The Creation and Evolution of Crystal Mush in the Upper Zone of the Rustenburg Layered Suite, Bushveld Complex, South Africa

Z. Vukmanovic 1*, M. B. Holness 1, M. J. Stock 1 and R. J. Roberts 2

1Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge CB2 3EQ, UK; 2Department of Geology, University of Pretoria, Hatfield 0002, South Africa

*Corresponding author. Tel: +441223 333474; E-mail: zv211@cam.ac.uk

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ABSTRACT

The Upper Zone of the Rustenburg Layered Suite of the Bushveld Complex contains the world’s largest Fe–Ti–V 6 P deposit and formed from the last major injection of magma into the chamber. Quantitative textural analysis of Upper Zone rocks was undertaken to constrain the processes operating during mush formation and solidification, focussing on horizons with the greatest density contrast to isolate the effects of gravitational loading. We examined three magnetitite layers, together with their underlying and overlying anorthosites. The similarity of microstructures in anorthosites above and below the dense magnetitite layers suggests that the rocks were not affected by viscous compaction driven by gravitational loading. The magnetitite cumulate layers formed by crystal accumulation from a mobile crystal slurry dominated by the Fe-rich conjugate of an unmixed immiscible liquid. We suggest a new mechanism of crystal nucleation in deforming crystal-rich systems, driven by undercooling caused by cavitation as grains slide past each other during simple shear. We propose that the super-solidus deformation recorded in these rocks was caused by prolonged regional subsidence of the magma chamber at Upper Zone times.

Key words: anorthosite; magnetitite; Bushveld Complex; crystal mush; cavitation creep; sub-solidus deformation; microstructure

INTRODUCTION

The fluid dynamical processes that occur in magma chambers are not well understood. Of particular importance is the physical behaviour of the solidifying magma. We still do not have a clear understanding of the mechanisms by which crystal mushes may form, with a variety of possibilities including: in situ nucleation and growth in the thermal boundary layers of cooling magma bodies (Hunter, 1996; Namur et al., 2015); crystal settling from a liquid-rich magma body (Woods & Stock, 2019); the accumulation of material derived from the redistribution of crystals grown elsewhere in the chamber (Holness et al., 2017b); or by settling from intruding batches of crystal-laden magma (Marsh, 1988; Hunter, 1996; Namur et al., 2015). The subsequent behaviour of such crystal mushes is also poorly constrained, in particular the extent to which gravitational loading results in expulsion of interstitial liquid. Such compaction affects the extent of fractionation in the bulk magma and has been invoked to explain the genesis of some magmatic ore deposits (e.g. Mathez et al., 1997; Boudreau & Meurer, 1999; Meurer et al., 1999; Barnes & Maier, 2002;) and the accumulation of large volumes of erupted crystal-poor magma necessary to feed volcanic eruptions (e.g. Bachmann & Bergantz, 2004).

Here we report the results of a microstructural study of cumulates from the Bushveld Complex, South Africa, combining electron backscatter diffraction (EBSD) analysis with mineral chemistry to address the extent to which gravitational loading by dense magnetite-rich layers affected their underlying low-density plagioclase-rich footwall. We demonstrate that these cumulates
preserve evidence of super-solidus deformation by dislocation creep: this cannot be attributed to internally-generated gravitational loading and most likely occurred in response to regional-scale events. We present evidence to support the concept of cavitation-driven nucleation in deforming crystal mushes and argue that cumulate microstructures may preserve evidence of whole-scale slumping of crystal-rich mushy layers.

**GEOLOGICAL BACKGROUND**

The Bushveld Complex

The Bushveld Complex was emplaced at 2.06 Ga into sedimentary host rocks of the Transvaal Supergroup, with its floor at ~12 km depth (Buick et al., 2001; Zeh et al., 2015), though some studies suggest that the floor might have been shallower at ~1–3 km (Wallmach et al., 1985). The Bushveld Complex comprises the mafic rocks of the Rustenburg Layered Suite (forming the largest layered intrusion on Earth, Fig. 1) which are capped by the Rooiberg volcanic suite, the Rashoop granophyres and the c.4000 m thick Lebowa Granite (South African Committee for Stratigraphy & Geological Survey (South Africa), 1980). The mafic magmas of the Bushveld Complex are thought to have been emplaced during regional extension (Carr & Groves, 1994). Extension-related subsidence was amplified by loading by the dense mafic magma, resulting in syn-magmatic deformation that continued into the subsolidus (Carr & Groves, 1994; Holness et al., 2017a).

