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Two expressions of the transition from mafic cumulates to granitoids in the Bushveld Complex, South Africa: Examples from the western and eastern limbs

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ABSTRACT

In the Bushveld Complex of South Africa, numerous granitic and granophyric rocks termed the Lebowa Granite and Rashoop Granophyre Suites, respectively, overlie the layered ultramafic-mafic rocks of the Rustenburg Layered Suite. Despite their close spatial and temporal association, the granites and granophyres are often interpreted as being unrelated to the Rustenburg Layered Suite. This paper describes the transition from the uppermost Rustenburg Layered Suite into overlying granite and granophyre at three locations in the western (the Bierkraal drill core) and eastern (Diepkloof farm and Stoffberg town) limbs of the Bushveld Complex. In the western limb, a ~60 m thick transition zone bridges the petrological gap between the overlying Nebo Granite (Lebowa Granite Suite) and Upper Zone (Rustenburg Layered Suite). Across the transition zone, the composition of olivine changes from Fo₆ to Fo₁, clinopyroxene from Mg#₂₅ to Mg#₂ and plagioclase from An₄₅ to An₁₆. At Stoffberg in the eastern limb, the Upper Zone dioritic cumulates grade into the overlying monzonitic Roof Zone. Across the Roof Zone, plagioclase compositions change from An₄₂ to An₄, clinopyroxene from Mg#₃₀ to Mg#₁₁ and olivine from Fo₉ to Fo₅. Based on geochemistry and petrography, we correlate the lowermost stratigraphy at the farm Diepkloof with the Stoffberg Roof Zone. At Diepkloof, the Roof Zone grades into the overlying Stavoren Granophyre (Rashoop Granophyre Suite). All units are indistinguishable in terms of bulk rock Nd ($\epsilon_{Nd} = -6.4$ to -5.4) and Hf ($\epsilon_{Hf} = -9.2$ to -6.6) isotopes (corrected to 2055 Ma). Similarly, the Upper Main Zone, Upper Zone and transition zone are indistinguishable in terms of bulk rock Sr isotopes ($(^{87}Sr/^{86}Sr)_{2055\text{ Ma}} = 0.7071$ to 0.7076 , except one transition zone outlier at 0.7058). We submit that the transition zone represents the fossil record of bulk and/or diffusional mixing between coexisting Upper Zone and Nebo Granite magmas. We test this hypothesis by combining field, petrographic and geochemical observations with forward modelling using the Rhyolite-MELTS algorithm. Our work on the Bierkraal core (western limb) shows that at least a portion of the granitic magma was emplaced before the residual liquid of the Upper Zone had solidified. At Stoffberg and Diepkloof in the eastern limb (where the granite is absent), the Roof Zone underwent uninterrupted fractional crystallization.

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1. Introduction

Layered mafic intrusions are usually associated with varying volumes of granites and granophyres (Wager and Brown, 1968), for instance the Sept-Iles (Namur et al., 2011), Muskox (Stewart and DePaolo, 1992) and Bjerkreim-Sokndal (Wilson et al., 1996) intrusions. Despite their common association, interpreting the genetic relationship between layered mafic intrusions and granites and granophyres can be complicated by poor constraints on their structural and temporal relations as well as ambiguous results from geochemistry. This is the case

in the Bushveld Complex of South Africa which comprises the World's largest layered mafic intrusion known as the Rustenburg Layered Suite, the Lebowa Granite Suite and the Rashoop Granophyre Suite (SACS, 1980). Moreover, the Rooiberg Group of mafic and felsic volcanics is considered part of the Bushveld Large Igneous Province (Rajesh et al., 2013).

Traditional views hold that the 7–8 km of Rustenburg Layered Suite cumulates crystallized from the base upwards in a repeatedly replenished magma chamber and it is generally accepted that large amounts of relatively evolved residual magma must have repeatedly escaped the Rustenburg Layered Suite magma chamber (e.g. Cawthorn and Walraven, 1998). Nuances in high-precision U–Pb ages has challenged the simple view of upsection

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younging of the cumulates of the lower part of the Rustenburg Layered Suite (Mungall et al., 2016; Robb and Mungall, 2020). However, the upper ~2.5 km of the Rustenburg Layered Suite (composed of the Upper Main Zone and Upper Zone) is thought to represent a single batch of magma that evolved by fractional crystallization as a closed system (Cawthorn et al., 1991; Kruger et al., 1987; Tegner et al., 2006). Mass balance considerations suggest that about 20% of evolved liquid is missing from the documented cumulate stratigraphy (Cawthorn and McCarthy, 1985; Cawthorn and Walraven, 1998; Tegner et al., 2006; VanTongeren et al., 2010). VanTongeren et al. (2010) attempted to locate the felsic residual magma that escaped from the Upper Main and Upper Zones by performing a MELTS forward model and pointed to either the felsic volcanic rocks of the Rooiberg Group or the Stavoren Granophyre (Rashoop Granophyre Suite) as representing the escaped magma. Cawthorn (2013) argued that the Rustenburg Layered Suite is intrusive into the Rooiberg Group. Instead, Cawthorn (2013) described a newly discovered zone of monzonite at the top of the Rustenburg Layered Suite, referred to as the Roof Zone and argued that it represents the residual liquid in the Rustenburg Layered Suite magma chamber.

Mathez et al. (2013) and VanTongeren and Mathez (2015) point to the Stavoren Granophyre as the most likely granitoid unit to have crystallized from the residual magma, partly because a gradual transition between the two units has been described at the farm Diepkloof in the eastern Bushveld (Walraven, 1987). These authors explained the Roof Zone as thermally metamorphosed and remelted roof material.

Here we revisit this problem by describing the transition from the Rustenburg Layered Suite to felsic roof rocks at three locations (Fig. 1). One location is the Bierkraal bore hole in the western limb, which intersects ~400 m of Lebowa Granite Suite and Rashoop Granophyre Suite before reaching Upper Zone mafic cumulates. The second location is the Diepkloof farm in the eastern limb, i.e. the location where Walraven (1987) described a gradual transition from Upper Zone cumulates to granophyre. The third location is near Stoffberg in the eastern limb representing the type locality of the Roof Zone described by Cawthorn (2013). We present major and minor element data for bulk rocks and minerals, mineral modes, whole rock Zr concentrations, and bulk rock Sr, Nd, and Hf isotope data. These new data add to an already extensive dataset for the Upper Zone and provide new constraints on the overlying granites and granophyres that permit more rigorous assessment of the connections between the granites, granophyres and the Rustenburg Layered Suite.

2. The Bushveld

2.1. Geology

The Bushveld Complex consists of the Rustenburg Layered Suite, Lebowa Granite Suite and Rashoop Granophyre Suite, intruding siliciclastic sediments of the Transvaal Supergroup and the volcanic rocks of the Rooiberg Group (Eales and Cawthorn, 1996). Palaeomagnetic data (Letts et al., 2009) show that the Rustenburg Layered Suite is a lens-shaped intrusion (Kleemann and Twist, 1989). The Lebowa Granite Suite is ~3 km thick and crops out in a circular shape over >30,000 km² in the center of the complex whereas the Rustenburg Layered Suite is 7–9 km thick and crops out over 65,000 km² in four main limbs surrounding the Lebowa Granite Suite (Fig. 1). The limbs are known as the eastern, northern, western and far western limbs, respectively. The distribution of the Rashoop Granophyre Suite is restricted to isolated occurrences embedded in granite or along the contact between the Lebowa Granite Suite and Rustenburg Layered Suite (Fig. 1). Gravity models (Cawthorn and Webb, 2001) and xenoliths equivalent to the Rustenburg Layered Suite found in kimberlites from the central part of the complex (Webb et al., 2011) indicate that the eastern and western limbs are connected below the granite. Because of post-crystallization subsidence in the central part of the Bushveld Complex, layering everywhere dips towards its center. Thus, the marginal part of the complex is not preserved and the original lateral distribution of the Rustenburg Layered Suite and Lebowa Granite Suite probably exceeded the present exposure.

The zonal subdivision of the Rustenburg Layered Suite is based on the appearance of cumulus minerals (e.g. Eales and Cawthorn, 1996). In contrast, the subdivision of the Lebowa Granite Suite and Rashoop Granophyre Suite is based on textural variations (SACS, 1980). The main part of the Lebowa Granite Suite is composed of the Nebo Granite, which is a coarse-grained hornblende or biotite granite (Hill et al., 1996; Kleemann and Twist, 1989) of ferroan composition (Frost and Frost, 2011). The remaining granite units are late-stage differentiates of the Nebo Granite or porphyritic varieties (SACS, 1980). The Rashoop Granophyre Suite is composed of three units. The Stavoren Granophyre Suite is the volumetrically dominant unit and its composition is also ferroan (Mathez et al., 2013). It consists of quartz and two feldspars as well as hornblende, biotite and minor phases (Walraven, 1985). The Rooikop Granophyre Porphyry and Zwartbank Pseudogranophyre are distinguished from the Stavoren Granophyre based on textural differences (see below).

