



Massive chromitites of the Bushveld Complex, South Africa: A critical review of existing hypotheses

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ABSTRACT

The controversy over the origin of massive chromitites in layered intrusions has recently become more contentious than ever before. At issue is whether they are produced via gravity settling/in situ crystallization of chromite directly on the chamber floor or by kinetic sieving, metasomatic replacement, or sill-like intrusions beneath the chamber floor, i.e., in deep parts of the cumulate pile. The latter group of models implies that massive chromitites have never been on the chamber floor. In this paper, we show that decisive clues to the 'chamber floor dilemma' come from field relations of massive chromitite with magmatic dropstones in the Bushveld Complex, South Africa - the largest fossilized magma chamber in the Earth's continental crust. The roof sequences in such basaltic magma chambers are known to be inherently unstable; therefore, crustal instability results in their breakdown and collapse as angular blocks (i.e., magmatic dropstones) through the resident melt column onto the upward-growing chamber floor. We have discovered such magmatic dropstones in the sequences that host massive UG1, UG2, and MG2 chromitites in the Critical Zone of the Bushveld Complex. The dropstones are easily recognizable in the field because they are composed of fine-grained melanorite/orthopyroxenite, which are texturally dissimilar from and have sharp contacts with adjacent cumulate rocks (chromitite, anorthosite, norite, etc.). The dropstones range in shape from angular to lenticular fragments a few centimetres in size to large tabular dropstones of ~0.5–1.0 m across and ~10 m long. The dropstones indent pre-existing layers of chromitite/anorthosite beneath them and are covered by subsequently deposited layers of the same rocks above them. Some dropstones appeared to have knifed into chromitite/anorthosite layers and cut them off, and a few dropstones appeared to have been driven into the floor cumulates and crumpled the layers outwards and upward. Also, some beheaded dropstones indicate the truncation of the floor cumulates by planar erosional surfaces. The physical relationships of dropstones with their host rocks indicate that: (1) there was almost invariably a sharp interface between the top of the inward-growing cumulate pile and the overlying resident melt; (2) the uppermost part of the cumulate pile was coherent and igneous layering, involving chromitites, was generally fully developed right up to the crystal-liquid interface; (3) by the time of dropstones' landing, the chromitite layers were already formed on the chamber floor, with the resident melt immediately overlying them; (4) the existence of chromitite layers at the crystal-liquid interface implies their formation through the deposition of chromite alone directly onto the chamber floor; and (5) such crystal deposition requires the resident melt in the chamber to be chromite-only saturated (i.e., no other liquidus phases in the chamber). The occurrence of magmatic dropstones thus rules out the formation of massive chromitites by processes that operate at some depth in the cumulate pile (e.g., kinetic sieving, metasomatic replacement, or sill-like intrusions) and rather indicate their deposition via in situ crystallization from a chromite-only-saturated magma directly on the upward-growing floor of a Bushveld magma chamber. A thorough review of this concept demonstrates its high potential in explaining most field, textural and chemical features of massive chromitites in the Bushveld Complex.

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1. Introduction

The current understanding of the origin of massive (stratiform) chromitites (60–90 vol% chromite) in mafic layered intrusions is facing challenges from multiple perspectives. Traditionally, the massive chromitites were considered to form by gravity settling of chromite onto the chamber floor from a chromite-only-saturated magma (Irvine, 1975; Cameron, 1980; Campbell and Murck, 1993; Lipin, 1993; Kinnaird et al., 2002; Cawthorn, 2005, 2011; Spandler et al., 2005; Naldrett et al., 2012; Holness et al., 2023). Following this school of thought, it has also been proposed that such a chromite-only-saturated magma may crystallize layers of massive chromitites directly on the instantaneous chamber floor (Latypov et al., 2015, 2017b, 2022a, 2023). The validity of this traditional model has been, however, recently questioned on various grounds and several alternative scenarios have been put forward. These models propose that massive chromitite form in deep parts of the cumulate pile, rather than on the chamber floor and that chromite-only-saturated magmas are not involved in this process. There are three major approaches here. A first approach implies gravity- and size-controlled separation of chromite from coexisting silicate phases (i.e., kinetic sieving) into a seam of massive chromitite at the base of a 20 to 100 m thick layer of a crystal-rich slurry (Mondal and Mathez, 2007; Eales and Costin, 2012; Maier et al., 2013; Jenkins and Mungall, 2018; Maghdour-Mashhour and Hayes, 2021; Barnes et al., 2023). A second approach envisages a situation when crystal-rich slurries are intruded as late-stage sills into the pre-existing cumulate pile well below the temporary chamber floor (Voordouw et al., 2009; Mungall et al., 2016; Mitchell et al., 2019; Scoates et al., 2021; Otto et al., 2024). While the slurry is still flowing along the sills, chromite grains are proposed to separate from silicate phases by kinetic sieving to form a layer of massive chromitite at the base of the sills (Mungall et al., 2016). In some models, such slurries are composed of only chromite crystals (Voordouw et al., 2009). Finally, a third approach implies that a large amount of upward migrating melts/fluids causes the complete remelting of mafic rocks leaving behind a restitic layer of massive chromitite in the cumulate pile (Mathez and Kinzler, 2017; Nikolaev et al., 2020; Marsh et al., 2021; Boudreau, 2024). To resolve this ‘chamber floor dilemma’ (i.e., the formation of massive chromitites on the chamber floor versus beneath the chamber floor) by various geochemical methods is quite challenging. We propose here, however, that this can be done using field relations of massive chromitites with ‘magmatic dropstones’, i.e., the blocks that collapse from the roof sequences on the floor of magma chambers.

A magmatic dropstone approach has not yet been applied to a problem of massive chromitites in mafic layered intrusions although it has been already employed to address a few petrological problems such as the origin of igneous layering (e.g., Irvine et al., 1998). Importantly, the vast majority of the layered intrusions lack any rocks that grew from the roof downward (Cawthorn, 1996; Charlier et al., 2015; Smith and Maier, 2021; Latypov et al., 2024b) even though the magma in the chamber predominantly cools through and crystallize at the roof (Irvine, 1970; Frenkel et al., 1988; Morse, 1988). The absence of such rocks has been attributed to breakdown of the initial roof sequences and their collapse onto the inward-growing chamber floor (Chistyakova and Latypov, 2022; Latypov and Chistyakova, 2024a; Latypov et al., 2024b). The blocks of the roof sequences (i.e., magmatic dropstones) are found among the rocks of the floor sequences in many layered intrusions. Among them are Skaergaard intrusion (Irvine et al., 1998), Akanvaara intrusion (Mutanen, 2005), Sudbury Complex (Latypov et al., 2019), Stillwater Complex (Hess, 1960), and Windimurra Complex (Nebel et al., 2013). Surprisingly enough, such magmatic dropstones have not yet been reported from the world’s largest layered intrusion – the Bushveld Complex in South Africa – in which the roof sequence is also absent, implying its complete destruction via transient episodes of tectonic activity. A recent activity in open pit mining of massive chromitites in the Critical Zone of the Bushveld Complex resulted in excavation of many large-scale, remarkable exposures in which magmatic dropstones

of roof-derived rocks have been discovered for the first time. In this paper, we present the first results of our small-scale mapping of physical relationships between these magmatic dropstones, and the floor cumulates which allow contributing to resolution of the controversy of whether or not massive chromitites in layered intrusions are formed by deposition of chromite crystals directly on the chamber floor of magma chambers. To facilitate the understanding of a magmatic dropstone approach, we start below with an interpretive framework that provides background information on the interpretation of our new field observations from the block-and-layering structures in the context of processes operating in large basaltic magma chambers.

2. Interpretive framework

There are apparent similarities between glacial dropstones falling out of melting icebergs and impacting the sediments at the bottom of the water column in sedimentary basins (Fig. 1a) (Harland et al., 1966; Ojakangas, 1988; Menzies, 2002; Melezhik et al., 2013) and the magmatic dropstones falling from the roof sequences and deforming the floor cumulates in magma chambers (Irvine, 1987; Irvine et al., 1998; Scoates, 2000; O’Driscoll et al., 2008; Brandriss et al., 2014). These similarities are worthwhile to emphasize and employ for the better interpretation of the processes operating in evolving magma chambers of layered intrusions.

In deep seas/lakes, glacial dropstones are isolated fragments of rocks that range in size from small pebbles to large boulders and usually penetrate lamination within fine-grained, water-laid sedimentary rocks (Menzies, 2002). The most characteristic feature of glacial dropstones is that due to density contrast with and relatively low viscosity of water, fragments fell nearly vertically through the seawater column to reach the bottom sediments. When deposited with high momentum, dropstones are able to deform, penetrate, and rupture bottom sediments (Fig. 1b), leaving a soft-sediment deformation structure (e.g., Bronikowska et al., 2021). In these structures, the muddy sediments may be squeezed up around the edges of the fallen dropstones. The continued sediment deposition will then result in blanketing of dropstones by younger sediment layers (Fig. 1c). Finally, the initial bending of sediment layers around the dropstones can be enhanced by load-induced compaction of poorly consolidated sediments. Although not explicitly formulated, the physical relationships of glacial dropstones with fine-grained, laminated sediments on which they deposited are normally taken to indicate:

- gravity-induced settling of dropstones from melting icebergs, floating at the surface of sedimentary basins;
- an existence of water column through which the dropstones were settled before reaching a sedimentary pile;
- an occurrence of a planar and distinctive sediment-water interface;
- an existence of well-developed laminations immediately at the top of a sedimentary pile.

In large layered intrusions, the roof-derived blocks impacting the floor cumulates of magma chambers (e.g., Irvine et al., 1998; Scoates, 2000; O’Driscoll et al., 2008; Brandriss et al., 2014) can be regarded as magmatic dropstones. These magmatic dropstones, when viewed in the context of their counterparts in sedimentary basins, can also be interpreted as indicating:

- their gravity-induced settling from the roof sequences that were broken by crustal instabilities, e.g., earthquakes associated with magma chamber replenishment;
- an existence of a thick column of low-viscosity resident melt in the chamber through which the ‘dropstones’ have been settling to reach the floor cumulate pile with a high momentum;
- an occurrence of a planar and sharp interface between the top of a cumulate pile and overlying resident melt;

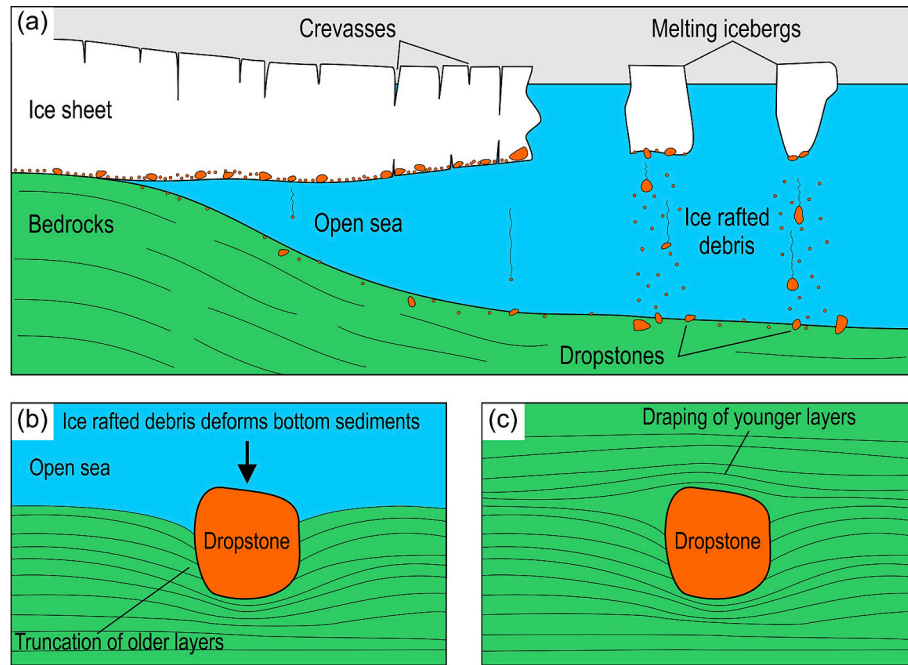


Fig. 1. A cartoon illustrates melting glacial icebergs that release dropstones, which fall and impact the sediments at the bottom of the water column in sedimentary basins. (a) The advance of glaciers can lead to the accumulation of significant sediment in the ice, which occurs due to the scraping and plucking of debris from the underlying substrate. The edges of the ice sheet offshore can calve to form icebergs. The edges of the ice sheet offshore can break off to form icebergs, which can travel hundreds of kilometres on the open sea. As they melt, the sediment that is trapped inside the ice becomes free and drops to the bottom of the sea, resulting in the formation of dropstones. (b) The layers beneath a dropstone are down warped and/or truncated, which occurs due to the dropping of the boulder onto originally soft mud. (c) Overlying younger sediment layers drape onto the sides of the dropstone, which occurs due to the continuous deposition of fine-grained sediments after the boulder was dropped by an iceberg. The figures are modified from drawings at the webpage <https://malayanoplia.wordpress.com/>.

- an existence of well-developed igneous layering immediately at the crystal-liquid interface of the chamber floor.

This interpretive framework is employed below for describing and

deciphering field relations of magmatic dropstones with the floor cumulates in the Bushveld magma chamber. One essential difference between sedimentary and magmatic environments is that glacial dropstones may penetrate bottom sediments much deeper because the

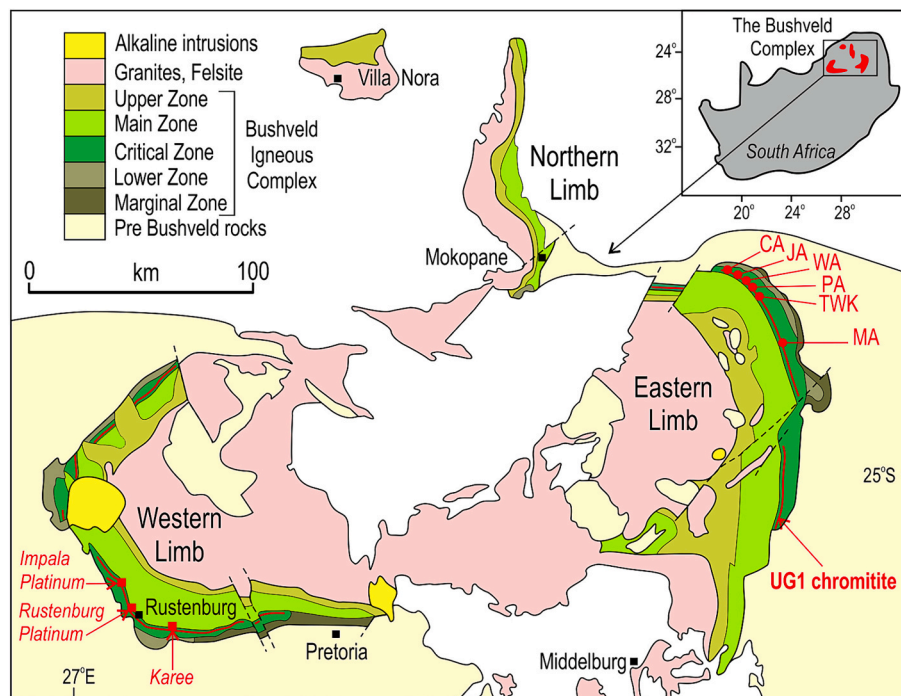


Fig. 2. Geographical location and a generalized geological map of the Bushveld Complex (Rustenburg Layered Suite) in South Africa. The places involved in this study are indicated on the map. MA - Maandagshoek, JA - Jagdlust, PA - Paschas Kraal, CA - Cameron, TWK - Twickenham. WA - Waterkop. Modified from Latypov et al. (2017a).

sediments are usually still loose and dropstones have a higher momentum at the impact point due to large difference in their density with water and low-viscosity of water (e.g., Bronikowska et al., 2021). The penetrating ability of dropstones in magma chambers is substantially lower because of much smaller difference in the density of cumulates and a basaltic melt and much higher viscosity of this melt (compared to water) and therefore dropstones are commonly arrested at or very close to a crystal-liquid interface of the cumulate pile (e.g., Irvine et al., 1998). This also happens because the crystal mush tends to be mostly solidified at the very top of the cumulate pile (Hess, 1960; Thompson and Patrick, 1968; Irvine et al., 1998; Latypov et al., 2015, 2024a; Holness et al., 2017a, 2017b; Kruger and Latypov, 2020) and therefore readily withstands the impact of slowly falling blocks. Another important difference is that, movement of some blocks towards the chamber floor may be partly guided by a streamline pattern of a convecting resident melt and they may, impact the floor cumulates obliquely, rather than sub-vertically (Irvine et al., 1998). Such a transportation of blocks may take place because of a relatively small difference in the density of cumulate rocks (blocks) and a resident melt in the chamber.

3. The Bushveld Complex

The 2.05 Ga Rustenburg Layered Suite of the Bushveld Complex in South Africa (Fig. 2) is the largest mafic-ultramafic layered intrusion in Earth's continental crust comprising about 600,000 km³ of igneous rocks (Cawthorn and Walraven, 1998; Cawthorn, 2015; Latypov et al., 2022a, 2022b). The complex consists of several parts, the western, eastern, and northern limbs being the largest, and is subdivided stratigraphically into five major units - the Marginal, Lower, Critical, Main, and Upper Zones, comprising a total thickness of 7 to 9 km (Fig. 3) (Cawthorn and Walraven, 1998; Cawthorn, 2015). The Bushveld Complex is widely considered to be a typical example of an open-system magma chamber (Kruger, 2005; Cawthorn, 2015; Yao et al., 2021; Latypov et al., 2022b). Its principal zones are attributed to major replenishing events, with numerous smaller magma recharges contributing to the formation of these zones. During this process, the magma chamber incrementally increased in size through both vertical and lateral inflation (Willemse, 1959; Eales, 2002; Kruger, 2005). All major replenishing events are marked by regionally extensive magmatic discontinuities, local erosive unconformities into previous strata, significant isotopic shifts, and notable changes in whole-rock and mineral compositions (Eales et al., 1990; Kinnaid et al., 2002; Kruger, 2005; Smith et al., 2013; Cawthorn, 2015). The Bushveld Complex hosts global resources of several critical metals. In particular, the Bushveld Complex contains >80% of the world's known chromium resources (Cawthorn, 2010) which are hosted within 14 principal layers of massive chromitites (60–90 vol% chromite), mostly confined to the Critical Zone (Fig. 3) (Scoon and Teigler, 1994; Schurmann et al., 1998; Naldrett et al., 2012). Three major groups of chromitites are recognised: the Lower (LG1 to LG7); Middle (MG0 to MG4); and Upper Groups (UG1 to UG3) (Schurmann et al., 1998). The thickness of individual chromitite layers ranges from a few centimetres to 2–3 m. Mining activities have allowed most of these layers to be traced across the entire Bushveld Complex (Schurmann et al., 1998; Hasch and Latypov, 2021) indicating that the process responsible for their formation has likely been working synchronously over lateral distances of up to 400 km to produce continuous, uniform blankets of chromitites (Kinnaid et al., 2002; Naldrett et al., 2012; Latypov et al., 2022a). Below we present a comprehensive set of field observations on magmatic dropstones from the sequences hosting UG1/UG2/MG2 chromitites in the Critical Zone (Fig. 3) that provide some strict constraints on the origin of these chromitites in the Bushveld Complex.

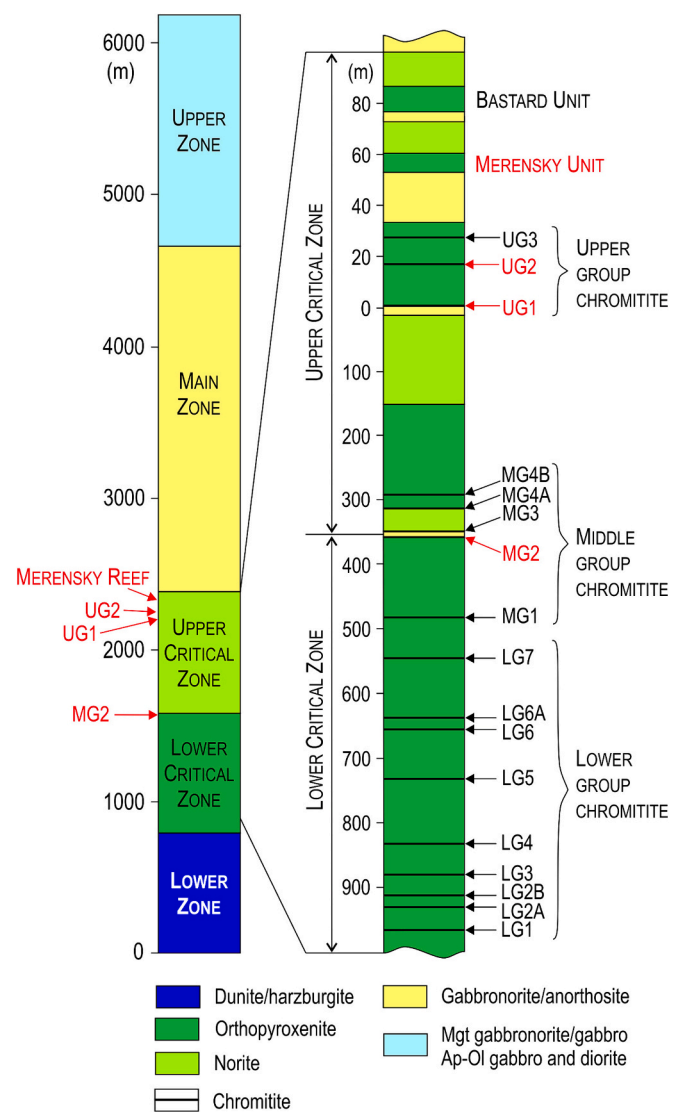


Fig. 3. Simplified stratigraphic section through the Bushveld Complex (Rustenburg Layered Suite) in South Africa and stratigraphic position of the Lower, Middle and Upper Group chromitites as well as the Merensky and Bastard Units in the Critical Zone. Also indicated by red scripts are objects (MG2, UG1, UG2, and Merensky Reef) involved in this study. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

4. General characteristics of magmatic dropstones

4.1. Spatial association of blocks with massive chromitite sequences

One of the first-order observations is that the fine-grained blocks (i.e., magmatic dropstones) appear to be not equally distributed throughout the floor cumulates of the Critical Zone (CZ) in the eastern Bushveld Complex but are rather confined to some specific horizons in the stratigraphy. They are mostly found in the rock sequences that host massive chromitites and are especially abundant in the UG1 sequence (Fig. 4). In one study area (Fig. 4), the UG1 sequence, exposed in a number of trenches, is composed of three chromitite sublayers (A, B, and C) with intervening layers of chromite-bearing norites and anorthosites/leuconorite. The UG1 sequence is underlain and overlain by orthopyroxenites, with or without leuconorite interlayers. A mapping of surface trenches reveals that blocks are closely associated with the UG1 sequence, with almost no such blocks in the underlying and overlying leuconorite and orthopyroxenite (Fig. 4). This observation may partly

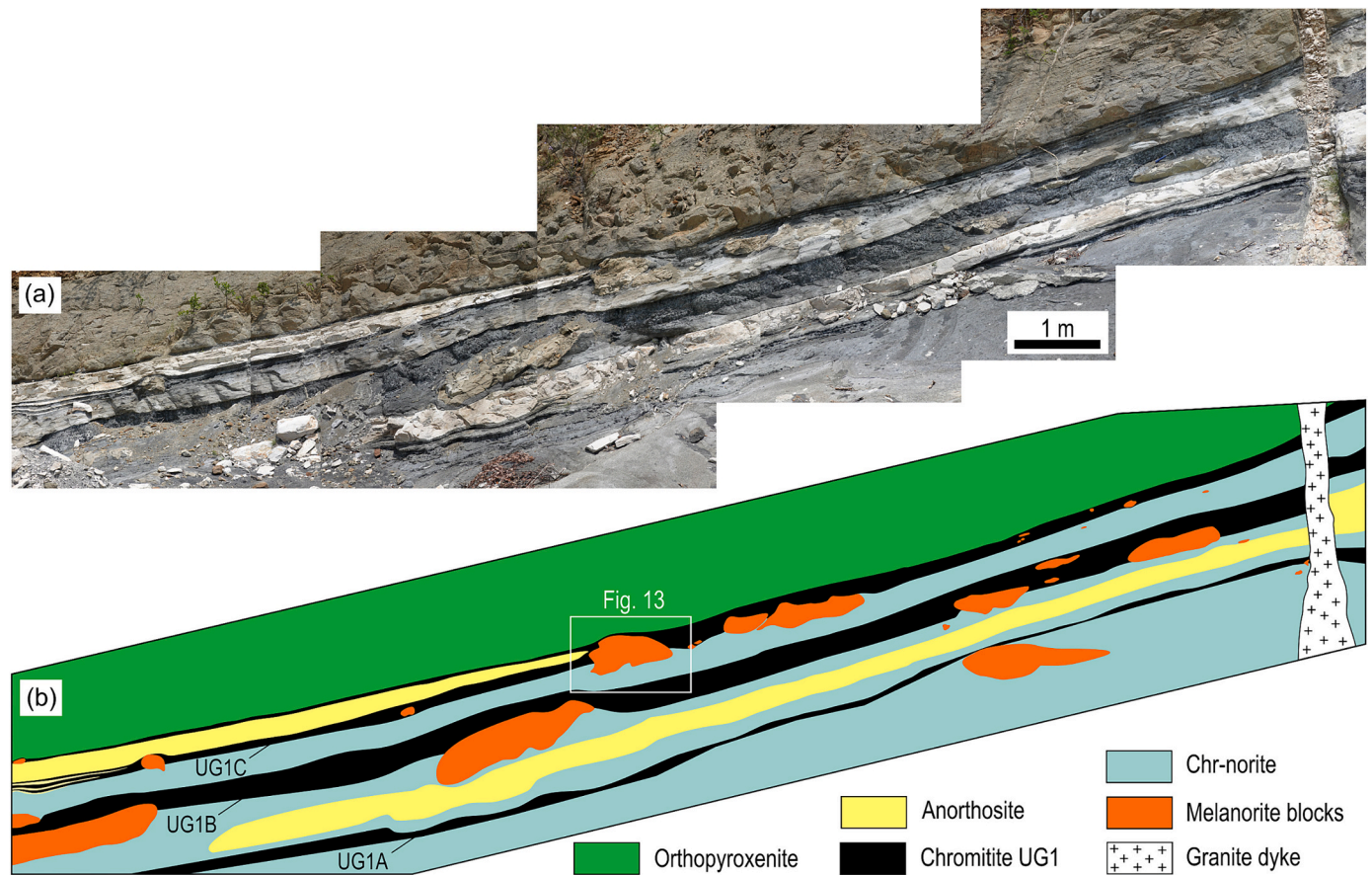


Fig. 4. Stitched photographs (a) and sketch (b) of a trench showing a section through the UG1 sequence. Twickenham section, Upper Critical Zone, eastern Bushveld Complex. In this area, the UG1 sequence comprises three chromitite sublayers (A, B and C) that are separated by layers of chromite-bearing norite and anorthosite. The sequence is both overlain and underlain (not shown here) by orthopyroxenite. A granitic dyke cuts the entire UG1 sequence and host orthopyroxenite on the right side of the exposure. Note that all melanoritic blocks (i.e., magmatic dropstones) are confined to the UG1 sequence and are absent from the overlying orthopyroxenite. The position of Fig. 13 is indicated by a white rectangular.

reflect the fact that dark-green blocks are much better visible on the contrasting background of black chromitite and white anorthosite/leuconorite rather than on the dark-green host orthopyroxenites. However, the search for fine-grained blocks in the exposures above and below the UG1 sequence have not met with success; only a few small blocks were found in host orthopyroxenite. We are therefore confident that the preferential occurrence of fine-grained blocks in the UG1 sequence is a solid observation that needs to be explained by any successful model for the formation of this sequence and associated massive chromitites. Similar blocks have been found in UG2, MG2 and LG6 sequences although they occur there at much lower abundance compared to the UG1 sequence. We think that such blocks may also be abundant in the Merensky Unit which can be technically referred as the UG4 sequence (Irvine and Sharpe, 1986). In this sequence, these blocks have been historically described as lenses of so-called ‘brown sugar norites’ (e.g., Leeb-Du Toit, 1986; Beukes et al., 2016). Other sequences hosting massive chromitites (e.g., LG1-LG5, MG1, MG3-MG4) need to be carefully re-examined in the future for the occurrence/absence of such blocks.