The Rustenburg Layered Suite is subdivided into five major zones (Fig. 1): the Marginal, Lower, Critical, Main and Upper Zones (Hall, 1932). The Marginal Zone forms the base of the complex and is dominated by heterogeneous noritic rocks (Engelbrecht, 1985; Wilson, 2012, 2015). The Lower Zone (LZ) is mainly composed of cyclic units comprising dunite, orthopyroxenite and harzburgite (Wilson, 2015). The Critical Zone (CZ) has been sub-divided into the pyroxenitic Lower Critical Zone and noritic Upper Critical Zone, both of which include major chromitite layers (Cameron, 1976; Eales & Cawthorn, 1996). Similarly, the chromite-free Main Zone (MZ) is sub-divided into the norite-dominated Lower Main Zone and gabbronorite-dominated Upper Main Zone. The two are locally separated in the Western Limb by a metre-thick pyroxenite layer (the Pyroxenite Marker), and are considered to have crystallized from a hybrid melt during magma chamber replenishment (Von Gruenewaldt, 1973; Cawthorn et al., 1991a; Mitchell, 1996; Van Tongeren & Mathez, 2013). The Upper Zone (UZ) has been divided into three sub-zones based on the primocryst assemblage (Von Gruenewaldt, 1973; Tegner et al., 2006): UZa contains cumulus magnetitite;

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**Fig. 1.** (a) A simplified geological map of the Rustenburg Layered Suite of the Bushveld Complex. (b) Schematic stratigraphy through the Rustenburg Layered Suite (after Pebane & Latypov, 2017), showing the location of the MZ sample SL12-1395.77, together with a sketch of the parts of the BK3 core used in this study. The abbreviation pmi denotes a plagioclase-magnetite-ilmenite cumulate. Depths in the core are shown in metres, with the suffix of the labels on the right of the core log showing the number of the magnetitite layer (numbers increase upwards stratigraphically). The positions of the samples studied by EBSD are shown by the yellow lines. See Supplementary Data Table S2 for the stratigraphic location of all BK3 samples examined for this study.
UZb additionally contains cumulus olivine; and the appearance of apatite defines UZc. UZa and UZb are dominated by magnetite gabbronorite (± olivine), whereas UZc comprises magnetite gabbros, magnetite troctolite and ferrodiorite, together with near-monomineralic magnetite layers and anorthosites. The UZ includes about 30 distinct magnetite and nelsonite layers (Cawthorn & Molyneux, 1986; Von Gruenewaldt, 1993; Lee et al., 1996; Cawthorn et al., 2005). Apatite-hosted melt inclusions, trace element geochemistry and mass balance calculations have been used to argue for a significant role of liquid immiscibility in the differentiation of UZ (VanTongeren & Mathez, 2012; Fischer et al., 2016; Yuan et al., 2017).

Together with the Upper Main Zone, the UZ is thought to have formed from the last major melt injection in the Bushveld magma chamber (Cawthorn et al., 1991b; VanTongeren & Mathez, 2013). The upward mineralogical and chemical evolution in this part of the stratigraphy has been interpreted as a product of closed-system fractional crystallization (Wager & Brown, 1968; Kruger et al., 1987; Tegner et al., 2006; VanTongeren et al., 2010). However, detailed examination of single borehole cores has shown reversals in mineral compositions and density in the UZ, which have been argued to result from replenishment by either primitive melts (Ashwal et al., 2005) or plagioclase-laden magmas (Yuan et al., 2017). Additionally, a regional examination of 5 boreholes showed variable mineralogy and chemistry along strike, which is inconsistent with closed-system fractionation (Scoon & Mitchell, 2012).

The world-class deposits of platinum group elements, vanadium and chromium in the Bushveld Complex have provided the impetus for extensive research over the last century (see Cawthorn (2015) for a review). However, the majority of this research is focussed on geochemistry, with little corresponding work on microstructures (e.g. Boorman et al., 2004; Williams et al., 2006; Vukmanovic et al., 2013; Holness et al., 2017a; Kaufmann et al., 2018). The existing microstructural work demonstrates ample evidence for dislocation creep (Vukmanovic et al., 2013; Holness et al., 2017a), with undeformed interstitial quartz in rocks containing strongly deformed plagioclase prismocrysts attesting to the syn-magmatic nature of this deformation (Holness et al., 2017a).

Plagioclase microstructures formed during deformation

In this contribution, we rely on quantitative microstructural interpretations in order to understand the behaviour of the crystal mush in the UZ chamber and its sub-solidus history. We focus particularly on plagioclase deformation microstructures, which are well understood (e.g. Rybacki & Dresen, 2004). Dislocation creep of plagioclase at high temperatures is most commonly achieved by slip on (010)[001] and (001)[110], with partial recovery leading to the creation of low angle boundaries (Passchier & Trouw, 2005). Whether a boundary represents a twist or a tilt in the crystal structure can be determined by analysis of the misorientation across it, which, for a tilt boundary, is a function of the slip system responsible (see the Supplementary Data; supplementary data are available for downloading at http://www.petrology.oxfordjournals.org for the method of determining slip systems).

The three main mechanisms involved in dynamic recrystallization of plagioclase undergoing dislocation creep are bulging recrystallization (BLG), subgrain rotation (SGR) and high-temperature grain boundary migration recrystallization (GBM) (Gerald & Stüinitz, 1993). Which of these mechanisms is dominant can be used to constrain the conditions of deformation (Passchier & Trouw, 2005). BLG and SGR are generally associated with deformation at lower temperatures than GBM and may operate in the subsolidus, whereas GBM may be associated with higher strain regimes (e.g. during regional deformation). Both SGR and, at lower temperatures, BLG, create neoblasts that are crystallographically closely related to their parent. GBM recrystallization is harder to recognise (Drury & Urai, 1990; Ji & Mainprice, 1990), but is usually associated with the formation of serrated grain boundaries and strain-free grains (Ji & Mainprice, 1990). In the absence of melt or fluid, neoblast compositions will be indistinguishable from their parent grains, but if melt or fluid were present at the time of deformation, the two grain populations may be chemically distinct (Rosenberg & Stüinitz, 2003).

