 14.9 | 15.8 | 17.1 |
| CaO | 7.47 | 8.69 | 7.51 | 6.24 | 8.68 | 7.22 | 10.9 | 11.7 | 13.4 | 8.33 | 9.76 | 11.3 | 10.9 | 11.8 | 10.4 |
| Na₂O | 0.05 | 0.12 | 0.68 | 0.49 | 0.58 | 0.87 | 1.65 | 1.48 | 1.06 | 1.27 | 1.53 | 1.61 | 1.23 | 1.14 | 1.00 |
| K₂O | 4.31 | 6.19 | 3.21 | 3.28 | 3.15 | 6.22 | 5.01 | 5.23 | 5.28 | 6.90 | 3.29 | 3.23 | 4.38 | 3.15 | 3.76 |
| P₂O₅ | 0.35 | 0.10 | 0.61 | 0.48 | 0.66 | 6.22 | 5.12 | 5.23 | 5.28 | 6.90 | 3.29 | 3.23 | 4.38 | 3.15 | 3.76 |
| Cr₂O₃ | 0.38 | 0.31 | 0.22 | 0.22 | 0.22 | 0.29 | 0.15 | 0.13 | 0.15 | 0.23 | 0.20 | 0.16 | 0.16 | 0.16 | 0.17 |
| NiO | 0.14 | 0.10 | 0.13 | 0.15 | 0.14 | 0.11 | 0.07 | 0.07 | 0.05 | 0.08 | 0.11 | 0.06 | 0.05 | 0.06 | 0.06 |
| LOI | 6.9 | 8.3 | 5.3 | 6.7 | 5.8 | 7.0 | 3.0 | 4.8 | 7.7 | 5.3 | 5.8 | 9.78 | 7.5 | 97.4 | 97.9 | 98.7 |
| CO₂ | 2.0 | 4.7 | 0.28 | 1.6 | 0.45 | 2.1 | 0.60 | 0.33 | 0.25 | 0.40 | 0.40 | 1.3 | 0.63 | 3.0 | 0.87 |
| SO₃ | 0.19 | 0.35 | 0.51 | 0.65 | 1.2 | b.d. | 0.38 | 0.44 | 0.76 | 0.10 | 1.0 | 0.29 | 0.12 | 0.32 | 0.21 |
| S | 37.1 | 34.4 | 51.6 | 40.6 | 51.9 | 47.6 | 66.4 | 65.3 | 59.6 | 76.4 | 74.6 | 82.8 | 85.8 | 88.5 | 82.0 |
| Sc | 8.6 | 7.4 | 11.5 | 10.2 | 11.3 | 19.6 | 6.0 | 64.3 | 96.4 | 59.6 | 47.6 | 23.1 | 23.2 | 22.9 | 21.1 |
| V | 112 | 17.0 | 123 | 96.4 | 115 | 27.9 | 51.3 | 59.9 | 110 | 115 | 129 | 215 | 287 | 144 | 286 |
| Ga | 6.5 | 6.7 | 6.9 | 6.5 | 6.6 | 11.6 | 10.0 | 10.2 | 10.4 | 15.0 | 11.9 | 15.9 | 16.3 | 17.0 | 15.9 |
| Ba | 871 | 655 | 807 | 930 | 850 | 776 | 441 | 432 | 302 | 579 | 798 | 430 | 341 | 427 | 454 |
| Zn | 38.8 | 5.9 | 37.5 | 29.2 | 37.1 | 8.5 | 96.5 | 94.6 | 39.6 | 52.9 | 49.1 | 63.5 | 63.2 | 63.3 | 59.7 |
| Sr | 8.6 | 7.4 | 11.5 | 10.2 | 11.3 | 19.6 | 6.0 | 64.3 | 96.4 | 59.6 | 47.6 | 23.1 | 23.2 | 22.9 | 21.1 |
| Li | 1.62 | 1.98 | 5.18 | 4.39 | 4.14 | 4.00 | 8.08 | 7.55 | 5.88 | 9.69 | 5.55 | 10.5 | 11.0 | 7.45 | 10.4 |
| Cs | 1.95 | 2.24 | 1.35 | 1.18 | 1.41 | 2.72 | 2.88 | 2.77 | 2.49 | 3.66 | 2.00 | 1.91 | 2.06 | 1.54 | 1.84 |
| Rb | 153 | 145 | 106 | 91.8 | 93.3 | 266 | 179 | 173 | 191 | 340 | 153 | 194 | 230 | 172 | 202 |
| Sr | 435 | 808 | 720 | 448 | 668 | 797 | 1061 | 1189 | 1417 | 1047 | 957 | 1884 | 2938 | 3327 | 3154 |
| Zr | 84.2 | 66.3 | 110 | 106 | 130 | 68.3 | 10.0 | 10.2 | 10.4 | 15.0 | 11.9 | 15.9 | 16.3 | 17.0 | 15.9 |
| Hf | 2.53 | 2.45 | 3.88 | 3.61 | 4.16 | 2.57 | 12.2 | 13.0 | 5.84 | 4.81 | 4.21 | 7.80 | 7.70 | 7.42 | 6.67 |
| Nb | 50.8 | 42.2 | 61.3 | 48.4 | 61.4 | 46.7 | 12.0 | 10.9 | 102 | 60.6 | 82.9 | 108 | 103 | 107 | 104.0 |
| Element | Concentration |
|---------|---------------|
| Ta      | 3.07          |
| Y       | 4.51          |
| Pb      | 8.84          |
| Th      | 5.53          |
| U       | 1.76          |
| W       | 5.3           |
| La      | 45.8          |
| Ce      | 91.6          |
| Pr      | 10.63         |
| Nd      | 38.9          |
| Sm      | 6.20          |
| Eu      | 1.66          |
| Gd      | 3.95          |
| Tb      | 0.43          |
| Dy      | 1.48          |
| Ho      | 0.20          |
| Er      | 0.51          |
| Yb      | 0.35          |
| Lu      | 0.05          |

**Sr**/**Sr** (m): 0.709390(6) - 0.708592(7)
**Sr**/**Sr** (i): 0.708741(6)
**Nd**/**Nd** (m): 0.511254(6) - 0.511956
**Nd**/**Nd** (i): 0.511932

**Hf**/**Hf** (m): 0.282633(7) - 0.282454
**Hf**/**Hf** (i): 0.282439

**Sr**/**Sr** (m) = total Fe as ferric iron; LOI, loss on ignition, is defined as the difference in sample weight after ignition at 930°C for 30 min.