Fig. 1. Sketch map of the Bushveld Complex (modified after Tegner et al., 2006, and Walraven, 1987).

2.2. Granophyre terminology

The Rashoop Granophyre Suite is a collective name ascribed to all granophyric rocks in the Bushveld Complex by the South African Committee of Stratigraphy (SACS, 1980) that recognized three granophyre units: Stavoren Granophyre, Rooikop Granophyre Porphyry and Zwartbank Pseudogranophyre. The distinction between these units is based strictly on petrography. The Stavoren Granophyre is dominated by relatively coarse-grained intergrown quartz and alkali feldspar, whereas the Zwartbank Pseudogranophyre consists of irregular quartz-feldspar intergrowths (SACS, 1980). The Rooikop Granophyre Porphyry differs by being porphyritic.

In addition to the units recognized by SACS (1980), Walraven (1985, 1987) defined the Diepkloof Granophyre. The Diepkloof Granophyre is texturally similar to the Stavoren Granophyre but is distinguished by the gradational contact between the Diepkloof Granophyre and the Rustenburg Layered Suite Upper Zone. Thus, the Diepkloof Granophyre sensu Walraven (1987) can be classified as Stavoren Granophyre sensu SACS (1980).

Cawthorn (2013) described a traverse across the uppermost c. 230 m of the Rustenburg Layered Suite in the Stoffberg area of the eastern limb as monzonitic rocks, and defined them as the Roof Zone. Walraven (1987, p. 35) mapped the same area as Diepkloof Granophyre and referred to it as a granodiorite variety (Walraven, 1987, p. 39; Fig. 2). Given that granophyric textures are poorly developed in the Stoffberg section we regard the Roof Zone of Cawthorn (2013) and Diepkloof Granophyre of Walraven (1987) as equivalent units.

3. Sampling and previous work

3.1. Bierkraal core

Three composite cores (BK1–3) drilled in the western Bushveld are collectively known as the Bierkraal cores. Together, the three cores intersect ~400 m of granitoids at the top, followed downwards by ~2.1 km of Upper Main Zone and Upper Zone cumulates and the top of the Lower Main Zone (Tegner et al., 2006; Walraven, n.d.). The Upper Zone and Upper Main Zone consist of layered cumulates with

varying proportions of cumulus plagioclase, olivine, clinopyroxene, orthopyroxene (and inverted pigeonite), magnetite, ilmenite, sulphides and apatite and intercumulus biotite, hornblende, quartz and potassium feldspar (Tegner et al., 2006). Concentrations of incompatible trace elements increase upwards in the stratigraphy (Cawthorn and McCarthy, 1985) and bulk rock Sr isotopes define an isochron with $(^{87}\text{Sr}/^{86}\text{Sr})_i = 0.7073 \pm 0.0001$ (Kruger et al., 1987), consistent with closed system fractionation of a single magma reservoir. However, mineral compositions record multiple reversals from a simple fractionation trend (Tegner et al., 2006; Yuan et al., 2017).

Walraven (1976) described the upper ~400 m of granitoids in BK1 as interchanging rock sequences of Nebo Granite and Zwartbank Pseudogranophyre with minor Stavoren Granophyre (Fig. 3a). The exact position of the contact between the Upper Zone and Nebo Granite lead to some confusion in the literature. One reason for this is that a number of quartzite xenoliths interrupt the magmatic stratigraphy near the contact (at 420.1–420.07 m, 419.9–418.4 m, 417.2–416.4 m, and 411.9–403.53 m depth, Fig. 3b). Tegner et al. (2006) placed the contact at 415 m (thus between the two upper xenoliths) and calculated the stratigraphic depth relative to 415 m assuming a 24° dip. Yuan et al. (2017) placed the upper contact at 411.8 m (i.e. at the base of the uppermost xenolith) and Kruger et al. (1987) placed it at 380 m. For consistency, we use the inferred stratigraphic heights of Tegner et al. (2006). Thus, samples above 415 m have negative heights (Fig. 3).

Our samples represent every ~15 m in the granitoids, although the sampling was more closely spaced between 358 and 420 m (see Table 1).

3.2. Diepkloof farm

On the Diepkloof farm (Fig. 2), Walraven (1987) described the contact between the Upper Zone and the overlying granophyre as gradational. From base to top, the transition begins within the Upper Zone with the occurrence of granophyric intergrowth in interstitial pockets between cumulus phases. Up through the granophyre, cumulus plagioclase and pyroxene give way to hornblende and abundant quartz and alkali feldspar in intergrowth (Walraven, 1987).

We also sampled the transition from Upper Zone to granophyre at the Diepkloof farm. Sampling was mainly done along the Kruis River

Fig. 2. Sketch map of an area in the Eastern Bushveld. The Stoffberg section (Traverse 1 of Cawthorn, 2013) is indicated. Simplified from Walraven (1987, p. 34–35) and Cawthorn (2013).

(Fig. 4a), which cuts the stratigraphy at a steep angle. The traverse is ~2 km long and covers ~50 m of altitude. Calculating the stratigraphic height poses a problem because the rocks are massive and their strike and dip directions could not be determined in the field. According to Molyneux (2008), the stratigraphy of the area strikes roughly east-west and dips to the south. One reported measurement from within the granophyre close to the Diepkloof traverse dips 40° to the south (Molyneux, 2008). In Table 1, we report stratigraphic heights referenced to sample 16SA-25 (Upper Zone) assuming that the stratigraphy strikes east-west and dips 40° to the south. Correcting for apparent thickness yields a stratigraphic thickness of c. 1 km. We also collected two samples from the overlying roof rocks (16SA-26 and 16SA-27; Fig. 2) located ≥2.5 km above sample 16SA-25 (Table 1). These rough estimates of stratigraphic thickness place sample 16SA-15 (a granophyre sampled off and slightly north of Kruis River traverse) c. 25 m below 16SA-25 (Upper Zone). This inconsistency reflects uncertainties in the strike and dip of the units.

Along the river bed, which was dry during our fieldwork, exposures are good and samples are generally fresh. The lowermost sample is a somewhat weathered Upper Zone ferrodiorite containing cumulus plagioclase and interstitial magnetite and clinopyroxene as well as inclusions of ultramafic (magnetite and clinopyroxenite) lithologies. Upsection, quartz and alkali feldspar become increasingly abundant and clinopyroxene, olivine, apatite and oxide gradually decrease in abundance. All samples except 16SA-25 (Upper Zone) contain granophyric intergrowth in varying proportions and the Upper Zone-granophyre boundary may be located a few hundred meters farther north than indicated in Fig. 4a. Light and dark xenoliths ranging from centimeters to meters are common in the granitoids (Fig. 4b). Some of these are cross-cut by granophyre, while others have mafic selvages suggesting reaction with the granophyric host. The light xenoliths are quartz + feldspar-rich lithologies and the dark xenoliths are rich in pyroxene and oxides.

Samples from the local roof (16SA-26 and 16SA-27) taken from a road cut west of the farm are meta-volcanics of the Rooiberg Group (Fig. 2). 16SA-26 is coarse-grained, whereas 16SA-27 is fine-grained. Both are rich in quartz and feldspar. Molyneux (2008) mapped 16SA-26 as Rooiberg Felsite and 16SA-27 as leptyte.

3.3. Stoffberg section

South of Stoffberg (Fig. 2), Cawthorn (2013) described the transition from Upper Zone ferrodiorite to felsic roof rocks across ~230 m. Cawthorn (2013) described the evolved rocks as hornblende quartz monzonites and modelled their major element compositions as cumulates, which contain increasingly higher proportions of evolved, interstitial liquid up through the stratigraphy. Near the top, mass balance modelling suggests that 80 wt% of the rocks represents residual liquid. Our sample set is identical to that of Cawthorn (2013) and includes Roof Zone samples from near Stoffberg and one sample from the Enkeldoorn farm (Fig. 2).

4. Methods

4.1. Major and trace element geochemistry

We obtained major and trace element data from two laboratories: Central Analytical Facility, University of Stellenbosch and the Bureau Veritas Commodities Canada, Ltd. In both laboratories, major elements were obtained by X-Ray fluorescence (XRF) methods.

17 g of powdered rock were sent the Bureau Veritas Commodities Canada, Ltd. Major elements were analyzed by X-ray fluorescence (XRF) methods on fused powder. The remaining powders were dissolved in aqua regia and analyzed by ICP-MS and ICP-ES methods or, for refractory elements, by lithium fusion ICP-MS. Reproducibility of replicate analyses and deviation from certified values of the secondary standards are usually better than 15% (with few exceptions of elements having concentrations only slightly higher than the reported limit of detection), and mostly better than 5% relative.