4.2. Shape, size and composition of blocks from sequences of massive chromitites

The dark-green blocks are easily identifiable in the field especially when they occur in the rocks of contrasting colour, texture, and mineral composition (e.g., anorthosite, norite, and chromitite). The blocks are much less visible when they are enclosed by dark green, medium- to

coarse-grained orthopyroxenites but are still recognizable owing to their fine-grained texture. The identity of the blocks is often emphasized by a few cm thick reaction rims that are developed along the margins of the blocks, especially when they are enclosed by massive chromitites (Fig. 5a). The rims are coarser grained and darker in colour (Fig. 5b) because they are richer in both orthopyroxene and chromite compared to internal parts of the blocks. In some cases, the rim-like material also occurs in the form of veins inside the blocks, apparently developing along some cracks/fractures in the blocks (see a description below and related Fig. 13). The blocks are composed of equant grains of orthopyroxene (60–70 vol%) cemented by interstitial plagioclase (20–25 vol%) and rarely clinopyroxene (10–15 vol%) (Fig. 6a) and are therefore referred here as melanoritic in composition. The fine-grained, non-cumulate texture of melanoritic blocks strongly contrasts them from all adjacent cumulus-looking, medium- to coarse-grained orthopyroxenites in the host sequences (Fig. 6b). In addition, the internal parts of the blocks may contain coarse-grained pods of irregular shape (Fig. 5) which are composed of plagioclase, orthopyroxene and occasionally clinopyroxene. Most medium- to large blocks are lenticular (convex) or tabular in shape, although the blocky and angular blocks are also common. In general, the angular blocks range in size from a few centimetres to more than one meter across whereas the tabular blocks may be up to ~0.5–1.0 m in height and up to ~10 m in length (Fig. 7). The long axis of most blocks is generally parallel to the direction of magmatic layering (Fig. 7). There are expectations that melanoritic blocks of much larger sizes may be found elsewhere in the sequence.

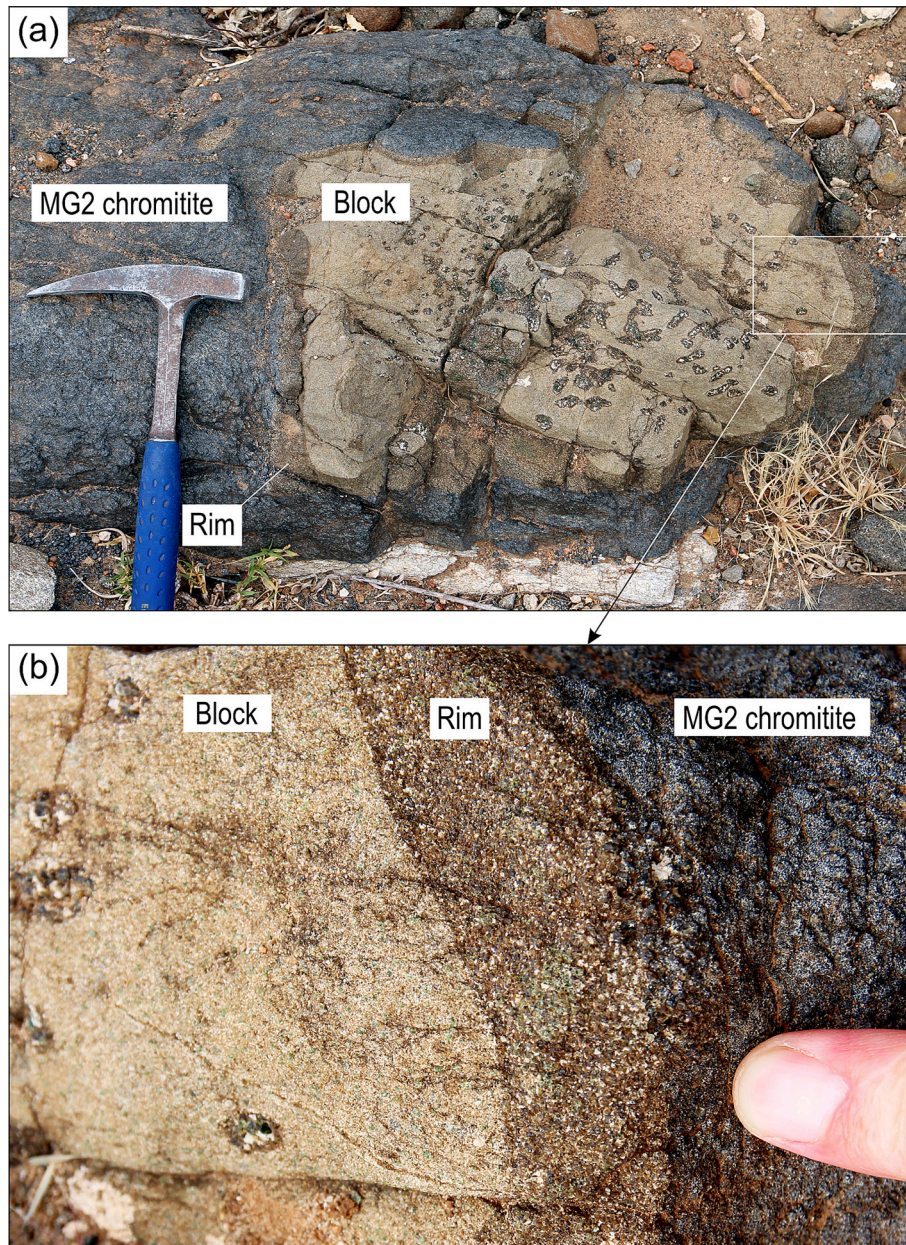


Fig. 5. Photographs of a melanorite block from the MG2 sequence. Cameron section, Upper Critical Zone, eastern Bushveld Complex. (a) a melanorite block enclosed by the MG2 chromitite. Note a rim around the entire block in contact with chromitite. (b) Close up view showing a rim that is darker and coarser grained compared to an internal part of the block. The internal part of the block contains coarse-grained pods consisting of plagioclase, orthopyroxene and occasionally clinopyroxene. Head of the hammer is 17 cm.

5. Field relationships of magmatic dropstones with impacted layered rocks

5.1. Preliminary notes on interpretation of field observations

Field relations in the block-and-layering structures are described/interpreted below keeping in mind that the falling blocks may be impacting the cumulate pile sub-vertically, obliquely or even sub-laterally (Irvine et al., 1998). Strong sub-vertical impacts may result in rupture, depression (i.e., deforming them downwards) and attenuation of mushy layers (i.e., making them thinner) beneath the blocks. In some cases, the underlying layers may escape depression and attenuation if they are already solid enough by the time of sub-vertical impacts. Such blocks commonly remain on top of the deformed layers of the chamber floor (as in Fig. 1b) and are subsequently covered by new layers (as in

Fig. 1c). Thus, these new overlying layers are normally younger than the blocks. It is important to realize, however, that this may not be true for the blocks that were able to penetrate into a mushy cumulate pile. After the penetration, these blocks may move through a cumulate pile for some time (sub-vertically or sub-laterally if impact was oblique) so that, when stopped, the blocks find themselves covered by the pre-existing layers (i.e., by older, not younger layers). In this process, the obliquely penetrating blocks may not only sharply rupture/truncate the adjacent layers but also push the overlying (older) layers outwards causing their upward bending. Importantly, the sub-laterally penetrating blocks may not impact the underlying layers so that they remain planar/non-deformed despite being located right beneath the blocks. Also, when such a block is located, for instance, within an anorthosite layer, it is evident that part of this layer (which is now occupied by the block) has been removed somewhere. A simple solution is that this missing part of

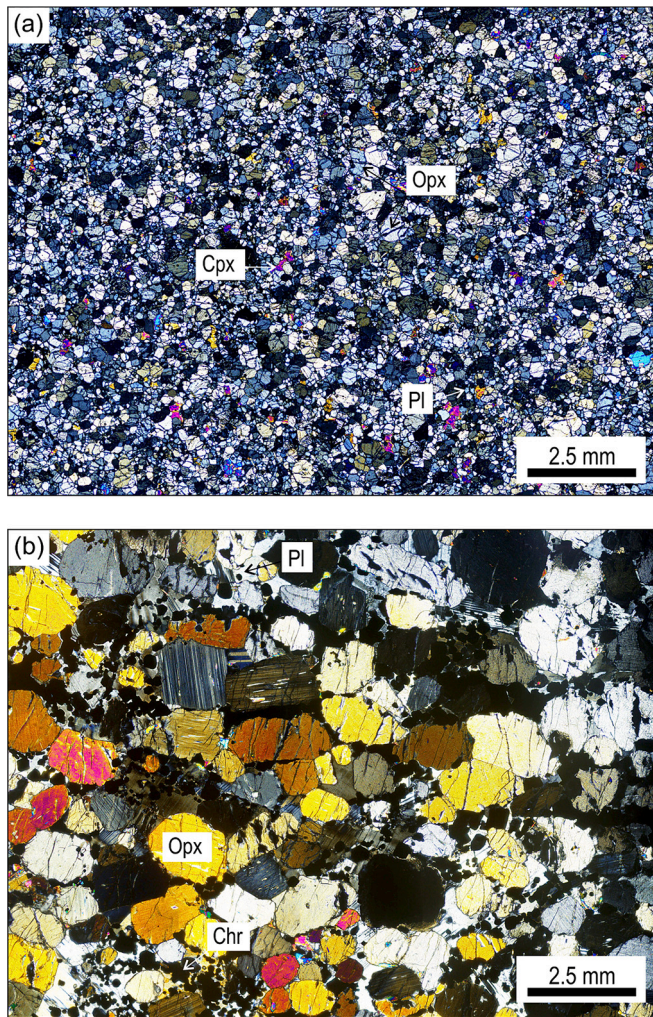


Fig. 6. Photomicrographs comparing the texture and mineralogy of melanorite blocks with host orthopyroxenites from the UG1 sequence. Upper Critical Zone, eastern Bushveld Complex. (a) fined-grained melanorite, sample TWK-6, Twickenham section; (b) coarse-grained chromite-bearing orthopyroxenite that overlies the UG1 chromitite, Maandaagshoek section. Both photomicrographs are in cross-polarized light. Abbreviations: Pl – plagioclase; Opx – orthopyroxene; Cpx – clinopyroxene, Chr – chromite.

the anorthosite layer has been pushed sub-laterally ahead of the moving block. Once again, the layers that overlie and are bent around such blocks predate the block and are lifted upwards by the laterally penetrating block. This lifting/pushing the overlying layers outwards is supported by the fact that they do not drape over the block but are rather folded/deformed.

5.2. Angular blocks that truncate underlying layering and lift overlying layering

Fig. 8a shows a medium-sized angular block of melanorite that occurs in the uppermost part of the UG1 chromitite, with its upper part extending upwards into the overlying orthopyroxenite. A thin layer of anorthosite (and likely part of chromitite immediately beneath it) is ruptured by the block; importantly, the edges of the layer are not depressed downwards (Fig. 8b, c), as would be expected if the layer was impacted by a vertically dropping block. Rather, the edges slightly concave upward, indicating that the impact was likely oblique, and involved not only a sharp truncation but also upwarding of older layers as the block knifed through the layers and pushed them outwards. The chromitite that overlies the block (Fig. 8a) can be technically interpreted

as a younger layer that drapes over the irregular morphology of the fallen block. But it seems more logical that this layer actually predates the block and was rather pushed upwards by the block that obliquely penetrated through the cumulate pile. In other words, the toe of the block could have “snowploughed” in and lifted the overlying chromitite layer. This interpretation is consistent with the fact that the overlying chromitite appears to be folded on the right side of the outcrop (Fig. 8c).

Fig. 9a illustrates another example of a medium-sized angular block of melanorite whose lower half sits within the UG1 chromitite while the upper half extends upwards into the overlying anorthosite. At the base of this anorthosite there is a thin sequence of interlayered anorthosite and chromitite which is ruptured by the block and slightly turned up at the edge of the block (Fig. 9b and c). Again, the relationships are not compatible with the vertical drop of the block and are rather indicative of the disruption of layers via the oblique impact of the block. The significance of chromitite on top of a block (Fig. 9a) and an anorthosite rim along the right margin of a block (Fig. 9c) to this story is not entirely clear.

Fig. 10 exhibits a lenticular block that sharply truncates the underlying sequence of a fine-scale anorthosite-chromitite layering. The layering is not depressed downwards, indicating that the block has likely impacted the layering obliquely rather than directly from above. The fact that the lowermost chromitite seam and underlying anorthosite are not deformed (i.e., depressed downwards) may also be due to the oblique nature of the impact and/or high rigidity of these layers by the time of the impact.

5.3. Angular blocks that indent igneous layering beneath them

Fig. 11a shows a small angular block (~20 cm in size) that indents a thin chromitite layer beneath it causing depression and attenuation of this layer. This block is covered by planar anorthosite and chromitite layers indicating no compaction of the overlying strata; the relationships are somewhat complicated by a reverse fault on the right side of the outcrop. Fig. 11b shows an angular block of a larger size (~1 m in size) that also indents an underlying chromitite layer causing its depression and substantial attenuation. The relationship of this block with the overlying strata is not clear because it consists of the non-layered massive anorthosite. In both cases, the anorthosites underlying the chromitite layers are probably also depressed beneath the blocks but this is not visible due to lack of clear markers in the anorthosites.

Fig. 12 illustrates the oval-like block that indents the underlying layered strata composed of thin interlayered chromitite and anorthosite. The deformation is particularly well evident from the chromitite layer that immediately underlies the block, and which indicates substantial depression and attenuation beneath the block. In addition, the underlying anorthosite layer A also shows a notable depression and almost three-fold decrease in its thickness beneath the block. Interestingly, the subsequent anorthosite layers B and C are also depressed but not attenuated beneath the block; they maintain nearly the same thickness throughout the entire outcrop. This likely suggests that at the time of impact the anorthosite layer A was still able to bend and flow whereas the layers B and C were only able to bend. The block is covered with the overlying strata that is composed of planar layers of chromitites, leuconorite and anorthosite, again indicating no compaction. It is notable that the first planar layer of chromitite (1–2 cm thick) occurs already at the level corresponding to the top of this block.

Fig. 13 depicts a large angular block that indents the underlying chromite-rich norites when landing from right to left. The impact is particularly evident from the folding and depression of the faint layering at the left bottom corner of the block. On the left side, the faint layering at the top of the chromite-rich norite sharply abuts the subvertical to overturned edge of the block, whereas, on the right side, the same layering drapes over the gently sloping top of the block. This suggests that the chromite-rich norite formed after the block's landing on the chamber floor and was able to deposit on the gentle slope but not on the

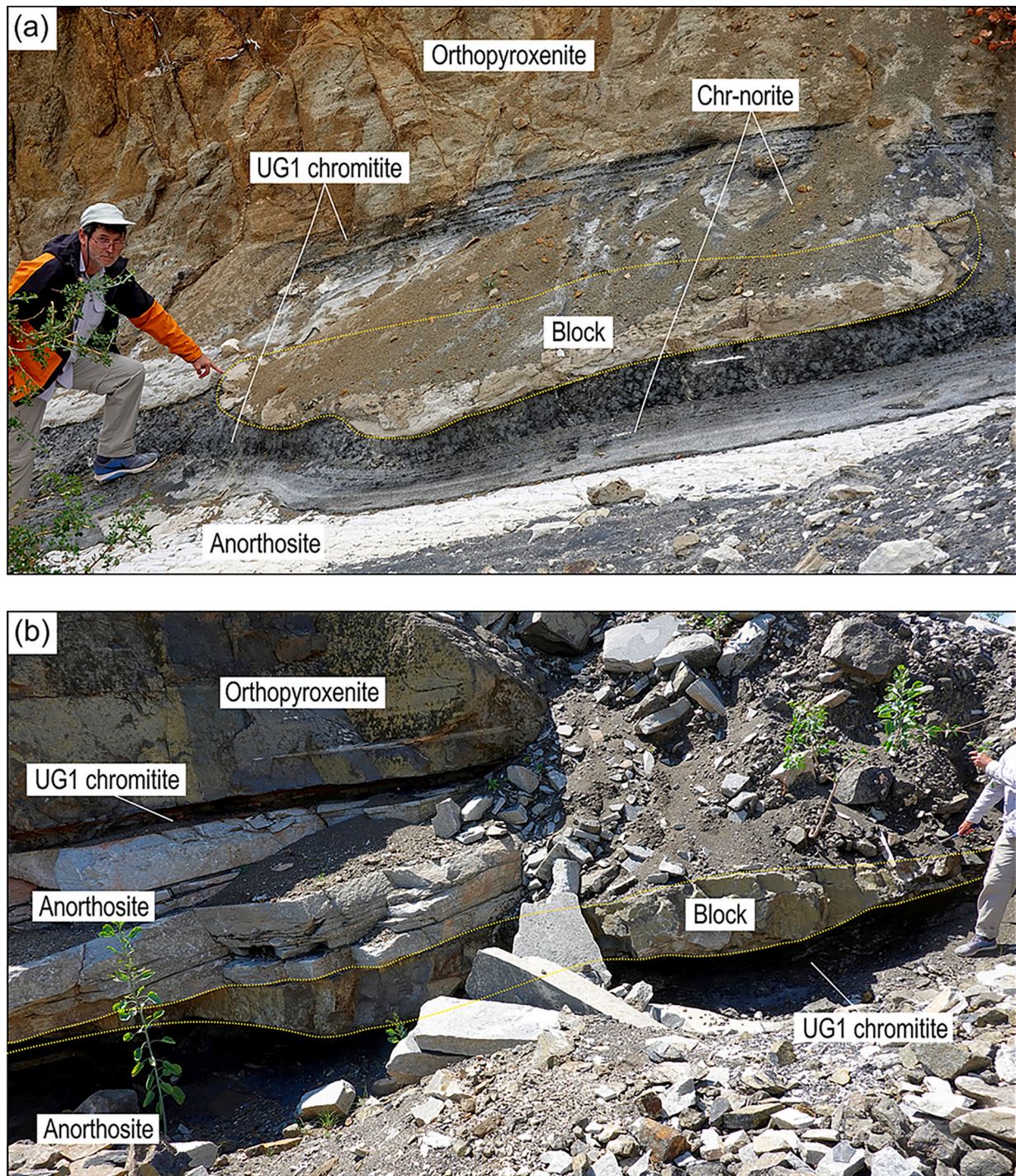


Fig. 7. Photographs of the largest tabular melanorite blocks from the UG1 sequence. Upper Critical Zone, eastern Bushveld Complex. (a) a tabular block in the Twickenham section. (b) a tabular block in the Paschas Kraal section. A person for scale in both photographs.

subvertical to overturned edge of the block (due to the high angle of repose). The faint layers in chromite-rich norite and overlying chromitite itself become thinner and are smoothed over the block. Reaction rims are developed along much of the block's periphery (Fig. 13b-d), including its interior where they look like veins that develop along the fractures.

5.4. Draping of layering over the blocks with or without depression of underlying layering

Fig. 14a illustrates a melanorite block that occurs entirely within the

upper part of the UG1 chromitite. The basal contact of the chromitite layer is planar (non-deformed) (Fig. 14b) whereas its upper contact is notably bent over the block's top. There is some indistinct layering within the chromitite itself (as indicated by a thin anorthosite layer) but physical relations of this layering with the block are not clear. A few overlying layers of anorthosite and chromitites are draped over the top of the block with no changes in their thickness. Again, this is likely indicative of deposition of these layers on the fallen block under conditions when crystals accumulating on the slopes of the 'hill' were gravitationally stable.

Fig. 15 exhibits two sections through the same block as it has been

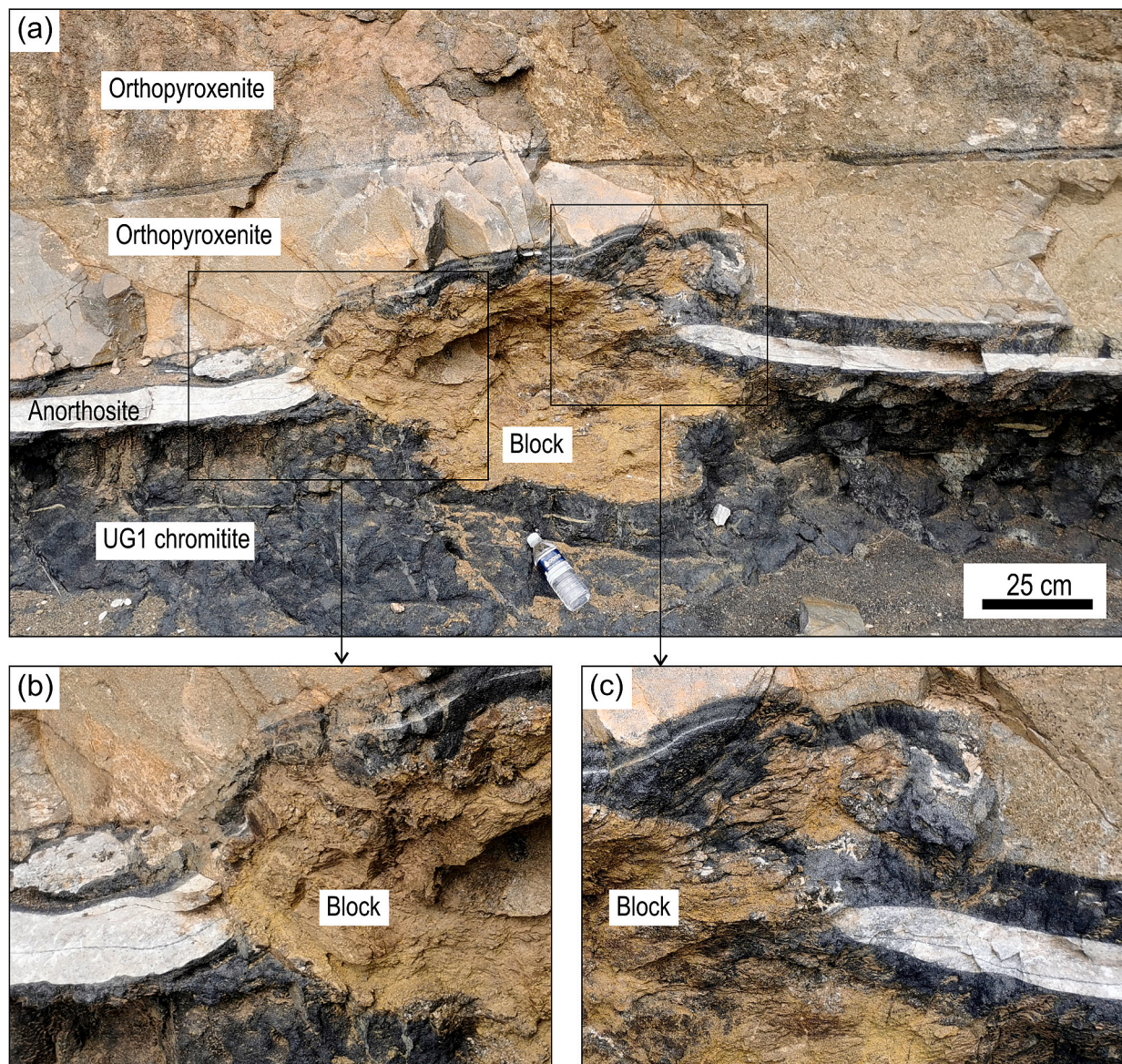


Fig. 8. Photographs of an angular melanorite block that impacts igneous layering of the UG1 sequence. Twickenham section, Upper Critical Zone, eastern Bushveld Complex. (a) A block ruptures a thin layer of anorthosite at the top of the UG1 chromitite; (b) Close up view of the edges of the anorthosite layer that slightly convex upward; (c) Close up of the folding of the chromitite layer that overlies the block.

exposed in the course of its progressive excavation during mining. The block is entirely enclosed within the upper part of a thick layer of the UG1 chromitite, which, unlike many other cases, lacks any associated anorthosite layers. The first (earlier) section (Fig. 15a) reveals that the uppermost part of the chromitite layer drapes over the top of the block. The physical relationships with the underlying part of the chromitite layer are not clear in this case due to lack of any markers. However, the second (later) section (Fig. 15b) has exposed a thin layer of melanorite/orthopyroxenite beneath the block that is clearly depressed relative to its original position. The uppermost part of the chromitite layer persists in draping over the top of the block in this section. The manner in which the layering is deformed above and below the block indicates that the block has likely impacted the cumulate pile sub-vertically from above.

Fig. 16a portrays a tabular block that landed at the very top of the UG1 chromitite and has been draped across by a thin layer of chromitite, resulting in a local increase in the overall thickness of the UG1 chromitite. Note that this block has a few cm thick reaction rim developed along its entire periphery. Fig. 16b-e shows four examples of relatively small rounded to lenticular blocks that are encapsulated from above and

below by two thin chromitite layers. The total thickness of a single chromitite layer away from the blocks appears to be equal to the sum of the thickness of these two smaller layers. A general interpretation of these relationships is that the lower chromitite layers are depressed beneath the blocks because of subvertical impacts whereas the upper chromitite layers are bent due to draping over the tops of the fallen blocks. It is conceivable that the original relationships have been somewhat accentuated by subsequent compaction of the cumulate pile.

5.5. Blocks truncated by a planar erosional surface

Fig. 17 shows an example of an erosional remnant of a block that was originally mantled by a thin chromitite rim. The rimmed block and its host anorthosites have been truncated from above by a planar erosional surface that has been subsequently covered by a chromitite layer. The erosion of the block is evident from an abrupt truncation of a chromitite rim around the block because it is separated from the overlying chromitite layer by a few mm thick anorthosite layer (Fig. 17b).

