

# Geological interpretations from the PGE distribution in the Bushveld Merensky and UG2 chromitite reefs

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The vertical distribution of platinum group elements (PGE) in the Merensky and UG2 chromitite reefs is highly variable, but does follow certain general patterns that allow the testing of geological models. In the Merensky Reef, historically popular geological representations and interpretations have suggested that the mineralization occurs in a pegmatitic pyroxenite bounded by two chromitite layers. However, there are now enough published exceptions to this relationship to expose its fallacy. The vertical distribution of the PGE depends largely upon the separation between these two chromitite layers or the absence of one chromitite layer. Where separation is small or only one chromitite layer is present, there is considerable PGE mineralization in the footwall below the pyroxenite and lower chromitite layer. With increasing separation between the two chromitites the mineralization occurs progressively higher in the succession, and tends to track the upper chromitite layer. The view presented here is that the entire Merensky package is comprised of three events of magma emplacement, in which each has produced variable reaction with, and erosion of, the footwall, causing recrystallization of pyroxenite into pegmatitic pyroxenite. The magma then produced a chromitite layer followed by pyroxenite. Each chromitite layer is associated with some mineralization which was extensively complemented by the subsequent formation of a sulphide liquid carrying PGE that trickled down through about 1 m of permeable crystal pile. The sulphide liquid formed after, not coincident with, the pyroxenite layers.

The distribution of PGE in the UG2 chromitite shows up to three upward depletion sequences of PGE, also suggesting three events. The PGE are associated only with the chromite. The contribution from a sulphide liquid is considered to have been minimal for two reasons. Cu and Ni are extremely low in the UG2 which would not be expected if sulphides had been present. Also, by analogy with the Merensky, downward trickling of sulphides would have caused some PGE to be present in the footwall. There are some PGE in the footwall, but only where remnants of chromitite occur, and the hangingwall is lacking in PGE.

In the northeastern Bushveld the UG2 chromitite is thinner than elsewhere, and there is a UG3 chromitite layer, not present elsewhere, that has a high Pt/Pd ratio. It is surmised that in the rest of the Bushveld the UG3 only appears to be absent because has merged with the UG2 to produce the high Pt concentration at the top of the composite UG2. The non-accumulation of the package of pyroxenite-norite-anorthosite between the UG2 and UG3 in the northeastern Bushveld, which is not seen elsewhere, creates an interesting challenge to the formation of silicate layering in the complex. The mineralization in the Platreef is not addressed here.

## Introduction

It is well known that the platinum group element (PGE) mineralization in the Merensky and UG2 chromitite reefs is restricted to a very narrow vertical interval of rocks (Wagner, 1929). In the UG2 chromitite there is essentially no PGE above or below the chromitite layer that rarely exceeds 1 m thick. In the Merensky Reef a very large proportion of the PGE is confined within 1 m vertically in many cases, although there are some thicker intervals, and occasionally the mineralization becomes split with impoverished intervals between. Even within the relatively narrow reefs there is considerable variation in the exact location of the highest grades with respect to different host rock types. Variation in the silicate mineral rock types and PGE distribution has led to the definition of different reef

facies (e.g. Wagner, 1929; Viljoen, 1999). This paper examines the published data on the distribution of PGE within the Merensky and UG2 reefs and uses this information to discuss models for the origin of the mineralization. There are geological and structural anomalies in the reefs, such as potholes, in which the mineralization may have a different distribution from the surrounding layered reefs, but these issues will not be discussed in detail here (see Carr *et al.*, 1994).

## Historical descriptions of the reefs and grade distribution

In the first five years after Merensky's discovery in 1924, the rate of acquisition and publication of information about the Merensky Reef was truly astounding. Wagner's

summary in (1929) contains an enormous wealth of information from the eastern, western and northern limbs. Details have been added, but no fundamental change in his overview has emerged. Because of the Great Depression further exploration and mining ceased and did not resume until 1933 and remained at a very low level until 1948 (Edwards and Silk, 1987). Thereafter, competition and secrecy meant that little further information on the reef was published. A major review by Vermaak (1976) provided much information on the host rocks, but the distribution of PGE within the reef was not included. The publication of the *Mineral Deposits of South Africa* by the Geological Society of South Africa in 1986 provided a further significant increase in knowledge from all the platinum mines, but it was only the Rustenburg Platinum Mines for which detailed distribution of PGE within the reef was presented (Viljoen and Hieber, 1986; Viljoen *et al.*, 1986a; Viljoen *et al.*, 1986b). Subsequently, more quantitative data have become available for most of the mines.