At the Central Analytical Facility, University of Stellenbosch, major oxides were obtained on a Phillip's XRF PW1404w on LaO-free glass beads using a Rh tube at 2.4 kW. Trace elements were obtained on the same glass beads using the method of Eggins (2003), on an Agilent 7500ce ICP-MS coupled with a Nd-YAG 223 nm New Wave laser ablation system, operating at a frequency of 12 Hz with a He–Ar carrier gas. The averaged results of three analyses (each comprising a 30 s blank followed by sample collection for 60 s) on each bead were

Fig. 3. a) Detailed log of the upper 450 m of the Bierkraal BK1 core. Sample positions are indicated by x. Stratigraphic height is relative to 415 m depth (Tegner et al., 2006). b) Details for the interval from 350 to 450 m. Partly based on Walraven (n.d.).

Table 1

New samples of this study.

Sample	Suite*	Unit†	Height (m)
Bierkraal drill cores			
1W15‡	RGS	ZP	–365.4
1W30‡	LGS	NG	–351.7
1W44.4‡	RGS	ZP	–338.6
1W60.1	LGS	NG	–324.2
1W74.9‡	LGS	NG	–310.7
1W89.9‡	RGS	ZP	–297.0
1W105.4	LGS	NG	–282.8
1W115‡	RGS	ZP	–274.1
1W130.4‡	LGS	NG	–260.0
1W146	RGS	ZP	–245.7
1W160.7‡	LGS	NG	–232.3
1W173.8	LGS	NG	–220.3
1W191.5	RGS	ZP	–204.2
1W209.3	LGS	NG	–187.9
1W215	RGS	ZP	–182.7
1W231.7	RGS	ZP	–167.5
1W246	LGS	NG	–154.4
1W259.7‡	RGS	ZP	–141.9
1W272.8	RGS	ZP	–129.9
1W285.8	RGS	ZP	–118.0
1W310‡	RGS	SG	–95.9
1W337	RGS	ZP	–71.3
1W347.2	RGS	ZP	–61.9
1W356.5	RGS	ZP	–53.4
BK1–359	LGS	tz§	–51.2
1W364.9	LGS	tz§	–45.8
1W377.3	LGS	tz§	–34.4
1W387.9‡	LGS	tz§	–24.8
1W395.0	LGS	tz§	–18.3
1W398	LGS	tz§	–15.5
1W400.3	LGS	tz§	–13.4
1W402.5	LGS	tz§	–11.4
1W411	–	x	–3.7
1W412.6	LGS	tz	–2.2
1W413.3	LGS	tz	–1.6
1W414.7	–	x	–0.3
1W417	–	x	1.8
1W418.3	RLS	tz‡	3.0
1W420	RLS	tz‡	4.6
1W422.2	RLS	UZ	6.6
1W423.7	RLS	UZ	7.9
1W431.8‡	RLS	UZ	15.3
1W732	RLS	UZ	289.6
1W908.6	RLS	UZ	450.9
3W742	RLS	UZ	1258.0
3W885	RLS	UZ	1388.6
3W1360	RLS	UZ	1822.5
Diepkloof farm (and vicinity)			
16SA-27	–	r (meta-Rooiberg?)	2616.5
16SA-26	–	r (meta-Rooiberg?)	2541.1
16SA-24	RGS	DG	985.8
16SA-23	RGS	DG	692.6
16SA-22	–	Light x	636.9
16SA-21	–	Dark x	634.4
16SA-20	–	Light x	628.9
16SA-19	RGS	DG	626.9
16SA-17	RGS	DG	418
16SA-18	–	Dark x	416.6
16SA-16	RGS	DG	366.3
16SA-15	RGS	DG	363.7
16SA-8	RGS	DG	270
16SA-9	RGS	DG	223
16SA-10	RGS	DG	222.3
16SA-12	RGS	DG	189
16SA-14	RGS	DG	–25.9
16SA-25	RLS	heterolithic UZ	0

* Suites recognized by SACS (1980): LGS, Lebowa Granite Suite; RGS, Rashaop Granophyre Suite; RLS, Rustenburg Layered Suite.

† The following units are recognized by SACS (1980): ZP, Zwartbank Pseudogranophyre; NG, Nebo Granite; SG, Stavoren Granophyre. In addition, we apply the following units: tz, transition zone; x, xenoliths; r, roof; UZ, Upper Zone; DG, Diepkloof Granophyre.

‡ Also sampled as BK1–[depth in core].

§ Logged as Nebo Granite by Walraven (n.d.).

‡ Logged as ferrodiorite and ascribed to Upper Zone by Walraven (n.d.).

obtained using a 100 µm diameter aperture. After every three samples standard NIST612 bead was analyzed, with NIM-G and BHVO-1 as secondary standards. Data were collected in time resolved mode and were reduced using an Excel spreadsheet using the SiO₂ content measured by XRF as internal standard. Reproducibility of replicate analyses and deviation from certified values of the secondary standards are better than 10%, and mostly better than 5% relative.

4.2. Modal analyses

Modal abundances were estimated by scanning a subset of thin sections with a M4 Tornado micro-XRF and exporting relative intensity maps of major elements and Zr. In the ImageJ software, intervals of color intensity that represent specific mineral phases were identified by comparing the intensity maps to thin sections. The areal proportions of individual minerals were measured in ImageJ and normalized to 100%.

4.3. Mineral chemistry

We obtained major element mineral compositions with a JEOL JXA-8600 superprobe electron microprobe at Aarhus University using either an energy dispersive (EDS) detector, accelerating voltage of 15.0 kV, beam current of 10.0 nA and a beam diameter of 5 µm or a wavelength dispersive (WDS) detector, a beam current of 20.0 nA and a beam diameter of 5 µm. When possible, we performed three analyses per grain in three different grains of plagioclase, olivine, clinopyroxene and hornblende in each sample. In samples with primocryst plagioclase, clinopyroxene or olivine, we targeted cores of primocryst grains. Analyses with compositional anomalies (e.g. unusually low sums or obvious outliers) were rejected. For the anhydrous phases, we use the following stoichiometric criteria to distinguish bad analyses, which are removed from the dataset: feldspar analyses should yield between 4.95 and 5.05 cations per 8 oxygen atoms and the difference between Si and Na (both normalized to 8 oxygen atoms) should be between 1.95 and 2.05; pyroxene between 3.95 and 4.05 cations per 6 oxygen atoms; and olivine between 2.95 and 3.05 cations per 4 oxygen atoms. Between analyses, two plagioclase, an olivine, a pyroxene and a hornblende in-house standard were analyzed and compared to reported concentrations. Data are reported as sample averages.

4.4. Bulk rock Sr, Nd and Hf measurements

Approximately 10 mg bulk rock powders were dissolved in HF and HNO₃ in proportions 10:1 in capped beakers for 24 h, and subsequently in H₃BO₃ in capped beakers overnight. Two samples had a very thin film-like surface and were further treated with hydrogen peroxide. After dissolution, the samples were split in two portions to be analyzed for concentration ratios (Lu/Hf, Sm/Nd, Rb/Sr) and isotopic ratios.

The concentration ratios were measured on an Agilent 7900 Quad ICP-MS at Aarhus Geochemistry and Isotope Research Platform, Aarhus University, and normalized to bulk rock standards. The concentration ratios generally agree with those obtained at the Central Analytical Facility, University of Stellenbosch and the Bureau Veritas Commodities Canada, Ltd. but one sample (1W732) yielded a high Lu/Hf ratio of ~1.4 compared to the Lu/Hf ratio of ~0.5 obtained at the Bureau Veritas. The discrepancy in that sample probably reflects residual zircon remained after dissolution.

The remaining split was taken through Fe removal, Hf separation, cation removal, and Sr and Nd separation columns. Hf, Nd and Sr isotopes were measured on a Nu Plasma II MC-ICP-MS at Aarhus Geochemistry and Isotope Research Platform, Aarhus University. Hf measurements were normalized to ¹⁷⁶Hf/¹⁷⁷Hf = 0.282170, Sr measurements to ⁸⁷Sr/⁸⁶Sr = 0.71025 in the standard NBS 987 and Nd measurements to ¹⁴³Nd/¹⁴⁴Nd = 0.512108 in standard JNdI. No blank correction was made. The standards AGV-1, G-2, AU AMES and BHVO-2 were repeatedly measured during the analytical sessions and yielded expected results.

Fig. 4. a) Sketch map after Walraven (1987, p. 35) showing the positions of samples along the Kruis River on the Diepkloof farm. b) Sample and xenolith positions in the stratigraphy. Note that lithological boundaries are adopted from Walraven (1987, p. 35). In reality, all samples above 0 m are granophyric.