Sr isotope ratios were determined by thermal ionization mass spectrometry, and Nd–Hf isotope ratios were determined by MC–ICP–MS at the University of Münster.

Major and minor element concentrations are X-ray fluorescence spectrometry data determined at the University of Johannesburg; b.d., below detection. Trace element concentrations are solution ICP–MS data determined at the University of the Witwatersrand. The CO2 concentrations were determined at the University of Münster.

Fe2O3 = total Fe as ferric iron; LOI, loss on ignition, is defined as the difference in sample weight after ignition at 930°C for 30 min.

2 SD uncertainties of the εNd and εHf values entail a full error propagation, where the uncertainties of the emplacement age and parent–daughter element ratios were set at 20 and 5%, respectively.

ΔεHf (i) is defined as εHf (i) − (1.59 × εNd (i) + 1.28), such that analyses with positive values fall by definition above the Nd–Hf isotope regression line of Chauvel et al. (2008).

Numbers in parentheses are 2-sigma-of-the-mean uncertainties for individual isotope ratio measurements.
exponential mass fractionation law and $^{88}\text{Sr}/^{86}\text{Sr}$ of 8.37521, $^{146}\text{Nd}/^{144}\text{Nd}$ of 0.7219 and $^{176}\text{Hf}/^{177}\text{Hf}$ of 0.7325, respectively. The measured Sr–Nd–Hf isotope ratios for all ‘unknowns’ including the secondary standards are reported relative to $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.710245 for NBS987, $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.512115 for JNdI-1 and $^{176}\text{Hf}/^{177}\text{Hf}$ of 0.282160 for AMES/ JMC–475 standard solutions, respectively. During the analyses in 2020, repeated measurements of $^{87}\text{Sr}/^{86}\text{Sr}$ for the NBS987 primary standard averaged 0.710282 ± 21 (2 SD, $n = 4$), $^{143}\text{Nd}/^{144}\text{Nd}$ for the JNdI-1 primary standard averaged 0.512069 ± 8 (2 SD, $n = 12$) and $^{176}\text{Hf}/^{177}\text{Hf}$ for the AMES primary standard averaged 0.282137 ± 17 (2 SD, $n = 14$).

We also processed the USGS basaltic reference materials BCR-2 and BHVO-2 alongside our samples, and the results are in excellent agreement with the recommended isotope ratios. For BCR-2 the measured values are: $^{87}\text{Sr}/^{86}\text{Sr}$ = 0.704996 ± 6; $^{143}\text{Nd}/^{144}\text{Nd}$ = 0.512634 ± 25; and $^{176}\text{Hf}/^{177}\text{Hf}$ = 0.282869 ± 17. For BHVO-2 the measured values are: $^{87}\text{Sr}/^{86}\text{Sr}$ = 0.703471 ± 6; $^{143}\text{Nd}/^{144}\text{Nd}$ = 0.512990 ± 9; and $^{176}\text{Hf}/^{177}\text{Hf}$ = 0.283100 ± 37. The results for this matrix-matched reference material are in excellent agreement with the recommended Sr–Nd–Hf isotope ratios for SARM-39 (Tappe et al. 2020a).

For our samples, the parent–daughter element ratios were calculated using the Rb, Sr, Sm, Nd, Lu and Hf concentrations determined by solution-mode ICP-MS analysis. The initial isotope ratios and notations are calculated at 140 Ma (Tappe et al. 2018) and the 2 SD uncertainties of the calculated initial epsilon Nd and Hf values reported in Table 1 entail a full propagation of ‘errors’. The error propagation comprises highly conservative uncertainty estimates, i.e. 20% for the c. 140 Ma magma emplacement age (140 ± 30 Ma) and 5% for the parent–daughter element ratios. The age corrections are based on the following decay constants: $^{87}\text{Rb} \times 1.42 \times 10^{-11}$, $^{147}\text{Sm} \times 6.54 \times 10^{-12}$ and $^{176}\text{Lu} \times 1.865 \times 10^{-11}$ per year.

**Results**

For convenience, results are described separately for the samples from the Impala and Sibanye mining concessions, despite the fact that they are derived from a single large potassic ultramafic dyke swarm. All data generated during this study (i.e. mineral and bulk rock compositions including Sr–Nd–Hf isotope ratios) are provided online in Supplementary Material 1. In addition, the complete bulk rock geochemical dataset is reported as Table 1 in the main article.

**Petrography and classification**

Our mineralogy and geochemistry data (see below) reveal that the highly micaceous ultramafic dykes intersecting the western Bushveld Complex represent ‘typical’ Kaapvaal orangeites (formerly Group 2 kimberlites), in contrast to their previous categorization as lamprophyres (cf. Hughes et al. 2016). A lamprophyric character can also be excluded on the basis of petrography (Fig. 3), with the absence or paucity of hydrous mafic phenocrysts/macrocrysts and phlogopite being largely confined to the groundmass (Rock 1991; Tappe et al. 2005). On a macroscopic scale, the orangeite dykes studied comprise olivine-rich and olivine-poor varieties; the latter are, hereafter, designated as ‘evolved’ orangeites (Fig. 2c).

The Impala orangeite samples have inequigranular textures with fairly abundant olivine macrocrysts (up to 15 vol%), ranging in size from 1 to 10 mm, as well as highly abundant microphenocrysts (100–500 µm) of phlogopite laths and clinopyroxene prisms (Fig. 3). These coarse to fine crystal populations are set in a very fine-grained groundmass (<100 µm) dominated by phlogopite and clinopyroxene with minor amounts of spinel-group minerals, apatite, carbonate and serpentine (Fig. 3). The olivine macrocrysts are largely serpentinitized but rare relics of olivine cores have been observed and analysed (see below). The phlogopite laths are typically zoned displaying pale yellow cores and deep orange rims under plane polarized light (Fig. 3a). Rare serpentinitized peridotitic microxenoliths up to 20 mm across also occur.