Chemical disequilibrium is known to significantly contribute to, or even drive, feldspar recrystallization (Vernon, 1975; White, 1975; Stüinitz, 1998; Rosenberg & Stüinitz, 2003). In the presence of melt and at temperatures >850°C, recrystallization by grain boundary migration is driven by internal strain energy related to differences in chemical potential (Lafrance et al., 1996, 1998; Rosenberg & Stüinitz, 2003). Microstructures resulting from this process are characterised by strain-free grains separated by lobate grain boundaries and residual grains (Passchier & Trouw, 2005).

METHODOLOGY

Electron backscatter diffraction analysis

Samples were prepared using the EBSD preparation routine described by Prior et al. (1999). Most EBSD analyses were performed using an FEI sFEG XL30 SEM in the Department of Physics, University of Cambridge, and one sample was analysed at the School of Environmental Sciences at the University of Liverpool. Collection of electron backscatter diffraction patterns (EBSDPs) was undertaken using AZtechKL 2.2 acquisition software. EBSD maps and pole figures were constructed using the Oxford Channel 5 software and the MTex MatLab toolbox (Hielscher & Schaeben, 2008).
For details of acquisition parameters see the
Supplementary Data Table S1.

Pole figures were constructed using a lower hemisphere, equal area projection. The fabric strength and type were determined by calculating the J-index, pole figure J-indices (PFJ), M-index and BA-index of the orientation distribution function (ODF) using the MTex MatLab toolbox. We calculated the J- and M-index using the de la Vallée Poussin kernel, and a half-width of 10°, which corresponds to a series expansion of 28. The J-index has a value of one for a random distribution and a value of infinity for a single crystal (Wenk et al., 1998). While the J-index can be used to assess the overall fabric strength, pole figure J-indices (PFJ) describe the characteristics of an individual pole figure. The value of the M-index increases with the strength of the fabric from 0 (random fabric) to 1 (single crystal) (Skemer et al., 2005). We also calculated the BA index which ranges from 0 to 1. The meaning of the BA index is illustrated by values for the three plagioclase fabric types defined by Satsukawa et al. (2013): an axial A fabric is defined by a strong point maximum concentration of [010] with parallel girdle distributions of (010) and (001); axial B is defined by a strong point alignment of (010) with a girdle distribution of [100]; and type P is defined by point maxima of [001], (010) and (001). The axial A fabric is associated with high values of BA index (>0.7), an axial B fabric is characterised by a low value of BA index (=0.2), and the P-type fabric has intermediate values. Axial B fabrics are characteristic of magmatic processes (e.g. crystal settling). Type P fabrics are formed as a result of deformation or magmatic flow, whereas axial A fabrics form by deformation.

Electron microprobe

Plagioclase was analysed using a 5WDS Cameca SX-100 Electron Microprobe in the Department of Earth Sciences, University of Cambridge. All major and trace elements were analysed with a 15 kV, 10 nA, defocussed (5 μm diameter) beam. Peak counting times were 10 s for major elements (Si, Al, Na, K, Ca), 60 s for minor (Fe, Mg), and 90 s for trace elements (Ti), with Si and Na analysed first to mitigate the effects of electron beam-induced sample damage. All EPMA data and associated representative uncertainties are given in the Supplementary Data Table S1.

SAMPLES

We chose our sample suite to enable us to focus on the drivers for deformation. We examined a leucogabbro from the lower MZ, previously described by Holness et al. (2017a). In the borehole-core SL12 (drilled by Lonplats Mining Company at Moooinooi in the Western Limb; Fig. 1), the base of the Giant Mottled Anorthosite (a marker used to designate the base of the MZ) occurs at a depth of 1546-45 m. The borehole-core is dominated by plagioclase-rich norite and is notable for the abrupt appearance of evidence for significant dislocation creep at around 1540 m depth, and the gradual reduction in the intensity of deformation above ~1300 m depth: rocks above ~1000 m are apparently undeformed. We examined an MZ sample from 1395-77 m depth in borehole-core SL12, within the most strongly deformed part of the stratigraphy. Although clinopyroxene first appears in the liquidus assemblage in the strongly deformed region (stratigraphically below our sample), there is no major change in the modal mineralogy (and, therefore, mush density) associated with the onset or cessation of deformation and, critically, the mush layer was only a few metres thick (Holness et al., 2017a). Our chosen MZ sample, therefore, provides an example of deformation that cannot be attributed to gravitationally-driven compaction.