5. Results

5.1. Petrography

5.1.1. Petrography of the Bierkraal core

The magmatic rocks of the Bierkraal core are diverse and vary between cumulates of the Rustenburg Layered Suite Upper Zone, Nebo Granite and granophyre rocks. Rocks from the core are generally fresh. In the following, we describe the uppermost 358 m of granitic rocks, the Upper Zone cumulates and, finally, the transition zone between these endmembers.

5.1.1.1. Granitic rocks (0–358 m depth). Figure 3 presents a detailed log of the Bierkraal BK1 core. According to the original core log by Walraven (n.d.) the uppermost 358 m is mainly composed of alternating Nebo Granite and Zwartbank Pseudogranophyre with minor Stavoren Granophyre. Pegmatite is developed at some levels and centimeter sized quartzite xenoliths are scattered throughout the granitic interval (see positions in Fig. 3a). The Nebo Granite consists mainly of subhedral to euhedral alkali feldspar and equidimensional quartz (Fig. 5b) whereas plagioclase is less abundant. Hornblende is the main mafic mineral and minor biotite, olivine, clinopyroxene, zircon, titanite, apatite and iron-titanium oxides are also present. In places, hornblende mantles and replaces clinopyroxene. The mineralogy of the rocks that were assigned by Walraven (n.d.) to Stavoren Granophyre and Zwartbank Pseudogranophyre are identical to that of the Nebo Granite, but their textures are different, and both classify as granite or alkali feldspar granite (Fig. 6a). Moreover, the contacts between the different units are gradational. However, the modal content of mafic phases is generally lower in the granophyre and pseudogranophyre (~5%) than in the Nebo Granite (~10%) (Fig. 6b). Texturally, the Stavoren Granophyre differs from the Nebo Granite by having abundant granophyric intergrowths that radiate from the center of an alkali feldspar grain (Fig. 5c). Intergrowth in the granophyre occurs on a scale of less than 1 mm. In the Zwartbank Pseudogranophyre, the intergrowth does not radiate from a central point, but resembles small (~1 mm) anhedral quartz inclusions in alkali feldspar that are optically continuous with neighboring quartz (Fig. 5a). Quartz may show intergrowth with multiple alkali feldspar grains and intergrown quartz can have grain sizes of >10 mm.

5.1.1.2. Ferrodiortite at the top of the Rustenburg Layered Suite (>420 m depth). Below 420 m, the core intersects Rustenburg Layered Suite Upper Zone diorites. The main cumulus phases are plagioclase, clinopyroxene, olivine, apatite, and magnetite, whereas biotite, quartz, alkali feldspar and hornblende are intercumulus (Fig. 5e). Zircon is a minor interstitial phase. Plagioclase is typically euhedral and lath-shaped. Greenish brown hornblende nucleates on and mantles clinopyroxene in a texture similar to the clinopyroxene-hornblende relations in the granite. In places, hornblende is oikocrystic in a poikilitic texture with any cumulus phase.

5.1.1.3. The transition zone (358–420 m depth). Four quartzite xenoliths of different thicknesses interrupt the magmatic stratigraphy between 420 m and 403 m depth (Fig. 3b) and multiple centimeter sized quartzite xenoliths are scattered through the interval. Walraven (1987) concluded that the xenoliths originated from the Transvaal Supergroup roof. In the following, we describe the stratigraphic modal and textural evolution of the magmatic rocks between ~358 and ~420 m and refer to this interval as the transition zone. The magmatic rocks are intermediate between the overlying Nebo Granite and underlying Upper Zone cumulates in terms of their modal composition and texture. The lower and upper boundaries of the transition zone are defined as the initiation and cessation of the transition from Upper Zone cumulates to Nebo Granite and pseudogranophyre. This transition is gradual and thus not marked by lithological contacts or first arrival of a new primocryst phase. Thus, the positions of the base and top of the transition zone are approximate.

The base of the transition zone is similar to the Upper Zone in terms of textures and modal mineralogy. Starting at the base of the transition zone (~420 m), the modal abundance of plagioclase and mafic phases gradually decrease and those of quartz increases, whereas alkali feldspar remains an interstitial phase with low modal abundance (Fig. 7a). This trend continues until ~358 m. Hornblende becomes increasingly dominant upwards as clinopyroxene, olivine, magnetite, and apatite abundances decrease. The grain size of the mafic phases notably decreases up through the transition zone with hornblende always being largest. Zircon first appears in quantifiable modal abundances (~0.2%) at 402 m (just above the uppermost quartzite, Fig. 3b). Between 402.5 m and 400.3 m (i.e., ~1 m

Fig. 5. Thin section photographs. **a)** Pseudogranophyre from BK1 (sample BK1–15.0). **b)** Nebo Granite from BK1 (sample 1W209.3). **c)** Stavoren Granophyre in BK1 (sample 1W310). **d)** Calcic (antecryst?) core in transition zone (sample 1W387.9). **e)** Interstitial alkali feldspar in the top of the Upper Zone (sample 1W423.7). **f)** Granophyric quartz and alkali feldspar in the top of the Stoffberg section (sample RGC21). **g)** Granophyric quartz and alkali feldspar in the base of the Diepkloof section (sample 16SA-10). **h)** Diepkloof sample completely dominated by granophyric intergrowth (sample 16SA-15). **i)** Fresh olivine and clinopyroxene in the top of the Diepkloof section (Sample 16SA-19). **g)** and **i)** in plane polarized light, remaining thin section photographs under crossed polarizers. Abbreviations: Afs, alkali feldspar; Qtz, quartz; Hbl, hornblende; Cpx, clinopyroxene; Pl, plagioclase; Ap, apatite; Ol, olivine.

above the zircon arrival), alkali feldspar increases from 2 to 20 modal % reflecting the change from its occurrence as an interstitial phase to primocryst (Fig. 7a). Above this level, the mode of alkali feldspar increases upwards. In contrast, the modal increase in quartz (from ~5 to ~18%) is gradual up through the transition zone. Plagioclase is subhedral to euhedral in the transition zone. Generally, plagioclase is not zoned but one plagioclase grain in sample 1W387.9 has a lath-shaped core and a rim (Fig. 5d). From base to top, the rocks evolve from quartz diorite through quartz monzodiorite to quartz monzonite (Fig. 6a).

5.1.2. Petrography of the Stoffberg section

Cawthorn (2013) has previously described the rocks from the Stoffberg section (Fig. 2). The rocks are variably altered with sporadic alteration of feldspars, clinopyroxene and olivine, whereas hornblende, apatite and oxides are fresh. Figure 7b presents Cawthorn's (2013) modal data from the Stoffberg section. Ten to fifteen percent quartz is present before alkali feldspar appears in the stratigraphy. The upsection increase in modal alkali feldspar is gradual. Hornblende mantles and replaces clinopyroxene in a magmatic resorption texture (Cawthorn, 2013) and is sometimes oikocrystic with chadacrysts of all other phases.

Fig. 6. a) Classification of Upper Zone and transition zone samples (modal data from this study and Yuan et al., 2017), Nebo Granite and granophyre/pseudogranophyre from the Bierkraal drill cores and Roof Zone at the Diepkloof farm (modal data from this study). The Roof Zone at Stoffberg (data from Cawthorn, 2013) and Diepkloof Granophyre (data from Lombaard, 1949, cited by Walraven, 1987) are also shown. **b)** Mafic content for the Upper Zone and transition zone, Nebo Granite and Granophyre/pseudogranophyre from the Bierkraal drill cores.

Granophyric intergrowth occurs in most samples but is limited to a few grains per sample (Fig. 5f) and tends to increase with stratigraphy. In summary, from base to top, the rock types change from quartz diorite to quartz monzonite (Fig. 6a).

5.1.3. Petrography of the Diepkloof section

The lowermost Diepkloof sample (16SA-25) from the top portion of the Upper Zone cumulates contains a variety of inclusions representing different lithologies. The major part is ferrodiorite dominated by cumulus plagioclase and interstitial magnetite, with smaller amounts of apatite, rutile, interstitial clinopyroxene and olivine that may be partially or completely altered. This lithology correlates with Upper Zone of the Rustenburg Layered Suite. The sample is also rich in millimeter- to centimeter-sized inclusions of different lithologies, e.g. pyroxenite and anorthosite.