The more evolved orangeite dykes from the Impala and Sibanye mining concessions mostly lack inequigranular textures because olivine macrocrysts are very rare or even absent (<2 vol%; Fig. 2c). However, the common occurrence of aggregates of phlogopite laths >500 µm in length results in a micro-glomeroporphyritic texture. Locally, phlogopite laths show preferred orientations as part of prominent flow structures (Fig. 3c). Rare olivine microcrysts displaying core–rim zoning are observed as inclusions within larger complexly zoned phlogopite plates (Fig. 3f). The phlogopite–diopside dominated groundmass of the evolved orangeites is petrographically similar to its counterpart in the more primitive orangeite dykes, but in addition it may contain subordinate amounts of K-feldspar (?sanidine) and andradite-rich magmatic garnet. Groundmass phlogopite in the evolved orangeite dykes studied may show a decussate texture with randomly oriented and interlocking laths (Fig. 3d–f).
Fig. 3. Photomicrographs and BSE images of representative samples from Bushveld-intersecting orangeite dykes. 
(a) Impala orangeite dyke IP20-1 exhibiting a serpentinized olivine macrocryst set in a phlogopite-clinopyroxene dominated groundmass (plane polarized light). (b) Impala orangeite dyke IP20-1 exhibiting abundant zoned phlogopite laths in the groundmass plus interstitial clinopyroxene, Cr-rich spinel and serpentine (?pseudomorphic after olivine microphenocrysts). (c) Impala evolved orangeite dyke IP20-9 exhibiting flow-aligned phlogopite laths that drape around serpentinized olivine microphenocrysts. Note the discrete groundmass Cr-rich spinel grains (cross polarized light). (d) Impala evolved orangeite dyke IP20-2 exhibiting complexly zoned phlogopite laths and plates in the groundmass plus interstitial acicular clinopyroxene prisms, equant Cr-rich spinel grains and calcite. Note also the serpentinized subhedral olivine microphenocryst. (e) Sibanye evolved orangeite dyke LM4 characterized by rare serpentinized subhedral olivine microphenocrysts set in a fine groundmass that is dominated by acicular phlogopite and clinopyroxene plus rare equant Cr-rich spinel grains (plane polarized light). (f) Sibanye evolved orangeite dyke LM5 showing a zoned and inclusion-bearing phlogopite microphenocryst set in a fine groundmass that is dominated by acicular phlogopite and clinopyroxene laths forming a decussate micro-texture. Cal, Calcite; Cpx, clinopyroxene; Ol, olivine; Phl, phlogopite; Spl, spinel; Srp, serpentine.
Mineral compositions

Olivine. Owing to widespread alteration of olivine to serpentine (Fig. 3a), olivine major and minor element analysis was only possible for orangeite dyke samples from the Impala mining concession (Supplementary Material 1). Olivine in the Impala orangeite dykes shows a rather restricted compositional range with high forsterite (91.9–93.6 mol%) and NiO (0.33–0.42 wt%) contents at <0.07 wt% CaO (Fig. 4). These compositions resemble those of peridotite-derived mantle xenocrysts reported from kimberlites and related rocks on cratons worldwide (Kamenetsky et al. 2008; Tappe et al. 2009; Bussweiler et al. 2015; Howarth and Taylor 2016; Giuliani 2018; Shaikh et al. 2019; Rooney et al. 2020; Ngwenya and Tappe 2021). Olivine in the evolved orangeite dykes has slightly lower forsterite contents (90.8–92.4 mol%) at elevated NiO (0.38–0.46 wt%), CaO (0.11–0.

Fig. 4. Olivine major and minor element compositions in Bushveld-intersecting orangeite and evolved orangeite dykes from Impala. (a) NiO v. forsterite contents. (b) CaO v. forsterite contents. Published olivine compositions for coarse and sheared peridotite xenoliths from the Kaapvaal craton are shown for comparison and the data are listed in Supplementary Material 1 together with the literature sources.
16 wt%) and MnO (0.11–0.16 wt%) concentrations (Fig. 4), which is more typical of magmatic olivine in ultrapotassic systems such as lamproites (Prelevic and Foley 2007; Jaques and Foley 2018; Howarth and Giuliani 2020). Olivine inclusions in phlogopite have the lowest forsterite (89.6–90.0 mol%) and NiO (0.17–0.21 wt%) contents at elevated CaO (0.21–0.23 wt%) and MnO (0.20–0.22 wt%) (Fig. 4), suggestive of a magmatic origin during the early stages of groundmass crystallization.

Phlogopite. Phlogopite is the dominant mica type in the Impala and Sibanye orangeite dykes, where it is largely restricted to the groundmass (Fig. 3). However, phlogopite microphenocrysts also occur in the form of laths and plates ranging in size between 0.3 and 1 mm. They show complex zoning and typically contain minute inclusions of spinel, olivine and carbonate (Fig. 3f). The phlogopite laths and plates are commonly intersected by prismatic groundmass diopside establishing their somewhat earlier crystallization during fast cooling of the narrow dykes (Fig. 3d).

Phlogopite laths and plates in both the olivine-rich and olivine-poor (or evolved) orangeite dykes from Impala have similar core compositions that are MgO-rich and relatively poor in Al₂O₃ and FeO, similar to the phlogopite core compositions in the evolved orangeites from Sibanye (Fig. 5a–c). Although phlogopite from the three sample subsets has evolved by Al-depletion and Fe-enrichment (i.e. a tetraferriphlogopite trend), we note subtle differences in the individual core–rim trends relating to the magnitude of the Al–Fe fractionation (Fig. 5a, b). Moreover, phlogopite from the olivine-rich Impala orangeites shows strong Ti-depletion towards the rims, whereas micas in the evolved orangeites from Impala and Sibanye have constant or increasing TiO₂ concentration levels, respectively, towards the rims (up to 4.8 wt% TiO₂; Fig. 5a). In general, the Al–Ti–Fe systematics of phlogopite in the potassic ultramafic dykes from the western Bushveld Complex are typical for Kaapvaal craton orangeites (Mitchell 1995), and only the most evolved dyke compositions from the Sibanye mining concession show a subtle lamproitic mica affinity (e.g. Ti-enrichment). An affinity of the Impala and Sibanye dykes to ultramafic lamprophyres can be largely ruled out because of their generally low Al₂O₃ contents of mica (<13.5 wt%), which is in contrast to the relatively Al-rich phlogopite compositions (>14 wt% Al₂O₃) of allilikes from localities worldwide (Tappe et al. 2004; Tappe et al. 2006; Nielsen et al. 2009; Nasir et al. 2011; Dalton et al. 2019; Fig. 5a).

