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## Immiscible iron- and silica-rich liquids in the Upper Zone of the Bushveld Complex



Lennart A. Fischer a,b,\*, Meng Wang a, Bernard Charlier a,c, Olivier Namur a, R. James Roberts d, Ilya V. Veksler e,f, R. Grant Cawthorn g, François Holtz a

- <sup>a</sup> Institut für Mineralogie, Leibniz Universität Hannover, 30167 Hannover, Germany
- b School of Physical Sciences, University of Tasmania, Hobart 7001, Australia
- <sup>c</sup> Department of Geology, University of Liege, 4000 Sart Tilman, Belgium
- <sup>d</sup> Department of Geology, University of Pretoria, Hatfield, Pretoria, 0002, South Africa
- <sup>e</sup> Department of Mineralogy and Petrology, Technical University Berlin, 13355 Berlin, Germany
- f Perm State University, Geological Department, Bukireva 15, 614990 Perm, Russia
- <sup>g</sup> School of Geosciences, University of the Witwatersrand, PO Wits 2050, South Africa

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#### ABSTRACT

The Bushveld Complex (South Africa) is the largest layered intrusion on Earth and plays a considerable role in our understanding of magmatic differentiation and ore-forming processes. In this study, we present new geochemical data for apatite-hosted multiphase inclusions in gabbroic cumulates from the Bushveld Upper Zone. Inclusions re-homogenized at high-temperature (1060–1100 °C) display a range of compositions in each rock sample, from iron-rich (35 wt.% FeO<sub>tot</sub>; 28 wt.% SiO<sub>2</sub>) to silica-rich (5 wt.% FeO<sub>tot</sub>; 65 wt.% SiO<sub>2</sub>). This trend is best explained by an immiscible process and trapping of contrasted melts in apatite crystals during progressive cooling along the binodal of a two-liquid field. The coexistence of both Si-rich and Fe-rich immiscible melts in single apatite grains is used to discuss the ability of immiscible melts to segregate from each other, and the implications for mineral and bulk cumulate compositions. We argue that complete separation of immiscible liquids did not occur, resulting in crystallization of similar phases from both melts but in different proportions. However, partial segregation in a crystal mush and the production of contrasting phase proportions from the Fe-rich melt and the Si-rich melt can be responsible for the cyclic evolution from melanocratic (Fe-Ti-P-rich) to leucocratic (plagioclase-rich) gabbros which is commonly observed in the Upper Zone of the Bushveld Complex where it occurs at a vertical scale of 50 to 200 m.

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#### 1. Introduction

Silicate liquid immiscibility and the unmixing of an iron-rich silicate melt and a silica-rich silicate melt has been identified in tholeiitic and andesitic magmas, both in volcanic settings (e.g. Philpotts, 1982; Charlier et al., 2013) and in plutonic environments (e.g. Jakobsen et al., 2005; Namur et al., 2012; Kamenetsky et al., 2013; Veksler and Charlier, 2015). In the case of the Bushveld Complex, South Africa (Fig. 1), the development of immiscibility has been suggested to occur in the Upper Zone (UZ). Reynolds (1985) and von Gruenewaldt (1993) suggested that the numerous (up to m scale) magnetitite and nelsonite layers in the UZ formed from an immiscible Fe-rich liquid. Scoon and Mitchell (1994) in-

E-mail address: l.fischer@mineralogie.uni-hannover.de (L.A. Fischer).

terpreted the occurrence of Fe-rich pegmatites in the Upper Critical Zone and the Lower Main Zone as having crystallized from an immiscible Fe-Ti-rich silicate melt derived from the UZ. This model was challenged by Cawthorn (2015) who presented several physical and chemical reasons for its implausibility. Based on a detailed study of rare earth element (REE) concentrations in apatite, VanTongeren and Mathez (2012) suggested a large-scale immiscibility process in the UZ, with a complete physical separation of the buoyant Si-rich melt from the dense Fe-rich melt. Data were re-interpreted by Cawthorn (2013a) who suggested that compositional variability in apatite results from re-equilibration with interstitial liquid in the crystal mush. The role of large-scale immiscibility on the differentiation of the Bushveld UZ has been further debated (Cawthorn, 2014; VanTongeren and Mathez, 2014) but consensus has not yet arisen, mainly because of the absence of any direct evidence for the existence of melts in cumulate rocks. This debate highlights the large uncertainty regarding the signifi-

<sup>\*</sup> Corresponding author at: Institut für Mineralogie, Leibniz Universität Hannover, 30167 Hannover, Germany.