The publication of such data merely confirmed the observations of Wagner (1929), who recognized various facies of the reef for which the distribution of the PGE was not confined to a specific rock type. The different distinct facies are shown in Figure 1. He did not quantify the detailed PGE abundance distribution, usually quoting average grade over a mining thickness. More quantitative data for analyses of short vertical intervals through the reef have been provided in many subsequent publications, and are included in Figure 1. Wagner (1929) produced much information for the Rustenburg area, since it was recognized as the area with the most consistent mineralization and most likely to be exploited. Vermaak's (1976) review of the reefs, again focused on the Rustenburg mines. As a result, many subsequent publications have considered the thin reef at Rustenburg to be typical of the Merensky around the entire Bushveld Complex. Possibly the most important omission in many of these subsequent contributions is the recognition about the distribution of the mineralization, summarized by Wagner (1929; page 102), as follows: 'Where [the reef is] thick, the platinum values are confined to its uppermost portion.' Wagner also stated (page 137) that where the reef is thin, as in the Kroondal facies, which can occur on any of the Impala, Rustenburg and Lonplats Mines, significant mineralization occurs in the footwall anorthosite. This variability in the vertical location of the PGE mineralization relative to the different rock types is fundamental to understanding the genesis of the ore as discussed below.

In contrast, to the non-publication of details of grade distribution for the Merensky, several important studies on the UG2 chromitite became available from the outset of mining (Hiemstra, 1979; 1985; 1986; McLaren and de Villiers, 1982), albeit mainly from one mine, Lonplats.

### Detailed illustrative vertical sections through the Merensky Reef

Assorted generalized plots of the grade distribution were compiled by Viljoen and Schürmann (1998) and Cawthorn *et al.* (2002; 2005), but to illustrate the irregularity of distribution vertically the following specific sections are described. Detailed vertical sampling needs to be undertaken to visualize the vertical distribution. Mining and exploration companies routinely analyse samples 20 cm thick, or occasionally slightly less, where obvious lithological contacts occur. More detailed studies are

usually undertaken only for research and academic studies. Publications from mining companies, compiled in Figure 1, tend to be more generalized, but are the average of a great many analysed sections. Examples of several very detailed studies of sections through the reef on different mines have been given by Mitchell and Scoon (2007) and Naldrett *et al.* (2009). What these different profiles show is that the mineralization can be very irregularly distributed vertically, especially for the thicker intersections of Merensky Reef, creating challenges to correct selection of the section to be mined. However, as discussed by Viljoen *et al.* (1986b), related to their Figure 13, the total PGEs within the Merensky Reef is much the same over large areas, regardless of reef width.

#### Contact or thin reef

Typical Rustenburg thin reef (Figure 1a) distribution has been shown by Viljoen and Hieber (1986). It appears to be the most common of all non-potholed reefs on Bafokeng Rasimone (Moodley, 2008) and becomes less frequent through Impala, Rustenburg and Western mining properties. Mining companies report that PGE grade continues into, and is mined from, the footwall. Note that in this section there is only one chromitite layer and no pegmatitic pyroxenite facies.

#### Pegmatitic pyroxenite reef

In this facies (Figure 1b and c), also found across the above-mentioned mines, there are two chromitite layers that may be separated by only a few cm to several tens of cm. Better grades are associated with the two chromitite layers, as can be seen as the separation increases (Figure. 1c). The interval between is usually a pegmatitic pyroxenite, with a normal grain-sized pyroxenite above the upper chromitite layer. The distribution of PGE may be seen (Figure 2a) in the detailed study (albeit for one section) from Barnes and Maier (2002) and so its representativity is not known. Unfortunately, they sampled to a depth of only 20 cm into the anorthosite below the chromitite layer and even there