The Cr₂O₃ contents of phlogopite in the orangeite dykes from the western Bushveld Complex are mostly <1 wt%. However, rare resorbed mica cores in the olivine-rich Impala dykes and more evolved Sibanye dykes reach higher Cr₂O₃ concentration levels of up to 1.8 wt% (Fig. 5c). The phlogopite BaO and F concentration levels are typically below 1 wt%, which presents a marked compositional difference to archetypal kimberlites (Giuliani et al. 2016) and cratonic lamproites (Ngwenya and Tappe 2021). These Ba–F systematics are similar to those of phlogopite in ultramafic lamprophyres such as type allilike (Tappe et al. 2006). Very rare biotite laths occur only in the evolved orangeite dykes from the Impala and Sibanye mining concessions, and they show some of the highest Al₂O₃ (up to 14.3 wt%) and F (up to 1.1 wt%) contents among the analysed micas at very low Mg# (<45) and Cr₂O₃ (<0.1 wt%) (Fig. 5c).

Clinopyroxene. Clinopyroxene is diopside-rich and occurs as small prismatic microphenocrysts (coeval with the phlogopite microphenocrysts) and needle-shaped crystals in the groundmass of the orangeite dykes that intrude the western Bushveld Complex. It also occupies interstitial space between slightly larger interlocking phlogopite laths (Fig. 3b, d). Although subtle compositional zoning has been observed for the clinopyroxene prisms, it is not as prominent as for the coexisting phlogopite laths and plates.

Clinopyroxene in the Impala orangeite dykes has an almost pure diopside composition (En₄₄.₃–₄₉.₆ W₀₉₀.₃–₇₁.₆Fs₀.₅–₃) with very low Al₂O₃ (<0.5 wt%) and TiO₂ (<1 wt%) contents (Fig. 5d). Diopsiderich clinopyroxene in the groundmass of the evolved orangeite dykes from both the Impala and Sibanye mining concessions (En₀.₀₉–₀.₄₁ W₀₉₇.₇–₅₁.₆Fs₀.₅–₁₀) is also Al₂O₃ poor (<0.5 wt%), but it has notably higher TiO₂ concentrations compared with diopside in the more primitive olivine-rich dykes from Impala, ranging between 1.6 and 2.6 wt% (Fig. 5d). The Cr₂O₃ and Na₂O contents of clinopyroxene are similarly low for the three sample subsets, reaching up to 0.2 and 1.36 wt%, respectively. These groundmass clinopyroxene compositions are typical for Kaapvaal craton orangeites (Mitchell 1995), whereas they differ greatly from the much more Al- and Ti-enriched groundmass diopside compositions of ultramafic lamprophyres from localities worldwide (e.g. Tappe et al. 2004, 2006; Dalton et al. 2019; Fig. 5d).

Spinel-group minerals. Spinel-group minerals are common accessory constituents of the petrographically diverse samples from the orangeite dyke swarm examined here. They typically occur as <50 µm euhedral to subhedral inclusions within microphenocrystic olivine and phlogopite, but are only rarely observed as <100 µm groundmass phase occupying textural interstices (Fig. 3). Atoll-
Fig. 5. Major and minor element compositions of petrogenetically important mineral constituents of Bushveld-intersecting orangeite dykes. (a) Al$_2$O$_3$ v. TiO$_2$; (b) Al$_2$O$_3$ v. total FeO; and (c) Cr$_2$O$_3$ and F v. Mg-number for abundant phlogopite and rare biotite in the groundmass. (d) Atomic Al v. Ti contents of diopside-rich groundmass clinopyroxene. (e) $\text{Fe}^{3+}/(\text{Fe}^{3+} + \text{Al} + \text{Cr})$ and (f) Ti/(Ti + Al + Cr) v. $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$ for groundmass spinel. Mineral compositional trends that are typical for kimberlites, lamproites and orangeites are taken from Mitchell (1995), whereas those for ultramafic lamprophyres or UML are adopted from Tappe et al. (2004, 2005, 2006).
textures, as commonly observed for groundmass spinel in archetypal kimberlite and aillikite (Mitchell 1986; Tappe et al. 2006), are notably absent in our orangeite samples. Spinel in the Impala orangeite dykes evolved from chromite to Ti-rich magnetite along a trajectory reminiscent of ‘Kimberlite Trend 2’, although intermediate compositions are conspicuously absent (Fig. 5e, f). Such an evolutionary trend at increasing Fe$^{3+}$/Mg is typical for Kaapvaal craton orangeites, but also for cratonic lamproites from localities worldwide (Mitchell and Bergman 1991; Mitchell 1995). In contrast, groundmass spinel compositional evolution in bona fide kimberlite magmatic systems occurs along relatively constant and low Fe$^{3+}$/Mg, the so-called ‘Kimberlite Trend 1’ (Mitchell 1986; Roeder and Schulze 2008). However, recent work has shown that evolutionary paths in between the two major ‘kimberlitic’ spinel trends may be followed by both archetypal kimberlites and aillikites (Tappe et al. 2009; Tappe et al. 2014; Dongre and Tappe 2019). This observation renders spinel compositions less diagnostic for the identification of volatile-rich ultramafic magma types than previously thought. Regardless, the evolved orangeites from the Impala and Sibanye mining concessions apparently contain only Cr-rich spinel grains (87.2–91.6 Cr#) that overlap with the compositions of chromite from the more primitive olivine-rich orangeite dykes at Impala (82.1–90.3 Cr#) (Fig. 5e, f).

**Andradite garnet.** Andradite-rich garnet occurs as <100 µm large groundmass grains within the evolved orangeite dykes of the Impala mining concession. The magmatic garnet grains exhibit very high CaO (32.0–32.5 wt%), TiO$_2$ (15.8–18.0 wt%) and FeO (17.5–18.2 wt%) contents at <0.01 wt% Cr$_2$O$_3$ (Fig. 6). They represent a solid solution series between the andradite, Ti-andradite and schorlomite garnet endmembers, with only a very small pyrope component. The Ti-rich andradite grains analysed here have similar compositions to the primary magmatic Ti-andradite reported from the groundmass of ultramafic lamprophyres on the North Atlantic craton in eastern Canada and West Greenland (Tappe et al. 2004; Tappe et al. 2006; Tappe et al. 2009), as well as from orangeites in the Dharwar craton in southern India (Dongre et al. 2016; Choudhary et al. 2020; Fig. 6). They are more Ti-enriched than the relatively pure andradite compositions reported from carbonatite dykes of the Premier kimberlite pipe on the central Kaapvaal craton (Fig. 6), which were interpreted to have a secondary origin (Dongre and Tappe 2019). We note that Hammond and Mitchell (2002) reported an ‘unidentified Ca–Ti–Fe–silicate’ phase from the Swartruggens orangeite dyke swarm some 60 km west of the Bushveld Complex on the central Kaapvaal craton (Fig. 1), and this phase possibly represents Ti-rich andradite of a primary magmatic origin (Fig. 6). Regardless, it appears as though magmatic Ti-andradite is a relatively common accessory mineral in evolved orangeites and, thus, less diagnostic for ultramafic lamprophyres than previously thought (cf. Tappe et al. 2005).