Between 0 and ~300 m, alkali feldspar is part of the assemblage and the rocks are monzonites. Granophyric intergrowth is common (Fig. 5g), and quartz, alkali feldspar and zircon increase upsection (Fig. 7c). Clinopyroxene, plagioclase, apatite and oxide are still relatively abundant. The main mafic phase is hornblende, which nucleates on clinopyroxene. Alteration of feldspars is common (Fig. 5g), but fresh

grains occur in some places. Above ~300 m, the modal abundance of intergrown quartz and alkali feldspar increases (Fig. 5h; Fig. 7c) and the rocks are granophyric granites (Fig. 6a). Olivine and clinopyroxene are present throughout the stratigraphy (Fig. 5i). Walraven (1987) considered the modal data of Lombaard (1949) from the farm Drievoet (just north of Fig. 4) also representative of the Diepkloof section (Fig. 6a). The xenoliths vary between almost pure quartzite (sample 16SA-21) to quartz and alkali feldspar rich granular rocks containing fresh pyroxene and amphibole.

5.2. Bulk rock geochemistry

Figure 8 illustrates the variations of selected major elements and Zr with increasing SiO_2 for the three study locations. The Stoffberg samples range from ~50 to ~65 wt% SiO_2 . The monzonitic to granitic Diepkloof samples range from ~60 to ~78 wt% SiO_2 and show a small compositional gap between 64.9 and 69.3 wt% SiO_2 . Upper Zone sample 16SA-25 has considerably lower SiO_2 (34.50 wt%). This is explained by a relatively high proportion of magnetite and ultramafic inclusions; this sample is therefore omitted from Figure 8. The Bierkraal samples range from ~45 (the uppermost Upper Zone at 431.8 m) to ~76 wt% SiO_2 . The Nebo Granite cluster between ~70 and 75 wt% SiO_2 and the Zwartbank Pseudogranophyre between ~74 and 76 wt% SiO_2 , whereas one Stavoren Granophyre sample has 74.77 wt% SiO_2 . The transition zone covers the compositional range between the Upper Zone and Nebo Granite (~50–68 wt% SiO_2).

In the Stoffberg and Diepkloof samples, K_2O (Fig. 8d) increases linearly from 1.5 wt% at ~50 wt% SiO_2 to 5.5 wt% at ~78 wt% SiO_2 , and is correlated with the gradual increase in the mode of alkali feldspar (Fig. 7b). In contrast, K_2O in the Bierkraal samples is constant at ~1 wt% below ~57 wt% SiO_2 , above which it abruptly increases to ca. 3 wt%. This corresponds to the first arrival of alkali feldspar in the stratigraphy (Fig. 7a). With further increase in SiO_2 , K_2O increases linearly to ~5 wt%.

In the Bierkraal samples, a similar abrupt increase is seen for Zr (Fig. 8f) (from ~200 to ~600 $\mu\text{g/g}$) occurs at ~58 wt% SiO_2 , corresponding to the level where zircon makes its first appearance (Fig. 7a). Zirconium in the Diepkloof and Stoffberg samples increase gradually from ~140 to ~500 $\mu\text{g/g}$.

At all locations, $\text{FeO}_{\text{total}}$ (Fig. 8a) and CaO (Fig. 8c) decrease linearly with SiO_2 . In the Bierkraal core, $\text{FeO}_{\text{total}}$ and CaO decrease from ~24 wt% $\text{FeO}_{\text{total}}$ and 8.6 wt% CaO at ~45 wt% SiO_2 to 2.5 wt% $\text{FeO}_{\text{total}}$ and 0.7 wt% CaO at ~76 wt% SiO_2 . For the Stoffberg samples, $\text{FeO}_{\text{total}}$ decreases from ~17 to ~9 wt% and CaO from ~7 to ~2 wt% between ~50 and ~65 wt% SiO_2 . At Diepkloof, FeO decrease from ~10 to ~1 wt% and CaO from ~4 to ~1 wt% between ~63 and ~78 wt% SiO_2 , although one outlier (16SA-17) at 59.50 wt% SiO_2 has lower $\text{FeO}_{\text{total}}$ and CaO than the linear trend suggests. Thus, all locations follow parallel and overlapping trends.

At all locations, MgO and P_2O_5 decrease with increasing SiO_2 . However, at a given SiO_2 , MgO (Fig. 8b) and P_2O_5 (Fig. 8e) are higher at Diepkloof and Stoffberg than in the Bierkraal core. At ~57 wt% SiO_2 , MgO in the Bierkraal samples decrease to ~0.1 wt% and remain constant at higher SiO_2 . In the Diepkloof and Stoffberg samples, MgO decreases with SiO_2 but never falls below ~0.2 wt%. In the Bierkraal core, P_2O_5 decrease from ~1 to ~0.1 wt% between ~45 and ~57 wt% SiO_2 . At Stoffberg, P_2O_5 decrease from ~0.7 to ~0.1 wt% between ~65 and ~51 wt% SiO_2 . At Diepkloof, P_2O_5 varies between ~0 and ~0.2 wt%.

In conclusion, the Stoffberg and Diepkloof samples follow similar and overlapping trends. In contrast, the Bierkraal MgO , K_2O , P_2O_5 and Zr trends differ from the Diepkloof and Stoffberg trends.

5.3. Mineral compositions

Figure 9 displays the cryptic variations of plagioclase, clinopyroxene, olivine and amphibole in the Bierkraal cores and at Stoffberg. Figure 9a–

Fig. 7. a) Modal data with depth in the BK1 core. Upper Main Zone and Upper Zone data mainly from Yuan et al. (2017). Stratigraphic column simplified from Fig. 3. b) Modal evolution with stratigraphy at Stoffberg (modified after Cawthorn, 2013). c) Modal evolution with stratigraphic depth at the Diepkloof farm. Stratigraphic column simplified from Fig. 4. Hbl: hornblende; cpx: clinopyroxene; ap: apatite.

d displays the evolution of plagioclase, clinopyroxene, olivine and amphibole throughout the Upper Main Zone and Upper Zone, across the transition zone and into the granite and granophyre. The Upper Main Zone and Upper Zone are characterized by an overall evolution from relatively high An in plagioclase (~72 mol%) and Mg# in mafic phases (~73 mol% in clinopyroxene and ~55 mol% in olivine) to lower values (~40 mol% An in plagioclase, Mg# ~16 mol% in clinopyroxene and ~3 mol% in olivine). However, multiple reversals from the general trend exists (Tegner et al., 2006; Yuan et al., 2017).

Figure 9e-h focuses on the transition zone from Upper Zone cumulates to granite and granophyre. In the uppermost Upper Zone (between 30 and 16 m depth), the published data shows a reversal in Mg#_{cpx} (from 5 to 25 mol%), Fo% (from 0.8 to 6 mol%) and An% (40 to 45 mol%). Within the basal few meters of the transition zone, Mg# in clinopyroxene decreases to ~2 mol%, Fo% to ~1 mol%, Mg# hornblende from 8 to 1 mol% and remain low up through the transition zone. In contrast, plagioclase An% decreases with a steep gradient of ~1 An% per meter in the base of the transition zone (from 42.6 mol% at 3 m to

Fig. 8. Bulk rock major element and Zr variations with SiO_2 . Bierkraal and Diepkloof data from this study; Stoffberg data from Cawthorn (2013). Different trends for Bierkraal and Diepkloof/Stoffberg sketched with arrows.

18.6 at -15.5 m). In the upper part of the transition zone, plagioclase An % continues to decrease with a smaller gradient of ~ 0.1 An% per meter and reaches 16 mol% at the top. One plagioclase core in sample 1W387.9 (Fig. 5d) is significantly more calcic (27 mol% An) than the remaining plagioclase grains in this sample (19 mol% An). In the granites and granophyres above the transition zone, the composition of mafic phases remains close to the iron-rich endmember. Plagioclase An% decrease further to 11 up through the lowermost 200 m of the granitoids.

At Stoffberg, the An content in plagioclase (Fig. 9i) decreases from 42 to 4 mol% across ~ 50 m of stratigraphy and remains low above this level. In the same interval, Mg# in clinopyroxene (Fig. 9j) decrease from 30 to 11 and Fo% in olivine (Fig. 9k) from 9 to 5. Above this level, the

composition of mafic phases stabilizes and never reaches the pure iron-rich endmember composition.

5.4. Sr, Nd and Hf isotopes

The Rb—Sr system in the Lebowa Granite Suite is known to be disturbed (e.g. Fourie and Harris, 2011; Hill et al., 1996; Walraven, 1981, 1987; Walraven et al., 1990), possibly by radiogenic ^{87}Sr loss from Rb sites in alkali feldspar (Walraven, 1981; Walraven et al., 1990). We have therefore screened the Sr isotopic data to remove from further discussion those samples having uncertainties in the calculated initial $^{87}\text{Sr}/^{86}\text{Sr}$ that exceed the age correction (calculated as $(^{87}\text{Rb}/^{86}\text{Sr}) \cdot (1 - e^{-\lambda \cdot \text{age}})$). See detailed description and justification in Supplementary file A.