Since the mushy layer through much of the Bushveld stratigraphy was only a few metres thick (Holness et al., 2017a), any effects of compaction must have been on only a local (metric) scale: therefore, to focus on identifying the microstructural record of compaction, we chose a suite of samples in which the density of the crystal mush changes significantly over length scales of a few metres. The many magnetitite layers in plagioclase-rich regions of the UZ present the ideal opportunity to constrain the role of compaction as they result in the greatest possible loading on underlying plagioclase-rich cumulates.

We chose 54 samples from the UZ, focussing on magnetitite layers and their plagioclase-rich footwall and hanging-wall. We sampled the Bierkraal borehole BK3, drilled in the western limb of the RLS (Fig. 1) in a region of stratigraphy containing multiple magnetitite layers (21 in total). These cores have been extensively described by Yuan et al. (2017). The sample suite includes three magnetitite layers ranging in thickness from 25 to 113 cm (Supplementary Data Table S2), together with the plagioclase-rich cumulates (either anorthosite or leucogabbro) from both ~25–50 cm above and ~10–150 cm below the magnetite layer (Table 1; Supplementary Data Table S2). A sub-set of 14 samples from this suite was analysed by EBSD, including one sample from each of the three magnetitite layers; five samples of footwall; and three samples of hanging-wall (Fig. 1). The thickest magnetitite layer (BK3-13, 113 cm thick) contains a thin (~10 cm) horizon of leucogabbro (Fig. 1b), but can be considered to have acted as a single layer during any gravitationally-driven compaction, since the mush thickness was of the order several metres (Holness et al., 2017a). The lower contacts of the magnetitite layers are generally sharp at the outcrop scale, but the upper contacts are commonly gradational over tens of centimetres (Supplementary Data Table S2). The mineral modes of all examined samples are shown in Table 1. Plagioclase compositions were determined in a suite of samples associated with one of the three magnetitite layers, including three footwall samples, one
hanging-wall sample, two samples from the lower contact, and one from the magnetitite layer itself (Supplementary Data Table S3). None of the samples were oriented, although the direction of the paleovertical orientation is known relative to the drillcore.

Petrography

Main Zone
The MZ leucogabbro is characterised by large (apparent size: 2500μm long axis) plagioclase grains that define a strong fabric. These large crystals commonly show evidence of lattice distortion (Fig. 2a) and stepped and irregular grain boundaries. They are typically surrounded by abundant smaller plagioclase grains < 500μm across (Fig. 2b–d). The large plagioclase crystals show evidence of lattice distortion at their margins (Fig. 2c) adjacent to the fine plagioclase grains (Fig. 2d).

Other cumulus phases, such as pyroxene and interstitial quartz, are undeformed (Fig. 2a, b).

Upper Zone

Plagioclase-rich cumulates
The anorthosites or leucogabbros stratigraphically above and below the magnetite layers are characterised by bimodal plagioclase grain size distributions (Fig. 3a–d; Fig. 4d–f). The larger crystals (apparent size: >1000μm long axis) are euhedral to subhedral and appear elongate in thin section, with an average apparent aspect ratio between 1.8 and 2.4 (Table 1). The euhedral large grains have straight grain boundaries parallel to the growth faces but are often stepped on a small scale (Fig. 3a, c, d). Some subhedral large grains show slightly serrated grain boundaries (Fig. 3b, Fig. 4e black dotted line). The larger crystals contain deformation twins (Fig. 3), low angle boundaries (Fig. 3d) and often show undulatory extinction (Figs 3b, d; Figs 4b, e). In contrast, the smaller grains (apparent size: <1000μm long axis) are equant (average apparent aspect ratio of 1.4, Table 1), and preserve less evidence of intra-grain deformation (Figs 3a–d, 4d, e). These smaller grains are generally distributed along the grain boundaries of larger grains (Figs 4d, e). The smaller grains commonly contain growth twins, but deformation twins are also present (Figs 3a, c). The thin leucogabbro horizon within the thick magnetitite layer BK3-13 does not show any observable differences to those above and below the magnetitites.

In leucogabbros, clinopyroxene is typically interstitial (Fig. 3d) and generally contains orthopyroxene exsolution lamellae (Fig. 4f). A few pyroxene crystals show undulatory extinction or rare low angle boundaries. Despite being part of the primocryst assemblage, interstitial overgrowth on oxides resulted in a subhedral habit (Fig. 3c). Rare interstitial quartz (<100μm) is present and appears strain-free (Fig. 3f). Reactive symplectites composed of plagioclase and augite are common in both anorthosites and leucogabbros, replacing plagioclase primocrysts and commonly spatially associated with undeformed, euhedral biotite (Fig. 3e, f). Following Holness et al. (2010), we interpret them as a product of reaction between an iron-rich immiscible conjugate liquid and the surrounding primocrysts, following the loss of the Si-rich conjugate.

Magnetitite layers
Magnetitite layers contain ~60 vol. % magnetite, with subsidiary plagioclase (~30–35 vol. %) and ilmenite (5–10 vol. %). On a thin section scale, the apparently sharp lower contact is revealed to be highly undulatory, with
mm-scale extension of oxides into the silicate footwall, outlined by abundant fine polycrystalline olivine (Fig. 4a).