**Bulk rock major and trace element compositions**

The Impala orangeite dykes have higher MgO (22.6–26.1 wt%), Cr (419–1694 ppm) and Ni (655–930 ppm) contents than the evolved orangeite dykes from this locality (15.1–20.2 wt% MgO, 415–1260 ppm Cr, 302–798 ppm Ni) and from the Sibanye mining concession (14.9–17.1 wt% MgO, 460–563 ppm Cr, 341–454 ppm Ni). The TiO$_2$ (0.78–1.24 wt%), CaO (6.24–8.69 wt%), Na$_2$O (0.05–0.68 wt%) and P$_2$O$_5$ (0.10–0.66 wt%) contents are notably lower for the Impala orangeite dykes compared with the evolved varieties from Impala (1.31–3.04 wt% TiO$_2$, 8.33–13.4 wt% CaO, 1.06–1.65 wt% Na$_2$O, 0.54–1.72 wt% P$_2$O$_5$) and Sibanye (1.78–1.93 wt% TiO$_2$, 10.4–11.8 wt% CaO, 1.00–1.61 wt% Na$_2$O, 0.89–0.97 wt% P$_2$O$_5$; Fig. 7). The other major and minor elements show
broad compositional overlap between the three sample subsets. For example, the SiO$_2$ (32.2–41.2 wt%), Al$_2$O$_3$ (4.41–6.64 wt%) and CO$_2$ (0.25–4.71 wt%) contents are variably low, whereas the K$_2$O (3.15–6.90 wt%) concentration levels are collectively high (Fig. 7b, e, f). The entire suite of samples from the Bushveld-intersecting orangeite dyke swarm has a truly ‘ultrapotassic’ character according to the criteria of Foley et al. (1987), with >3 wt% MgO and K$_2$O, respectively, at K$_2$O/Na$_2$O ≥2 (2.0–86.2)
Compared with other potassic ultramafic rock types, the Bushveld-intersecting ultrapotassic dykes show a good compositional match to typical Kaapvaal craton orangeites (Fig. 7), which is in agreement with their mineral assemblages and compositions (see above). However, there is also some overlap in terms of major element compositions with cratonic lamproites and ultramafic lamprophyres from continental shields worldwide (Figs 7 & 8).

The orangeite dykes analysed here are strongly enriched in incompatible trace elements and they have similar primitive mantle normalized distributions compared with other Kaapvaal craton orangeites (Becker and le Roex 2006; Coe et al. 2008) (Fig. 9). Whereas the olivine-rich Impala dykes fall at the lower end of the incompatible element concentration range of Kaapvaal orangeites, the olivine-poor or evolved orangeites from the Impala and Sibanye mining concessions cover the middle and upper ranges (Fig. 9), but the trace element patterns for the three sample subsets are very similar (Figs 9 & 10). A characteristic feature of Kaapvaal craton orangeites is their strong LILE (Cs–Rb–Ba–K) enrichment coupled to a marked relative depletion of certain high field strength elements (HFSE; Th–U–Nb–Ta; Coe et al. 2008), and the Bushveld-intersecting orangeite dykes studied here are no exception (Fig. 9). This geochemical feature also displays prominently in the Ba/Nb v. La/Nb diagram (Fig. 11a), in which the Impala dykes even extend the level of LILE/HFSE fractionation that is
commonly observed for South African orangeites (Becker and le Roex 2006).

**Bulk rock Sr–Nd–Hf isotopic compositions**

The initial Sr–Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes from the central Kaapvaal craton are calculated for a magma emplacement age of 140 Ma (Table 1; Supplementary Material 1). The initial $^{87}\text{Sr}/^{86}\text{Sr}$ of the three sample subsets are all elevated and fall within a relatively narrow range between 0.707012 and 0.707410. The range displayed by the corresponding initial $^{143}\text{Nd}/^{144}\text{Nd}$ is relatively wide, with subchondritic initial $\varepsilon\text{Nd}$ values from $-10.6$ to $-5.8$, although the majority of samples fall between $-10.6$ and $-9.6$ (Fig. 12). The olivine-rich orangeite dykes from Impala tend to have more radiogenic Nd isotopic compositions compared with the olivine-poor or evolved orangeite dykes from the Impala and Sibanye mining concessions (Figs 12 & 13). The corresponding initial $^{176}\text{Hf}/^{177}\text{Hf}$ ratios show even larger variations, with subchondritic initial $\varepsilon\text{Hf}$ values from $-14.4$ to $-2.5$ (Fig. 13). Similar to the Nd isotope systematics, the olivine-rich orangeite dykes from Impala have more radiogenic Hf isotopic compositions than the evolved varieties. The Bushveld-intersecting orangeite dykes fall firmly within the terrestrial Nd–Hf isotope array (Chauvel et al. 2008; Fig. 13), with initial $\Delta\varepsilon\text{Hf}$ values between $+0.4$ and $+5.5$ (Table 1). The positively correlated Nd–Hf isotope systematics are in contrast to the commonly observed isotopic decoupling that characterizes many archetypal kimberlite sample suites from cratons worldwide (for a review see Tappe et al. 2020a). In general, the combined Sr–Nd–Hf isotope systematics of the Bushveld-intersecting orangeite dykes reveal moderately to strongly enriched source compositions relative to primitive mantle, very similar to those of other orangeite occurrences from across the Kaapvaal craton (Figs 12 & 13).