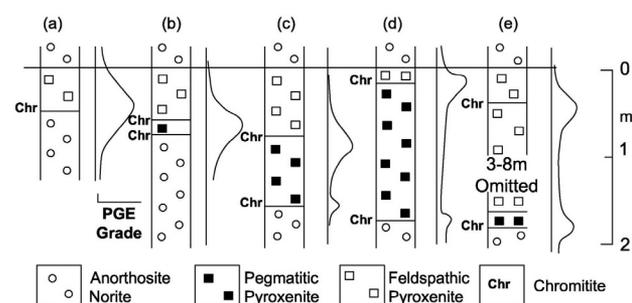
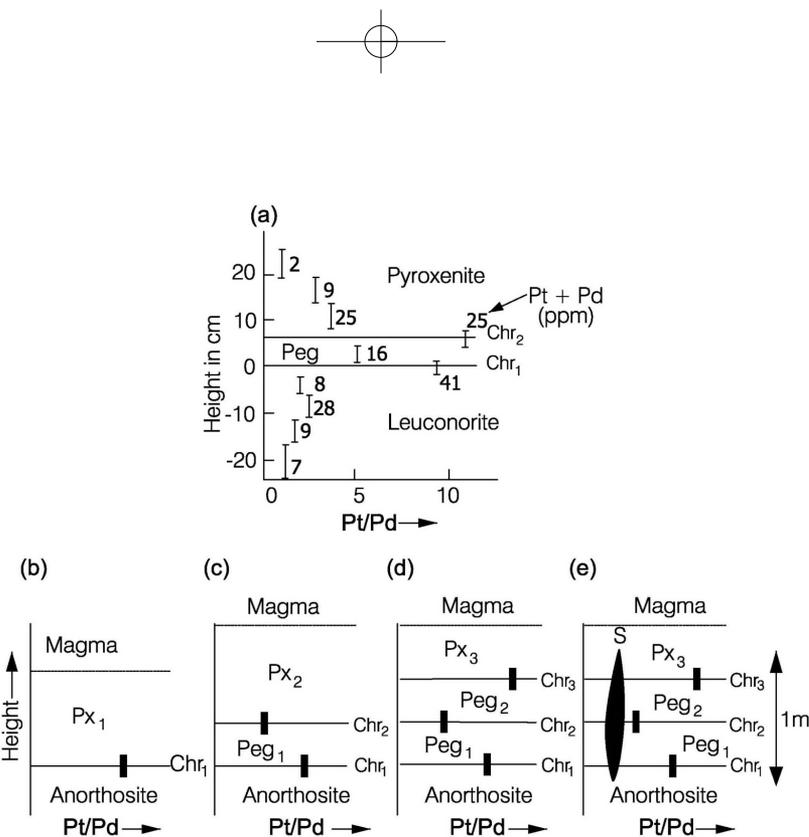


Figure 1. Facies variations in the Merensky unit on different mines. (a) thin reef, also called contact reef on Bafokeng Rasimone (Moodley, 2008), on northwestern parts of Impala Mines (Leeb-du Toit, 1986), and also occurs in places on Rustenburg Platinum Mines and Lonplats; (b) and (c) pegmatitic pyroxenite reef in which the thickness of the pegmatitic unit can vary from a few cm up to 1 m, traditionally viewed as 'typical' Merensky (Viljoen and Hieber, 1986); (d) thick pegmatitic reef, typical of Amandelbult and Union Mines (Viljoen *et al.*, 1986a; b); (e) thick pyroxenitic reef or Westplats type on Western Platinum mines and also at Lebowa and Marula Mines in the eastern Bushveld (Mitchell and Scoon, 2007). Semi-quantitative PGE distribution is shown



**Figure 2. (a) Pt + Pd grade and Pt/Pd ratio in very short vertical samples from Impala Platinum Mine (from Barnes and Maier, 2002). The vertical bar indicates the location and length of each sample. The number beside each bar is the Pt+Pd grade in ppm. (b) to (e) show the development of this succession, with up to 3 chromitite layers, each of which has its own pulse of mineralization with its own specific Pt/Pd ratio (shown by short, vertical, thick bars). The sulphide-carrying PGE mineralization is then superimposed in (e) by downward percolation, and has its own Pt/Pd ratio**

the grade was still 7 g/t combined Pt and Pd. Hence, the total depth of significant mineralization below the chromitite was not assessed, but can be traced to a depth of 1.5 m below the chromitite layer (Cawthorn, 1999a).