Plagioclase within the oxide layer is commonly aligned, forming a foliation (Fig. 4b). It is generally separated from the surrounding oxide by a thin film of polycrystalline biotite or olivine, and/or is partially replaced by reactive symplectites (Fig. 4b, c). Some plagioclase grains appear entirely recrystallized to form aggregates of randomly oriented grains that preserve the elongated shape of the original grain (Fig. 4b, c). While most plagioclase grains within magnetite layers are elongated tabular crystals or part of a recrystallized cluster replacing either single isolated laths or clusters of original laths, some are much smaller and form isolated rounded grains (Fig. 4c). Magnetite grain size inversely correlates with plagioclase mode, with relatively fine-grained magnetite (<500 μm in diameter, compared to the usual 5000 μm diameter) in samples with >20 vol. % plagioclase (Supplementary Data Fig. S1).

QUANTIFICATION OF PLAGIOCLASE FABRICS

Main Zone

The foliation defined by larger plagioclase crystals in the MZ leucogabbro is parallel to the floor of the magma chamber (Fig. 5) and is defined by a preferred grain orientation with [100] and (010) planes parallel, and (001) perpendicular to the foliation (Fig. 5). The relationship between the (010) and (001) planes attests to a predominantly tabular plagioclase grain shape (Morales et al., 2011, Holness et al., 2017b). A preferred orientation of the small plagioclase crystals (<800 μm) defines the same crystallographic fabric (Fig. 5b). In both cases, the fabric BA index (e.g. 0.17, Table 2) attests to its magmatic origin. The misorientation axes are parallel to foliation and correspond to the [100] axis (Fig. 5b).

The small plagioclase grains that surround the larger grains form groups with a very similar crystallographic orientation, but with an angular difference of ~40° compared to the adjacent large grain (Figs 2c, 6). The large grains commonly show evidence for lattice distortion (~15–20°) and contain low angle boundaries, as well as subgrains (Supplementary Data Fig. S2). Of the 15 low
angle boundaries analysed in this sample, more than half are tilt boundaries created by slip on (010)[001].

**Upper Zone**

The orientation of the plagioclase crystals in the UZ samples that we examined partly correlates with their grain size (Figs 7a, b). The orientation of large (>1000 µm) and small (<1000 µm) grains is almost identical (Fig. 7c, f), with [100] and poles to the (010) plane parallel to foliation, whereas poles to the (001) plane are randomly distributed (Fig. 7c, d). The relationship between the (010) and (001) planes attests to a predominantly tabular grain shape (Morales et al., 2011; Holness et al., 2017b). However, both M- and J-indices show that the fabric defined by the larger grains is stronger than that defined by the smaller grains (see Supplementary Data Figs S3, S4). The BA index is within the range 0.17 to 0.64 (Table 2). Our UZ samples record three types of fabric: P, axial B and axial A (Satsukawa et al., 2013). Most samples record a P-type fabric (e.g. defined by point maxima for all three axes), indicative of crystal lineation (Table 2, Supplementary Data Figs S3, S4). This is most strongly defined by the smaller grains. In other samples, the second most abundant fabric is axial B-type (e.g. a foliation defined by...
alignment of (010) planes), which is typical of undeformed layered intrusions (e.g. Holness et al., 2017b) and ophiolites (Morales et al., 2011; Satsukawa et al., 2013; VanTongeren et al., 2015). This fabric is most strongly defined by large grains (Table 2, Fig. 7, Supplementary Data Figs S3, S4). Axial B and P-type fabrics are not systematically confined to either hanging wall or footwall layers (Table 2). A small number of samples, mainly those dominated by magnetite, contain plagioclase with an axial A-type fabric, characteristic of deformation (Table 2, Supplementary Data Figs S3, S4). Misorientation axes for the bulk samples (i.e. for both grain size populations) show that misorientation axes coincide with [100] axes (i.e. in the same plane as the foliation; Fig. 7c, f).

The most common microstructure shows no crystallographic relationship between neighbouring plagioclase grains of different size (Fig. 8a, b). The pole figures of the larger grains generally show unambiguous evidence for crystal deformation, with strong dispersion of the orientation of (010) and (001) around the [100] axis (Fig. 8b). The nearby small grains are either only weakly deformed or undeformed, with clustered orientations of the [100] axis and (010) planes (Fig. 8b).
The few small grains in clusters have a close orientation to the neighbouring large grains (Fig. 8c, Supplementary Data Fig. S5). In rare cases, we observe very similar crystallographic orientations in neighbouring grains of different size. An example is shown in Fig. 8c, where a gradual increase in crystallographic misorientation is observed across a low angle boundary associated with a subgrain along the profile A–A’. Dispersion of (010) and (001) occurs around the [100] axis, consistent with operation of the (010)[001] slip system during dislocation creep (Supplementary Data Fig. S6). The smaller crystals were, therefore, produced by either BLG or SGR recrystallization, both of which produce neoblasts with a similar orientation to their parent (Rosenberg & Stünitz, 2003; Passchier & Trouw, 2005; Svahnberg & Piazolo, 2010).
The extent of crystallographic misorientation within large plagioclase crystals can vary from ~10° to >35° (Fig. 8c, Supplementary Data Fig. S4a). Low angle boundaries are often parallel within a single crystal and the predominant slip system is {010} [001]. There is no systematic variation in the amount of crystal plasticity relative to the position of magnetite layers (Supplementary Data Fig. S6a).