**Discussion**

**Mesozoic orangeite magmatism on the central Kaapvaal craton**

The Swartruggens dyke swarm as a benchmark. The c. 145 Ma potassic ultramafic dyke swarm near Swartruggens is one of the best-studied orangeite occurrences on the central Kaapvaal craton and thus provides a benchmark against which to compare the petrogenesis of the Bushveld-intersecting orangeite dykes near Rustenburg (Fig. 1). The Swart-ruggens dykes, exploited at Helam Diamond Mine, have been variably referred to as kimberlites, lamprophyres, minettes, lamproites and, more recently, as orangeites (Fourie 1958; Skinner and Scott 1979; Mitchell and Bergman 1991; Mitchell 1995; Gurney and Kirkley 1996; Klump and Gurney 1998; Hammond and Mitchell 2002; McKenna et al. 2004; Coe et al. 2008). Although some studies used this terminology interchangeably, several workers pointed out that the Swartruggens dyke system is structurally and compositionally highly complex, comprising multiple intrusive phases and showing evidence of internal differentiation processes that may have created the highly variable petrographic characteristics and diverse mineralogy...
from the highly diamondiferous dykes (Klump and Gurney 1998; McKenna et al. 2004). On the basis of a detailed geochemical investigation, Coe et al. (2008) concluded that the compositionally highly variable Swartruggens orangeite dykes are related to a common parental magma, which suggests that differentiation processes have a strong control on primitive potassic magma type diversification upon ascent, including redistribution of any possibly existing diamond cargo.

The c. 140 Ma orangeite dykes on the western Bushveld Complex studied here are structurally and compositionally very similar to the Swartruggens dyke swarm (Fig. 1). Our sample suite from these Bushveld-intersecting orangeite dykes allows for an evaluation of the role of crystal fractionation in the evolution of these potassic ultramafic magmas, which is an often-neglected process that operates in the plumbing system of deep-sourced volatile-rich melts on and around cratons worldwide.

**Ultrapotassic magma evolution at Impala and Sibanye.** The Bushveld-intersecting orangeite dykes comprise relatively olivine-rich and very olivine-poor varieties, illustrated by the example of drillcore intersections in Figure 2c. Importantly, both varieties contain compositionally overlapping Cr-spinel inclusions and discrete groundmass grains (Fig. 5e, f), which suggests a common parental magma to the entire orangeite dyke swarm on the central Kaapvaal craton, similar to the interpretation by Coe et al. (2008) for the Swartruggens dyke system.

Although a significant portion of the macrocrystic olivine in the olivine-rich orangeite dykes from Impala is derived from disaggregated cratonic mantle peridotites (i.e. xenocrystic), magmatic olivine is present in these rocks and reveals the diagnostically high Ni and elevated Ca contents commonly observed in olivine that crystallized from ultrapotassic magmas such as lamproites (Prelevic et al. 2013; Jaques and Foley 2018). These Ni- and Ca-enriched magmatic olivine compositions partially overlap with those of rare olivine microphenocrysts in the more evolved orangeite dykes. The evolved orangeite dykes contain olivine inclusions in groundmass phlogopite laths and they record magmatic evolution towards lower forsterite and Ni contents at elevated CaO (Fig. 4). Hence, olivine compositions in both the olivine-rich and olivine-poor or evolved orangeites provide evidence for magmatic olivine fractionation within this ultrapotassic system, superimposed on the effect of relatively minor xenocrystic olivine addition.

The geochemical variation diagrams in Figure 14, using MgO as a ‘differentiation’ index, show that the olivine-rich orangeite dykes from Impala have slightly higher MgO contents between 22.6 and...
Fig. 12. Initial Sr–Nd isotopic compositions of Bushveld-intersecting orangeite dykes. Analytical uncertainties including a full propagation of errors are smaller than or similar to symbol size (Table 1). Fields for comparisons are based on data from the following literature sources: southern African Group 1 kimberlites – Smith (1983), le Roex et al. (2003), Nowell et al. (2004), Becker and le Roex (2006), Griffin et al. (2014), Sarkar et al. (2014), Tappe et al. (2020a, b); southern African Group 2 kimberlites or orangeites – Fraser et al. (1985), Nowell et al. (2004), Becker and le Roex (2006), Coe et al. (2008); global kimberlites including groundmass perovskite – Tappe et al. (2017); global OIB – Stracke (2012). CHUR, Chondritic uniform reservoir; OIB, oceanic island basalts.

Fig. 13. Initial Nd–Hf isotopic compositions of Bushveld-intersecting orangeite dykes. Analytical uncertainties including a full propagation of errors are smaller than or similar to symbol size (Table 1). Fields for comparisons are based on data from the following literature sources: southern African Group 1 kimberlites – Nowell et al. (2004), Tappe et al. (2020a, b); southern African Group 2 kimberlites or orangeites – Nowell et al. (2004), Coe et al. (2008); global kimberlites – Tappe et al. (2017); global OIB – Stracke (2012). The terrestrial array regression line is taken from Chauvel et al. (2008).
26.1 wt% compared with the estimated close-to-primary magma composition of the Swartruggens orangeite dyke swarm at c. 21 wt% MgO (Coe et al. 2008). Given the similar age and Sr–Nd–Hf isotopic compositions, plus the close geographic proximity on the central Kaapvaal craton (Fig. 1), it is reasonable to infer that a compositionally similar parental magma supplied both dyke systems. If correct, up to 10% of xenocrystic olivine addition to the parental Impala orangeite magma can explain their higher MgO contents relative to the close-to-primary ultrapotassic magma composition of the Swartruggens dyke swarm (Fig. 14). Regarding the more evolved orangeite varieties from the Bushveld-intersecting dyke swarm, their MgO contents range from 20.2 down to 14.9 wt%, which suggests between 2 and 12% of olivine fractionation, arguably controlled by the removal of magmatic olivine. Alternatively, flowage-segregation and filter-pressing may have contributed to the observed

Fig. 14. Bulk rock major (a–c) and trace (d–f) element variation diagrams for Bushveld-intersecting orangeite dykes using MgO as a differentiation index. The data compilation for type orangeites from the Kaapvaal craton is taken from Ngwenya and Tappe (2021). The estimated close-to-primary magma compositions of Kaapvaal orangeites (error bars at 1-sigma) and Swartruggens orangeites are adopted from Becker and le Roex (2006) and Coe et al. (2008), respectively. The inferred olivine control line is guided by olivine compositions from the Impala orangeite dykes (Supplementary Material 1).
geochemical ‘olivine control’, but such intra-dyke differentiation mechanisms are difficult to quantify in kimberlitic magmatic systems (Kjarsgaard et al. 2009; Zurevinski and Mitchell 2011; Tappe et al. 2014; Abersteiner et al. 2019). Regardless of the circumstances of olivine control, it is generally accepted that olivine and diamond crystals are transported in a similar manner within flowing and erupting kimberlitic magmas, mainly because of their similar densities (Grunsky and Kjarsgaard 2008; Field et al. 2009; Moss et al. 2010). Thus, identification of olivine fractionation helps to explain the lack of diamonds in the Bushveld-intersecting orangeite dykes on the central Kaapvaal craton, a geological domain that has a highly diamondiferous lithospheric mantle root (Viljoen et al. 2004; Field et al. 2008; Korolev et al. 2018; Tappe et al. 2018; Smart et al. 2021; Fig. 1).