Fig. 10 presents bulk rock Sr, Nd and Hf isotopic values corrected to 2055 Ma. In the Bierkraal cores, the granite and granophyre bulk rock Nd and Hf isotopes are indistinguishable from the Upper Main Zone and Upper Zone at $\epsilon_{\text{Hf}}^{2055 \text{ Ma}} = \sim 8$ to ~ 6 and $\epsilon_{\text{Nd}}^{2055 \text{ Ma}} = \sim 6$ to ~ 5.5 . In the transition zone, the values are slightly lower but identical within uncertainty to the Upper Main Zone and Upper Zone values.

The entire Upper Main Zone and Upper Zone is homogenous in terms of bulk rock Sr isotopes, yielding an initial value of 0.7073 ± 0.0001 (Cawthorn et al., 1991; Kruger et al., 1987; Tegner et al., 2006). A similar value (0.7074) was found by in-situ measurements of plagioclase from the Upper Zone (Karykowski et al., 2017). Bulk rock Sr isotopes in the transition zone are generally indistinguishable from the Upper Main Zone and Upper Zone, although one sample (BK1–359) yields a lower value (Fig. 10).

In the Diepkloof section, bulk rock Nd (Fig. 10e) and Hf (Fig. 10f) isotopes (corrected to 2055 Ma) are similar to the Upper Main Zone and Upper Zone in the Bierkraal core. In the xenoliths, $\epsilon_{\text{Nd}}^{2055 \text{ Ma}} (-5.6 \pm 0.5$ to $-4.7 \pm 0.4)$ and $\epsilon_{\text{Hf}}^{2055 \text{ Ma}} (-7.2 \pm 0.6$ to $-6.9 \pm 0.6)$ are slightly high relative to the magmatic rocks although only one xenolith is outside of uncertainty of the magmatic rocks. In contrast, the bulk rock Sr isotopes vary between 0.7064 ± 0.0011 and 0.7317 ± 0.0017 with no stratigraphic trend (Fig. 10d). The roof meta-rhyolite is similar to the Upper Main Zone and Upper Zone at Bierkraal.

6. Discussion

6.1. Correlations and subdivisions in the top of the Bushveld Complex

6.1.1. Distinction between granite and granophyre/pseudogranophyre in BK1

Walraven (1976, 1987) argued that the characteristic irregular granophyric intergrowth in the Zwartbank Pseudogranophyre is indicative of replacement of quartz by alkali feldspar and interpreted the Zwartbank Pseudogranophyre as a metasomatized equivalent of the sedimentary country rocks to the Bushveld Complex. In that model, alkali feldspar grew as K-rich fluids infiltrated quartzite xenoliths.

Walraven's (1976) model strongly depends on the textural interpretation of the pseudogranophyre and is contradicted by structural and geochemical observations presented here. Firstly, quartzite xenoliths are present in the core, indicating that complete alteration of quartzite to pseudogranophyre did not occur. Further, the quartzite xenoliths are hosted in both pseudogranophyre and Nebo Granite, and there is no obvious reason why the metasomatic alteration of large (i.e., tens of meters thick) quartzite xenoliths would leave small (centimeters thick), rounded fragments observed here unaffected.

Secondly, we observe no difference in initial Nd and Hf isotopes between the Nebo Granite and pseudogranophyre. If metasomatic alteration of quartzite by fluids originating in the Nebo Granite magma produced the pseudogranophyre, the pseudogranophyre would be expected to yield isotopic ratios intermediate between those two end members. Thus, the isotopic similarity between Nebo Granite and pseudogranophyre would require that the quartzite and Nebo

Fig. 9. Results of microprobe analyses for the Bierkraal cores (a–h) and Stoffberg section (i–l). Data from this study, Tegner et al. (2006) and Yuan et al. (2017). Stratigraphic column simplified from Figure 3.

Granite were initially isotopically identical. The Transvaal sediments are extremely heterogeneous in terms of Nd (bulk rock varying from $\epsilon_{\text{Nd}}^{2055 \text{ Ma}} = -18$ to -4 , Harris et al., 2003; Jahn and Condie, 1995) and Hf (where $\epsilon_{\text{Nd}}^{2055 \text{ Ma}}$ in zircon range from -38 to -24 , Zirakparvar et al., 2014) isotopes. Thus, the required isotopic similarity seems fortuitous.

Thirdly, the contacts between the Nebo Granite, Zwartbank Pseudogranophyre and Stavoren Granophyre are gradual. This does not suggest that the Nebo Granite is intrusive into the Zwartbank Pseudogranophyre.

In light of the discussion above, we propose that the granite, pseudogranophyre and granophyre are cogenetic and all crystallized from a single magma reservoir. The modal and textural differences reflect differences in magma composition, redistribution of residual

liquid and thermal history while the magma body solidified. Granophytic textures commonly develop in ferroan granites under magmatic conditions where degassing or decompression cause undercooling (e.g., Lowenstern et al., 1997; Morgan and London, 2012) causing rapid simultaneous crystallization of quartz and alkali feldspar. Pegmatites within the Nebo Granite and pseudogranophyre (Fig. 3a) show that volatiles did exsolve and accumulated locally, possibly leading to volatile depletion and slight undercooling elsewhere in the cooling magma body.

Regardless of whether the repetitive development of granite and granophyre in the Bierkraal core (Fig. 2a) represents horizontal layering or a complex three-dimensional structure, the textural variations are consistent with local variations in volatile concentrations. In addition,

Fig. 10. Stratigraphic variation in initial Sr, Nd and Hf isotopes in (a–c) the Bierkraal cores (stratigraphic height after Tegner et al., 2006) and in the (d–i) Diepkloof section (heights calculated assuming that stratigraphy strikes east-west and dips 40° to the south). Bulk rock data from this study and Kruger et al. (1987). Stratigraphic columns simplified from Figs. 3 and 4.

the lack of visible contacts between the different lithologies, the similar mineral compositions between neighboring granite, granophyre and pseudogranophyre samples, the similar elemental and isotopic compositions and the absence of xenocrystic zircon inheritance in the Zwartbank Pseudogranophyre (Skursch, 2019) are consistent with crystallization of the Nebo Granite, Zwartbank Pseudogranophyre and Stavoren Granophyre from the same body of granite magma.

6.1.2. Correlation of rock units in the Diepkloof and Stoffberg areas

In the eastern limb the uppermost rocks of the Rustenburg Layered Suite and their transition to the overlying roof rocks have also been debated. Walraven (1987, p. 35) mapped the Diepkloof Granophyre between the uppermost Upper Zone and the Rooiberg Group roof. As seen in Figure 2, the greater part of the Diepkloof Granophyre crops out in an area striking east to west. About 10 km farther to the east,

Cawthorn (2013) studied the stratigraphically equivalent rocks and described them as monzonites that were interpreted as the most evolved rocks of the Rustenburg Layered Suite. Cawthorn (2013) referred to these as the Roof Zone. Alternatively, VanTongeren and Mathez (2015) interpreted the rocks in the Stoffberg area as thermally metamorphosed and remelted roof material. In terms of textures and geochemistry the Stoffberg Roof Zone and the Diepkloof Granophyre share many similarities. They both follow a gradual modal evolution from quartz diorite to granite, although the Diepkloof Granophyre extends to almost plagioclase free granite (Fig. 6a). Further, both show an increasing tendency for granophyric intergrowth up through the stratigraphy (Fig. 5f–i). Finally, the major element compositions follow overlapping trends (Fig. 8), although the Diepkloof rocks (60–78 wt% SiO₂) extends to more evolved compositions than the Roof Zone at Stoffberg (50–65 wt% SiO₂). Such trends are not consistent with an origin of partial melting. These observations indicate that the top of the Roof Zone at Stoffberg is equivalent with the base of the Diepkloof Granophyre (Fig. 4), as originally proposed by Walraven (1987). In the following we therefore refer to these rock units as the Roof Zone (Cawthorn, 2013), and discuss their formation below.

6.2. Two expressions of the uppermost Rustenburg Layered Suite

6.2.1. Formation and distribution of the transition zone

6.2.1.1. Formation of the transition zone. A number of gradual changes occur in the transition zone between 358 and 420 m in BK1. The modal content of quartz increases upwards, alkali feldspar and zircon make their first appearances as primocryst phases, and the modal content and grain size of all mafic phases decrease (Fig. 7a). In the same interval, the mafic phases (olivine, clinopyroxene and hornblende) reach their Fe-rich endmember compositions, whereas plagioclase compositions change with a steep gradient (plagioclase An% change by 1 unit per meter in the lowermost 20 m of the transition zone, Fig. 9). The gradual modal and geochemical changes begin at ~420 m, i.e. ~20 m below the first arrival of alkali feldspar primocrysts (Fig. 9e). This observation is not consistent with the suggestion by Walraven (1976) that the quartz monzodiorite and quartz monzonite proportion of the transition zone (358 to 400 m in the core) represents a Nebo Granite dyke intruding into the contact between the already crystallized Upper Zone and its roof. Instead, the stepwise appearance of zircon and then alkali feldspar in the primocryst assemblage, the continuous change in mineral composition towards more evolved compositions and the continued crystallization of olivine and clinopyroxene up through the granite and granophyre (Fig. 7a) are, to a first order, in agreement with fractional crystallization trends. However, a mass balance consideration would rule out that the 3 km thick Nebo Granite represents residual magma produced by fractional crystallization of the 2.1 km thick Upper Main Zone and Upper Zone magma.