Clinopyroxene primocrysts commonly contain low angle boundaries, with lattice distortion of <25°. Interstitial clinopyroxene is generally less deformed than the associated primocrysts, with no low angle boundaries (Supplementary Data Fig. S6b). Both plagioclase and clinopyroxene crystals within the late reactive symplectites show no evidence of crystal plasticity (Fig. 9).

PLAGIOCLASE MICROSTRUCTURE IN THE MAGNETITITE LAYERS

Plagioclase crystals within the magnetite layers show a different microstructure to those in anorthosites and leucogabbros, falling into four types: (1) isolated round grains (Fig. 4c); (2) isolated, subhedral, elongate, grains that record evidence of minor plastic deformation (~10° of crystal distortion) with only minor marginal recrystallization (Fig. 10a); (3) grains that preserve their original shape but with extensive recrystallization along grain boundaries with other plagioclase grains (Fig. 10b), the orientation of these neoblasts progressively changes from that of the adjacent larger crystal with increasing distance from the latter; and (4) completely recrystallized grains, comprising neoblasts with no evidence for a retained magmatic CPO (Fig. 10c). The third and fourth categories of grains are most common in plagioclase-rich zones (Fig. 10b, c), suggesting that recrystallization is most common where the original large laths formed clusters.

PLAGIOCLASE MINERAL CHEMISTRY

Plagioclase compositions (core and rim) were measured in large and small grains from six UZ samples (both above, below and within, a magnetite layer; Supplementary Data Table S3). The plagioclase anorthite number (An# = Ca/(Ca+Na) x 100) varies between 55 and 77 (Fig. 11a) and correlates with grain size (quantified using the Feret diameter). The small crystals (<1000 μm) have An# in the range 55–77 (mean ~60) and include the most primitive (i.e. highest An#) crystals (Fig. 11a). Coarser plagioclase grains (>1000 μm) typically have more evolved compositions (i.e. lower An#) that fall in a narrow range with An# typically 50–62 (mean ~58; Fig. 11a). Individual crystals have homogeneous anorthite contents, with An# varying by <2 between the cores and rims of single large grains (Supplementary Data Table S3).

Table 2: CPO characteristics of the anorthosites studied here

<table>
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<tr>
<th>Sample</th>
<th>Layer</th>
<th>Grain Size</th>
<th>N</th>
<th>BA</th>
<th>J-index</th>
<th>PIJ 100</th>
<th>PIJ 010</th>
<th>PIJ 001</th>
<th>M-index</th>
<th>CPO</th>
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<td>1269</td>
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<td>3.80</td>
<td>1.26</td>
<td>1.70</td>
<td>1.13</td>
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<td>B</td>
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N, number of grains measured; BA, index; PIJ, pole figure index for three main axes; type of CPO shown are axial–B, type P and axial–A fabrics (see text for detailed explanation of CPO types). Other abbreviations as in Table 1.
DISCUSSION

Chemical differences between the two populations of plagioclase grains

According to Yuan et al. (2017), ilmenite first crystallizes in the UZ when the plagioclase An# is ~58. Although some grains in our UZ samples have high An# (<77) and likely crystallized before ilmenite joined the liquidus, most of our feldspar measurements have An# within analytical uncertainty of ilmenite saturation (i.e. ~58; Fig. 11a), consistent with their proximity to the magnetitite layers. The smaller plagioclase grains are often more anorthitic than larger grains, suggesting that they either crystallized from a more primitive liquid or record more extensive reaction with an interstitial (chemically isolated) Fe-rich conjugate liquid (which would contain abundant Ca and P) than the larger grains (e.g. Humphreys, 2011). The latter hypothesis is supported by the spatial association between small anorthite-rich plagioclase grains and reactive symplectites (Fig. 9d). However, a more detailed study of the plagioclase chemistry (e.g. zoning and precise trace element measurements) is required to fully understand the An# relationship with grain size.

The cause of plastic deformation in the leucogabbros and anorthosites

Both the MZ leucogabbro and the UZ anorthosites and leucogabbros preserve abundant evidence of deformation by dislocation creep. The misorientation axes in all samples are uniformly parallel to the foliation, suggesting that each of our samples experienced a single deformation event (cf. Reddy & Buchan, 2005; Michels et al., 2015; Vukmanovic et al., 2018b). Furthermore, the absence of plastic deformation recorded by interstitial
quartz and the reactive symplectites, and only weak deformation of interstitial clinopyroxene, suggests that this single deformation episode was confined to the super-solidus, when a significant volume of melt was present in the crystal mush (in agreement with Holness et al., 2017a).