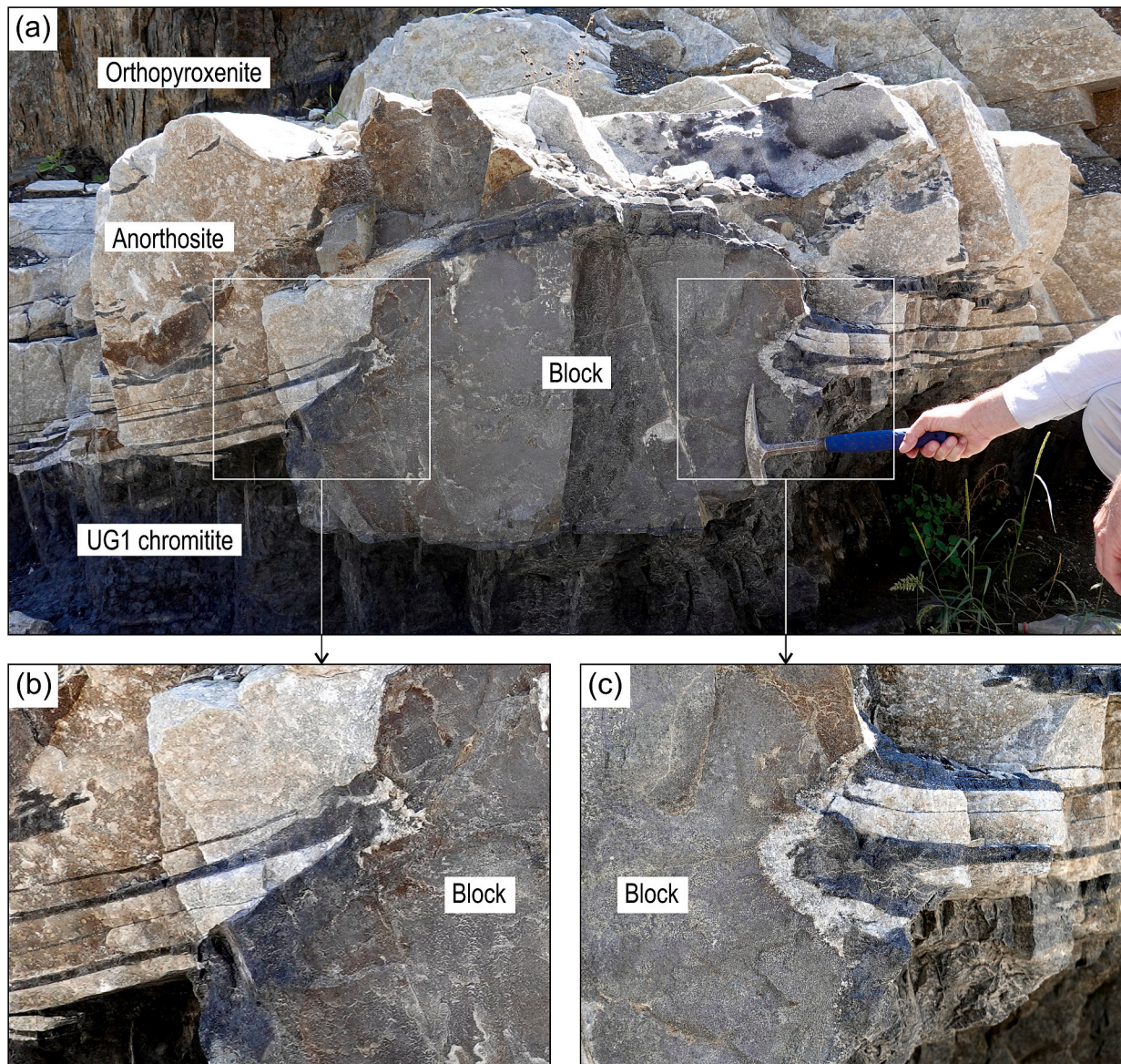


Fig. 9. Photographs of an angular melanorite block that impacts igneous layering of the UG1 sequence. Outcrop PA-14, Paschas Kraal section, Upper Critical Zone, eastern Bushveld Complex. (a) A block ruptures a thin sequence of interlayered anorthosite and chromitite that overlies the UG1 chromitite; (b-c) Close up views of interlayered anorthosite and chromitite that are slightly turned up at the edge of the block. Head of the hammer is 17 cm.

6. Implications for the architecture and internal dynamic processes in the Bushveld magma chamber

The field relations from block-and-layering structures of the CZ of the Bushveld Complex are in most respects similar to those described by Neil Irvine and his co-workers in such structures from the Skaergaard intrusion (Irvine et al., 1998) as well as to such structures documented by James Scoates in the layered series of the Poe Mountain anorthosite (Scoates, 2000). For this reason, our general inferences from the field observations about magma chamber processes are in line with those from the mentioned studies. The conclusions pertain to three principal parts of the evolving Bushveld magma chamber, namely (1) a roof sequence of cumulates, (2) a floor sequence of cumulates, and (3) an internal magma reservoir (Fig. 18).

6.1. Where do blocks come from?

There are three potential sources of the blocks that should be examined here. The blocks may represent: (1) fragments of eroded/

disrupted layers from the pre-existing cumulate stratigraphy, (2) fragments of rocks that formed in staging chamber/feeder conduits and were brought into the chamber with the ascending magmas and (3) fragments of rocks crystallized along the chamber roof and dislodged and collapsed on the chamber floor.

6.1.1. Fragments of the pre-existing layers from the cumulate stratigraphy

Fragments of the pre-existing layers from the cumulate stratigraphy are quite common in the CZ. For instance, lenses of orthopyroxenite of various sizes commonly occur in massive chromitites but are interpreted as in situ erosional remnants of the previously continuous layers (Latypov et al., 2017b). Unlike the blocks in question, these erosional remnants are cumulus-type, medium- to coarse-grained in texture and do not show any deformational impacts on layering beneath them. This option can therefore be excluded from further considerations. There are also angular to rounded fragments of various rocks (e.g., anorthosite, norites, orthopyroxenite) that occur in some massive chromitites, particularly in the UG1 (Pebane and Latypov, 2017) and MG2 (e.g., Maier et al., 2013). These fragments are likely derived from disruption

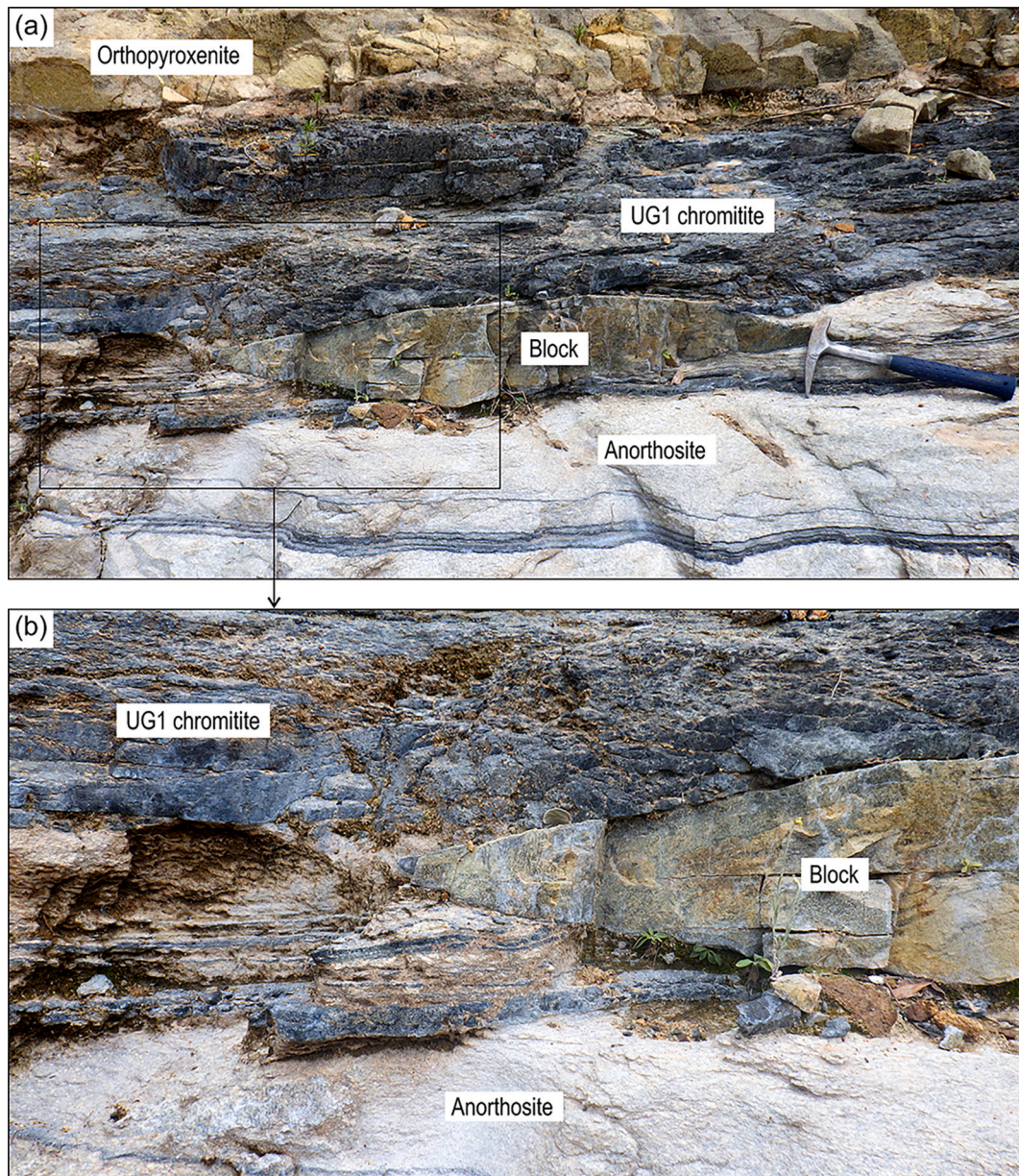


Fig. 10. Photographs of a lenticular melanorite block that impacts igneous layering of the UG1 sequence. Maandaagshoek section, Upper Critical Zone, eastern Bushveld Complex. (a) A block sharply truncates the underlying layering of anorthosite and chromitite. The underlying layering is not depressed downwards. (b) Close up view of the anorthosite-chromitite layering truncated by a block. Hand of the hammer is 30 cm.

of floor cumulates and their transportation/dragging along the chamber floor by basal flows of magmas replenishing the chambers (Latypov et al., 2024a). The fragments are also of cumulus-type, medium- to coarse-grained in texture and show field evidence for the lateral transportation (preferred elongation parallel to the igneous layering; deflection of layering around the fragments, etc.), rather than the subvertical/oblique impacting of layers from above as is commonly the case with magmatic dropstones (e.g., Irvine et al., 1998). This option can, therefore, also be ruled out. Notably, the fine-grained and non-cumulate texture observed in melanoritic blocks (Fig. 6a) is not characteristic of any cumulate rocks in the entire sequence of the CZ. This indicates that the blocks cannot be derived from the local stratigraphy, e.g., as parts of some overlying layers that were broken and settle into the underlying crystal mush. The UG1 sequence is overlain by ~10 m thick cumulus-looking, medium- to coarse-grained orthopyroxenites (Fig. 6b) that are texturally very distinct from the fine-grained melanorites. In addition, the fact that the blocks are transformed into coarser-grained variety

along the reaction rims (Fig. 5b) indicates that the blocks were already fine-grained upon arrival into their current place. This means that the blocks acquired their fine-grained texture at the place of their origin.

6.1.2. Fragments of cumulate rocks from a deeper staging chamber

The blocks may have been ripped up and entrained from an underlying staging chamber with ascending magmas. Such an interpretation has been recently proposed for rectangular blocks of noritic rocks up to ~6 m in size that are found in the mottled anorthosite sequence of the Main Zone in the eastern Bushveld Complex (Bourdeau et al., 2022). Numerical calculations show that such large blocks cannot be brought into the chamber with crystal-free magmas because this would require unrealistically high magma velocities (Bourdeau et al., 2022). Therefore, it has been postulated that the blocks have been entrained into the Bushveld chamber with plagioclase-rich slurries (with 5–30 vol% of plagioclase) derived from crustal staging chambers. A key piece of evidence for the emplacement of plagioclase-rich slurries is the pervasive,

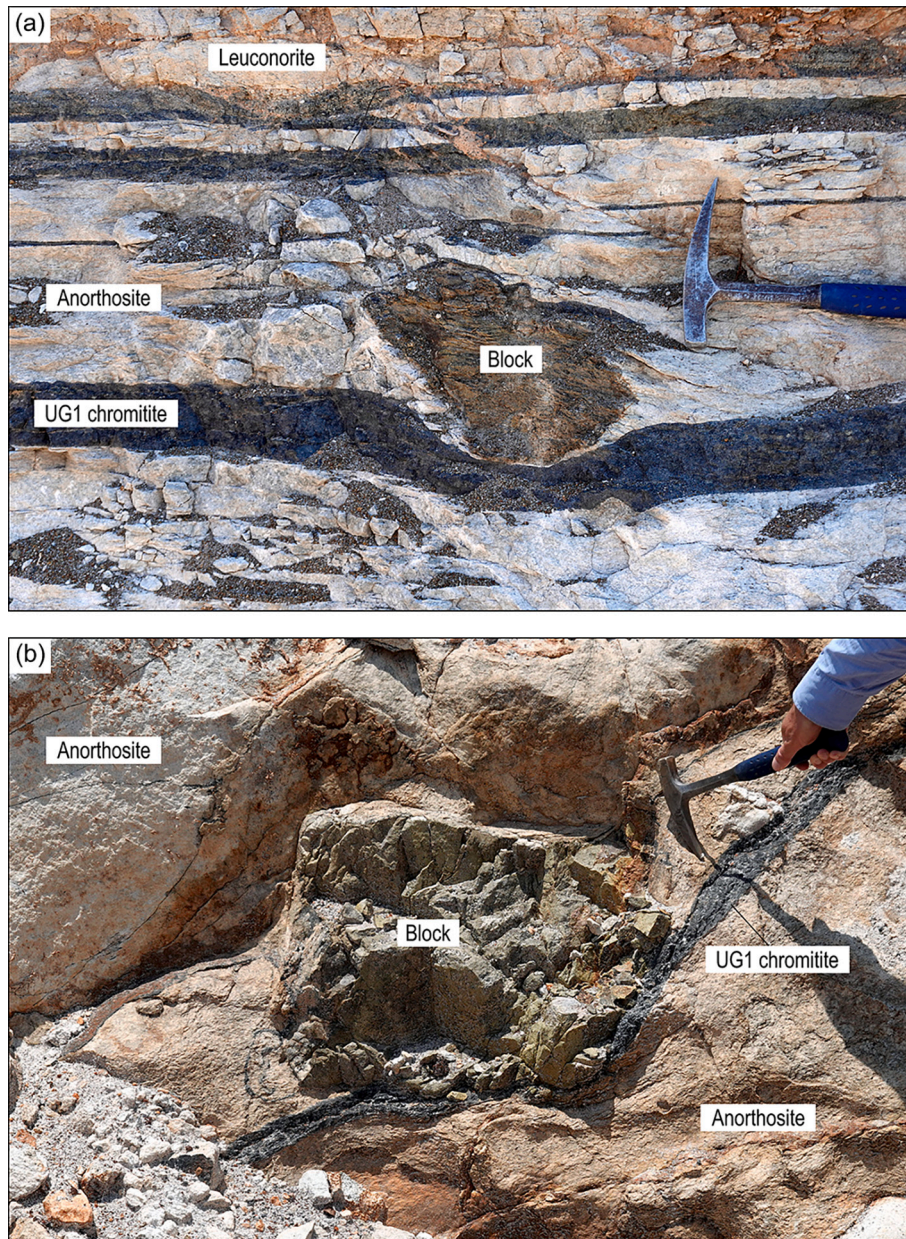


Fig. 11. Photographs of two melanorite blocks that indent igneous layering of the UG1 sequence. Upper Critical Zone, eastern Bushveld Complex. (a) A small block that depresses the underlying chromitite layer in the Jagdlust section. (b) A larger block that depresses the underlying chromitite layer in the Maandaagshoek section. Outcrop MA-12 A. Head of the hammer is 17 cm.

moderate to strong foliation of plagioclase in mottled anorthosites hosting the noritic blocks. This foliation is attributed to magmatic flow of plagioclase-rich slurries upon their arrival into the chamber (Bourdeau et al., 2020). The interpretation makes perfect sense because nearly all igneous rocks that are produced from flowing crystal-rich mushes – be it stratiform or massive anorthosites (e.g., Ashwal, 1993), gabbroic rocks in layered intrusions (e.g., Vukmanovic et al., 2018), or granitic rocks (e.g., Vernon, 2000) – display various degree of alignment (e.g., lamination and/or lineation) of plagioclase and other minerals reflecting passive rotation of elongated crystals during magmatic flows (Vernon, 2000).

However, the above explanation fails to appreciate one important textural feature of mottled anorthosites: the plagioclase foliation is present only in their matrix but is absent from the pyroxene oikocrysts (i. e., mottles) occupying up to 30–50 vol% of these rocks. These oikocrysts enclose numerous plagioclase chadacrysts that are randomly oriented,

show a wide range of sizes and reveal irregular grain boundaries that are generally not parallel to expected growth faces (Bourdeau et al., 2020; their Fig. 5b and 8a-b). These are typical features of a texture that results from plagioclase nucleation and growth from crystal-free melts in a boundary layer on the chamber floor (McBirney and Noyes, 1979; Higgins, 1991; McBirney and Hunter, 1995; Marsh, 2006; Holness et al., 2020). Oikocrysts in cumulate rocks are generally thought to capture and preserve the texture of the early mushy layer (Mathison, 1987; Barnes et al., 2016, 2021; Latypov et al., 2020). Pyroxene oikocrysts in mottled anorthosites can therefore be regarded as snapshots of the initial in situ growth that produced an open framework of randomly oriented plagioclase grains. The slight to strong plagioclase lamination observed in the matrix between the pyroxene oikocrysts and deflecting around these oikocrysts (Bourdeau et al., 2020; their Fig. 5b-d) is produced after the oikocrysts fossilized this primary texture. The plagioclase lamination has likely resulted either from compaction (Meurer and Boudreau,

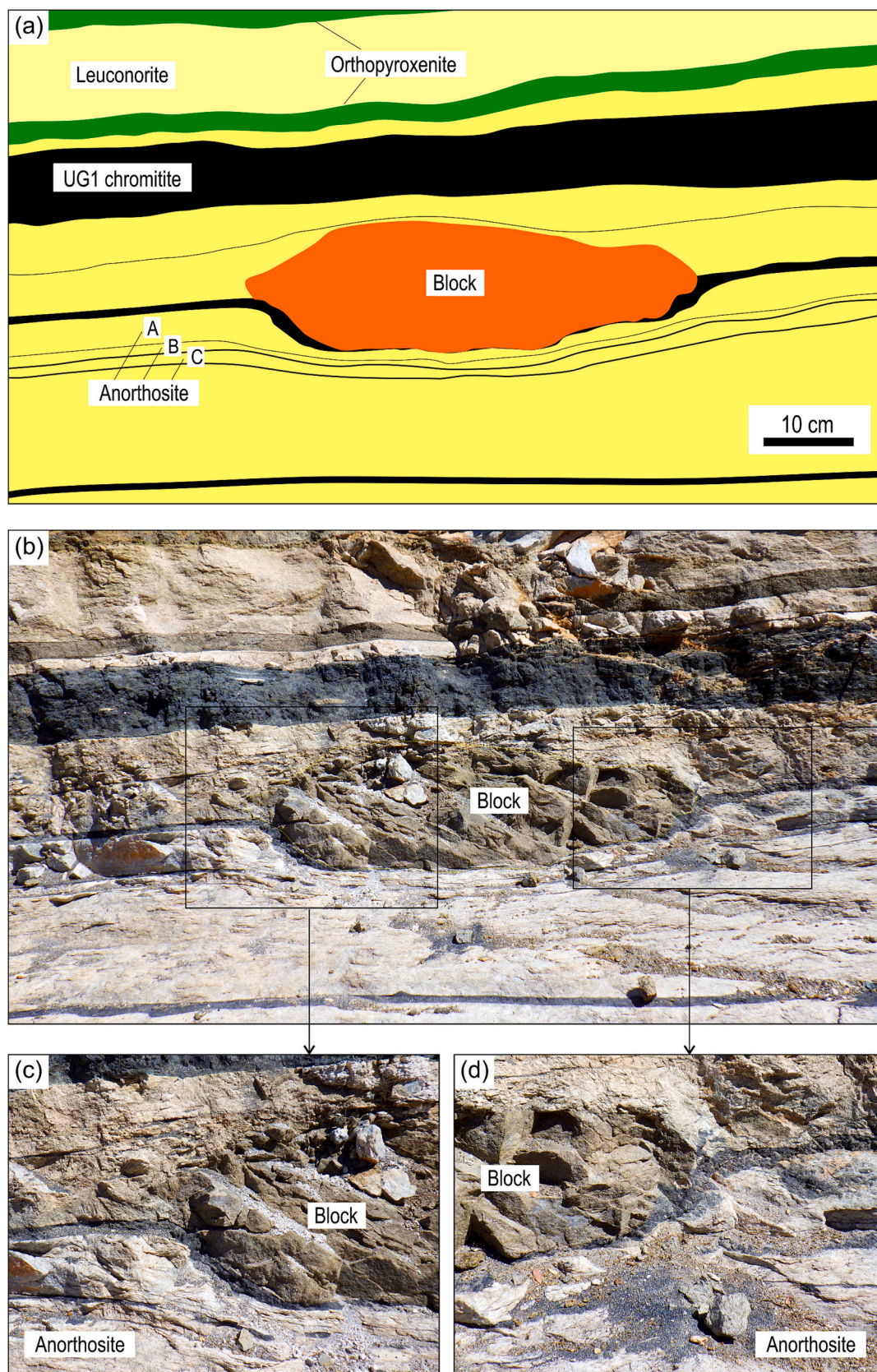


Fig. 12. Sketch (a) and photographs (b-d) of a melanorite block that indents igneous layering of the UG1 sequence. Jagdlust section, Upper Critical Zone, eastern Bushveld Complex. Note that igneous layering that underlies the block is depressed whereas the layering that overlies it is almost planar (i.e., non-deformed). The significance of anorthosite layers (A, B and C) is discussed in the text.

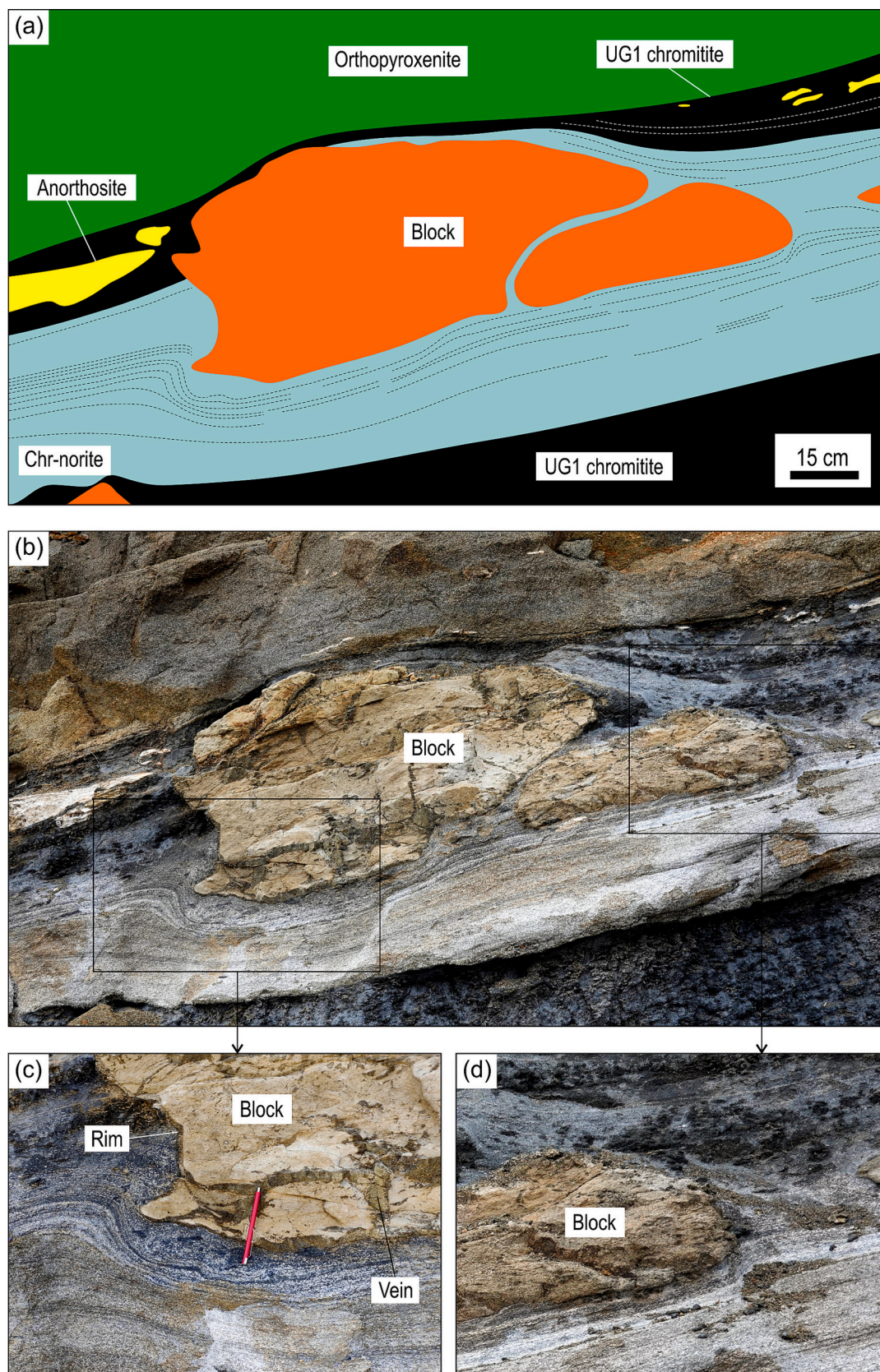


Fig. 13. Sketch (a) and photographs (b-d) of a large melanorite block that indents igneous layering of the UG1 sequence. Twickenham section, Upper Critical Zone, eastern Bushveld Complex. The impact occurred from right to left as evident from the folding and depression of the faint layering at the left bottom corner of the block. Pen for scale is 13 cm long. A position of this outcrop in the stratigraphy of the UG1 sequence is indicated in Fig. 4.