Additionally, the geochemical gradients across the transition zone (e.g. the An% in plagioclase change by c. 0.01 unit per meter in the Upper Main Zone and Upper Zone, 1 unit per meter in the lower part of the transition zone and 0.1 unit per meter in the upper part of the transition zone) are difficult to reconcile with simple fractional crystallization and such radical changes in geochemical gradients are not observed in natural magmatic systems that appears to have undergone closed system fractional crystallization. The Skaergaard Intrusion of East Greenland exemplifies a system that underwent uninterrupted fractional crystallization. Here, plagioclase continuously change composition from An₇₁ in the Hidden Zone to An₃₀ in the Upper Zone C (e.g. Salmonsén and Tegner, 2013; Thy et al., 2009) but does not show intermediate intervals with elevated geochemical gradients. This point is illustrated in Figure 11, where we present the result of fractional crystallization modelling in the Upper Zone using the Rhyolite-MELTS software

(Gualda et al., 2012). Tegner et al. (2006) and VanTongeren et al. (2010) have previously presented fractional crystallization models for the Upper Main Zone and Upper Zone. As starting composition, we use Tegner et al.'s (2006) modelled magma composition at 70% crystallization ($F = 0.3$, where F is the fraction of magma remaining), which most closely match the top of the Upper Zone in terms of plagioclase composition. The model runs from 1075 °C (liquidus temperature at $F = 0.3$) and is terminated at $F = 0.12$ (823 °C, Fig. 11a) where crystallization of alkali feldspar is predicted (Fig. 11e) and the modelled magma is rhyolitic. Rhyolite-MELTS predicts that plagioclase and magnetite crystallize immediately and are joined by clinopyroxene at $F = \sim 0.25$ (1015 °C) and apatite at $F = \sim 0.2$ (963 °C). Thus, the predicted phase assemblage is similar to the cumulates in the Upper Zone (Fig. 11c). The predicted anorthite content of plagioclase decreases linearly from 51 mol% at $F = 0.29$ to 19 mol% at $F = 0.12$ (823 °C). Thus, the steep gradient observed across the transition zone (An₄₅ to An₁₅) is not predicted in the fractional crystallization model. Following this discussion, we consider other possible origins for the transition zone.

Given the limited thickness of the transition zone (~60 m), the pressure can be assumed constant during its crystallization. Therefore, the decrease in anorthite content must reflect a large gradient in composition and potentially also temperature. Large compositional gradients can develop at the interface between compositionally contrasting magmas where diffusional processes and/or bulk mixing occur. During bulk mixing, the concentration of major and trace elements varies linearly between the two endmembers. Isotopic ratios vary either as hyperbolae or, if the daughter element ratios are the same in the two endmembers, linearly. By contrast, diffusional exchange leads to transient compositional states that can be very different from the bulk mixing scenarios because diffusion rates vary between different elements (e.g. Lesher, 1990; Watson and Baker, 1991). However, the transition zone consists of cumulate rocks whose major element compositions do not represent magma compositions. Thus, compositional variations cannot distinguish between these two end-member processes. Further, the initial Sr, Nd, and Hf isotopes do not vary across the transition zone and thus cannot distinguish between diffusional and bulk mixing.

Even though the geochemical variation across the transition zone cannot distinguish between diffusional and mechanical mixing, theoretical considerations suggest that static diffusion is known to be ineffective for homogenizing silicic magmas (Baker, 1990). Where rhyolite and dacite magma bodies are in contact, compositional boundary layers of ≤1 m thickness develop (Baker, 1990; Clark et al., 1987). Thus, the ~60 m thick transition zone probably does not represent a static zone of mixing. However, double diffusive convection might be much more efficient (Clark et al., 1987). We note that numerous reversals in mineral compositions characterize the Upper Zone (Tegner et al., 2006; Yuan et al., 2017). Tegner et al. (2006) interpreted these reversals as zones where double-diffusive convective layers within the Upper Zone magma chamber broke down due to density inversions. Thus, the lower layer, which had evolved through fractional crystallization, mixed with the non-differentiated overlying layer. If the ~60 m thick transition zone represents the uppermost double-diffusive layer in the Upper Zone magma chamber, it would be expected to undergo normal fractional crystallization. A ≤1 m thick compositional boundary layer could develop between the uppermost convection layer and the Nebo Granite magma, similar to the rhyolite-dacite boundaries described above (Baker, 1990; Clark et al., 1987). Evolved components could continuously diffuse through the boundary into the transition zone. In this scenario, the transition zone cumulates would be increasingly contaminated with granitic components up through the stratigraphy.

Alternatively, the transition zone may represent a zone of predominantly bulk mixing between the residual Upper Zone magma and Nebo

Granite magma. Bulk mixing can occur when the magmas both behave as liquids at the same temperature (e.g. Sparks and Marshall, 1986). Temperature estimates for the uppermost Upper Zone (925 °C; Tegner et al., 2006) and Nebo Granite magma (>900 °C; Kleemann and Twist, 1989) are similar. Thus, the temperature estimates are consistent with bulk mixing in the transition zone.

Figure 11 shows the results of a Rhyolite-MELTS model for isenthalpic bulk mixing between Upper Zone residual magma and a Nebo Granite crystal mush. For the granite endmember, we use the median composition of the Bierkraal granites and granophyres. We chose a mushy endmember to keep the enthalpy of the mix low enough to allow crystallization. This approach is equivalent to mixing the two endmembers at their liquidus temperatures and subsequently remove enthalpy by cooling. Compared to the fractional crystallization scenario, the temperature interval is relatively limited (Fig. 11b) and the change in mineralogy and plagioclase composition is to a large extent controlled by the mixing proportions of Upper Zone and granitic magmas. With progressive mixing of granite magma into the dacitic residual magma of the Upper Zone, the plagioclase composition changes from An₄₀ (90% Upper Zone magma) to An₁₄ (100% granitic mush) (Fig. 11d). The models fit well with the observed changes in the transition zone. In addition, they are consistent with the observations that plagioclase crystallize throughout the transition zone and is joined by quartz and alkali feldspar as the granitic endmember is approached (Fig. 11f).

In light of the discussion above, we interpret the transition zone as a zone of mixing between residual Upper Zone magma of dacitic composition and granitic magma or mush. We cannot constrain the extents of diffusional exchange and mechanical stirring across the interface between the Upper Zone magma and a Nebo Granite crystal mush, but do present compelling evidence that the transition zone was the product of interaction between coexisting magmas of contrasting composition.

6.2.1.2. Lateral extent of the transition zone. The Rustenburg Layered Suite underlies the Nebo Granite (Fig. 1) but the actual contact between the two suites is generally poorly exposed (e.g. VanTongeren and Mathez, 2015). Thus, interpretations on the structural relationship between the two suites are largely based on drill core observations (e.g. Walraven, 1976, 1987; Walraven and Hattingh, 1993). Likewise, we have defined the transition zone in the Bierkraal drill core but its lateral extent is unknown. We note, however, that Mac Caskie (1983) described a several hundred meters thick zone of fayalite (Fo₂) granite with plagioclase compositions (An_{18–8}) similar to the top of the transition zone and base of the Nebo Granite in the Bierkraal core. This zone stretches at least 50 km along strike of the contact between the Upper Zone and Nebo Granite in the eastern limb. We speculate that the fayalite granite in the eastern Bushveld may represent the transition zone.