### Thick reef

To the east of the Lonplats properties, and in the Marula (Mitchell and Scoon, 2007) and Lebowa mines the reef package becomes thicker and lithologically more variable (Figure 1d and e). There is a basal chromitite above an anorthosite or norite. The chromitite usually carries a small amount of mineralization. Above the chromitite there can be a variable package of pyroxenite within which there may be pegmatitic lenses, bounded at the top by another chromitite layer. Similarly, at Union and Amandelbult mines the entire interval between the two chromitites may be pegmatitic and often contains olivine (Viljoen *et al.*, 1986a, 1986b). Very occasionally there is an intermediate chromitite layer that will also carry minor mineralization. Above the top chromitite is a pyroxenite grading into a norite. In most cases the best mineralization occurs in the top metre associated with, and below, the top chromitite layer.

### Genetic models for the Merensky Reef

Various hypotheses have been proposed for the origin of the PGE mineralization and these are reviewed in the light of these variable profiles.

#### Upward infiltration of fluid

The model of upward migration of a fluid (Boudreau, 2008) involves the following stages. A great thickness of footwall rocks accumulated. Final crystallization of the residual magma deep in this pile of crystals produced a water-rich

vapour. This vapour percolated upward, dissolving a portion of any sulphide and PGE present. When this vapour encountered a layer of crystal mush where the interstitial magma was undersaturated in water, the vapour (and its sulphur and PGE) dissolved into this interstitial magma. The high water content acted as a flux and caused significant remelting in that zone with the subsequent formation of coarse-grained textures (pegmatitic pyroxenite) and precipitation of PGE-rich sulphides. A quantitative geochemical model for this process was described by Boudreau (2008). The PGE distribution profile that he was aiming to produce is shown in Figure 3. This model requires that the PGE mineralization be associated entirely with the pegmatitic layer, and occupies an extremely thin zone. As shown in the various actual vertical sections in Figure 1 the PGE mineralization may occur either below or above this pegmatitic pyroxenite. It may be that there is no pegmatitic pyroxenite present anywhere in the succession, but there is still the same PGE grade as elsewhere. A further petrological issue also arises. If the precursor layer is significantly remelted then the overlying cumulate package ought to have collapsed into it, or the volatile-enriched melt ought to have migrated upward because of its lower density than the overlying cumulate rocks. Wagner (1929) commented on this same debate, and concluded: 'No instances were noted in all the hundreds of sections which the writer has examined of the normal Merensky Reef or feldspathic pyroxenite or harzburgite sending off apophyses into the hanging wall.' I know of one example of a short vertical protuberance of Merensky pegmatitic pyroxenite into overlying pyroxenite, but that is at the bottom of a pothole where considerable slumping and disruption of the succession has occurred (Carr *et al.*, 1999). Hence, it is not clear if this event represents a magmatic upward incursion or merely deformation of a crystal mush.

In summary, the observed geological relations are not consistent with this model of upward infiltration of a PGE-rich fluid. This conclusion does not deny the existence and migration of fluids, but to suggest that they did not transport or redistribute the PGE.

#### Downward accumulation of a PGE-enriched sulphide liquid

This model assumes that the PGE were concentrated in an immiscible sulphide liquid that sank to the instantaneous floor of the magma chamber (the anorthosite-norite below the Merensky unit). This model has had many variations that have emphasized different aspects, mainly in trying to produce an immiscible sulphide liquid (Campbell *et al.*, 1983; Naldrett and von Gruenewaldt, 1989), in trying to explain the high PGE content of the sulphide which is so radically different from many other magmatic sulphide ores (Campbell and Naldrett, 1979), and simultaneously producing the distinctive geochemical features associated with this mineralization.