Drivers for super-solidus deformation are either internally-generated, such as gravitational loading by the overlying mush (Sparks et al., 1985; Meurer & Boudreau, 1998; McKenzie, 2011; Holness et al., 2017b), or externally-generated and related to regional tectonics (Carr & Groves, 1994). We consider it unlikely that
gravitationally-driven compaction was the main cause of the deformation observed in the MZ leucogabbro, because dislocation creep is confined to a particular stratigraphic horizon in the SL12 drill core, with no correlation between the strength of deformation and the modal mineralogy of the crystal mush (Holness et al., 2017a).

Our comparison of both the footwall and hanging-wall of the UZ magnetitite layers leads us to the same conclusion. Critically, our results show that the amount of plastic deformation experienced by plagioclase-rich cumulates is the same both above and below magnetitite layers, regardless of the thickness of the magnetitite. Hence, if compaction had occurred it cannot have involved dislocation creep. Furthermore, if the footwall had been affected by viscous compaction by pressure-solution processes we might expect to see smaller modal proportions of interstitial phases compared to the hanging-wall, but this is generally not observed (Table 1). There is no consistent relationship between fabric strength and stratigraphic position relative to the magnetitite layers; footwall samples may have a weaker fabric than those above (Table 2), inconsistent with the fabrics forming by pressure-solution (c.f. Meurer & Boudreau, 1998).

As our microstructural data are inconsistent with internally-driven deformation by viscous compaction by either dislocation creep or pressure-solution, we instead suggest that the high-temperature deformation recorded by our MZ and UZ leucogabbros and anorthosite is related to external, progressive and long-lasting regional subsidence of the magma chamber (e.g. Carr & Groves, 1994; Maier et al., 2013). Interestingly, the absence of evidence of deformation in grains formed during the last stages of solidification (i.e. the reactive symplectites and the interstitial quartz) suggests that such subsidence only affected the crystal mush while it contained more than a few vol. % liquid.

Fig. 8. (a) Photomicrograph and grain size map, using ECD, of two large plagioclase grains and their fine-grained neighbours. (b) Pole figure data for the two large grains (teal) and their fine-grained (red) neighbours. (c) Misorientation map of a large plagioclase grain and two smaller neighbours. The map is coloured using the inverse pole figure (IPF) key referenced to the mean orientation of a grain. The colour scale (top left) is adjusted so that white represents 0° and black represents 10° of misorientation. Misorientation profile A–A’ showing an increase of misorientation towards two small neighbouring grains. Pole figure data for the large grain and its neighbours, shows that the dispersion axis (i.e. axis that experienced a minimum of dispersion) is [100], whereas dispersion between the small grains and the large grain is ~ 20°.
The cause of plagioclase deformation in the magnetitite layers

Plagioclase grains in the UZ magnetitite layers experienced a different deformation history to those in the over- and underlying anorthosites and leucogabbros. Of particular importance is the observation that the degree of deformation recorded by each plagioclase grain correlates with the local plagioclase mode. Isolated crystals surrounded by magnetite record only minor lattice distortion (Fig. 10a), whereas nearby groups and clusters of plagioclase grains are either partially or fully recrystallized (Fig. 4b, c; Fig. 10b, c). The greater extent of deformation for clustered compared to isolated plagioclase grains suggests that the deformation took place in an essentially liquid environment, in which isolated grains were able to move freely while clustered grains experienced local jamming (e.g. Bergantz et al., 2017). In this case, the magnetitite layers must have originally been very liquid-rich, rather than representing crystal accumulations. Such a liquid would necessarily be the Fe-rich immiscible conjugate of a fully-unmixed immiscible magma. This interpretation is consistent with the suggestion of Maier et al. (2013) that the magnetitite layers formed from liquid-rich slurries, containing plagioclase and magnetite grains in an Fe-dominated liquid. It is also consistent with the presence of abundant reactive symplectites that partially replace the plagioclase grains, both within the magnetitite layer and at its base, indicating reaction with an Fe-rich liquid (e.g. Holness et al., 2010). The numerous small, rounded plagioclase grains may have originally formed part of recrystallized laths that subsequently disintegrated during flow of the liquid-rich mush. The generally sharp and planar lower contact of the magnetitite layers is consistent with a generally thin (~1 m) mushy layer in the Bushveld (Holness et al., 2017a); we envisage these mobile slurries moved across an almost completely solidified plagioclase-rich floor into which interstitial (and reactive) liquid locally percolated only a few centimetres (Fig. 4a).

A new phenomenon: cavitation-driven nucleation?