Fig. 11. Comparison of closed system fractional crystallization to an isenthalpic mixing scenario in terms of temperature (a–b), plagioclase composition (c–d) and predicted phase assemblage (e–f). Both systems were modelled with the oxygen fugacity fixed at FMQ and a pressure of 2 kbar using Rhyolite-MELTS v.1.1.0 (Gualda et al., 2012). Fractional crystallization (a, c, e) was modelled by using Tegner et al.'s (2006) proposed magma composition at $F = 0.3$ (where F denotes the fraction of magma remaining) as the starting composition. The reported F is relative to the entire Upper Main Zone and Upper Zone. The model is started at $F = 0.3$ with 1 wt% H₂O added. Isenthalpic mixing (b, d, f) was modelled between a granitic crystal mush and an Upper Zone liquid. For the granitic mush, we use the median composition of the Bierkraal Nebo Granite and Zwartbank Pseudogranophyre with 1 wt% H₂O added. This composition was cooled to 866 °C, where it is 50% crystalline. For the Upper Zone end member, we use the melt composition and temperature of the fractional crystallization model at $F = 0.207$. Here it is in equilibrium with plagioclase of composition An₄₀ and thus resemble the top of the Upper Zone. f denotes the fraction of granitic crystal mush in the mix. Abbreviations: pl: plagioclase; Ksp: alkali feldspar; qz: quartz; mag: magnetite; cpx: clinopyroxene; ap: apatite; opx: orthopyroxene.

6.2.2. Formation of the Roof Zone

Walraven (1985, 1987) originally favored a partial melting origin for the rocks at Stoffberg and Diepkloof that were later ascribed to the Roof Zone (Cawthorn, 2013). However, Cawthorn (2013) showed that the Roof Zone rocks at Stoffberg are coarse-grained monzonites showing cumulate textures. Moreover, it was shown that the major element evolution can be modelled by fractional crystallization with upwards increasing amounts of trapped, evolved interstitial liquid. The continuous geochemical evolution up through the Roof Zone monzonites at Stoffberg is further substantiated by our microprobe results (Fig. 9j–l). Finally, VanTongeren and Mathez (2015) also described monzonite from Stoffberg. They reported on one, fine grained sample and found that it was similar to the meta-felsite of the local roof (Rooiberg Group) in terms of grain size, phase assemblage and mineral compositions. VanTongeren and Mathez (2015) therefore argued that this sample represents thermally metamorphosed and remelted roof material (felsite). However, the monzonite sample of VanTongeren and Mathez (2015) is not representative of our sample set, in so far as our samples are coarse-grained (similar to the Upper Zone), show cumulate textures, and are geochemically significantly more primitive (e.g. 62 to 50 wt% SiO₂, 19 to 12 wt% Fe₂O₃, and 7 to 3 wt% CaO) than the roof felsite (see discussion in Cawthorn, 2013).

The parallel modal and geochemical trends for the Roof Zone rocks at Stoffberg and Diepkloof (Figs. 6a, 8) indicate a similar origin for the two sections as discussed above, whereas it is also evident that the Diepkloof samples extend to more evolved compositions. The multiple xenoliths exposed up through the Roof Zone at Diepkloof showing back-veining and reaction rims (Fig. 4b) suggest contamination of the magma by the xenoliths. The Nd and Hf isotopic compositions of the xenoliths are slightly higher than (although most samples overlap within uncertainty with) the Roof Zone rocks (Fig. 10e–f) but local contamination cannot be resolved in the data set, which is everywhere identical within uncertainty to Upper Main Zone and Upper Zone of the Rustenburg Layered Suite. Thus, the simplest interpretation is that the Roof Zone at Stoffberg and Diepkloof represents residual magma from fractionation of the Upper Zone (as suggested for the Stoffberg Roof Zone by Cawthorn, 2013) with the possibility of an unconstrained amount of assimilation from the partially melted xenoliths.

6.2.3. Magma chamber model

A key observation is that the geochemical evolution of the Roof Zone at Stoffberg and Diepkloof sections contrasts that observed in the transition zone in the Bierkraal core. This is schematically illustrated in Figure 12. As discussed above, the stratigraphic evolution of the transition zone in the Bierkraal core is consistent with crystallization across a steep geochemical gradient between two contrasting magmas. This explains the distinctive first arrivals of new primocryst phases (zircon, then alkali feldspar and quartz, Fig. 7) and a steep decrease in

plagioclase anorthite content across the lower part of the transition zone (Fig. 9e, 12b). This evolution stops at the top of the transition zone, above which the rocks crystallized from a largely homogenous magma (Fig. 12b). In the Roof Zone as described for Stoffberg and Diepkloof, the arrival of alkali feldspar primocrysts is harder to locate because of the gradual modal and textural evolution (Fig. 7), consistent with increasing amounts of interstitial magma in a cumulate rock (Cawthorn, 2013). Moreover, plagioclase evolves to almost pure albite at Stoffberg (Fig. 9, 12c), consistent with extreme differentiation. In contrast to the transition zone, the Diepkloof and Stoffberg sections lack the geochemical characteristics of crystallization in an interface between two contrasting magmas.

In summary, we consider that the uppermost rocks of the Rustenburg Layered Suite have two main expressions and explanations as illustrated in Figure 12. Where the top of the Rustenburg Layered Suite is in contact with the Lebowa Granite Suite, a distinct transition zone develops between co-existing granitic and evolved mafic melts – possibly involving diffusional exchange and mechanical mixing (Fig. 12). This is presumably the dominant scenario for the central portion of the Bushveld Complex. In the eastern limb, the uppermost rocks of the Rustenburg Layered Suite appear mainly to be in contact with the meta-felsite of the Rooiberg Group (Fig. 12). In this setting, a Roof Zone representing extreme differentiation through fractional crystallization developed. Our observations suggest that this scenario is dominant where the Nebo Granite is absent.

7. Conclusions

In the Bushveld Complex, granite and granophyre overlie mafic cumulates over thousands of square kilometers. The top of the mafic cumulates and the transitions into granite and granophyre have been studied at three locations. In the western Bushveld (the Bierkraal drill cores), the transition occurs over a ~60-m-thick zone where alkali feldspar and zircon become primocryst minerals and the amount of mafic minerals and plagioclase decrease. This zone is transitional between the Rustenburg Layered Suite below and Nebo Granite above in terms of mineralogy and mineral compositions, but all these units show identical Sr–Nd–Hf isotope compositions. We therefore interpret this transition zone as having crystallized from a mix between the magmas from which the Upper Zone and granitoids formed. This is also supported by Rhyolite–MELTS modelling results. The absolute age of the Lebowa Granite Suite is relatively poorly constrained (e.g. Walraven and Hattingh, 1993) but roughly similar to the Rustenburg Layered Suite at 2055–2056 Ma (Mungall et al., 2016; Scoates and Wall, 2015; Zeh et al., 2015). Our data from Bierkraal indicate that the granitic magma began to be emplaced before the residual liquid of the Upper Zone had solidified. Interaction between the two liquids occurred producing a hybrid sequence some 60 m thick that indicated a complete continuity

Fig. 12. a) Sketch of the difference between Bierkraal and Stoffberg/Diepkloof. b) At Bierkraal, the Upper Zone magma coexists and is in contact with the granite magma. The two magmas interact through diffusive processes and/or bulk mixing. Steep geochemical gradients characterize the interface. c) At Stoffberg and Diepkloof, there is no granite magma overlying the Upper Main Zone and Upper Zone magma. Thus, fractional crystallization can proceed until evolved compositions are reached.

of crystallization between the two, not a later (however short the time gap) intrusion of granite into or above the completely solidified Upper Zone.

Following the textural classification of the South African Committee of Stratigraphy (SACS, 1980), the granitoids in the Bierkraal core belong to the Nebo Granite, Zwartbank Pseudogranophyre and Stavoren Granophyre units. The distinction between these units is purely textural and we find that there is no reason to invoke different magmas. Instead, we explain the textural variations as a consequence of local accumulations of fluids or residual magma during crystallization.

In the eastern Bushveld, the studied sections of the Roof Zone at Stoffberg and Diepkloof show continuous evolution from Upper Zone mafic cumulates with relatively calcic plagioclase (An_{42}) and magnesium-rich mafic phases ($Mg\#_{-28}$ in clinopyroxene, Fe_9 , $Mg\#_{-18}$ in hornblende) to granophyric granites with relatively albitic plagioclase (An_{-3}) and iron-rich mafic phases ($Mg\#_{-11}$ in clinopyroxene, $Mg\#_{-7}$ in hornblende). Unlike the Bierkraal core, volcanic host rocks top the Stoffberg and Diepkloof sections. Thus, Nebo Granite magma was not available for mixing with the residual Upper Zone magma, which evolved through fractional crystallization, rather than mixing with granite magma. The Nd and Hf isotope compositions at Diepkloof are similar to the Upper Zone, the transition zone, and the Lebowa Granite Suite in the Bierkraal core of the western Bushveld. Following Cawthorn (2013), we interpret these sections as the final products of differentiation of the mafic cumulates. The sections at Stoffberg and Diepkloof are very similar in terms of geochemistry and textures, supporting the conclusion of Walraven (1987) that they represent one geological unit. Hence, the nature of the uppermost Rustenburg Layered Suite varies from a transition zone where it is in contact with granite to a residual Roof Zone where the granite is absent.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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