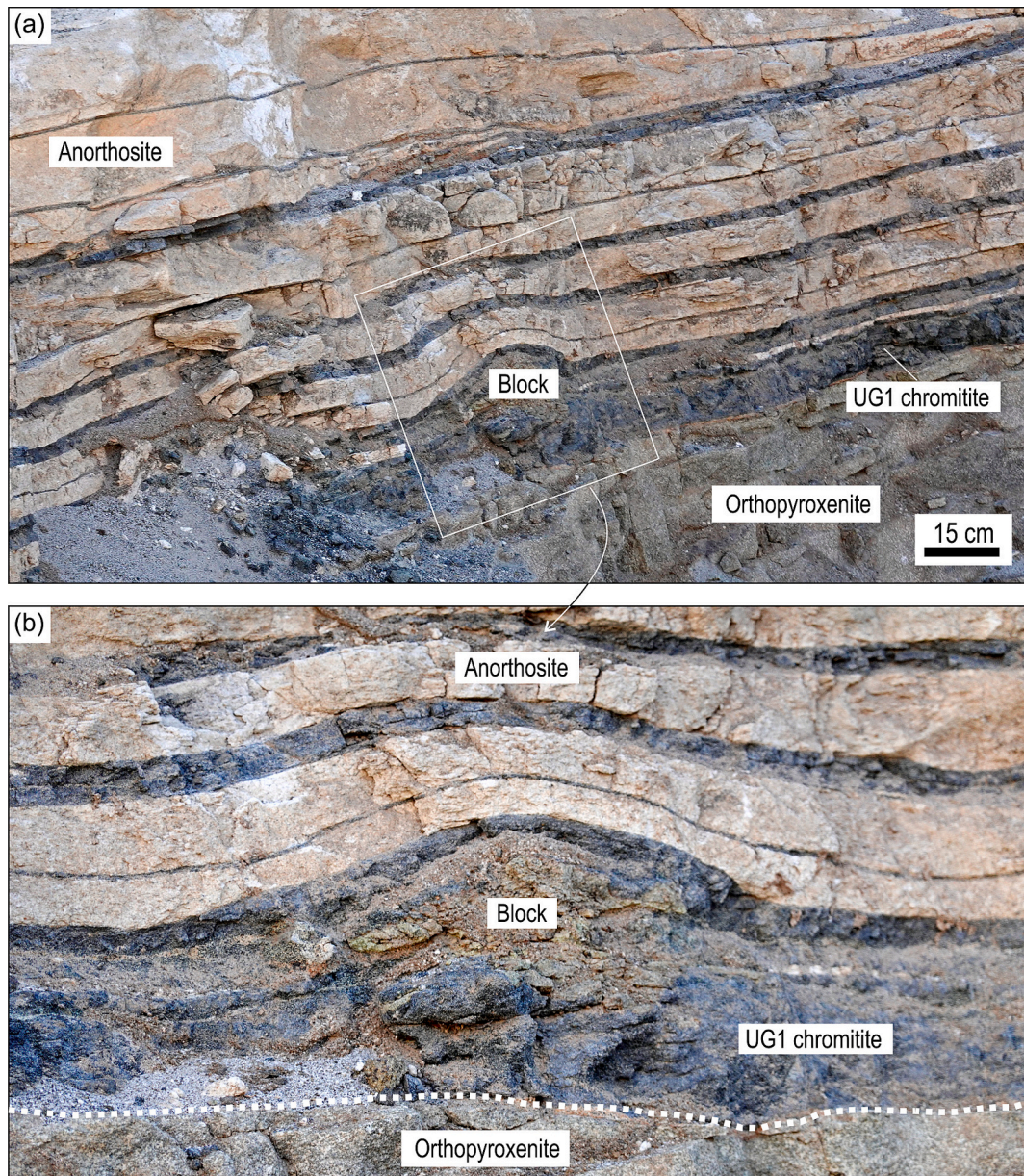


Fig. 14. Photographs (a-b) of a melanorite block that is draped over by igneous layering in the UG1 sequence. Jagdlust section, Upper Critical Zone, eastern Bushveld Complex. Note that the base of the UG1 chromitite (b) is planar (not depressed).

1998a, 1998b) and/or pure shear (Higgins, 1991) of a mushy matrix of poorly-consolidated anorthosites. Central to our discussion is that the above textural features of plagioclase crystals in pyroxene oikocrysts indicate that mottled anorthosites formed from crystal-free magmas. The absence of crystals in these magmas prevents them from transporting large blocks of noritic rocks from a deeper staging chamber (Bourdeau et al., 2022).

It is worthwhile to stress that a whole idea about the transportation of rock fragments from a deeper staging chamber is not consistent with the shape and size of the mafic blocks observed in the Bushveld Complex. Such fragments – usually referred to as nodules or crystalline enclaves in the literature – are common in association with basaltic scoria in all major tectonic environments (e.g., ocean ridges, hotspots and subduction zones). As a rule, the nodules are ovoid because they are subjected to breakage, disaggregation, and erosion during the long transportation in the ascending magmas (Sparks et al., 1977; Russell et al., 2019; Russell and Jones, 2023). In contrast, all medium- to large-size blocks from CZ (Figs. 7, 10) and MZ (Bourdeau et al., 2022) of the

Bushveld Complex are lenticular, rectangular or tabular; to the best of our knowledge, such a non-ovoid nodules have never been described from magmatic complexes. In addition, the nodules are commonly not larger than 10–20 cm in diameter, with maximum sizes reaching 80 cm (Sparks et al., 1977; Fodor and Vandermeijden, 1988; Shamberger and Hammer, 2006; O'Reilly and Griffin, 2010; Russell et al., 2019; Russell and Jones, 2023). Even the largest nodules are thus nowhere near to the size of tabular blocks (up to ~10 m long; Fig. 7) from our study and, therefore, the scenario of the block's transportation from a deeper staging chamber should be questioned. Whether magmas replenishing the Bushveld chamber were emplaced as basal flows into the chamber itself (Campbell and Murck, 1993; Naldrett et al., 2012; Latypov et al., 2017b) or as sills into the solidified cumulate pile (Mungall et al., 2016; Mitchell et al., 2019; Scoates et al., 2021), it seems highly unlikely that these magmas may have been able to transport mafic blocks of such shapes and sizes.

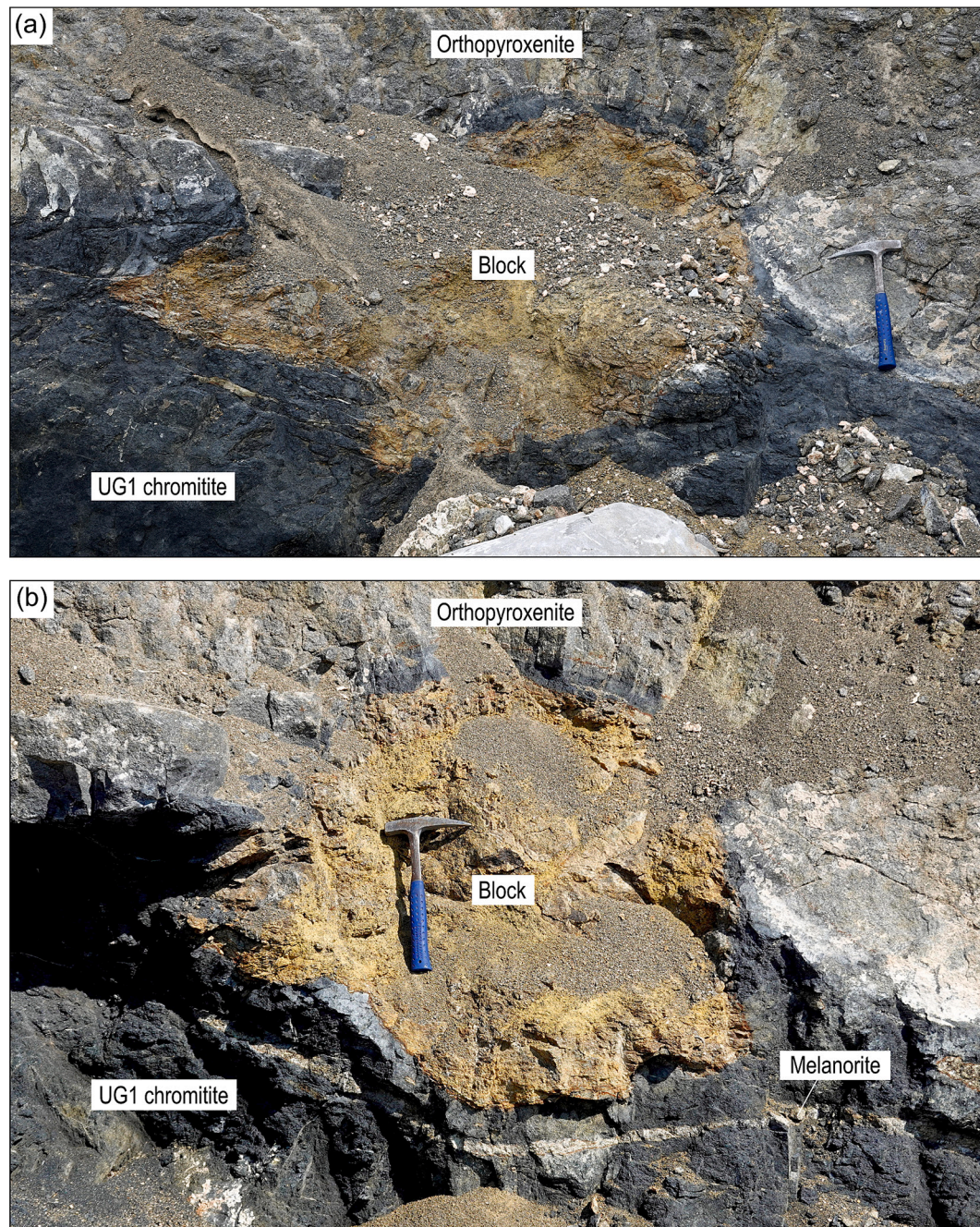


Fig. 15. Photographs of the same melanorite block (a-b) during its progressive excavation in the UG1 sequence. Cameron section, Upper Critical Zone, eastern Bushveld Complex. The uppermost part of the chromitite layer drapes over the top of the block in both sections (a and b). In addition, a section (b) exposes a thin melanorite layer that is depressed beneath the block. Hand of the hammer is 30 cm.

6.1.3. Fragments from a roof rock sequence of the Bushveld chamber

This last option appears to be most attractive because it may ensure the derivation of the blocks (i.e., magmatic dropstones) of any size and shape from the broken roof sequence. There is, however, one intriguing problem here. Like all magmatic bodies in Earth's crust, the Bushveld chamber is supposed to have been mostly cooled through the cold overlying rocks (Irvine, 1970; Frenkel' et al., 1988; Morse, 1988), and, if so, the unidirectional crystallization of cumulates from the roof downwards is almost unavoidable. However, the Bushveld Complex shows no such cumulates (Eales and Cawthorn, 1996; Cawthorn, 2015), a problem that has never been rigorously addressed. At issue is thus what has happened with the initial roof sequence of the Bushveld chamber? A simplest answer is that the roof cumulates formed but were later

destroyed and collapsed as blocks on the floor rock sequence (Latypov and Chistyakova, 2024a). A partial destruction of the roof sequence has been documented in the Skaergaard intrusion (Irvine et al., 1998), and a nearly complete destruction of such a sequence has been recently reported for the Sudbury Complex (Latypov et al., 2019; Chistyakova and Latypov, 2022) and has been earlier envisaged for some other layered intrusions (Hess, 1960; Mutanen, 2005; Nebel et al., 2013). We therefore suggest that the roof sequence of cumulates in the Bushveld Complex is likely absent due to its breakdown and collapse onto the inward-growing chamber floor (Fig. 18). If so, the fragments of the destroyed roof sequence must be scattered among the rocks of the floor sequence and it is therefore logical to conclude that the blocks in this and previous studies (Bourdeau et al., 2020, 2022, 2023) are these long-missing

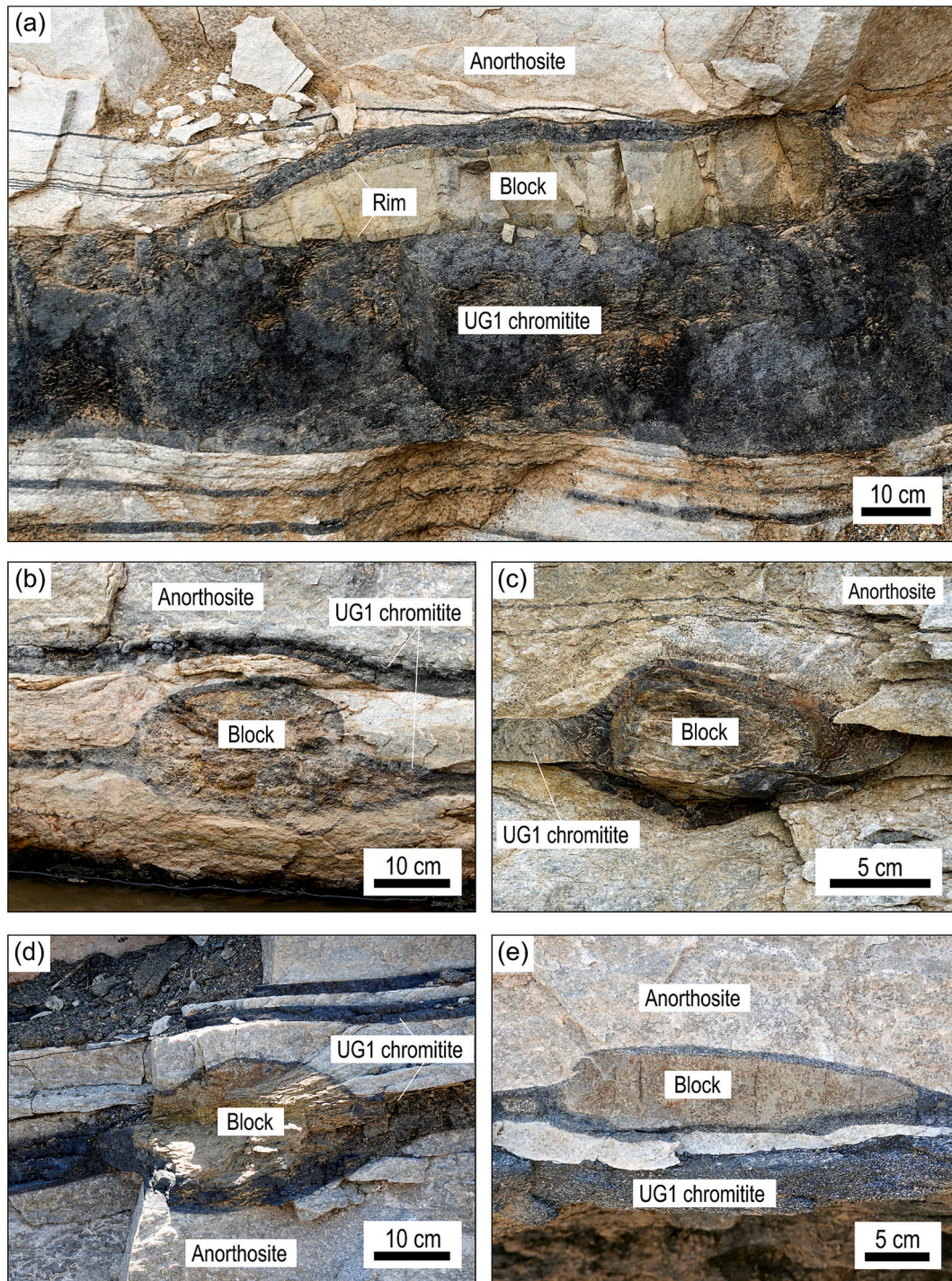


Fig. 16. Photographs of melanorite blocks enclosed in chromitite layers of the UG1 sequence. Upper Critical Zone, eastern Bushveld Complex. Underlying chromitite layers are depressed beneath the blocks (apart from a block in (a)) and overlying chromitite layers are draping over the tops of the blocks. (a, c) – Maandaagshoek section; (b) – Jagdlust section; (d, e) – Paschas Kraal section.

fragments from the destroyed roof sequence of the Bushveld Complex.

The fine-grained texture of the melanoritic blocks (Fig. 6a) in our study remains unclear at this stage. One possibility is that the blocks are fine-grained because of their crystallization under strong supercooling that is common along the roof zone of magma chambers (e.g., in response to chilling effect of cold country rocks or magma degassing). It should be mentioned that due to local variations in the extent of the roof disruption (from partial to complete; Fig. 18), the blocks can be derived at any particular time from any stratigraphic level in the roof sequence (i.e., not only from its lowermost part). This means that the blocks are

not expected to be compositionally similar to the floor rocks on which they have landed. For instance, the blocks sitting in the CZ may be well derived from the contact zone of the Bushveld chamber with the roof country rocks. Alternatively, the fine-grained texture of melanorite may be related to some not yet specified process of re-crystallization of the originally cumulus-textured medium- to coarse-grained rocks that happened when these rocks were still at the roof (i.e., before being dislodged). Mineralogical, geochemical and isotopic studies of these blocks are currently under way and may help shedding light on the nature of melanorite blocks.

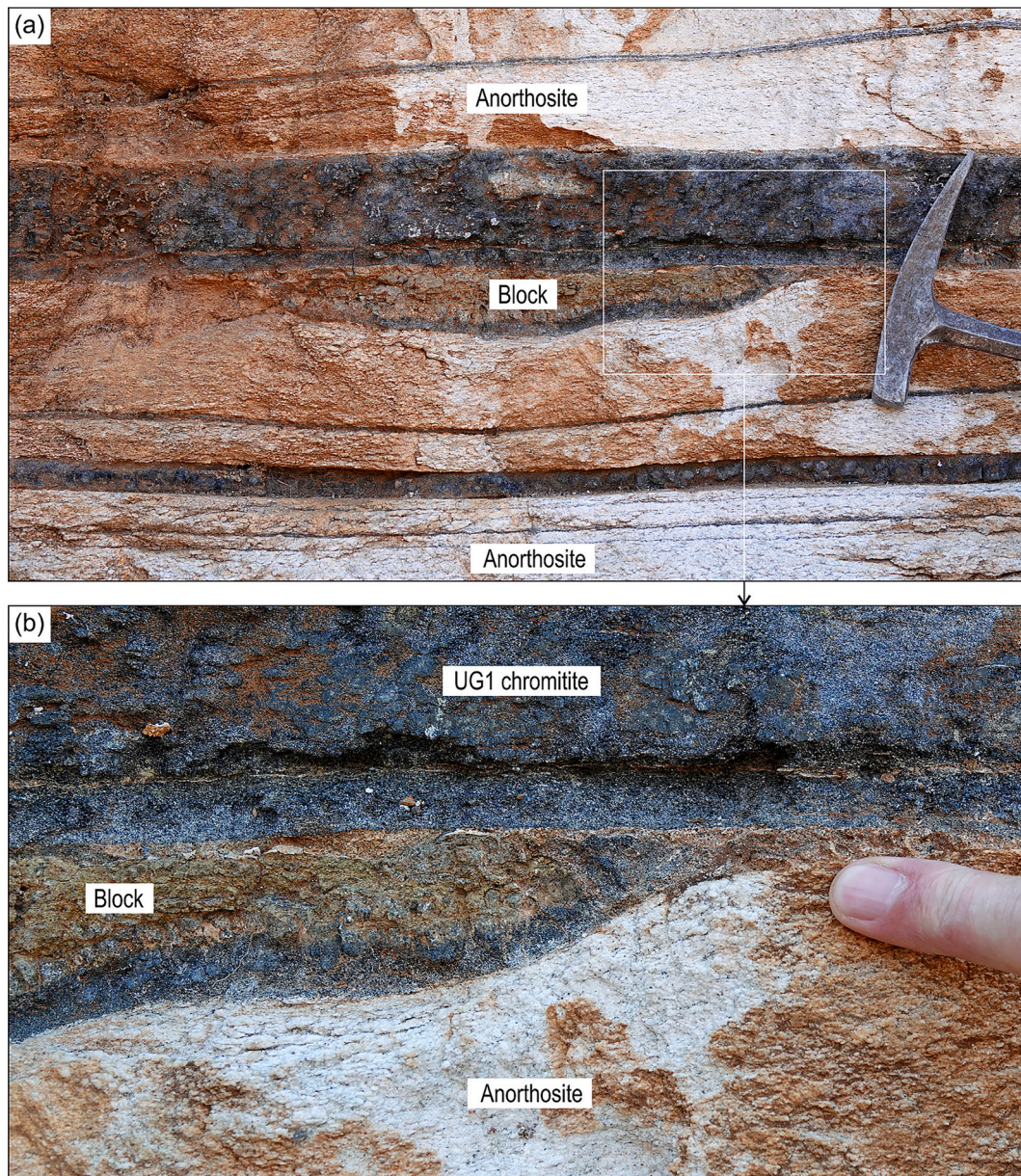


Fig. 17. Photographs (a-b) of the eroded melanorite block from the UG1 sequence. Maandaagshoek section, Upper Critical Zone, eastern Bushveld Complex. The block has been rimmed by thin chromitite seam. The rimmed block has been truncated by a planar erosional surface (a) which has been subsequently overlain by a UG1 chromitite layer. Head of the hammer in (a) is 17 cm.

6.2. What do blocks tell us about the rheological state of the floor sequence?

The melanorite blocks caused a localized disruption and deformation of the cumulate pile on which they landed. The boundary between the blocks and impacted layers of rocks is always notably clear and sharp (Fig. 7–16) allowing to conclude that – while the Bushveld chamber was crystallizing – there was almost invariably a sharp interface between the resident melt and the top of the upward-growing cumulate pile (Fig. 18). This conclusion is in line with Irvine et al., (1998), who arrived at the same idea using a similar set of field observations from the Skaergaard intrusion. One alternative interpretation needs to be considered here, however. One may argue that the blocks sunk through a thick pile of mushy cumulates before reaching the rigid layers (e.g. chromitite) that terminated their sinking. For instance, in the Windimurra Complex, there is a large anorthositic block that rests on top of a curved magnetite layer and is overlain by planar layers of magnetite gabbro and

magnetite (Nebel et al., 2013). It was proposed that the magnetite layer was curved due to impact by the sinking block, whereas all the overlying layers are planar because they were mushy and able to merge after the anorthositic block penetrated them (Nebel et al., 2013). Such an interpretation is not applicable, however, to our case. Figs. 15 and 16 show a few examples of blocks that are enclosed in chromitite layers. One can hardly imagine a situation when a block sinking through the mushy pile may become enclosed by two pre-existing chromitite layers (Figs. 15–16). The settling block may only deform/depress the pre-existing layer of chromitite, but it cannot become overlain by such a layer. A simplest interpretation is that these blocks impacted the older chromitite layer on the top of the cumulate pile and then were covered by the younger chromitite layer from above resulting in their encapsulation within a seemingly single layer of chromitite (see a full story in Fig. 20 below). For this reason, a first scenario with a sharp interface between the resident melt and the top of the cumulate pile (Fig. 18) appears to be more realistic for our case. This interface was highly

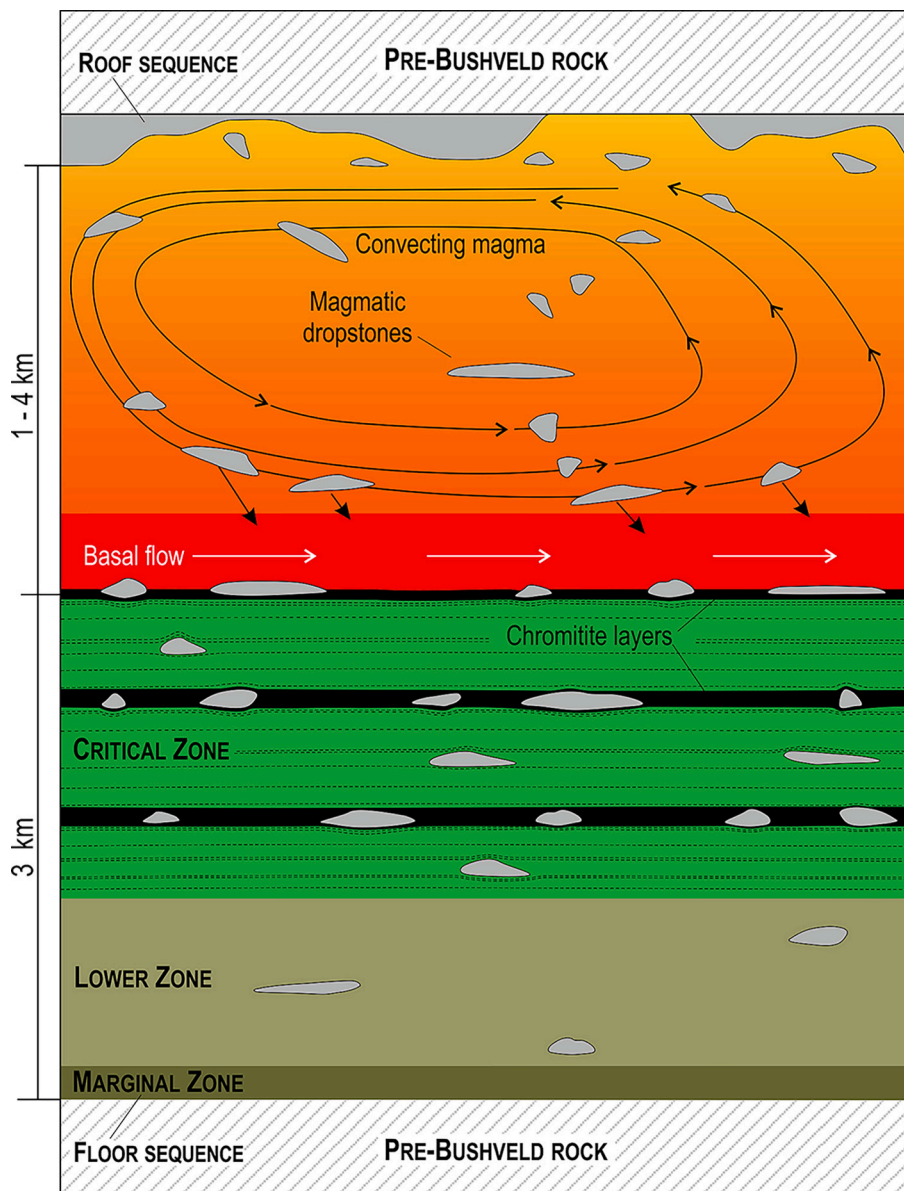


Fig. 18. Schematic cartoon of the Bushveld chamber at the time of its CZ crystallization as deduced from the block-and-layering structures. The magma chamber develops as a central body of the crystal-free, convecting resident melt which nucleates and crystallizes in situ along the roof and base of the chamber. The resident melt passes to solidifying rocks at the margins through an inward advancing solidification front. The roof sequence of cumulates is being destroyed by tectonics and its blocks (i.e., magmatic dropstones) are collapsed onto the inward-growing chamber floor (the lower solidification front). This most commonly happens when the chamber is replenished by new magma pulses that enter and spread along the chamber floor as basal flows. In the CZ, these basal flows are responsible for the formation of massive chromitites on the chamber floor. This explains the close association between the chromitites and roof-derived magmatic dropstones. There is a sharp interface between the chamber floor and overlying resident melt. Igneous layering in the floor cumulates is fully developed right up to crystal-liquid interface and is entirely coherent so that magmatic dropstones cause disruption and deformation of the layering. It is assumed that the roof sequence is destroyed at the later stages of internal differentiation. The main stratigraphic zones and magmatic dropstones are drawn out of scale. The morphology of the magma chamber is partly based on Irvine et al. (1998) and Latypov et al. (2022a). The streamline pattern in the resident melt is based on Irvine et al. (1998).

dynamic, and it was repeatedly struck by blocks (Figs. 8–16) and locally eroded, sometimes together with the blocks themselves (Fig. 17) when the chamber was replenished by new magma pulses. The conclusion about the sharp crystal-liquid interface is at odds with some current petrological models implying that such a boundary (i.e., a temporary chamber floor) may not exist in magma chambers at all. In these models, the crystal-liquid boundary is absent because the magma in the chamber strongly convects and retains all crystals in suspension (i.e., entirely preventing crystal settling) so that when it finally comes to halt a crystal content reaches the critical crystallinity of about 45–50% in the entire chamber (e.g., Bachmann and Bergantz, 2004; Bea, 2010).