A firm olivine control on the petrogenetic link between the Impala orangeites and the more evolved varieties from Impala and Sibanye is also supported by both compatible (e.g. Ni) and incompatible (e.g. Zr, Ce) trace element variations with varying MgO contents (Fig. 14d–f). Furthermore, the similar primitive mantle normalized incompatible element patterns of the olivine-rich and olivine-poor orangeite varieties, with a systematic offset to higher trace element abundances in the samples with the lowest modal olivine content, are best explained by olivine fractionation (Fig. 9). This effect is also evident in the chondrite-normalized REE diagram (Fig. 10), where complete compositional overlap exists between the most primitive Swartruggens and Impala orangeite dykes, whereas the more evolved varieties from Impala and Sibanye exhibit up to an order of magnitude higher REE concentrations, at broadly parallel element distributions for all rock suites discussed here (Fig. 10).

*K-metasomatized cratonic mantle sources. Similar to global anorogenic lamproite volcanism, type orangeites from the Kaapvaal craton and adjoining mobile belts are characterized by strongly enriched Sr–Nd–Hf isotopic compositions (Nowell et al. 2004). The long-term enriched geochemical signature of the mantle source to orangeite magmatism stands in contrast to the more primitive and moderately depleted isotopic compositions of archetypal kimberlites in southern Africa (Figs 12 & 13). The striking isotopic difference between the two major diamond ‘delivery’ systems on the Kaapvaal craton forms the basis of petrogenetic models that invoke ancient metasomatized lithospheric mantle sources for orangeites as opposed to athenospheric sources for kimberlites (Smith 1983; Fraser et al. 1985; Nowell et al. 2004; Tappe et al. 2020a, b).

The Sr–Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes from the central Kaapvaal craton are similar to those of other orangeite localities in South Africa, including the Swartruggens dyke swarm (Coe et al. 2008; Figs 12 & 13). For the Impala and Sibanye dykes, the olivine-rich and more evolved orangeite varieties have broadly overlapping isotopic compositions, but we note that dyke IP20-1 shows significantly higher Nd and Hf isotope ratios at similarly radiogenic $^{87}$Sr/$^{86}$Sr, when compared with the other orangeite dykes (Figs 12 & 13). Dyke IP20-1 from Impala also shows more extreme HREE depletion and, altogether, these geochemical features suggest a more notable interaction of the ultrapotassic melt with much less enriched lithologies within the Kaapvaal cratonic mantle root (Figs 9–13). This material was probably clinopyroxene bearing, because among common cratonic mantle minerals it is the only phase that can significantly influence the Nd and Hf budgets of passing melts alike, retaining isotopic coupling (Griffin et al. 2000). Alternatively, the detected variability in the Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes may reflect lithospheric mantle source heterogeneity. For example, MARID-type metasomes have frequently been invoked as source components to potassic ultramafic magmas (Foley 1992; Ulmer and Sweeney 2002; Tappe et al. 2008; Smart et al. 2019), and isotopic investigations of such heavily K-metasomatized lithospheric mantle materials revealed extreme heterogeneity down to the centimetre scale (Kramers et al. 1983; Grégoire et al. 2002; Choukroun et al. 2005; Fitzpayne et al. 2019).

A ‘stored’ subduction signature in Kaapvaal cratonic mantle metasomes. Type orangeites from South Africa collectively exhibit a strong Cs–Rb–Ba–K enrichment that is coupled to a relative depletion in Th–U–Nb–Ta (Becker and le Roex 2006; Coe et al. 2008; this study). This distinctive trace element signature is associated with remarkably low TiO$_2$ contents of 2 wt% for the most primitive orangeite varieties (Figs 7a & 9), much lower than what is typically observed for other potassic ultramafic magma types on cratons, such as anorogenic lamproites and aillikites, which reach up to 8 wt% TiO$_2$ (Figs 7a & 9). The high LILE/HFSE ratios of Kaapvaal craton orangeites are reminiscent of a classic subduction zone signature (Fig. 11), where residual titanate minerals (e.g. rutile) in the downgoing oceanic slab retain in particular Ti–Nb–Ta while the LILE (and LREE) become enriched in extracted melts/ fluids and enter the lithospheric mantle of the overriding tectonic plate (Foley and Wheller 1990; Pearce and Peate 1995).

As Coe et al. (2008) pointed out, the Kaapvaal craton was last subjected to subduction and collisional events in its immediate vicinity during the Mesoproterozoic Namaqua–Natal orogeny at
1220–1090 Ma (Spencer et al. 2015). Although the geometry of these ancient subduction zones is far from understood (Jacobs et al. 2008; Van Schijndel et al. 2020), it is reasonable to assume that oceanic slab remnants temporarily stalled beneath the thick Kaapvaal lithosphere and released small volumes of metasomatic melts/liquids by which the widespread ‘subduction’ signature was introduced into the lowermost cratonic mantle. Structural responses of the Kaapvaal lithosphere to these Mesoproterozoic collisions are evident from its burial and unroofing history (Baughman and Flowers 2020), and also from the prolonged emplacement history of the Premier kimberlite cluster in the central part of the craton between c. 1155 and 1135 Ma (Tappe et al. 2020a).

A direct origin of potassic ultramafic magmas from slab remnants, including subducted sedimentary components (Murphy et al. 2002; Rapp et al. 2008), within the deeper convecting mantle is excluded here. The main reason for this is that the phlogopite/richterite-carbonate bearing peridotitic source assemblages required to explain orangeites and related rocks are largely restricted to the pressure–temperature–$\Delta$O$_2$ conditions of the lower cratonic mantle lithosphere (Tappe et al. 2007; Foley 2011). Ulmer and Sweeney (2002) pointed out that even under such cratonic mantle conditions, mixed phlogopite/richterite–carbonate assemblages are only metastable and probably do not persist for prolonged periods of geological time. This led Tappe et al. (2008) to propose a model in which ancient phlogopite–richterite-bearing cratonic mantle metasomes are fused by small volumes of infiltrating CO$_2$-rich melts from the underlying asthenosphere, giving rise to K-rich carbonated ultramafic silicate magmas such as aillikites and orangeites.