In the model by Campbell *et al.* (1983), addition of new magma and its mixing with resident magma caused an immiscible sulphide liquid to form that scavenged the PGE. The added magma was assumed to have had a similar composition to that which had formed the previous succession. The problem is that there is a very abrupt increase in the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  value from 0.706 to over 0.708 within a very short vertical interval (Hamilton, 1977; Kruger and Marsh, 1982). This change in ratio suggests that a totally different magma had been added. The nature of this added magma creates a great deal of uncertainty to the model, because the composition, and hence density and temperature of this added magma are all unknown, and so whether it could mix with the resident magma in the way described by Campbell *et al.* (1983) must be considered uncertain. Further, this model requires the complete and thorough mixing of an extremely large volume of magma. The total thickness of the combined magmas (new and resident) has never been stated in these models, but using the likely PGE of basic magmas and its extractability into the sulphide liquid many hundreds of metres to kilometres are required (Cawthorn, 1999b). Furthermore, this mixing

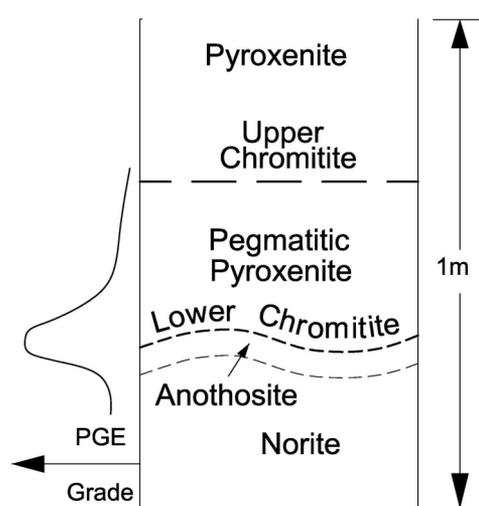


Figure 3. Lithology and PGE grade distribution as reported by Boudreau (2008). Note that the entire mineralization is assumed to be hosted within pegmatitic pyroxenite

must occur over an area of 30 000 km<sup>2</sup> since the Merensky is found in the eastern and western Bushveld. Such enormous areas and thicknesses of mixing have been questioned by Cawthorn (1999b), and more recently has been accepted as a major flaw to this model by Naldrett *et al.* (2009, p. 654).

Because of the implausibility of mixing such large volumes, it was suggested by Scoon and Teigler (1994) that the added magma was only a thin layer. However, neither its thickness nor the PGE contents of either magma was stated, and so it was not possible to quantify this model. A rather similar model was developed by Naldrett *et al.* (2009) who did attempt to quantify it. They suggested that the added layer was in the order of 12 m thick. The problem of the huge mass of PGE that had to be sequestered from the liquid was recognized in their model and so they suggested that the new magma contained 200 ppb Pt. The authors admitted that such values far exceeded those reported for the maximum solubility of PGE in basaltic magmas.

There are a number of issues that these models fail to recognize. The emplacement of a thin (12 m) layer of liquid places implausible constraints on the geometry of the instantaneous floor of the magma chamber (see Figure 4). In both models, the new magma is considered to have been denser than the resident magma and so must have flowed along the floor of the chamber. If there had been a topographic variation in height of this footwall succession of more than 12 m then the new liquid would not have been able to flow over the footwall, but would have flowed round such topographic elevations. Thus, no Merensky Reef would have developed on any topographic high greater than 12 m in the footwall rocks. There are no reported examples of Merensky pinching out against small-scale or irregular topographic highs of footwall succession. Hence, for this thin magma layer model to have operated the entire floor to the chamber from east to west must have had a topographic relief of less than 12 m over an area of 30 000 km<sup>2</sup>. I suggest that is very implausible, and question the thin-magma-layer model.