Another important aspect is that the blocks disrupt (Figs. 8–10) and

indent (Figs. 11–13) the well-developed layers of anorthosite and chromitite in the floor cumulate pile indicating that the igneous layering was fully developed right up to this interface and was already entirely coherent (and sometimes even rigid) (Fig. 18). The truncation indicates that the igneous layering was not formed by any late-stage process such as fluid-induced recrystallization of deeply buried non-layered rocks as argued in some studies (e.g., Boudreau and McBirney, 1997). Instead, the data suggest that the igneous layering was fully developed very close to or even directly at the crystal-liquid interface of the inward-growing chamber floor. The igneous layering is thus considered as a primary magmatic feature. Following Pebane and Latypov (2017), we suggest considering the igneous layering in the UG1 sequence as consisting of

several chromitite-anorthosite cyclic units, with each being produced from a separate pulse of magma saturated in spinel (chromite) only. Each pulse may have been emplaced as a basal flow with little mixing with the resident melt taking place. An inferred crystallization sequence of each pulse is chromite (to form chromitite) followed by chromite and plagioclase (to form chromite-bearing anorthosite) (Pebane and Latypov, 2017).

In addition, the blocks were only able to impact the uppermost portion of the cumulate pile. This supports the recent realization that, in the Bushveld chamber, only a few first metres of the cumulate pile were poorly consolidated, whereas all other underlying rocks were nearly totally solidified (Latypov et al., 2015, 2024a; Holness et al., 2017a) (Fig. 18). An interesting insight comes from the outcrop in Fig. 12 where a first anorthosite layer (A) beneath the melanorite block is both depressed and attenuated, whereas the second and third ones (B and C) are only depressed with almost no change in their thickness. This can be taken as an indication that only the first layer was mushy and able to plastically flow outwards from the place of the impact. The subsequent two layers were likely already much more coherent and not able to flow and, as a result, they were only depressed, but not attenuated by the impact. If this interpretation is correct, then the mushy layer in this place was at best 20 cm thick. This is in line with data from other intrusions. In particular, field observations on deformation of igneous layering beneath blocks fallen from the roof onto the inward-growing chamber floor, has been taken to indicate that a mushy layer was only 1 to 3 m thick in the Stillwater Complex (Hess, 1960) and a few meters thick in the Sarqata Qaqa Complex (Thompson and Patrick, 1968) and the Skaergaard intrusion (Irvine et al., 1998).

6.3. Did the magma chamber contain a large, long-lived, and totally molten melt body?

The disruption of the roof sequence into angular blocks which collapsed downwards and impacted the floor cumulates implies the existence of a molten magma body that was sandwiched between the two inward-propagating solidification fronts. In other words, this suggests that the Bushveld chamber was developing as a ‘big magma tank’ (Fig. 18), i.e., a largely molten, long-lived, and slowly fractionating magma body that mostly grew from the base upwards through the thin solidification front (Kruger, 2005; Cawthorn, 2015; Kruger and Latypov, 2020; Latypov et al., 2022a, 2022b). It should be noted that our field observations do not provide evidence on whether crystals have been settling on the chamber floor from the overlying magma or crystallized in situ, i.e., directly on the chamber floor. However, for a variety of reasons discussed later, we favour the formation of cumulates in the Bushveld chamber by in situ crystallization. The existence of such a liquid-dominated magma body (i.e., ‘big magma tank’) is straightforwardly demonstrated by the mere presence of blocks that impact the CZ’s floor cumulates. The thickness of the magma body at the time of the formation of the CZ is not possible to evaluate from the field observations in this study. However, the previously reported field data indicate that the overlying Main Zone was formed from a melt column of ~5 km thick (Latypov et al., 2022b), and it would not be entirely unreasonable to assume that the thickness of the CZ’s magma column may also have been in the same range. This conclusion is at variance with a newly emerged mushy paradigm that denies the existence of such totally molten magma bodies in Earth’s crust and postulates, instead, the existence of transcrustal mushy systems which contain small melt lenses for only a very short period of time before accumulating and erupting as lavas on the Earth’s surface (Cashman et al., 2017; Jackson et al., 2018; Bachmann and Huber, 2019; Edmonds et al., 2019; Sparks et al., 2019; Blundy, 2022).

7. Implications for the origin of massive chromitites in the Bushveld Complex

We will now focus on the observations from the block-and-layering structures that involves layers of stratiform chromitites (60–90 vol% chromite). The origin of these industrially important igneous rocks (as a major source of chromium) in layered intrusions has recently attracted much interest from igneous petrologists causing lively debates in the literature (Mondal and Mathez, 2007; Latypov et al., 2017b, 2018, 2022a; Mathez and Kinzler, 2017; Mukherjee et al., 2017; Pebane and Latypov, 2017; Jenkins and Mungall, 2018; Kaufmann et al., 2018, 2019; Hepworth et al., 2020; Nikolaev et al., 2020; Marsh et al., 2021; Otto et al., 2024). This topic is of particular relevance to the Bushveld Complex because it contains >80% of the Earth’s known chromium resources (Cawthorn, 2010). Our novel observations on physical relationships of magmatic dropstones with layers of massive chromitites provide a unique opportunity to draw some solid constraints and conclusions with regards to the formation of these petrologically and economically important magmatic rocks. Let us start with a short review of the existing ideas on their origin.

7.1. A classical approach to the origin of massive chromitites in layered intrusions

A traditional class of models (Fig. 18) considers massive chromitites as a product of crystallization of resident, newly emplaced, or hybrid magmas that are saturated in chromite as the only liquidus phase (e.g., Irvine, 1975; Campbell and Murck, 1993; Lipin, 1993; Kinnaird et al., 2002; Spandler et al., 2005; Cawthorn, 2011; Marques et al., 2017; Latypov et al., 2018; Friedrich et al., 2020; Langa et al., 2021, 2023). The chromite is proposed to form either throughout the entire magma chamber with subsequent gravity settling on the chamber floor either as separate grains (e.g., Wieser et al., 2019) or their clusters (Holness et al., 2023) or, alternatively, the chromite is considered to crystallize from the basal layers of replenishing magmas in situ, i.e., directly on the chamber floor (Latypov et al., 2017b, 2022a, 2023; Mukherjee et al., 2017; Pebane and Latypov, 2017; Hasch and Latypov, 2021). Associated with this class is also a model by Veksler and Hou (2020) which envisages the generation of chromite-only saturated melts through large-scale melting of the floor orthopyroxenites by a basal layer of hot/volatile-rich melt followed by in situ growth of chromitite layers on the eroded chamber floor. A key feature of the traditional class of the models is that massive chromitite form directly on the inward-growing chamber floor (Fig. 18).

7.2. Three alternative approaches to the origin of massive chromitites in layered intrusions

There is a wide spectrum of alternative models for the formation of stratiform chromitites which can be broadly subdivided into three major categories: (1) a crystal-rich slurry class, (2) a sill emplacement class and (3) a reactive porous flow class (Fig. 19).

A first class of alternative hypotheses questions the necessity of chromite-saturated magmas for the origin of massive chromitites. Instead, this class emphasizes the crucial role of crystal-rich slurries which are either formed directly within the chamber or brought into the chamber from some deep staging reservoirs (Fig. 19). The slurry models envisage that chromitite layers form by gravity- and size-controlled separation of chromite grains from silicate minerals in crystal-rich slurries that either flow or slump along the chamber floor (Mondal and Mathez, 2007; Eales and Costin, 2012; Maier et al., 2013; Jenkins and Mungall, 2018; Maghdour-Mashhour and Hayes, 2021; Barnes et al., 2023). A distinguishing feature of this class of models is that individual chromite grains congregate into layers of massive chromitites underneath ~20–100 m thick crystal-rich slurries (Fig. 19); thus, at no time in their history, the chromitite layers occur on the chamber floor, i.e., at crystal-liquid interface (see a recent review in Latypov et al., 2023).

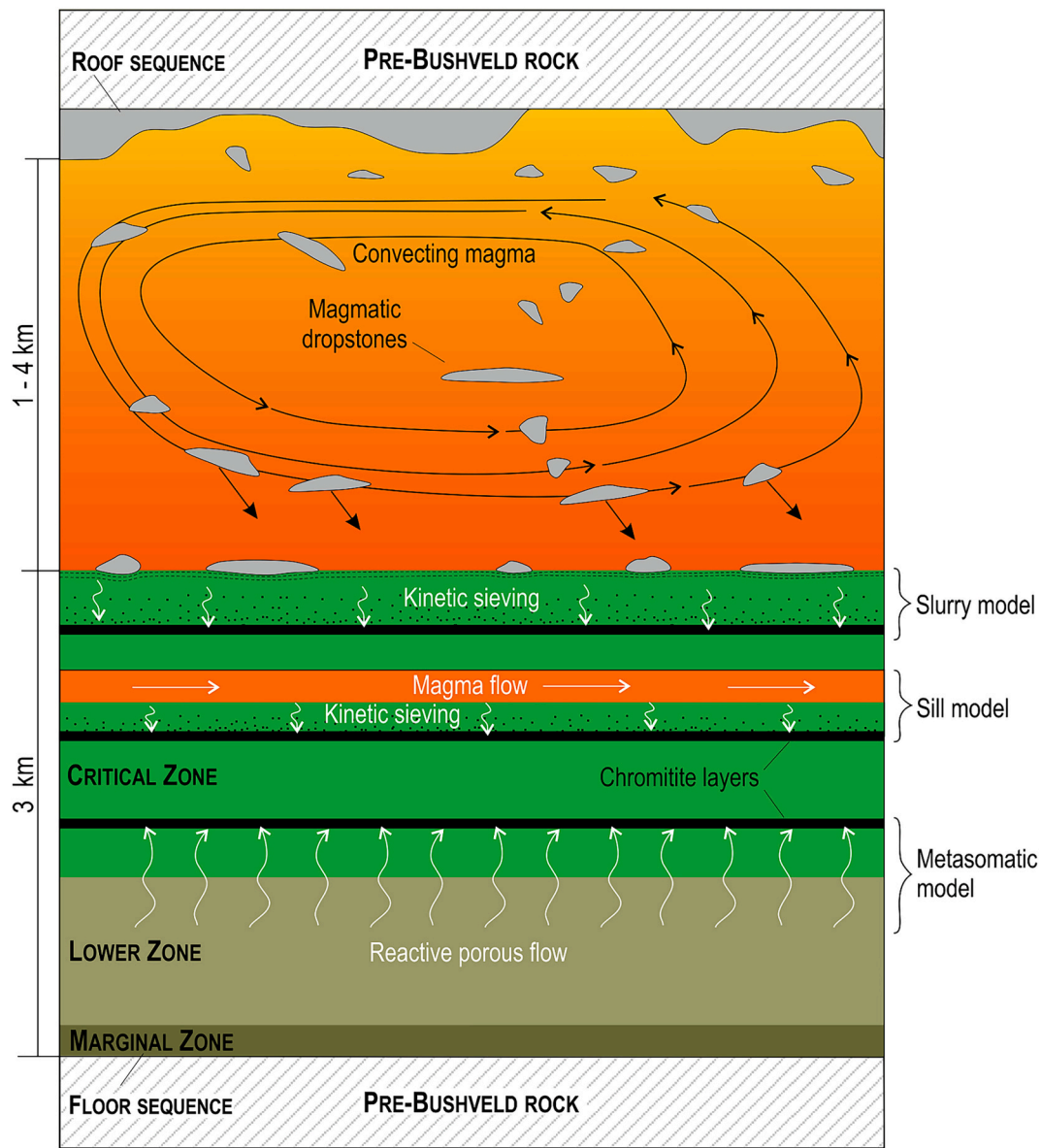


Fig. 19. Schematic cartoon of the Bushveld chamber at the time of its CZ crystallization as deduced from the block-and-layering structures. The magma chamber develops as a central body of the crystal-free, convecting resident melt which nucleates and crystallizes in situ along the roof and base of the chamber. The resident melt passes to solidifying rocks at the margins through an inward advancing solidification front. The roof sequence of cumulates is being destroyed by tectonics and its blocks (i.e., magmatic dropstones) are collapsed onto the inward-growing chamber floor. The cartoon indicates that three alternative hypotheses (crystal-rich slurry, sill emplacement and reactive porous flow) envisage the formation of layers of stratiform chromitites at some depth in the cumulate pile. If so, these layers cannot be impacted by roof-derived magmatic dropstones that settle on the chamber floor. For this reason, these three hypotheses are ruled out as potential explanations for the origin of massive chromitites in the Bushveld Complex. The morphology of the magma chamber is partly based on Irvine et al. (1998) and Latypov et al. (2022a). The streamline pattern in the resident melt is based on Irvine et al. (1998).

A second class of alternative models can be regarded as a variety of the previous one in which crystal-rich slurries emplaced as late-stage sills into the pre-existing cumulate pile, rather than onto the chamber floor (Voordouw et al., 2009; Mungall et al., 2016; Mitchell et al., 2019; Scoates et al., 2021; Otto et al., 2024) (Fig. 19). In one of these models, it is assumed that chromite separates from silicate phases by kinetic sieving while the crystal-rich slurry is still flowing along the sill and this results in the formation of a layer of massive chromitite at the base of this sill (Mungall et al., 2016). In another model, such slurries are proposed to consist of only chromite crystals so that no crystal sorting is required to form a layer of massive chromitite (Voordouw et al., 2009). A depth of the sill emplacement into the cumulate pile is not specified in this class of models but in any case, the chromitite layers are thought to form in the late-stage sills well below the temporary chamber floor

(Fig. 19).

Finally, a third and most recently emerged class of alternative models challenges the importance of both the chromite-only-saturated magmas (in a traditional class) and crystal-rich slurries for the chromitite petrogenesis (in two above classes). More specifically, the models postulate a large-scale reactive porous flow of interstitial liquid/fluid through a km thick mushy zone of the floor cumulate pile (Fig. 19). A large amount of upward migrating melts/fluids is proposed to induce at some stratigraphic levels the complete remelting of norite/orthopyroxenite cumulates and reprecipitation of released chromium in the form layers of massive chromitites (Mathez and Kinzler, 2017; Nikolaev et al., 2020; Marsh et al., 2021; Boudreau, 2024). Again, just like in the above two classes of models, these stratiform chromitites have never seen/been to the chamber floor; they are produced via metasomatic

transformation of silicate rocks located at some depth in the cumulate pile (Fig. 19).

7.3. Crystal-rich slurry, sill emplacement and reactive porous flow hypotheses are inconsistent with field observations

A distinguishing feature of alternative classes of models is that layers of massive chromitites are envisaged to form deep in the cumulate pile (Fig. 19). For this reason, these layers cannot be physically impacted by magmatic dropstones that land on the temporary chamber floor and may penetrate, at best, only its uppermost part (perhaps, not >1 m). The physical relationships in the block-and-layering structures clearly show, however, that chromitite layers are ruptured by (Figs. 8–10) or indented (Fig. 11–13) beneath the blocks and are draped over the blocks (Figs. 14–16). In other words, the blocks are deforming the chromitite layers on which they have fallen, with the younger chromitite layers being draped over their tops. These observations indicate that chromitite layers have already been on the chamber floor when the blocks landed on this interface. If so, this means that stratiform chromitites have been forming directly at the chamber floor (i.e., at the crystal-liquid interface) rather than somewhere deep in the cumulate pile. This is one of the most significant inferences that can be drawn from our field observations for the formation of massive chromitites. To the best of our knowledge, none of the other existing field, textural or chemical observations is able to provide such a strict constraint on the origin of massive chromitites.

In addition, there is no logical explanation for the block's association with massive chromitites (Fig. 4) in the frame of the crystal-rich slurry and reactive porous flow models. More specifically, there is no obvious reason as to why mm-size chromite grains should be accumulated within the cumulate pile into separate layers of chromitites together with meter-size blocks (even if they brought into the chamber together with crystal-rich slurries). Similarly, there is no reason as to why metasomatic transformation of norite/orthopyroxenite into chromitite should have occurred at the stratigraphic level where the roof-derived blocks have been earlier deposited. In addition, metasomatic models face a serious challenge to explain why angular blocks of melanorite/orthopyroxenite may have survived (i.e., remain intact) when many dozens of meters of surrounding norite/orthopyroxenites have been completely transformed into massive chromitites (Boudreau, 2024). Thus, crystal-rich slurry and reactive porous flow models fail the most basic test against the field observations from the block-and-layering structures and may therefore be excluded from further considerations in this study. In addition, there are several other field, textural and chemical arguments for questioning these models (Latypov et al., 2017b, 2022a, 2023). For this reason, only the first class of traditional models that envisage the origin of chromitites through deposition of chromite crystals on the chamber floor will be subjected to further testing/examination below.

7.4. Blocks testify against the out-of-sequence emplacement of late-stage sills into the cumulate pile

The Bushveld Complex has been traditionally interpreted as having formed from numerous magma pulses that successively replenished the evolving chamber and deposited crystals on a temporary floor, building the layered sequence from the base upwards (Wager and Brown, 1968; Eales and Cawthorn, 1996; Cawthorn, 2015). There are, however, several recent attempts to question this view, with the suggestion that some of the layers (e.g., UG2 chromitite layer) or even entire units (e.g., Merensky Unit) are late-stage sills intruded into the nearly solidified cumulate pile (Voordouw et al., 2009; Mungall et al., 2016; Mitchell et al., 2019; Scoates et al., 2021; Otto et al., 2024). For instance, Mungall et al. (2016) reported that ultramafic units composed of chromitite and orthopyroxenite in the upper CZ have out-of-sequence U–Pb zircon and baddeleyite ages. On this basis, they questioned the traditional, sequential formation of the macrolayering of the upper CZ. Instead, they

suggest that the noritic rocks of the upper CZ formed first and were then intruded by sills of crystal-rich slurries, which internally differentiated into the chromitite-orthopyroxenite units (e.g., UG1 or UG2 chromitite layer). The discovery of the large melanorite blocks that impact the igneous layering of UG1, UG2 and MG2 sequences challenges this interpretation. Clearly, to be impacted by roof-derived magmatic dropstones, the UG1, UG2 and MG2 chromitites need to be located on the chamber floor (Fig. 18), not in the late-stage sills intruded in the cumulate pile well below the chamber floor (Fig. 19). One can perhaps argue that the blocks may have been brought into the sills with crystal-rich slurries. To prove the physical feasibility of this idea is a great challenge, however, given particular attention to the non-ovoid shape of the blocks and their very large size (up to 10 m in length, Fig. 7). In addition, if blocks were brought into the sills as a component of crystal-rich slurries, one would expect the large blocks to drop out first, resulting in lateral sorting of blocks by size. This phenomenon has not been documented in our study. One would also expect to see imbrication of blocks (e.g. Irvine et al., 1998, their Fig. 9c) and other sedimentary features due to flow separation around the blocks, such as greater thickness of chromitite in 'trap sites' immediately downstream. These features were also not observed in our study. Overall, it seems highly unlikely that such blocks can be brought into the sills even with crystal-rich slurries, especially considering that the crystal-rich nature of transporting magmas is postulated, rather than inferred from any specific observations. In contrast, our field and textural data indicate that the magmas replenishing the CZ were likely superheated (i.e., with no crystals) to be able to erode a large amount of pre-existing floor cumulates (Latypov et al., 2015, 2017b, 2017c, 2022a, 2022c).

7.5. Origin of massive chromitites by deposition of chromite on the chamber floor

The fact that the blocks are landed on and/or indent the chromitite layers (i.e., with no layers of other silicate rocks present in between; Figs. 7–16) indicates that only chromite alone has been depositing on the chamber floor at the time of the block's impact. In many cases, the deposition continued after the impacts as follows from the draping of chromitite layers over the tops of the fallen blocks. This results in complete encasing of blocks into massive chromitites (Figs. 15–16). These relations allow us to infer that the parental melt was saturated in chromite alone (i.e., no other liquidus phases were present in the magma). This is another extremely important inference that can be drawn from our observations and which should be built into any successful model for the origin of the Bushveld massive chromitites.

Fig. 20 summarises a scenario (mostly to explain the case in Figs. 15–16) in which deposition of chromite on the chamber floor from chromite-only-saturated melt produces a layer of massive chromitite. This layer together with the underlying strata are then subjected to sub-vertical impact that causes depression and attenuation of the layers beneath the fallen block. Subsequently the block is draped over its top by another chromitite layer that continues to deposit from the same parental melt. Further crystal deposition on the chamber floor produces anorthosite and chromitite layers that become increasingly less distorted above the block until they become completely planar. Fig. 21 shows another scenario (mostly to explain the case in Fig. 13) for the indentation and depression of the layered strata by an angular block that falls on the chamber floor at moderate angle. The difference in morphology of the block results in some deposition of crystals on its gentle slope but no deposition on its subvertical to overturned edge. No deposition can be best explained by poor adhesion of settling crystals on the overhanging edges. It can also, however, be attributed to an unstable position of in situ grown crystals so that they can be torn loose from such edges and re-deposited on the planar chamber floor by flowing/convecting magma.

Field observations also suggest that the melt parental to massive chromitites was likely derived from some external source(s), rather than internally. This follows from the fact that the blocks are confined to

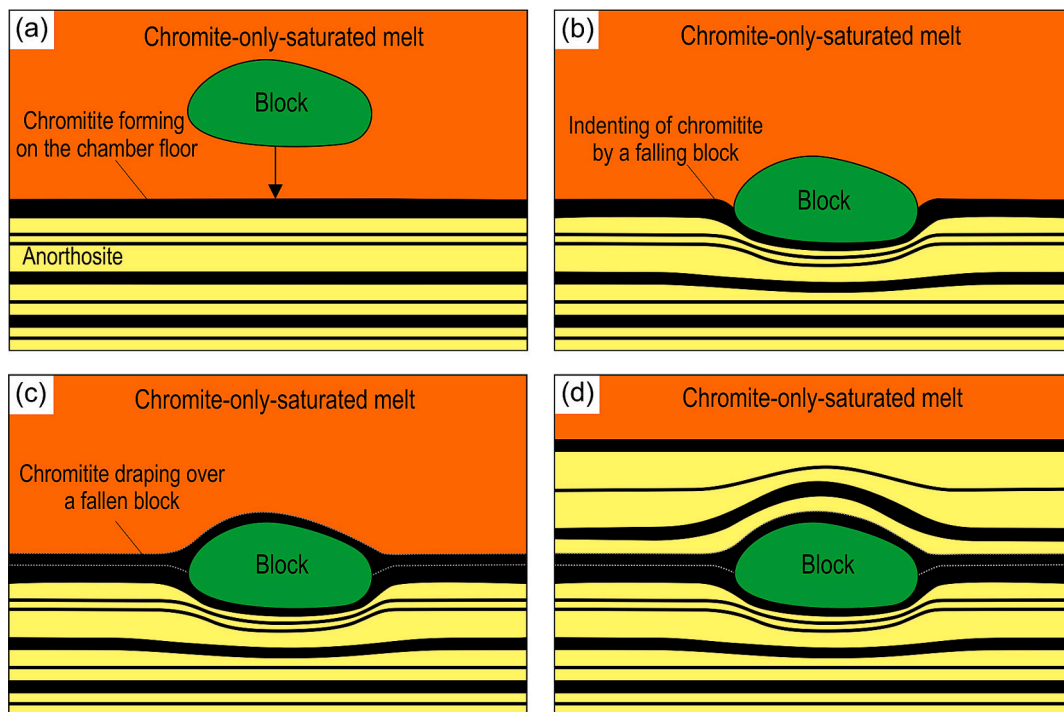


Fig. 20. A cartoon showing a melanocratic block that is enclosed by two layers of massive chromitites. (a) Deposition of chromite from a chromite-only-saturated melt results in the formation of a chromitite layer on the chamber floor. (b) This layer, along with the strata beneath it, is subjected to sub-vertical impact, causing the layers beneath the block to depress and thin. (c) The block is then covered by another layer of chromitite that continues to deposit from the same parental melt. (d) Further deposition leads to the formation of anorthosite and chromitite layers that become progressively less bent above the block until they become completely planar. The scenario explains field relations in Fig. 16b-e.

some specific horizons in the stratigraphy of the CZ and are particularly abundant in rock sequences with massive chromitites (Fig. 4). The preferred concentration of blocks likely results from the repeated transient episodes of crustal instabilities – such as earthquakes – that are commonly associated with magma chamber replenishments and may trigger the disruption and collapse of the roof sequence. It is tempting to conclude, therefore, that it is the earthquakes that caused by the emplacement of the magma parental to massive chromitites, likely as the basal flows (Latypov et al., 2017b), and this led to the close association of blocks with massive chromitites (Fig. 18). In other words, the blocks concentrate in horizons with massive chromitites because those horizons record the earthquakes that accompanied the magma chamber replenishment events. In some respects, the blocks can be viewed as a smoking gun of the process that was responsible for the formation of massive chromitites, which is the replenishment of evolving magma chamber by chromite-only-saturated melts. There are different ways to produce such magmas but we prefer a mechanism in which the ascending basaltic melts find themselves in the field of the chromite stability in response to a drop in lithostatic pressure (Latypov et al., 2018; Drage and Brenan, 2023; see discussion below).

Our field observations are unable, however, to give us a clue to the exact mechanism of chromite accumulation on the chamber floor. In the class of traditional models, there are two contrasting options – gravity settling of chromite from the overlying magma onto the chamber floor (Irvine, 1975; Campbell and Murck, 1993; Lipin, 1993; Kinnaird et al., 2002; Spandler et al., 2005; Cawthorn, 2011; Holness et al., 2023) and in situ growth of chromite directly on the chamber floor (Latypov et al., 2017b, 2022a, 2023; Mukherjee et al., 2017; Pebane and Latypov, 2017; Hasch and Latypov, 2021). The second option seems to be more consistent with a range of recently obtained field (Latypov et al., 2015, 2017b, 2017c), textural (Kaufmann et al., 2018; Latypov et al., 2022a) and chemical (Kaufmann et al., 2018; Latypov et al., 2023) observations on the Bushveld chromitites. However, the basic tenets of this model

have been recently challenged on various grounds (Cawthorn, 2018; Veksler and Hou, 2020; Maier et al., 2021; Barnes et al., 2023; Drage and Brenan, 2023; Holness et al., 2023), necessitating careful examination of the validity of the objections raised against this mechanism.