Microstructures in the MZ leucogabbro are a result of super-solidus dislocation creep accompanied by dynamic recrystallization, creating neoblasts on grain boundaries between the larger grains, regardless of boundary orientation. There are also bimodal crystal size populations in the UZ leucogabbros and anorthosites, but there are key microstructural differences between these UZ samples and the dynamically recrystallized MZ sample, suggesting a different mode of super-solidus deformation. Critically, in contrast to the homogeneous spatial distribution expected for dynamic recrystallization, the UZ samples show a preferred distribution of the smaller grains on grain boundaries parallel to the plane of foliation (Fig. 3a–c; Fig. 4d, e; Fig. 7b, e). The small crystals often have a very different crystallographic orientation (misoriented by more than 60°) to their immediate larger neighbours, without the progressive change in orientation with increasing distance from the large grain that would be expected if the small crystals formed during dynamic recrystallization (Fig. 8b, d). Additionally, the small grains in the UZ samples are euhedral, in contrast to the rounded irregular shapes expected for dynamic recrystallization (Table 1, Fig. 3a–c, Fig. 4d–e). There is also a compositional difference between large and small plagioclase grains in the UZ, with the small crystals...
typically less evolved (higher An#) than the larger grains (Fig. 11). We would not expect small grains to form during slow, uninterrupted, cooling of a partially solidified mush, since it is energetically more favourable for the interstitial liquid to form overgrowths on existing grains than to nucleate new ones (Vernon, 2004). We suggest instead that the small crystals in the UZ samples formed during an episode of enhanced undercooling related to deformation of the mush, triggering a burst of nucleation in the interstitial liquid. We term this process ‘cavitation-driven nucleation’.

Deformation by diffusion creep is always accompanied by grain boundary sliding (Passchier & Trouw, 2005). Where deformation is sufficiently rapid to create an imbalance between grain boundary sliding and the necessary crystal plasticity or diffusional mass transport to accommodate it, micro-cavities open along grain boundaries. This process is called ‘creep cavitation’ (Kassner & Hayes, 2003; Dimanov et al., 2007). Précigout & Stünitz, (2016) showed that olivine can nucleate in such micro-cavities during sub-solidus deformation. We suggest that the rapid localised decompression accompanying creep cavitation in super-solidus crystal mushes may also create sufficient undercooling (i.e. departures from chemical equilibrium) to promote nucleation in the newly-formed cavities. Although the localised reduction in pore pressure associated with dilatation is strain-dependent (i.e. Reynolds dilatancy) and we might, therefore, expect only minor pore pressure reduction in slowly-deforming mushes in a regionally-subsiding magma chamber, John & Stünitz (1997) showed that fracturing parallel to foliation requires very small effective stress. The predominant localisation of the smaller plagioclase crystals parallel to the foliation defined by large plagioclase grains is consistent with this, suggesting that cavitation-driven nucleation was confined to cavities formed by separation of plagioclase grains within the foliation during deformation at low strain rates. The newly-formed plagioclase grains nucleated as a result of cavitation in a deforming mush are likely to be rotated into alignment with the shear direction, consistent with the
foliation and weak lineations recorded by the small plagioclase grains in our UZ magnetitite layer (Fig. 8b; Table 2). The low aspect ratio of these small grains (Table 1) suggests that post-nucleation crystal growth occurred in a slow-cooling environment (Holness, 2014).

The driver for shearing of the UZ mush, in common with that of the MZ sample investigated here, is likely to be gravitational instability of the floor mush, perhaps induced by regional-scale deformation (e.g. Maier et al., 2013). That we see significant differences in the microstructures formed during super-solidus, deformation of the MZ and UZ plagioclase-rich cumulates may be linked to differences in the amount of liquid present in the deforming mush (with higher liquid content in the UZ cumulates) or to differences in the strain rate during deformation.

The thickness of the plagioclase-rich mushy layer undergoing shearing in the UZ is not well-constrained, as the method of Holness et al. (2017a) only provides a precise measure of mush thickness at points in the stratigraphy where the liquidus assemblage changes. While the sharp lower boundaries of the magnetitite layers suggest that the Fe-rich liquid slurry flowed across an essentially rigid, low-permeability floor with a very thin mush, it is possible that a plagioclase-dominated mush may have reached several metres in thickness. Future work should focus on finding stratigraphic horizons containing both evidence of our newly-proposed cavitation-driven nucleation and field or microstructural evidence that can be used to precisely constrain mush thickness.

CONCLUSIONS

Despite a focussed examination of regions of the Bushveld stratigraphy containing mineralogical contrasts providing the greatest possible driver for internally-generated gravitationally-driven compaction of a crystal mush (i.e. thick, dense magnetitite layers), we found no convincing evidence that the deformation preserved in these rocks was a consequence of compaction. The thickest magnetitite layer we examined (113 cm) did not provide sufficient gravitational load to drive plastic deformation by dislocation creep, or to reduce the amount of interstitial liquid in the plagioclase-rich footwall compared to the hanging-wall. Instead, we suggest that the abundant evidence for super-solidus deformation in these rocks was a consequence of regional-scale events such as slumping and subsidence of the crystal mush.

Our microstructural examination of the UZ cumulates supports a history of mush mobility and a significant role of Fe-rich immiscible liquids in the formation of the magnetitite layers. Our microstructural observations of abundant fine-grained plagioclase grains in the UZ leucogabbros and anorthosites points to the possibility that shearing of a relatively liquid-rich crystal mush may drive crystal nucleation by localised undercooling within the cavities that form as the grains slide...
past each other. This suggestion requires further detailed work (e.g. experiments and numerical modelling) to test this hypothesis and to place tight constraints on the relationship between strain rate, cavitation and localised departures from chemical equilibrium.

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SUPPLEMENTARY DATA
Supplementary data are available at Journal of Petrology online.

REFERENCES