It needs to be mentioned that the above statement implies that the eastern and western limbs of the complex were part of one large chamber, certainly by the time the Merensky package was forming. This was the original concept for the Bushveld lopolith (e.g. Hall, 1932). It was challenged by Cousins (1959) who used gravity data to suggest that each limb was well separated from the others. However, Cawthorn *et al.* (1998) showed that Cousins' model had neglected the effect of isostasy, and that the gravity data

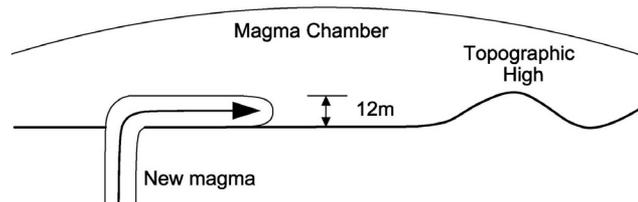


Figure 4. Schematic cross-section of a magma chamber, illustrating the implications of the addition of a thin, dense layer of magma, as proposed by Naldrett *et al.* (2009). This magma layer must form a perfectly horizontal upper surface. If there had been topographic relief on the floor cumulates thicker than the magma layer, there could have been no new magma above it, and so no Merensky package could have formed

were consistent with a single connected lopolith between east and west. Connectivity between these and the northern limb is less certain, and so the correlation of the Merensky Reef and Platereef is moot. Scoon and Mitchell (2009) continued to suggest that the limbs are discrete. However, it should be noted that both limbs contain a number of very distinctive, verging on unique, layers and sequences of rocks. Thus, the LG6, LG7, MG1, MG2, MG3, MG4, UG1, UG2, Merensky and Bastard packages occur in both limbs (Cawthorn *et al.*, 1998). Also the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in both limbs follow the same sequence of changes in values and at the same stratigraphic levels. The idea that two discrete intrusions could have produced such identical sequences and diagnostic layers is considered totally implausible.

The second issue concerns the Sr isotopic data referred to above. In this model the new magma added had the composition and isotopic ratio of the lower rocks of the Bushveld Complex, i.e. type 1 magma. It was intruded into a chamber which contained magmas already influenced by the magma that made the Main Zone according to Mitchell and Scoon (2007) and Naldrett *et al.* (2009). Thus, addition of this basal surge of primitive, type 1 magma ought to have lowered, not raised, the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio in the Merensky unit package.

In all these models there is a geochemical paradox that arose from the study of the Cr content of orthopyroxene throughout the entire Bushveld Complex, as summarized by Eales (2000). He noted that there was a massive decrease in this Cr content close to the Merensky Reef, and attributed it to addition of a fundamentally chemically different magma type. A more detailed examination of this break showed that it occurred several metres above the Bastard Reef (Cawthorn, 2007). Thus there is a chemical anomaly, namely that the Cr in orthopyroxene data suggest that new magma was added several metres above the Bastard Reef, whereas the Sr isotope break suggests that addition was close to the top of the ultramafic rocks of the Merensky Reef. Seabrook *et al.* (2005) resolved this paradox by noting that the Sr is largely held in plagioclase, and so suggested that the orthopyroxene and plagioclase had been derived from two coexisting, but stratified, magmas. In this model, the added magma (type 2) was of a different composition from that existing in the chamber (type 1), and that it underflowed the existing magma because it was denser. No mixing of magmas occurred. Orthopyroxene, chromite and sulphide separated from the upper (resident) magma (type 1) and because they are all denser than the magma sank through the underlying magma layer. The underlying magma layer (type 2) ultimately produced the interstitial plagioclase. This stratified magma column is the exact reverse of that proposed by Naldrett *et al.* (2009) who arbitrarily added 1% of water into the resident magma that had Main Zone affinities (type 2), but no water to the added primitive magma (type 1) so that the relative densities fitted their thin, dense, basal layer geometry.

Cawthorn *et al.* (2002) and Mitchell and Scoon (2007) argued that what is considered typical Merensky by many authors, the thin pegmatitic pyroxenite reef (Figure 1), has been so condensed that it is not possible to identify all the events that actually occurred. They both suggested that the thicker reef facies preserved a greater proportion of the evolving succession and allowed the entire sequence to be recognized and deciphered. They suggested the following sequence of events. Magma of the type 1 composition, which is relatively siliceous and with about 13% MgO