8. A review of objections against in situ growth of chromite on the chamber floor from chromite-only-saturated melts

8.1. Can a pressure decrease produce a chromite-only-saturated melt?

In our recent study (Latypov et al., 2018), we have proposed using three lines of evidence – graphical phase equilibria, thermodynamic modelling and experimental data – that some basaltic melts lying on the orthopyroxene(olivine)-chromite cotectic may become saturated in chromite alone in response to a decrease in lithostatic pressure. The saturation may happen when the melts from some deep-seated reservoirs ascend towards the Earth's surface with subsequent stalling and cooling in shallow-level chambers. A key role in this process is played by plagioclase whose stability field tends to substantially expand with a decrease in pressure causing some corresponding changes in the topology of phase equilibria diagrams. More specifically, the expansion of the plagioclase field (Fig. 22a) results in the shift of a chromite stability field to the left side of the phase diagram so that the melt originally located on orthopyroxene(olivine)-chromite cotectic finds itself in the chromite stability field, i.e., becomes saturated in chromite alone. Importantly, our model does not imply that this happens in response to a change in the size of the chromite field itself – i.e., its expansion or shrinking – but rather because of its shift to the left side of the phase diagram (Fig. 22a).

Drage and Brenan (2023) has recently undertaken an experimental study to verify the basics of our hypothesis. This experimental study has fully confirmed a key point of our proposal – a decrease in pressure does result in saturation of some basaltic melts in chromite alone. A question that remains to address is what is responsible for this saturation? Drage

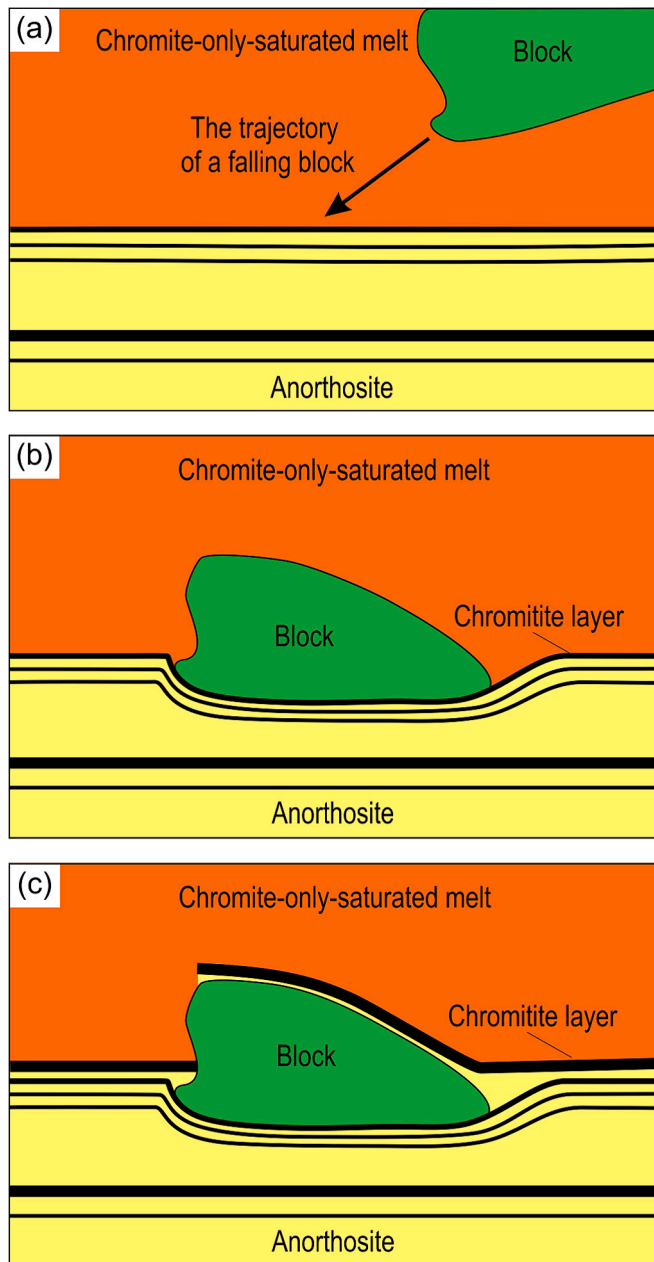


Fig. 21. A cartoon showing a melanocratic block that indents and is partly draped over by igneous layering. (a) An angular block falls from right to left towards the temporary chamber floor with anorthosite-chromitite layering. (b) This layer, along with the strata beneath it, is subjected to sub-vertical impact, causing the anorthosite layers beneath the block to depress and thin. (c) Further deposition leads to the formation of anorthosite and chromitite layers that can deposit on the gentle slope of the block (a right side) but not on its subvertical to overturned edge of the block (a left side) likely due to the high angle of repose. The scenario explains field relations in Fig. 13.

and Brenan (2023) concluded that the chromite-alone saturation is due to a decrease in the modal abundance and Cr-content of orthopyroxene (as the major Cr reservoir) with a decrease in pressure. We have depicted results of their experiments using the Irvine's type diagrams (Fig. 22b-c). The ternary and binary diagrams illustrate three major results of Drage and Brenan (2023): as pressure decreases from 10 kbar to 1 kbar, (a) the stability field of chromite expands, (b) modal abundance of orthopyroxene in cotectic with chromite (compare points X and X' in Fig. 22c) declines and (c) Cr-content in orthopyroxene (compare solid solutions of Opx_{ss} and Opx_{ss}' in Fig. 22c) decreases. From the phase relations, we are

not convinced that a decrease in modal abundance and Cr-content of orthopyroxene with a decrease in pressure is a major reason for the saturation of a melt in chromite. It seems more logical to conclude that the saturation is simply caused by the expansion of the chromite stability field with a pressure decrease. Moreover, it is also quite possible that the saturation may be caused by a shift of chromite stability field as proposed in our original study (Fig. 22a; Latypov et al., 2018). The experimental results by Drage and Brenan (2023) do not exclude this possibility.

To conclude, Drage and Brenan (2023)'s study has confirmed that some basaltic melts may become saturated in chromite alone due to a decrease in lithostatic pressure. How this exactly happens remains an open question. It may occur either due to expansion of a chromite field with decreasing pressure as implied by phase equilibria considerations (Fig. 22b-c) or in response to a shift of a chromite stability field with decreasing pressure as advocated in our study (Fig. 22a; Latypov et al., 2018). These two approaches are not mutually exclusive and may work together hand in hand. A key point is that – one way or another – a basaltic melt may become saturated in chromite alone in response to a decrease in lithostatic pressure and this melt may be parental to massive chromitites. The major strength of this proposal is in its inherent simplicity – all ascending melts are inevitably subjected to a reduction in lithostatic pressure. It is quite natural, therefore, that some of these melts, compositionally located alongside the orthopyroxene(olivine)-chromite cotectic, may reach chromite-only-saturation. We think that this scenario is conceptually the simplest and physically most plausible explanation for a generation of chromite-only-saturated melts parental to massive chromitites in layered intrusions. Finally, a short note with regards to the parental magma composition which we have used for our thermodynamic modelling (Latypov et al., 2018). This composition has never been meant as a real magma parental to Bushveld's massive chromitites, but rather as an example to illustrate our idea that some basaltic melts may become saturated in chromite alone with pressure reduction. The true composition of parental magma remains to be identified and rigorously justified. An experimental study by Drage and Brenan (2023) is an important step towards addressing this challenge.

8.2. Are there any alternative solutions for a mass balance problem of massive chromitites?

The stratiform chromitites in layered intrusions can be up to 2 m thick and contain 40–50 wt% Cr_2O_3 , and are commonly considered to have crystallized from a basaltic melt that have contained no more than ~0.10 wt% Cr_2O_3 (Murck and Campbell, 1986). Inevitably, the mass balance dictates that the formation of chromitite layers requires extraction of Cr from a large volume of melt which must be present either as a thick melt layer in the chamber or as the melt flowing through the chamber, or both. Based on their experimental study, Drage and Brenan (2023) indicate that the formation of a 1 m thick layer of chromitite from a chromite-only-saturated melt (e.g., such as B1 magma commonly considered as parental to CZ) requires an ~2 km thick melt column at a minimum. The estimation seems reliable and is broadly in the same range with those from several earlier studies (Murck and Campbell, 1986; Veksler and Hou, 2020). Applying this logic to the entire CZ, the formation of its most prominent chromitites would have required a column of 13 to 15 km of a parental chromite-only saturated melt (Cawthorn and Walraven, 1998; Naldrett et al., 2012). This enormous volume of required melt is at odds with the size of the CZ that is just over 1 km thick (~1260 m). To resolve this paradox, many authors proposed that the large volume of residual melt that remained after the formation of massive chromitite was removed from the chamber by lateral or vertical expulsion (Cawthorn and Walraven, 1998; Naldrett et al., 2012; Latypov et al., 2022a). The solidification products of this enormous volume of escaped magma (i.e., basalt lavas) have not yet been found and therefore several researchers refer to this scenario as geologically unreasonable/beyond imagination (Veksler and Hou, 2020;

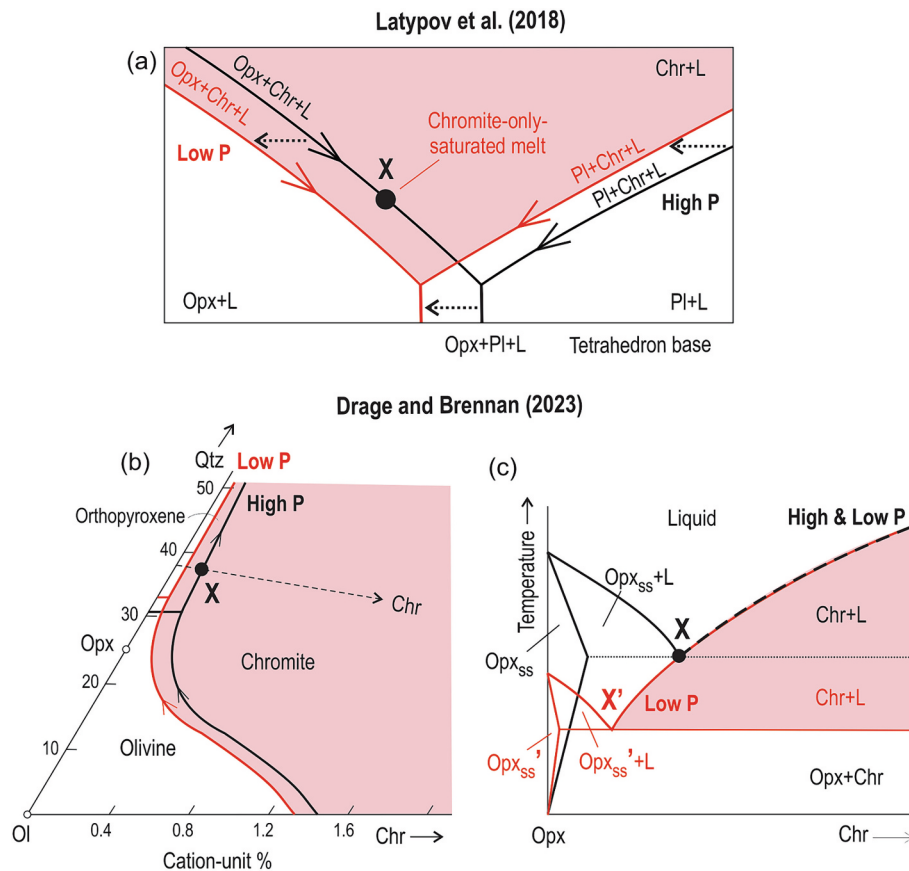


Fig. 22. Two changes in liquidus phase equilibria associated with pressure reduction that may potentially induce saturation of basaltic melts in chromite as the sole liquidus phase. (a) Latypov et al. (2018) proposed that expansion of the plagioclase stability volume with decreasing pressure results in a shift of the Opx-Chr-saturated melt (X) into the chromite-only stability field. Note that in this process the chromite stability field does not expand but merely changes its position. (b–c) Our interpretation of the Drage and Brennan (2023) experiments using a ternary liquidus diagram Ol-Chr-Qtz (Irvine, 1977) and a binary pseudosection Opx-Chr (indicated by a dotted line in b). We propose that the Opx-Chr-saturated melt (X) becomes saturated in chromite alone due to expansion of the chromite stability field with a decrease in pressure. The expansion is associated with a decrease in modal abundance of orthopyroxene in cotectic (X vs X') and in Cr-content in orthopyroxene (Opx_{ss} vs Opx_{ss}'). Note in (c) that liquidus temperature of chromite is a function of temperature; pressure has almost no effect on the liquidus temperature.

Drage and Brennan, 2023).

To solve this mass-balance issue, two alternative approaches have been proposed that are not dealing with chromite-only-saturated melts. These are crystal-rich slurry and reactive porous flow models which have already been discussed above. The slurry models attempt to solve the issue by invoking crystal-rich slurries (Mondal and Mathez, 2007; Eales and Costin, 2012; Maier et al., 2013; Mungall et al., 2016; Jenkins and Mungall, 2018). As an example, Mondal and Mathez (2007) have proposed that the magma chamber can be replenished by an ~56 m thick crystal-rich slurry (with 45% Opx and 1–2% chromite) from a deeper magma reservoir. After emplacement, chromite separates from coexisting pyroxenes by kinetic sieving to form an ~1 m thick chromitite layer, overlain by an ~55 m thick column of an orthopyroxene-rich mush (Mondal and Mathez, 2007). Needless to say, that, in terms of the mass balance issue a ~56 m thick crystal-rich slurry seems much more appealing than ~2 km thick column of chromite-saturated melt in the traditional concept. The reactive porous flow models appeal to remelting of Cr-rich norite/orthopyroxenite at some depth in the cumulate pile by upward migrating fluids which leaves behind a restitic layer of massive chromitite (Boudreau, 2016, 2024; Mathez and Kinzler, 2017; Nikolaev et al., 2020; Marsh et al., 2021). As an illustration, the formation of ~1 m thick chromitite layer would require remelting of only ~50 m of orthopyroxenite or ~100 m of norite (Boudreau, 2024). Again, these numbers seem much more inviting than ~2 km thick column of chromite-saturated melt. Despite their apparent attractiveness, the above models cannot be considered as viable solutions to a mass

balance issue because they imply the formation of chromitites at some depth in the cumulate pile (Fig. 19). This is not consistent with indenting and rupturing of chromitite layers by magmatic dropstones (Figs. 7–16) which indicate that massive chromitites form directly on the chamber floor (Figs. 18, 20). In addition, it should be mentioned that despite a mathematical model to simulate the formation of chromitite layers via remelting of norite/orthopyroxenite is theoretically elegant (Boudreau, 2024), it is not supported by any independent evidence (e.g., field and textural observations). We have not yet found in the field anything that would be indicative of the partial transformation of 50–100 m thick pile of silicate rocks into 1 m of massive chromitites.

There is another interesting suggestion that is sitting between metasomatic and magmatic models and envisages the formation of chromitite directly on the chamber floor (Veksler and Hou, 2020). The model implies that the resident melt at the time of the formation of the CZ was nearly dry. At some point, the cumulate pile was tectonically fractured and fluids from the sedimentary rocks underlying the Bushveld Complex entered the magma chamber. The fluids were not, however, transferred into the entire magma chamber but rather somehow concentrated at the chamber floor forming a basal layer of melt with 3–4 wt% H_2O . Such a water-rich melt is chemically aggressive and causes hydration melting of the floor cumulates. The melting of cumulates is accompanied by in situ crystallization of chromite on the uneven erosional surface from a basal stagnant layer. Like in reactive porous flow models, ~1 m thick chromitite layer is supposed to form from ~100 m thick orthopyroxenite pile. This again seems very attractive

from a mass balance point of view. Moreover, the concept can be reconciled with field observations because the chromitite layer forming directly on the chamber floor may be subjected to indentation and rupturing by falling magmatic dropstones. However, the model appears to be unrealistic from a physical point of view. The study itself shows that addition of 3–4 wt% H₂O to a dry melt will cause a dramatic decrease in its density from 2607 kg/m³ to 2429 kg/m³ at 1125 °C and 300 MPa (Veksler and Hou, 2020). This would make a basal layer extremely buoyant. As a result, the basal layer will be immediately destroyed by compositional convection, thereby precluding hydration melting of floor cumulates and formation of a chromite-only-saturated melt. In fact, there seems to be even much more likely that the fluids entering the chamber through fractures in solidified cumulates will be immediately transferred into and mixed with the main magma body, leaving no chance for the formation of a basal layer from the very beginning.

We thus seem to have no viable alternative for resolving the mass balance problem other than in the context of the models involving chromite-only-saturated melts. However, as indicated above, this approach has its own difficulties. In particular, the huge amount of melt that needs to be involved is particularly troubling. In this regards, Naldrett et al. (2012) have come up with a revealing idea that basaltic magmas parental to massive chromitites may contain more Cr₂O₃ than it is usually assumed in the mass balance calculations (~0.10 wt%; normal basalts). Indeed, the vast lateral extent and uniformity of chromitite layers across the entire the Bushveld Complex (Naldrett et al., 2012) indicate that the processes responsible for their formation were dramatic events that occurred synchronously over lateral distances of up to 400 km. Such special events may well be associated with introduction into the chamber of basaltic magmas with the higher-than-normal amount of Cr₂O₃. A combination of data including their MELT-based modelling and available experimental data (Poustovetov and Roeder, 2001) led Naldrett et al. (2012) to a conclusion that the Cr₂O₃ content of the magma parental to chromitites was not <0.25 wt%. This estimation does not look unreasonable because it is sitting at the lower end of the range of Cr₂O₃ content from the chill/quenched rocks in the western (0.23 wt%; Maier et al., 2016) and eastern (0.32 wt%; Wilson, 2012) Bushveld Complex. If we accept the Naldrett et al. (2012) idea, then a mass balance issue become much less severe. For instance, based on phase equilibria experiments and modelling by Drage and Brenan (2023), for a chromite-saturated magma with 0.3 wt% Cr₂O₃, one would need only ~0.4 km thick melt column to produce ~1 m thick chromitite layer (Appendix Table 1).

As for the magmas that escaped from the Bushveld chamber, it should be noted that these magmas should not have erupted as basalt lavas right above the Bushveld Complex as it is commonly believed (Eales, 2000, 2002). Recent seismic reflection- and field-based studies indicate that most basaltic magma travel through the lithosphere mostly sideways as interconnected sill complexes rather than upwards as vertical dykes (Townsend et al., 2017; Magee et al., 2018). It has been shown that such sill complexes can facilitate the magma transport over lateral distances as much as ~4100 km (Magee et al., 2016). If so, the Bushveld-related volcanoes fed by such sill complexes should not necessarily overlie the Bushveld Complex (i.e., a melt source). Rather, the magma that went through the Bushveld chamber may have ended up many hundreds if not thousands of km away as lateral sills or erupted lavas. An interesting possibility is that these magmas may have travelled well beyond the current boundaries of the Kaapvaal craton (into which the Bushveld Complex was intruded) and may now occur as sills/lavas on cratonic parts of other continents (e.g., Wyoming or Pilbara cratons) unless they were later entirely eroded away. Therefore, the chances to find the sills/lavas formed from the Bushveld-related magmas are quite low. For this reason, the current absence of evidence for such sills/lavas should not be taken as evidence of their absence in the past.

To conclude, none of the current approaches attempting to solve a mass balance issue of massive chromitites appears to be perfect, but

some look more problematic than others. In particular, a model by Veksler and Hou (2020) appears to be physically implausible because a basal layer of a water-rich melt cannot be gravitationally stable when it occurs below the dry resident melt. All crystal-rich slurries and sill emplacement models (Mondal and Mathez, 2007; Maier and Barnes, 2008; Voordouw et al., 2009; Eales and Costin, 2012; Maier et al., 2013; Mungall et al., 2016; Jenkins and Mungall, 2018; Maghdour-Mashhour and Hayes, 2021; Barnes et al., 2023; Otto et al., 2024) come into a conflict with the occurrence of magmatic dropstones that rupture and indent chromitite layers. Such field relations are physically impossible for these models because they envisage the formation of chromitites in deep parts of the cumulate pile (Fig. 19). The reactive porous flow models (Mathez and Kinzler, 2017; Nikolaev et al., 2020; Marsh et al., 2021; Boudreau, 2024) face the exactly same problem (Fig. 19) plus, in addition, they are not reconcilable with the lack of silicate rocks that would reveal a state of partial transformation into massive chromitite. Moreover, if many dozens of meters of norite/orthopyroxenites were remelted to produce a layer of chromitites, then angular melanorite dropstones would simply have no chance to survive in this process. But the dropstones exist and show no evidence of metasomatic replacement by chromitites. In such circumstances, the traditional depositional models (Fig. 18) (e.g., Campbell and Murck, 1993; Naldrett et al., 2012; Latypov et al., 2022a) appear to be much more attractive because, even though some of their aspects (such as the removal of a large volume of melt from the chamber) may seem difficult to imagine, they are, at least, physically not impossible. We hope that future research in this field will bring us some better solution(s) to the mass balance issue (i.e., Cr-paradox) in the context of the traditional models.

8.3. Alternative interpretations for chromitite development on the overhanging/ undercutting parts of the chamber floor?

One of the most unexpected recent discoveries in the Bushveld Complex is that some layers of silicate rocks and chromitite may develop on subvertical to overturned sidewalls of the potholes (Latypov et al., 2015, 2017b, 2017c; Chistyakova et al., 2019a, 2019b). As an example, one newly found pothole in which a UG1 chromitite layer passes from the planar floor to vertical sidewalls without changing its thickness is shown in Fig. 23. Even more surprising was a discovery of so-called undercutting sill-like bodies of the Merensky Reef (Latypov et al., 2017c) and UG1 chromitite (Mukherjee et al., 2017) which occur entirely within the footwall rocks (i.e., a few meters below the temporary chamber floor at the time of their formation) (Fig. 24). Again, note that a chromitite seam at the base of the MR does not changes its thickness as it transitions from the planar floor through a subvertical sidewall to completely overturned roof of the undercutting bodies (Fig. 24). The MR undercutting bodies may extend outwards from a pothole for about 300 m at about 20 m below the normal position of the MR. The apophyses vary in thickness from a few cm to one meter and usually have 1–2 cm thick chromitite seams along their both margins (Latypov et al., 2015; their Fig. 19; Latypov et al., 2017b; their Figs. 8, 9). Since mechanical accumulation of individual or clustered chromite grains on “gravity-settling shadows”, especially in the undercutting bodies, is physically impossible/problematic, we have used these findings as a key evidence for the crystallization of these rocks in situ, i.e., directly on the chamber floor (Latypov et al., 2015, 2017b, 2017c; Mukherjee et al., 2017; Chistyakova et al., 2019a, 2019b). This straightforward interpretation of the field observations have not gone unchallenged, however, and several recent studies have come up with alternative explanations (e.g., Cawthorn, 2018; Holness et al., 2023) which are discussed below.