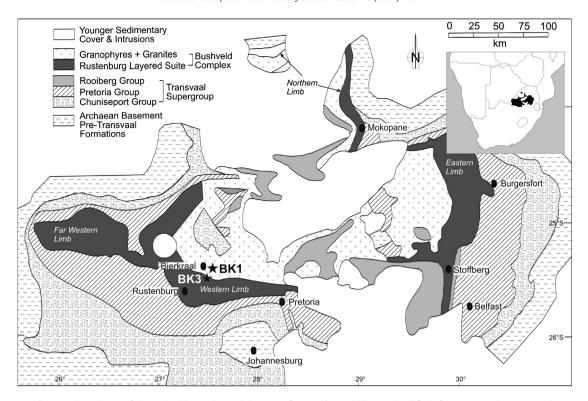


Fig. 1. Geological map of the Bushveld Complex with location of BK1 and BK3 drill-cores (modified after Barnes and Maier, 2002).

cance of liquid immiscibility for magma differentiation in plutonic settings.

In this study, we investigated the petrography of polycrystalline apatite-hosted melt inclusions that we interpret as crystallized melt inclusions. For selected samples, we re-homogenized the inclusions at high temperature and measured their major element compositions. Based on these data, we present the first evidence for the coexistence of immiscible melts during the crystallization of the UZ of the Bushveld Complex. The coexistence of immiscible melts within gabbroic rocks formed by magma at the transition between basaltic and rhyolitic magmatism has implications for the dynamics of magma chambers, the density distribution of silicate melts and cumulate rocks, and the ore-forming processes of Fe–Ti–P-rich layers. This has also implications for the formation of evolved lavas (quartz monzonitic and rhyolitic compositions), which are closely associated to the Bushveld (Mathez et al., 2013; Cawthorn, 2013b).

#### 2. The Upper Zone of the Bushveld Complex

The Bushveld Complex, South Africa (Fig. 1), includes a 7 km thick mafic cumulate sequence emplaced in three limbs. These cumulates are divided into the Marginal, Lower, Critical, Main, Upper and Roof Zones and their corresponding subdivisions (Wager and Brown, 1968). The base of the UZ (subzone UZa) is defined by the appearance of cumulus titanomagnetite, which is followed by the appearance of olivine (UZb) and finally apatite (UZc). The initial emplacement of magma in the Bushveld occurred at 2055.91  $\pm$  0.26 My, and the whole intrusion cooled to below 650 °C in 1.02  $\pm$  0.63 My (Zeh et al., 2015). Cawthorn and Walraven (1998) proposed an even shorter crystallization time of 200 000 yr.

The Upper Zone and Upper Main Zone above the Pyroxenite Marker (hereinafter referred to as UUMZ) are generally considered as having crystallized from a single batch of magma following a large event of magma chamber replenishment (Cawthorn et

al., 1991; Tegner and Cawthorn, 2010; VanTongeren and Mathez, 2013).

The UUMZ interval is famous for the occurrence of numerous layers of magnetitite and nelsonite (Molyneux, 1974; von Gruenewaldt et al., 1985; von Gruenewaldt, 1993), the major World resource for vanadium. Tegner et al. (2006) identified 26 magnetite and 6 nelsonite layers in the UZ in the western limb of the Bushveld Complex. Crystallization of abundant Fe-rich minerals drove residual liquids towards SiO<sub>2</sub>-enrichment (Tegner et al., 2006; Tegner and Cawthorn, 2010). The roof sequence of the Bushveld is poorly defined and the uppermost rocks of the layered intrusion could be either quartz monzonites (Cawthorn, 2013b) or mafic cumulates if evolved residual melts were erupted from the magma chamber (Tegner et al., 2006; VanTongeren et al., 2010).

Although there is a distinct overall differentiation up-section in the UUMZ (Fig. 2), compositional reversals in minerals and significant changes in bulk rock chemistry were described, possibly indicating multiple events of magma replenishment (von Gruenewaldt, 1973; Molyneux, 1974; Ashwal et al., 2005; Scoon and Mitchell, 2012). Tegner et al. (2006) defined nine cyclic units based on whole-rock and mineral compositions (whole-rock  $P_2O_5$  content, anorthite in plagioclase, Mg# in pyroxenes and olivine, and  $V_2O_5$  in magnetite). Six cycles with apatite-bearing rocks (nelsonites, gabbros) occur in the upper half of the UZ (Fig. 2) of the western Bushveld (subzone UZc). In each of these cycles (50 to 200 m-thick),  $P_2O_5$  in the whole-rock first increases from very low concentrations (<0.2 wt%) up to 10 wt.% (23 wt.% apatite) and then decreases continuously upwards to 0.5 wt.% (Fig. 2).

It is commonly assumed that evolved, rhyolitic magma escaped from the Bushveld magma chamber during the late stages of differentiation (Cawthorn and Walraven, 1998; Tegner et al., 2006; VanTongeren et al., 2010). The most recent investigation of the parental magma (VanTongeren et al., 2010) of the UUMZ is based on a careful summation of cumulate compositions to which 15–25 vol.% of rhyolitic magma (average compositions of the Damwal, Kwaggasnek, Schrikkloof and Rashoop Formations) was added. This amount of rhyolite is required to stabilize orthopy-

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