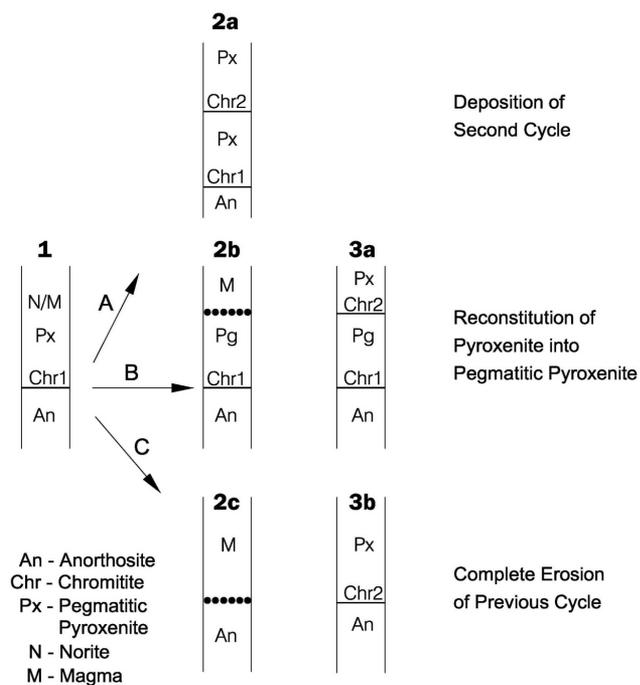
(Davies *et al.*, 1980), crystallized numerous rhythmic units of chromitite through pyroxenite and norite to anorthosite up to the level of the footwall to the Merensky package. The top of this succession may have been undulating due to primary accumulation effects, or to later erosion or redistribution of the uppermost few metres of anorthosite (Leeb-Du Toit, 1986).

Another similar cycle was initiated at the beginning of the Merensky package. There may have been addition of more of type 1 magma or there may have been convective overturn of an already stratified magma column as has been proposed for the Upper Zone (Tegner *et al.*, 2006). Alternatively, changes in pressure due to tectonic activity may have changed the mineral assemblage crystallizing from the magma (Lipin, 1993; Naslund and McBirney 1996). An increase in pressure in the chamber would have stabilized chromite (Cameron, 1982), pyroxene (Cawthorn, 2003) and sulphide (Mavrogenes and O'Neill (1999)). As a result of this postulated pressure increase, a chromitite and thin (possibly 1 m or slightly more) pyroxenite layer accumulated. A pressure decrease caused a cessation in crystallization. The pyroxene crystal mush was then in contact with a magma that was slightly superheated. Three possible processes could then have occurred as shown in Figure 5 (Cawthorn and Barry, 1992; Cawthorn *et al.*, 2002; Cawthorn and Boerst, 2006). Column 1 in Figure 5 shows the partially formed cycle of chromitite (the lowest layer, Chr 1) and pyroxenite. Another succession could have formed on top of it (shown by the arrow A), producing a chromitite layer (Chr 2) and more pyroxenite (column 2a, Figure 5). The second option (shown in column 2b by the arrow B), was that because the magma was slightly superheated the layer of pyroxenite was thinned and coarsened in texture to produce the pegmatitic pyroxenite.

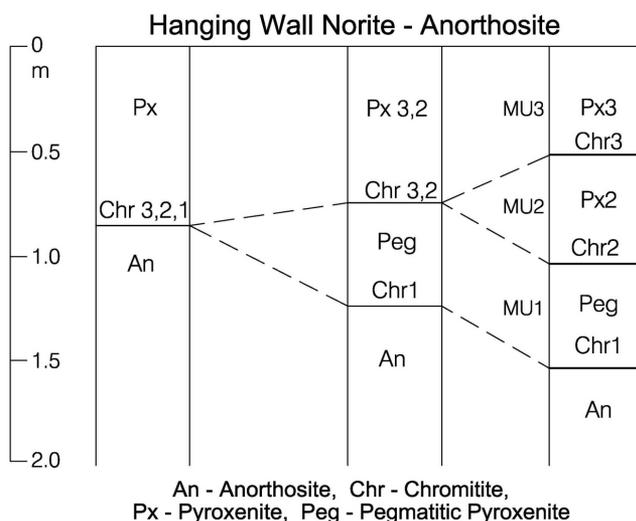
A similar coarsening process was advocated by Viring and Cowell (1999) and Viljoen (1999) who referred to it as reconstitution. It also has similarities with the process proposed by Roberts *et al.* (2007), but they envisaged the replacement of pre-existing minerals and textures a very short distance into the footwall below chromitite layer 1, whereas the process here is considered to have occurred mainly at the crystal-magma interface. A second injection of magma could have produced a second chromitite above this pegmatitic pyroxenite, as shown in column 3a, Figure 5.