Cawthorn (2018) argued that the viewing of gravity settling as simple downward sinking of crystals in a magma chamber is an oversimplification of the process. Following Schmidt (1952), he proposed that magma in potholes may undergo turbulent convection (a swirling eddy) and therefore sinking and swirling chromite grains could be

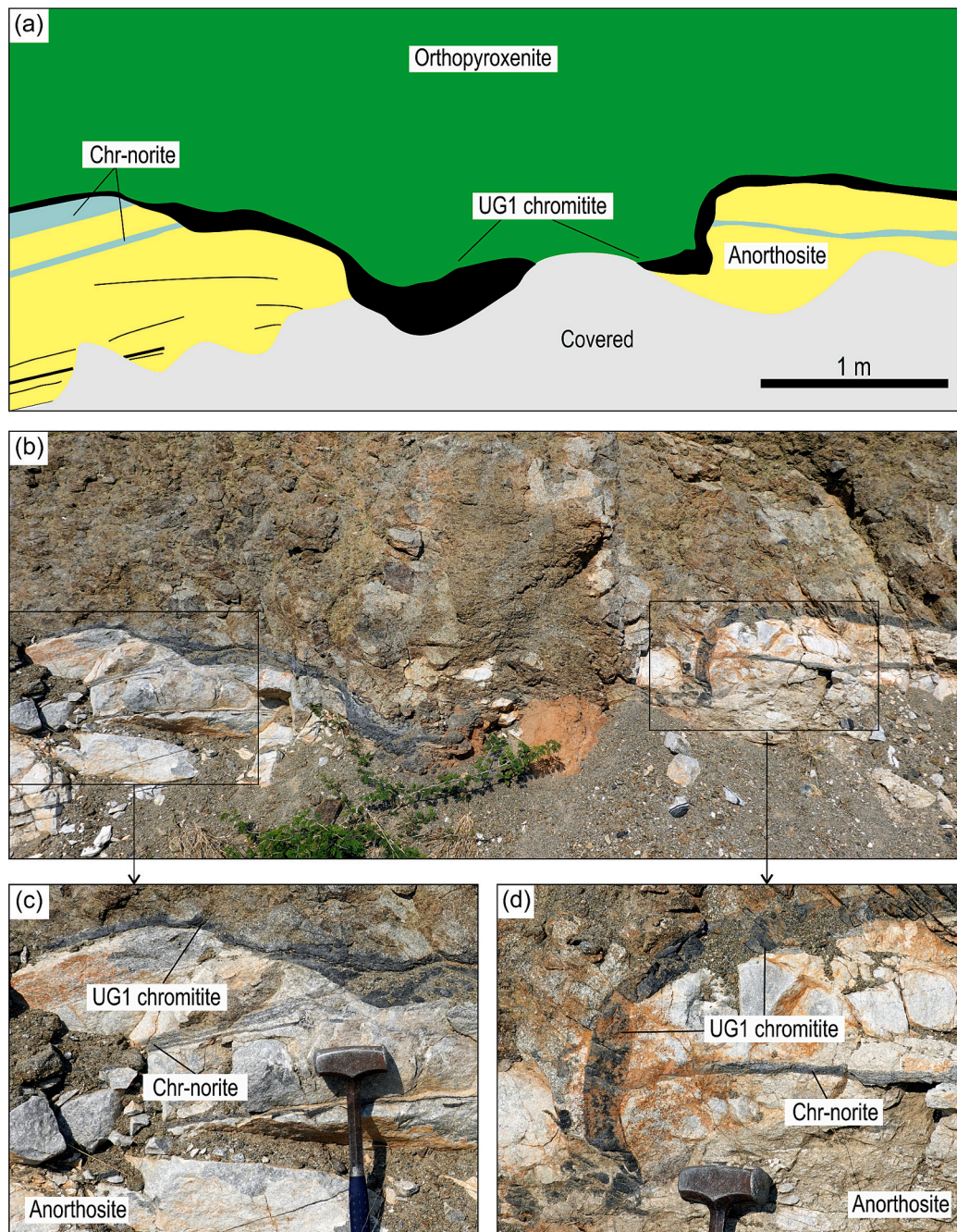


Fig. 23. Sketch (a) and photographs (b-d) a shallow pothole from the UG1 sequence. Waterkop section, Upper Critical Zone, eastern Bushveld Complex. Note that on the right side of outcrop (d) the UG1 chromitite layer transitions from the planar floor to vertical sidewalls without changing its thickness. Some increase in the thickness of UG1 chromitite in the basal part of the pothole (a-b) is attributed to the transport and subsequent redeposition of crystals that originally grew in situ by magma flowing along the floor. The pothole acts as a trap for the re-deposited chromite grains. Note that field relations are not consistent with the origin of the pothole by downward slumping of cumulates, because in this case the UG1 chromitite in the basal part of the pothole would become thinner, rather than thicker.

plastered against any solid surface, essentially by a form of synneusis. He suggested that chromite grains with a low sinking velocity due to their very much smaller grain size could become attached to any surface whatever its spatial orientation. From his point of view, this may explain the occurrence of chromitite layers on vertical and overhanging sidewalls of potholes (e.g., as in Fig. 23). A proposal by Cawthorn (2018) is a very strong claim that cannot be accepted without providing equally strong justification for its physical feasibility. In particular, the segregation of chromite grains from enclosing magma into a layer of monomineralic chromitite implies that the tiny chromite crystals should move independently from (and much faster than) the swirling magma. At our

request, Oleg Melnik (unpublished data, 2018) has done a pilot modelling of convection of a typical basaltic melt (55 wt% SiO₂) within a pothole of ~20 m in diameter using Comsol Multiphysics. The results showed that magma flow in potholes is expected to be laminar and quite slow. Under these conditions, chromite crystals (1 mm in size) would not be able to move separately from the magma to bump against the sidewalls of potholes. For this process to occur the rotational speed of magma in potholes needs to be about 30 rev/s (1.9 km/s, tangential velocity) which is unrealistically high for any natural magmas. Of course, much more rigorous modelling needs to be done to examine this process, but we doubt that the general conclusion of this pilot study

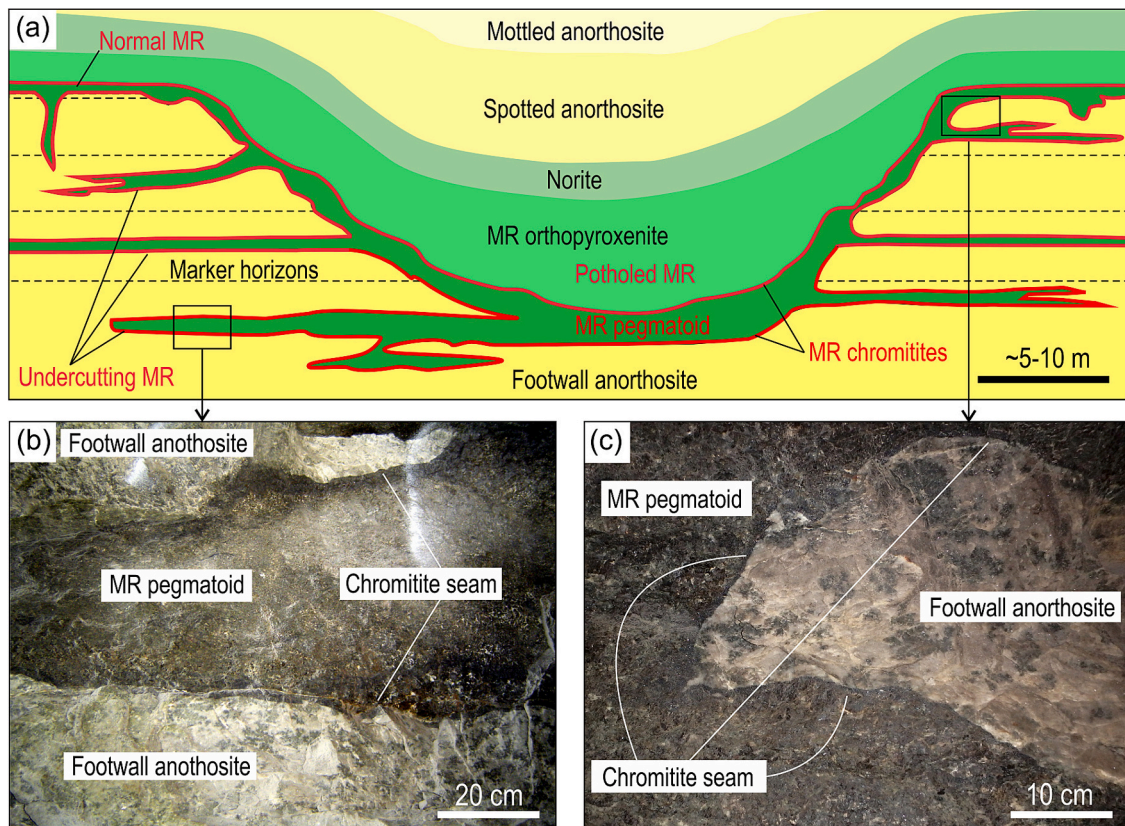


Fig. 24. A generalized morphology (a) of a typical Merensky Reef (MR) pothole at the Impala Platinum Mine, western Bushveld Complex illustrating the transgressive relationship of the MR to its footwall. Note that the potholed MR along the edge of the pothole is accompanied by undercutting MR bodies that are sill-like sulphide- and chromite-mineralized protrusions extending laterally from pothole margins into footwall rocks. Modified from Latypov et al. (2017c). (b) Photograph of an underground exposure showing the undercutting MR with chromitite seams developed along both margins of the sill-like body. Brakspruit pothole, Rustenburg Platinum Mine, western Bushveld Complex. Modified from Latypov et al. (2015). (c) Photograph of an underground exposure showing the undercutting MR with the basal chromitite seam which remains of the same thickness irrespective of the pothole morphology. Karee mine, Lonmin Platinum, western Bushveld Complex. Modified from Latypov et al. (2017c). The assumed position of both underground exposures is shown in (a). Such development of the chromitite seams indicate that chromitite seams are likely produced by in situ crystallization against the sidewalls of the pothole and its undercutting bodies.

would be changed. As for the fact that orthopyroxene crystals are aligned parallel to the sides of MR potholes ('plastering effect') – an observation that was originally taken by Schmidt (1952) as textural evidence for swirling magma – it may be alternatively interpreted as resulting from re-alignment (lamination & foliation) of in situ growing crystals parallel to the margins of a pothole by shear effect of convecting melt (Higgins, 1991). Another weakness of Cawthorn (2018)'s concept is that the swirling eddies in the potholes cannot deposit chromitite seams along the upper and lower contacts of the undercutting MR bodies (Fig. 24b-c) that extend outwards from pothole's margins for many dozens of meters (Latypov et al., 2015).

Based on the textural data, Holness et al. (2023) have proposed that Bushveld massive chromitites are produced by gravity settling of chromite clusters on the chamber floor. To address an issue about chromitites that develop on the pothole's overhangs (Fig. 24; Latypov et al., 2017a, 2017b), they proposed that chromite clusters can be deposited on the overturned margins if they would be brought there by convective magma currents and stick to the sidewalls by cohesive forces. In support of this idea, they mentioned natural examples of the deposition of crystal clusters along the roof of ~130 m Shiant sill and along the steep (up to 80°) margins of the Skaergaard intrusion. However, both cited examples appear to be irrelevant to the problem in question. There is a strong disparity between deposition of crystal clusters along the Shiant's roof and the Skaergaard's margins versus deposition on sidewalls of potholes at the floor of the Bushveld Complex. Both the Shiant and Skaergaard are relatively small intrusions that must have cooled relatively fast along their margins. In these small intrusions, crystals and their clusters can

be, therefore, easily captured by (relatively) rapidly advancing solidification fronts. In contrast, the Bushveld Complex is a supergiant intrusion, and sidewalls of the CZ potholes cannot be regarded as the cooling margins because these potholes occur at the chamber floor ~3–4 km away from the cold country rocks. In fact, the chamber floor may represent, instead, the hottest part of the magma chamber if one accepts an idea about the existence of heat maximum at a crystal-liquid interface (Morse, 1986). A typical solidification rate for such large layered intrusions is only ~1 cm/year (Morse, 2011). If so, crystal clusters, even if they are brought to the overturned sidewalls by convective magma, can hardly be captured by such slowly growing solidification fronts. It seems much more likely that after bumping against the walls the crystals will settle downwards to the base/deepest parts of potholes.

The above conclusion can be perhaps questioned by referring to trough bands in the Skaergaard intrusion which resulted from crystal deposition in channels formed by long-lived ridges perpendicular to the walls and likely created by convective patterns in the main magma body (Irvine and Stoeser, 1978; Irvine, 1983a, 1983b). The steep sidewalls of these trough bands dip at up to 80° but still hold onto the deposited grains from magmatic currents suggesting that crystals of magmatic phases are fairly cohesive and may easily stick to the subvertical sidewalls. The trough bands are almost 3 km above the cold floor of the intrusion and are in the highly evolved part of the intrusion which would have cooled slowly due to the very high contribution of latent heat to the overall enthalpy budget. It could be argued, therefore, that the cooling rate of these rocks was likely similar to that of Bushveld's potholes and

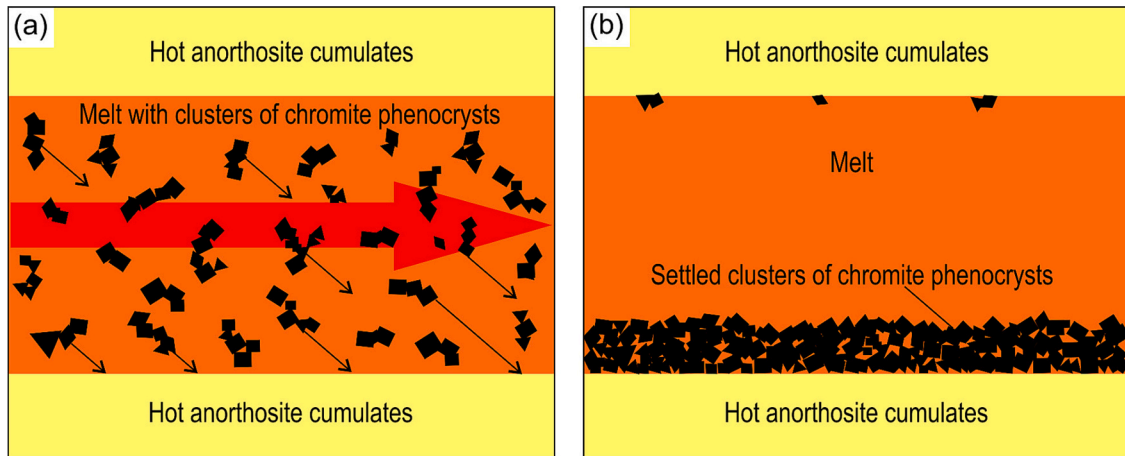


Fig. 25. A cartoon illustrating an idea that if the undercutting MR sill-like bodies are formed from a magma that brings chromite clusters, then most of them will deposit on the floor of the undercutting bodies forming a basal chromite seam. In reality, chromite seams that are developed along the both margins of the sill-like bodies are of the same thickness (Fig. 24).

so could be analogous. However, this analogy is again not relevant because the steep margins of the troughs are not the places where rare individual crystals or clusters from the freely convecting melt are

bumping against the pothole's sidewalls – a scenario that is considered above for the origin of chromitites. Rather, the trough margins are the places where a large mass of crystal-rich mush that collapsed downward

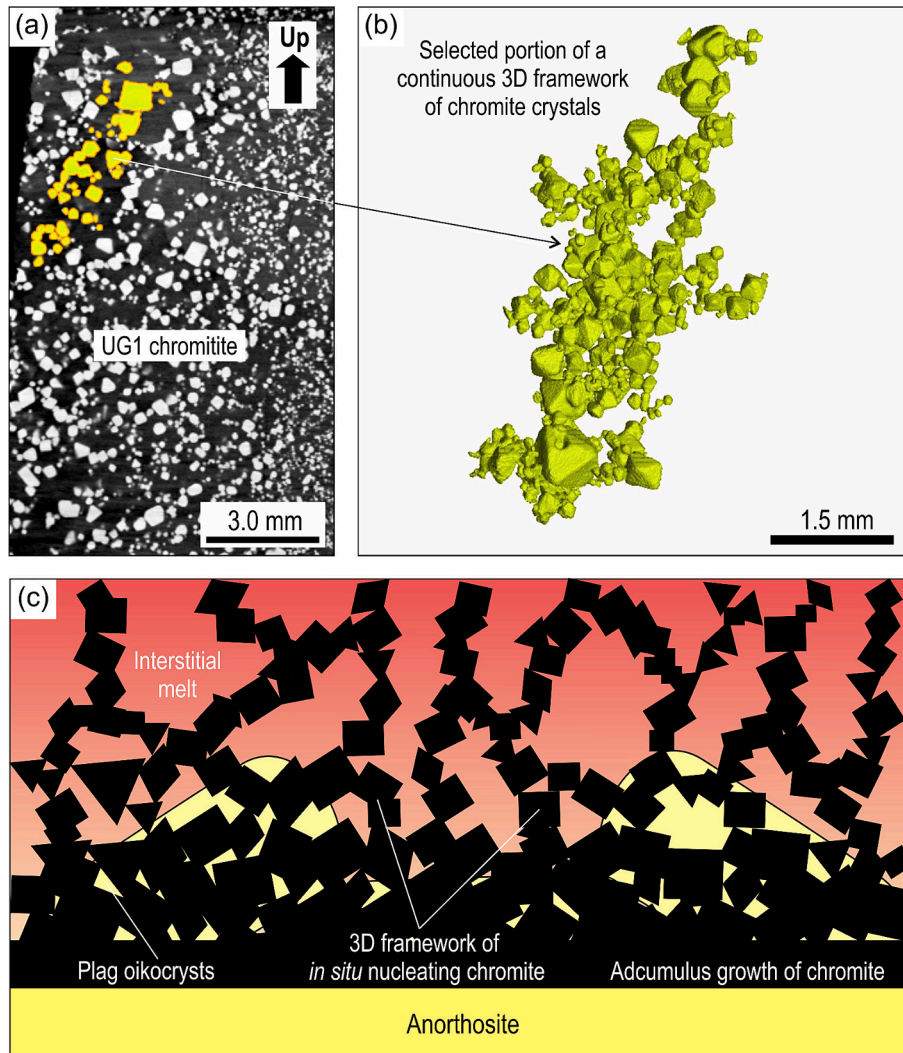


Fig. 26. X-ray computed tomography shows that almost all chromite grains in the UG1 chromitite (a) are interconnected into a continuous 3D framework that consists of many thousands of grains (b). This framework has been attributed to in situ nucleation and growth of chromite grains directly on the chamber floor (c) The framework becomes stable due to early crystallization of and capturing by plagioclase oikocrysts from interstitial melt. Modified from Latypov et al. (2022a).

from the pluton's walls came in contact with the floor cumulates due to filling in the entire trough volume (Vukmanovic et al., 2018; Holness et al., 2022). As a result, the crystals at the margins of the flowing crystal-rich mush were physically pushed (in essence, pressed downwards/outwards) by the entire crystal mush's mass towards the cumulates that form the floor and walls of the troughs, and these marginally

located crystals may, therefore, be easily glued to these rocks by cohesive forces.

The analysis by Holness et al. (2023) has been limited to the case of chromitites that occur on overhangs of the potholes but there are more challenging cases for gravity setting of chromite clusters. These are chromitite seams of the same thickness that develop along both the roof

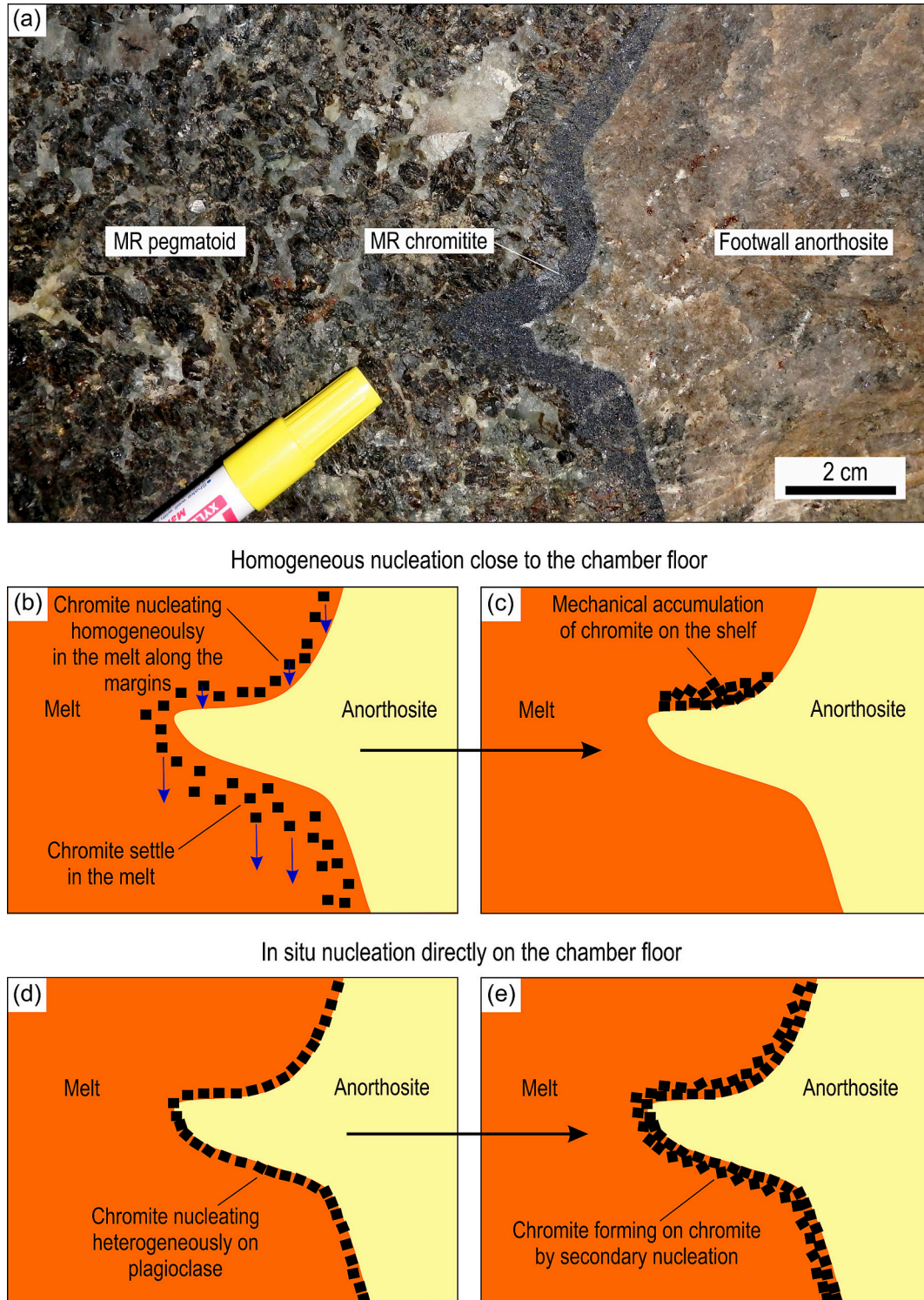


Fig. 27. A chromitite seam developed along the vertical to overturned sidewalls of the MR pothole in the western Bushveld Complex. Modified from Latypov et al. (2015). Homogeneous nucleation of chromite along the chamber floor (b) will result in the accumulation of chromite only on the shelf in the topography of the pothole (c). In contrast, heterogeneous nucleation of chromite on anorthosite (d) followed by secondary nucleation on already grown chromite grains (e) may produce chromitite seams along the pothole margins of any morphology.

and basal contacts of the MR undercutting bodies (Fig. 24; Latypov et al., 2017b). Even if chromite clusters are part of the crystal cargo of inflowing magmas, it seems physically impossible to deposit a similar amount of chromite clusters along the roof and basal contacts of the cavities. In a sill intruded into country rocks, nearly all dense phenocrysts deposit on the chamber floor; only a very small portion of them is captured along the cold roof by a downward-moving solidification front (if cooling is strong enough). This is what is expected in the undercutting MR sill-like bodies if they are formed from a magma that brings chromite clusters into hot cumulates. Most of these clusters, if not all of them, should settle to the base of the undercutting bodies forming a thick basal chromite seam (Fig. 25). But this is not what is observed. The thickness of the chromitite seam (1–2 cm) tends to be the same along the base and roof of the MR undercutting bodies (Fig. 24b). Moreover, even the chromitite seam on the planar chamber floor that is exposed to (i.e., it can ‘see’) the entire overlying magma and, therefore, is expected to be thickest, has the thickness identical to ones along subvertical to overturned position along the roof of the undercutting bodies (Fig. 24c). The similar thickness of chromitite seams irrespective of their spatial orientation testifies against their formation by gravity settling of chromite clusters and can be better explained by in situ nucleation and growth of chromite directly on the chamber floor (Figs. 20; Latypov et al., 2017a, 2017b, 2022a).

8.4. Can a secondary nucleation be a potential solution for in situ growth of chromite?

Based on several theoretical considerations, Holness et al. (2023) have questioned an idea about the formation of chromitite layers by in situ nucleation and growth of chromite on the chamber floor (Latypov et al., 2022a). A major theoretical objection deals with a mechanism of nucleation and growth of chromite on the chamber floor. We have shown using X-ray computed tomography that nearly all chromite grains in the UG1 chromitite are interconnected into a continuous 3D framework composed of many thousands of grains that extend across multiple plagioclase/pyroxene oikocrysts (Fig. 26a–b). Following Campbell (1978, 1987) we have attributed this feature to the preferable appearance of new nuclei on the existing crystals (chromite on chromite) along the base, a process referred to as ‘heterogeneous self-nucleation’ (Fig. 26c). Holness et al. (2023) have shown, however, that this nucleation mechanism is implausible because kinetically it is more favourable to continue growth of existing crystals rather than to nucleate and grow new grains of the same phases. The objection is well justified and, therefore, inspired us to search for a new in situ nucleation mechanism. We tentatively suggest that one of the most likely candidates is so-called secondary nucleation that is normally caused by the seed crystal surfaces - a process that has never been invoked in igneous petrology - but which plays a key role in the formation of crystals in industrial crystallizers where new crystals generally emerge in the presence of pre-existing crystals of the same solute (Qian and Botsaris, 1997; Ahn et al., 2022a, 2022b; Bosetti et al., 2022). Since a secondary nucleation is induced by surfaces of pre-existing parent crystals which act as catalysts for further nucleation, one may logically expect it to actively operate along the crystal-rich solidification fronts of evolving magma chambers (Campbell, 1978; McBirney and Noyes, 1979; Marsh, 1996), such as the chamber floor of the Bushveld Complex (Kruger and Latypov, 2020; Latypov et al., 2022a). We speculate that secondary nucleation may become a potential way to reconcile a concept of in situ crystallization of Campbell (1978, 1987, 1996) for layered intrusions with basic principles of the classical nucleation theory. The detailed discussion of this new working hypothesis is reserved for a separate study.

8.5. Can a homogeneous nucleation at the chamber floor be a potential solution for in situ growth of chromite?

Another way of addressing the nucleation issue raised by Holness

et al. (2023) is to attribute the formation of massive chromitites to a homogeneous nucleation of chromite near the chamber floor. Zoja Vukmanovic (2023, pers. comm) has endorsed this idea to explain the occurrence of chromitite seams along steeply inclined to subvertical sidewalls of potholes (Fig. 24). It should be noted that this type of in situ nucleation and growth has been originally proposed by McBirney and Noyes (1979). In their model, crystals originate by homogeneous nucleation within a magma but at some small distance from the chamber floor (rather than directly on the chamber floor as in the model by Campbell, 1978, 1987, 1996). The model envisages some gravity settling of crystals that are not captured by inward-propagating solidification fronts (McBirney, 1995). Let us consider, as an example, a situation when in situ nucleation and growth of chromite takes place along the vertical to overturned margins of a pothole produces chromite of the MR (Fig. 27a). Chromite may perhaps nucleate homogeneously in response to some cooling effect of the footwall anorthosites on the hot melt emplaced along the relatively colder margins (Fig. 27b–c). However, it is not clear if the footwall rocks may indeed be sufficiently cold to cause cooling and crystallization of the melt inflowing into the chamber. Some cooling may perhaps occur if the erosion will expose the deep (and colder) portions of the cumulate pile (e.g., Kruger and Latypov, 2021). However, a major problem here is that chromite grains are twice denser than the melt and, therefore, they are supposed to settle downwards though the melt. There is no physical reason that may keep chromite grains/clusters hanging in the melt. If so, the settling chromite grains may only be accumulated on the sub-horizontal shelves in the topography of the pothole (Fig. 27c) which is not consistent with field relations (Fig. 27a). To resolve this issue, an alternative scenario may be proposed (Fig. 27d–e). Namely, first chromite grains may nucleate heterogeneously on plagioclase crystals of the footwall anorthosites and then subsequent chromite grains may form by secondary nucleation on already existing chromite grains. In this case, chromite may form

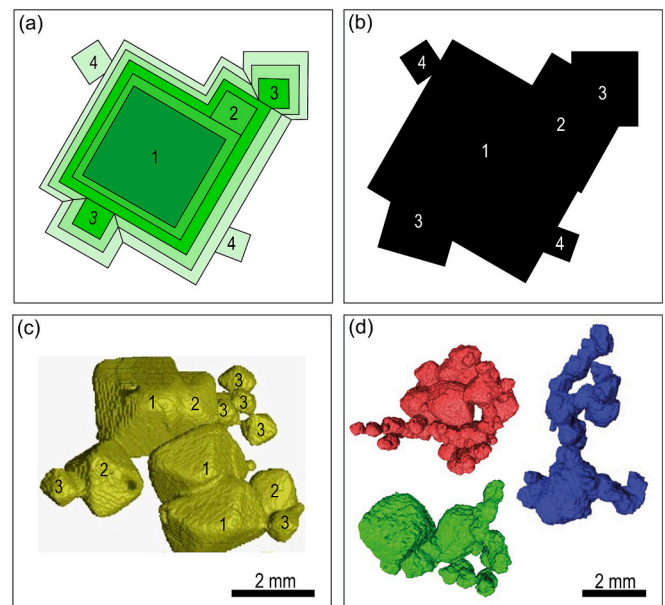


Fig. 28. Cartoon showing the progressive change (a) in grain shape and compositional zonation in a crystal cluster to which grains of silicate minerals are added incrementally during active growth (with the numbers indicating the order of crystal addition to the cluster). In massive chromitite, this cluster is expected to look as in (b) due to opaque nature of and lack of chemical zonation in chromite grains. Modified from Holness et al. (2023). Morphology of chromite clusters in the UG1 chromitite of the eastern Bushveld Complex (c–d) as revealed by high-resolution X-ray computed tomography. Note that the change in shape and size of clustered grains is consistent with the incremental addition of chromite grains to the clusters as indicated for illustration by numbers in (c). Modified from Latypov et al. (2022a).