An inherited subduction signature manifested in cratonic mantle metasomes is supported by $^{40}$Ar/$^{39}$Ar age results for phlogopite from strongly metasomatized peridotites along the southern Kaapvaal craton margin, yielding ages of 1.25–1.0 Ga (Hopp et al. 2008). In contrast, U–Pb geochronology of zircon in MARID-type xenoliths from the western Kaapvaal craton revealed exclusively Late Mesozoic ages (Konzett et al. 1998; Giuliani et al. 2015; Hoare et al. 2021), but it is uncertain whether these dates represent the timing of K-rich mantle metasomatism or simply the time of isotopic closure as a consequence of lithospheric mantle cooling after a short-lived, yet impactful regional heating event during the Early Mesozoic (Bell et al. 2003). Regardless of the timing of lithospheric mantle K-enrichment, it is remarkable how uniform this enriched domain apparently was in terms of its mineralogical and geochemical compositions, as evident from the similar incompatible element systematics (Figs 9–11) and Sr–Nd–Hf isotopic compositions (Figs 12 & 13) of type orangeites. These deep-sourced ultrapotassic magmas erupted in pulses on and off the Kaapvaal craton, across an area that was 1000 $\times$ 750 km in size, during progressive breakup of the Gondwana superterrane between 200 and 110 Ma (Tappe et al. 2018).

**Orangeite v. lamproite terminology: what’s in a name?**

The potassic ultramafic dykes on the central Kaapvaal craton in South Africa highlight the problems surrounding naming conventions for kimberlites and related rocks. They have been variably referred to as kimberlites, lamprophyres, minettes, lamproites and orangeites (Skinner and Scott 1979; Leeb-Du Toit 1986; Mitchell and Bergman 1991; Mitchell 1995; Gurney and Kirkley 1996; Hammond and Mitchell 2002; McKenna et al. 2004; Coe et al. 2008), even by the same authors over short periods of time (compare Mitchell and Bergman (1991, p. 72) with Mitchell (1995, p. 30)). There appears to be wide consensus within the alkaline rock petrology community that the term Group 2 kimberlite should be abandoned in favour of these potassic rocks being called orangeites (Mitchell 1995), but in recent years there has been some development to fully assimilate the term orangeite into the lamproite clan. What began with notions about orangeites being the ‘lamproite variety Kaapvaal’ (Mitchell 2006) has now culminated in orangeites being considered as ‘CO$_2$-rich lamproites’ or ‘carbonate-rich lamproites’ (Pearson et al. 2019). This development is regrettable for three main reasons: first, lamproites from classic potassic igneous provinces around the globe are mostly free from primary magmatic carbonate but tend to have a silicate glass matrix instead, such that the term CO$_2$-rich lamproite presents an oxymoron in a resemblance classification scheme that relies on similarities to standard members (Foley et al. 1987). Second, type orangeite from the Kaapvaal craton commonly comprises primitive and evolved varieties, the latter of which may approach CO$_2$ concentration levels of zero (Fig. 71) and thus cannot be referred to as CO$_2$-rich lamproites. Third, the introduction of a carbonate-rich lamproite subgroup obstructs the classification of ultramafic lamprophyres (Tappe et al. 2005), because the carbonate-rich lamprophyre variety aillikite would, by the same logic that was applied to type orangeite, also belong to the newly proposed lamproite class. Recently, Sarkar et al. (2018) referred to carbonate-rich potassic ultramafic rocks, akin to aillikite or orangeite, as lamproites, which highlights the need for a more coherent classification scheme that can satisfactorily accommodate the petrographical variability of mantle-derived volatile-rich igneous rocks (Mitchell and Tappe 2010).
Here we argue to maintain the status quo of orangeite as a discrete type of mantle-derived potassic magma with occurrences worldwide (Chalapathi Rao et al. 2011; Donatti-Filho et al. 2013; Kargin et al. 2014; Dalton et al. 2019; Chalapathi Rao et al. 2020; Choi et al. 2021; Krmíček et al. this volume, in press). The recognition that orangeites or Group 2 kimberlites may show closer compositional affinities to cratonic lamproites and allilikes than to archetypal Group 1 kimberlites was a milestone in alkaline rock petrology (Dawson 1987; Mitchell 2005), clearly serving a petrogenetic purpose (Tappe et al. 2005). However, modifications of and changes to the terminology of igneous rocks without any obvious advance in our understanding of magma petrogenesis are not endorsed here. Although the names of rocks do not affect what they really are and how they formed – alkaline rock terminology is highly complex and does not appear to benefit from incoherently applied changes – this only complicates matters further.

Conclusions

- The mineralogical and geochemical compositions of 15 individual potassic ultramafic dykes, intersecting the western Bushveld Complex, show that these Mesozoic minor intrusions represent ‘typical’ Kaapvaal craton orangeites, formerly known as Group 2 kimberlites. A strong olivine control is recognized in the magmatic evolution of the orangeite dykes, with olivine-poor more evolved varieties petrographically resembling lamprophyres. Olivine fractionation and physical sorting are held responsible for the absence of significant quantities of diamond, even though the orangeite magmas traversed one of the richest diamond repositories in the world, as represented by the central Kaapvaal craton mantle lithosphere.

- The enriched Sr–Nd–Hf isotopic compositions of the Bushveld-intersecting orangeite dykes suggest a magma origin from ancient K-metasomatized mantle lithosphere, similar to the source of other Kaapvaal craton orangeites. Despite the highly enriched incompatible trace element contents, the orangeite samples studied exhibit pronounced depletions in Th–U–Nb–Ta as well as Ti, resulting in high LILE/HFSE ratios. Such a subduction zone geochemical signature is common among the Mesozoic orangeites from across the Kaapvaal craton. The ‘stored’ subduction signature is best ascribed to K-rich metasomatism of the cratonic mantle lithosphere in the vicinity of Mesoproterozoic collision zones associated with the Namaqua–Natal orogeny during Rodinia supercontinent formation.

- Our study reinforces that orangeites represent a discrete type of mantle-derived potassic magma. Although compositional similarities with cratonic lamproites and ultramafic lamprophyres do exist, type orangeite from South Africa is sufficiently distinct such that different magma formation processes are implied.

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