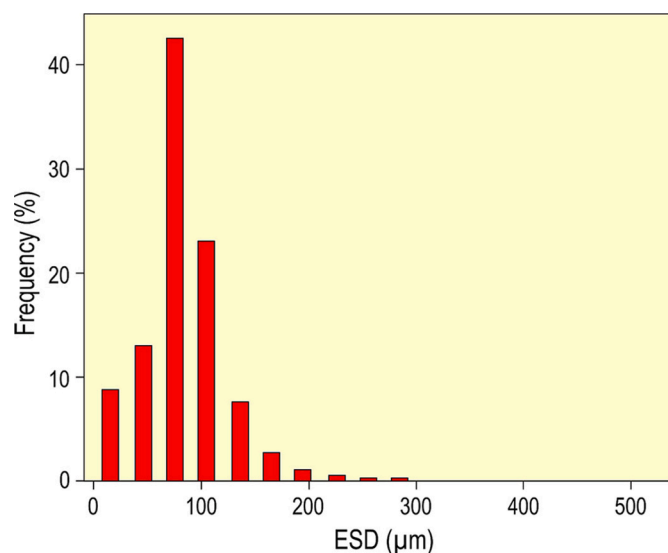


Fig. 29. Histogram showing the size distribution of chromite grains in the UG1 chromitite of the eastern Bushveld Complex (ESD: equivalent sphere diameter) calculated from about 500,000 grains within 1 cm³. Note strong variations in chromite grain sizes. Modified from Latypov et al. (2022a).

crystals that are physically attached to the pothole margins of any morphology (i.e., planar, subvertical or overturned). Thick layers of massive chromitites may be formed in exactly the same way on the planar portions of the chamber floor.

8.6. Are textural data consistent with in situ nucleation and growth of chromite?

Holness et al. (2023) have also used textural observations to question an idea about the formation of chromitite layers by in situ nucleation and growth of chromite on the chamber floor (Latypov et al., 2022a). A major textural objection relates to size and shape of chromite grains in massive chromitites. Holness et al. (2023) have reasonably argued that, if chromite nucleates and grows in situ (i.e., chromite on chromite), then progressively forming chromite grains in the 3D framework are supposed to show corresponding changes in shape and size (Fig. 28a–b). A basic idea is that during solidification the chromite growth may only occur on the interfaces that are exposed to a surrounding liquid. The addition of new chromite grains prevents further growth of the earlier-formed chromite grains (i.e., substrate) on that part of their surfaces, and as a result, the substrate chromite grains are expected to have a different shape (become increasingly non-equant) compared to those that joined the cluster later (Fig. 28a). This also implies that chromite grains must show a wide variation in grain sizes, with the earlier-formed grains being larger and the younger ones smaller in size. Our re-examination of the 3D XRF tomography data of the UG1 chromitite (Latypov et al., 2022a) shows, however, that the expected changes in shape and size are observed (Fig. 28c–d). In a similar way, the histogram summarizing the size distribution of chromite grains from the UG1 chromitite (Fig. 29) clearly show that chromite reveals a wide variation of grain sizes – from 10 to 20 μm to 200 μm, with the maximum being at about 100 μm (and some even are up to 200–300 μm). If upscaled to cm level of silicate mineral sizes (e.g., plagioclase or orthopyroxene), then the chromite grains would range from 0.1 to 0.2 mm to 2–3 cm, with the maximum being at about 1 cm. These grain sizes are quite substantial. One more textural objection is that in situ nucleating chromite grains are supposed to show epitaxial relations which are not observed in the Bushveld chromitites (Zoja Vukmanovic's pers. comm., 2023). This seems to be a valid objection against 'heterogeneous self-nucleation', but not against a secondary nucleation which does not imply such crystallographic

relations between crystals. We thus argue that a wide range of chromite grain sizes and systematic changes in their shapes in clusters are fully consistent with progressive in situ nucleation and growth of chromite in the solidification front at the Bushveld chamber floor.

8.7. Some challenges of a cluster model in explaining the origin of massive chromitites

Textural studies have revealed the extremely low packing density of chromitites (e.g., ~27% in UG1 chromitite) which is more than twice lower the density that is expected from mechanical accumulation of separate, non-interacting chromite grains (~60%) (Latypov et al., 2022a). For this reason, the models involving accumulation of isolated grains, either via crystal settling (Irvine, 1977; Kinnaird et al., 2002; Leshner et al., 2019; Spandler et al., 2005; Wieser et al., 2019) or by kinetic sieving in a crystal-laden slurry (Maier et al., 2013; Mondal and Mathez, 2007; Mungall et al., 2016) cannot be correct. There is, however, one potential solution for this issue in the context of gravity settling models. This is to assume that the low packing density of chromitites has been achieved via mechanical accumulation (e.g., gravity settling) of chains or aggregates of crystals, rather than single crystals (Holness et al., 2023). We have already discussed above that the cluster model has a problem to explain field relations of chromitites, particularly, their development on overhanging/undercutting parts of potholes (Figs. 24–25). Here we will focus on some other issues that this model needs to address to become a viable model for the origin of massive chromitites.

The model assumes that chromite clusters are formed *before* their settling on the chamber floor. It should be stressed that the formation of chromite clusters by aggregation of initially isolated grains in the convecting melt (or synneusis; Dowty, 1980) has been only logically inferred to explain the low packing density of chromitites and the lack of fining-upwards in grain size in chromitite layers (Holness et al., 2023). Textural evidence for the chromite clustering before their settling is not yet available. It should also be realized that rounded shape of chromite grains does not allow the EBSD method used in this study to get any meaningful conclusions regarding the timing of chromite clusters formation – i.e., *before* their gravity settling on the chamber floor (Holness et al., 2023) versus *during* in situ nucleation and growth of chromite grains on the chamber floor (Latypov et al., 2022a). In other words, the synneusis of chromite crystals cannot be confirmed by the conventional EBSD method. Thus, some independent field, textural or chemical data are strongly required to support the postulate that chromite clusters form first in the convecting magma and only then settle on the chamber floor. This is a crucial issue especially in the view of the field evidence (e.g., Figs. 23–24; Latypov et al., 2017a, 2017b) that are not easily reconcilable with gravity settling and can be explained with less contradiction by the formation of chromite clusters directly on the chamber floor (e.g., Fig. 27).

One also needs to demonstrate that the formation of chromite clusters by synneusis is a physically viable process in basaltic magma chambers. There is no question that normal silicate minerals, such as plagioclase, may form clusters in this way (Philpotts et al., 1998; Philpotts and Dickson, 2000). This is because a large amount of separate plagioclase crystals forming from a basaltic melt (it contains ~50 wt% of normative plagioclase) ensure high chances for these crystals to bump against each other in a convecting melt. This logic is not valid, however, for chromite because basaltic melts contain only 0.1–0.3 wt% of Cr₂O₃ (Murck and Campbell, 1986; Naldrett et al., 2012; Drage and Brenan, 2023) and may only produce a negligible amount of small chromite crystals. For this reason, in a large magma body, the scarcely nucleating chromite crystals are expected to be widely separated from each other, perhaps, by several meters of chromite-free melt. Under the low Stokes Law velocities, these rare and small chromite crystals will follow the flow lines in the convecting magma, and so unless the magma is flowing turbulently, there is very limited chance for the tiny grains in the dilute

suspension to bump into one another to form a cluster. Nonetheless, chromite clusters are indeed occasionally observed in basaltic lavas. How can they be formed? There are two potential possibilities here. Firstly, these chromite clusters may be part of a crystal cargo derived from disrupted chromitite cumulates in the staging chamber and entrained with ascending magmas towards the Earth's surface. This would be in line with the current realization that many of the 'crystal clots' and glomerocrysts observed in volcanic rocks are samples of crystal-rich mushes from transcrustal magmatic systems (e.g., McIntire et al., 2019). Secondly, chromite clusters may, perhaps, form in the feeder channels (Latypov and Chistyakova, 2020, 2024b) if magma would have been flowing turbulently to provide higher chances for chromite grains to meet each other. In both cases, this would mean that Bushveld's chromitites are produced from chromite-bearing slurries replenishing the chamber, an idea that needs to be built into the cluster model (Holness et al., 2023) and reconciled with constraints imposed by field observations (Figs. 23–24) and other problematic aspects of slurry-rich models.

This logically brings us to a list of petrogenetic issues that have not yet been addressed in the cluster model (Holness et al., 2023) compared to other existing concepts for the origin of massive chromitites (Irvine, 1977; Campbell and Murck, 1993; Naldrett et al., 2012; Maier et al., 2013; Latypov et al., 2017b, 2018, 2022a; Jenkins and Mungall, 2018). These are: (a) how and where a parental magma saturated in chromite-

alone has been produced? (b) how does the cluster model solve mass balance issue regarding a low Cr-content in melts? (c) where exactly have the chromite clusters been produced (in a deep staging chamber, feeder conduits or a shallow-level chamber), (d) which re-occurring process is responsible for the origin of multiple chromitite layers in the CZ and how this process is related/reconciled with the formation of cluster chromite, and (e) how can the cluster model explain the lack of chemical reversals at the level of the chromitite layers? We encourage authors of the cluster model to consider incorporating a discussion of these essential topics into their concept, thereby ensuring that it is comparable with other models in terms of the comprehensiveness of the treatment of massive chromitite formation in layered intrusions.

8.8. Was the chamber floor non-horizontal during the formation of massive chromitites?

Most petrogenetic models, including ours (Latypov et al., 2017b, 2022a), implicitly assume that the inward-growing chamber floor on which chromite has been deposited was nearly horizontal. For the case of the Bushveld Complex, this assumption seems to be well justified by palaeomagnetic data indicating that the intrusion solidified close to horizontally (e.g., Letts et al., 2009). Cawthorn (2018) has, however, questioned this assumption indicating that local topographic irregularities of the instantaneous chamber floor exist in the Bushveld chamber

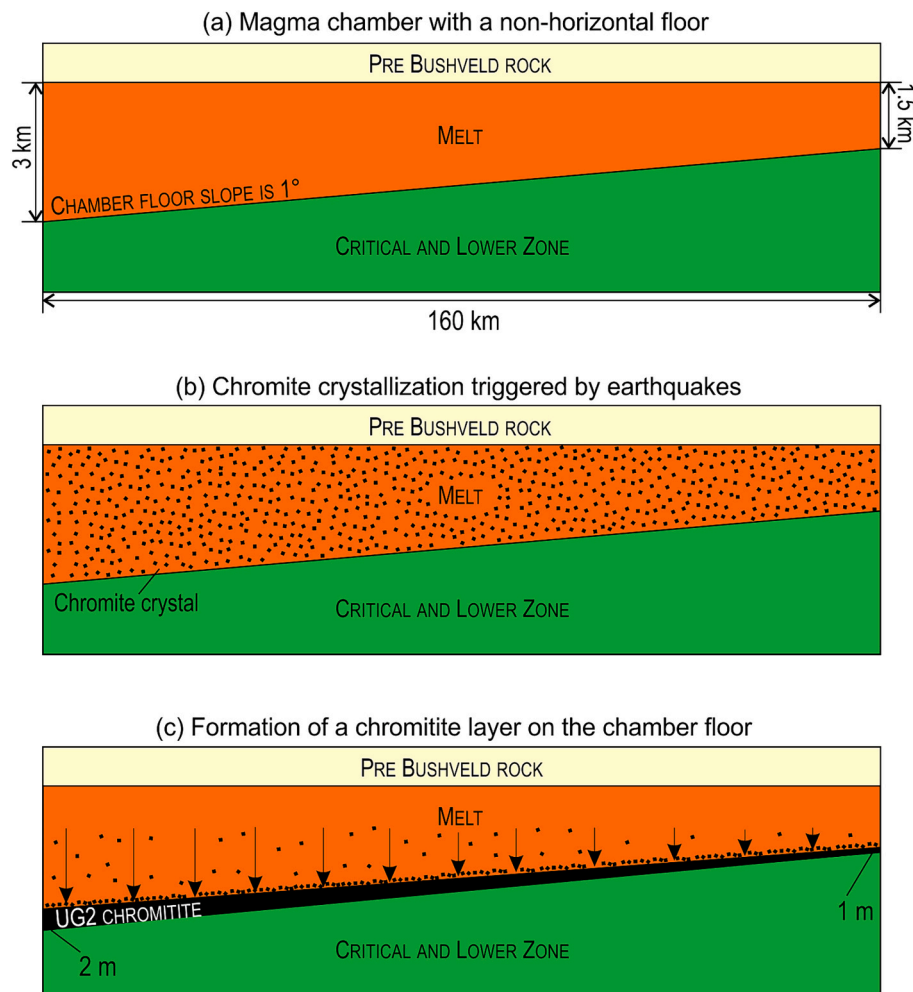


Fig. 30. Graphical testing of an idea by Cawthorn (2018) that the temporary chamber floor at the time of the UG2 chromitite formation has topographic relief >1.5 km along the 160 km strike length in the Bushveld chamber (a). If this would be the case, then gravity setting of chromite from the overlying magma column (b) would result in a systematic increase in the thickness of UG2 chromitite with depth (c). In reality, however, the UG2 chromitite shows uniform thickness of ~1 m across the entire Bushveld Complex (Cawthorn, 2018).

despite its solidification as a nearly horizontal body. In particular, he used the chemistry of orthopyroxene to identify one of such irregularities. Based on the original data by [Maier and Eales \(1997\)](#), he showed the Al_2O_3 content of orthopyroxene in the UG2 chromitite unit change from 1.5 to 1.1 wt% along a strike length of 160 km in the western Bushveld Complex. Since the Al_2O_3 content of orthopyroxene is sensitive to pressure (0.2% increase per km depth), such changes were taken by [Cawthorn \(2018\)](#) to indicate that there was topographic relief >1.5 km along the 160 km strike length in the temporary floor of the western Bushveld chamber ([Fig. 30a](#)). Our model implies that the UG2 chromitite (and MR) are likely produced by replenishment of the chamber by dense magma that spread along the chamber floor as basal flows of a few hundred meters thick ([Latypov et al., 2017b, 2022a](#)). [Cawthorn \(2018\)](#) has reasonably argued that such basal flows would fail to flood over >1.5 km of topography of the chamber floor and, therefore, could not have produced UG2 chromitite.

It is instructive to start with a closer look at the predictions of the model put forward by [Cawthorn \(2015\)](#) to explain the origin of UG1/UG2 chromitites. The model does not rely on replenishment of the chamber by chromite-only-saturated magma. Instead, it prefers to transform the resident melt in the chamber into chromite-only-saturated one by shock waves associated with tectonic instability triggering earthquakes. The shock waves are supposed to initiate homogeneous nucleation of chromite grains (as a mineral with the simplest structure; [Wager, 1959](#)) everywhere in the magma chamber followed by settling of chromite grains onto the chamber floor to produce a continuous chromitite layer ([Cawthorn, 2015](#)). The claimed advantage of this model is that gravity settling of chromite from a very thick column of magma may blanket the instantaneous chamber floor regardless of its topographic elevation. The story seems logical, but it comes into a serious conflict with one of the most fundamental features of the UG2 chromitite, namely its uniform thickness of ~1 m across the entire Bushveld Complex ([Cawthorn, 2018](#)). This uniformity apparently means that the same amount of parental melt was involved in the formation of the UG2 chromitite everywhere in the chamber. This is at odds, however, with the postulated sloping floor of the magma chamber which predicts that the amount of parental magma (its vertical extent) must increase with moving towards the deeper parts of the chamber ([Fig. 30b](#)). If so, the thickness of the UG2 chromitite to form by setting of chromite from the overlying column of magma ([Fig. 30c](#)) must also systematically increase with depth (by a factor of at least 2 if the total magma column is about ~3 km). This is not, however, the case ([Cawthorn, 2018](#)).

We conclude therefore from the above analysis that an idea about the topographic relief of over 1.5 km at the temporary chamber floor in the western Bushveld Complex cannot be correct. Some other reason, not necessarily pressure-related, must be responsible for the reported systematic variations in the Al_2O_3 content in orthopyroxene along a strike length. One potential reason is the co-crystallization of orthopyroxene with an increasing amount of plagioclase that may also result in a decrease the Al_2O_3 content in orthopyroxene; this alternative possibility has been not evaluated by [Cawthorn \(2018\)](#). The model in question has several other problematic aspects. For instance, if the chamber was closed during the formation of massive chromitite, then it is not clear how the mass balance issue can be addressed and how the model with turbulently convecting magma in the potholes fits into this story ([Cawthorn, 2018](#)). In fact, it is not even clear how in the closed system one can produce the potholes in the first place. No erosional force is envisaged in the model to produce the potholes in the chamber floor ([Cawthorn, 2018](#)). We would thus argue that the uniform thickness of the UG2 chromitite across the entire extent of the Bushveld Complex should be regarded as strong evidence for its formation from the same volume of parental melt (likely flowing through the system) throughout the entire chamber. This can be best understood if the temporary floor was sub-horizontal across the entire chamber, with numerous pulses of the inflowing magma entering the chamber and spreading across its entire floor in a matter of few weeks so that in situ crystallization of

chromite would have been taking place at the same time everywhere ([Latypov et al., 2017b, 2022a](#)).

8.9. Does chromite chemistry provide evidence against the formation of chromitites from chromite-only saturated melts?

[Barnes et al. \(2023\)](#) have recently examined minor and trace element contents of chromite across layers of massive chromitites from several layered intrusions, including the Bushveld Complex. They have found that, in UG1/UG2 chromitite layers, the concentrations of elements that behave compatibly in chromite (V, Ga, Mn, Co, Ni, Zn, etc.) do not systematically decrease from the base towards the top of these layers as would be expected from fractional crystallization of a chromite-only saturated melt. Very little systematic variations in the concentrations of these elements are found across the layers. From this they concluded that the formation of the UG1/UG2 chromitites cannot be explained in the frame of the models that invoke parental magmas saturated solely in chromite (e.g., [Irvine, 1977](#); [Murck and Campbell, 1986](#); [Lipin, 1993](#); [Spandler et al., 2005](#); [Naldrett et al., 2012](#); [Latypov et al., 2017a, 2022a](#)). Instead, they favour a model where chromite and silicate minerals crystallized somewhere at a staging chamber in cotectic proportions (~2:98). Subsequently, the crystal-rich slurry consisting of chromite, silicates, and melt were emplaced into the shallow Bushveld magma chamber. During the emplacement the chromite and silicate minerals were separated from each other due to viscous particle flow to form massive chromite layers buried beneath a thick pile of a silicate-rich mush. A justification behind this choice is that chromite grains from the same crystal-rich slurry would show no systematic variations in chemistry across the chromitite layers ([Barnes et al., 2023](#)). However, indentation and rupturing of UG1/UG2 chromitite layers by magmatic dropstones ([Figs. 8–16](#)) indicate that these layers were forming directly on the instantaneous chamber floor, rather than at some depth in a crystal mush. It is also clear that this should have happened from chromite-only saturated melt so that chromite deposition would not have hampered by any other minerals in the chamber.

The chemical objection by [Barnes et al. \(2023\)](#) against the models involving chromite-only-saturated melts appears to be a logical mistake. The lack of systematic upward evolution in compatible elements across the chromitite layers is only valid for the closed system in which chromite crystallizes from a single pulse of chromite-only-saturated melt. But this is certainly not the case. All the available data – field ([Latypov et al., 2017b](#); [Pebane and Latypov, 2017](#)), textural ([Kaufmann et al., 2019](#); [Hasch and Latypov, 2021](#)) and chemical ([Hiemstra, 1985, 1986](#); [Cawthorn, 1999](#); [Junge et al., 2014](#)) observations show that the UG1/UG2 chromitite layers (and all other chromitite layers from MG/LG group) are composed of multiple distinct sublayers crystallized from separate magma pulses. In addition, a mass balance dictates that each of these chromitite layers may only be produced from a large volume of basaltic magma, likely flowing through the systems (i.e., entering the chamber, depositing chromite on the chamber floor and are then pushed away from the chamber by new magma pulses). Under such open-system conditions, one cannot expect any systematic trends in chemistry of chromite in these layers, especially in terms of trace elements. We would thus contend that no variations across the layers of chromitites are not only expected for crystallization from chromite-only-saturated melts but is an absolute must of this model. A lack of variations may be considered as a hallmark of the formation of chromitite from a large volume of melt in a flowing through system. Some vertical variations in V ([Naldrett et al., 2012](#)) and Ti ([Kaufmann et al., 2019](#)) that are reported from massive chromitites of the Bushveld Complex are likely related to some changes in composition of the flowing-through melt, rather than internal differentiation of the parental melt in the chamber.

9. Conclusions

The discovery of magmatic dropstones and their physical

relationships with the floor cumulates in the Bushveld Complex allow us to draw some important conclusions with respect to the architecture of a magma chamber and its internal magma dynamics as well as the nature of the processes responsible for the formation of layers of massive chromitites. Most important among them are as follows:

- (1) The Bushveld chamber had initially the sequence of cumulates that grew from roof downwards but was subsequently destroyed and collapsed as angular blocks (i.e., magmatic dropstones) on the inward-growing floor of the magma chamber; this study reports such blocks from the CZ, but they also occur at other horizons throughout much of the stratigraphy of the complex, e.g., in the MZ and UZ;
- (2) The Bushveld chamber contained an internal reservoir of a resident melt through which the magmatic dropstones were settling downwards on the chamber floor. The reservoir was bounded by the inward-propagating solidification fronts at the floor and roof sequences. The thickness of the internal reservoir at the time of the formation of the CZ is not possible to evaluate from the current data, but considering that the MZ's melt column was ~5 km thick (Latypov et al., 2022b), it may have been in the similar range;
- (3) The way the magmatic dropstones indent older layers beneath and are covered by younger layers above indicate that the interface between the top of the cumulate pile and the overlying resident melt at the time of the CZ formation, was almost invariably sharp, clean and well-defined; in other words, there was mostly a solidified hardground at the chamber floor (Latypov et al., 2024a);
- (4) Field observations involving magmatic dropstones that deform, penetrate, and rupture layers on the chamber floor provide abundant evidence for the igneous layering to have been fully developed right up to the crystal-liquid interface and for the cumulates beneath this interface being usually coherent (perhaps >50% crystalline) and are able for both ductile and brittle deformation;
- (5) The localized indentation and disruption of chromitite layers by magmatic dropstones is clear evidence for the formation of massive chromitites through deposition of chromite crystals directly on the chamber floor (as part of igneous layering formation) in the absence of any other phases in the chamber; this implies that the resident melt must have been saturated in chromite as the only liquidus phase;
- (6) Field relations do not allow to infer how exactly chromite has been deposited on the chamber floor – by gravity settling of chromite grains/clusters or in situ crystallization on the chamber floor. However, field and textural observations from previous studies (e.g., development of chromitite seams on overturned sidewalls of potholes and especially along the margins of undercutting bodies) as well as theoretical arguments in this study are more indicative of in situ nucleation and growth of chromite on the chamber floor;
- (7) The occurrence of magmatic dropstones at certain stratigraphic horizons (e.g., UG1/UG2/MG2 sequences) is consistent with repeated transient episodes of crustal instabilities that triggered the disruption of the roof sequence. The instability is likely associated with earthquakes that usually accompany magma chamber replenishment events. We infer therefore that it is the replenishment of the chamber with new pulses of chromite-only-saturated melt that is likely responsible for the formation of massive chromitites;
- (8) The ascending basaltic melts likely become saturated solely in chromite due to a decrease in lithostatic pressure. The saturation may be caused by either a shift (Latypov et al., 2018) or expansion (Drage and Brenan, 2023) of a chromite stability field, or both. The pressure decrease scenario is advantageous over other available mechanisms because no melts can escape a reduction in lithostatic pressure while ascending from the deep magma sources towards the Earth's surface;
- (9) The block-and-layering structures provide one of the strongest piece of evidence against the formation of massive chromitites through hydrodynamic sorting of crystal-rich slurries (e.g., Maier et al., 2013), sill-like emplacement (e.g., Mungall et al., 2016) and reactive porous flow (e.g., Boudreau, 2024). The indentation and disruption of chromitite layers by magmatic dropstones rule out the formation of chromitites in deep parts of the cumulate pile. A problem of massive chromitites in the Bushveld Complex can therefore be considered as firmly resolved – they were produced from the chromite-only-saturated magma directly on the temporary chamber floor;
- (10) Large, roof-derived magmatic dropstones impacting the igneous layering in the UG1, UG2, MG2 sequences reject the possibility of the formation of these units as the late-stage sills emplaced into pre-existing cumulate pile. This means that high precision out-of-sequence ages of the UG1, UG2, MG2 and some other units (Mungall et al., 2016; Scoates et al., 2021) do not reflect their emplacement ages and require some alternative interpretation (Latypov et al., 2017a, 2024b; Latypov and Chistyakova, 2022);
- (11) The recently proposed cluster model for the formation of massive chromitite (Holness et al., 2023) have some support from textural observations but needs to be reconciled with field observations (e.g., undercutting chromitite bodies), and address several petrogenetic issues (e.g., origin of parental chromite-only-saturated melt, a mass balance issue, a mechanism for the formation of chromite clusters) in order to attain a status of a viable hypothesis;
- (12) Finally, the above conclusions are primarily relevant for the thick massive (stratiform) chromitites of the Bushveld Complex and some other layered intrusions. Some millimetre-thick chromitite seams along the contact of layers of contrasting composition (Scoon and Costin, 2018; Marsh et al., 2021) and reaction rims around the xenoliths in the Boulder Bed (Smith et al., 2023) in the Bushveld Complex may be of non-magmatic origin (e.g., reaction/restitic/metasomatic). The origin of massive chromitite layers and thin rim-like chromitite seams may therefore be considered separately.

In conclusion, our study re-emphasizes the obvious fact that ground truth information from layered intrusions is a very powerful tool for improving our understanding of the architecture and internal dynamics of basaltic magma chambers (Wilson and Larsen, 1985; Irvine et al., 1998; Latypov et al., 2022b). In particular, our new field observations and inferences from the block-and-layering structures in the Bushveld Complex provide strong evidence for the classical concept that considers layered intrusions as big magma tanks, i.e., large, long-lived, and crystal-poor melt bodies that gradually crystallize in situ from margins inwards through thin solidification fronts (Wager and Brown, 1968; Wilson and Larsen, 1985; Parsons, 1987; Morse, 1988; Cawthorn, 1996; Naslund and McBirney, 1996; Irvine et al., 1998; Charlier et al., 2015; Kruger and Latypov, 2020; Latypov et al., 2022a, 2022b). The field and textural observations from layered intrusions will likely continue to occupy geologists' imagination and will be a continuing source of fascination for igneous petrologists from many generations to come (Latypov et al., 2024b).

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.earscirev.2024.104858>.

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