The third possibility (shown by arrow C in column 2c, Figure 5) is that complete dissolution of the pyroxenite and chromitite might have occurred. An uneven upper surface of the footwall anorthosite may have resulted if continued dissolution had occurred of the footwall. In this case, the next injection of magma would have produced a chromitite layer on the footwall anorthosite (column 3b, Figure 5), but it was the second chromitite, not the first, that survived at the contact with anorthosite. These processes were repeated to give all variations from a contact reef where dissolution had removed components of the first and second Merensky packages, leaving only the last event, to a complex and thick reef in which up to three chromitites could be recognized, as shown in the actual examples in Figure 1. These variations and problems with correlation are shown in Figure 6. By this sequence of events, all the pyroxenites (and their pegmatitic reconstitutions) have the initial Sr isotopic ratio of the first magma type.

I suggest that evidence for this remelting and reconstitution can be seen in the Cr contents of pegmatitic and pyroxenitic parts of the reef. In a study of many sections of these rock types from Impala mine, Boerst



**Figure 5. Schematic models for the formation of repeated chromite – (pegmatitic) pyroxenite sequences, of which there may be three in the Merensky package. The three sequential events are numbered 1, 2 and 3 (from left to right). Three possible processes, denoted A, B and C may occur during each event. In A there is simply the deposition of a subsequent cycle that totally preserves the previous cycle. In B there is reconstitution of pyroxenite into pegmatitic pyroxenite during a period of non-deposition. In C there is variable to complete dissolution of the previous cycle. The line of solid circles represents the boundary between cumulate grains and magma**



**Figure 6. Different sections can be preserved in the Merensky package depending on the degree of preservation of each of the cycles as shown in Figure 5. On left, removal of layers formed by previous cycles result in the formation of thin or contact reef. As a result of erosion it is not known to which cycle the chromitite layer ought to be attributed (cycle 1, 2 or 3). On right is the other extreme, where most of the three cycles have been preserved, and are called Merensky Unit 1, 2 and 3 (MU1, MU2 and MU3)**

(2001) showed that the pegmatitic facies reef had a much higher Cr content than pyroxenitic facies reef (see Figure 7). The pegmatitic reef also often has a higher olivine content than pyroxenitic reef. Both of these features can be explained by the well-known incongruent melting of orthopyroxene. The primary pyroxene grains would have had a Cr content of about 3 000 ppm. However, as they were redissolved olivine was produced, containing essentially no chromium, and the Cr was left as refractory chromite grains.

The final event involved addition of type 2 magma, which was denser than the existing magma (Seabrook *et al.*, 2005). This magma was not in equilibrium with orthopyroxene or chromite, and erosion, both chemical and physical associated with the emplacement of the magma could also have ensued. The increase in pressure associated with the injection of the second magma triggered chromite, pyroxene and sulphide formation in the upper magma which sank into the lower magma layer, making the pyroxenite of normal grain size above the upper chromitite. This process resulted in the highly anomalous situation in the pyroxenite above the Merensky Reef that the cumulus crystals of orthopyroxene with high Cr content formed from type 1 magma (the upper stratified layer), whereas the matrix plagioclase with high Sr isotopic ratio that solidified round these grains was from a different, type 2 magma (the lower stratified layer). In this way, the apparent contradiction that the Cr content of pyroxene indicates type one magma, whereas the Sr isotope ratio indicates type 2 magma, becomes explicable. It should be emphasized that the Sr isotopic break becomes significant only at the top of the pyroxenite. It does not occur at the lower chromitite as would be expected if the major magma addition and Merensky forming event were represented by that lower boundary. In thin reef this distinction in the elevation of the isotopic change is difficult to demonstrate, although it should be noted that in the very first isotopic study, Hamilton (1977) stated that the break occurred 3 m above the reef. However, his sampling was not as close as in many subsequent studies. The isotopic succession through a thick Merensky package has been presented by Shelembe (2006) and is shown in Figure 8. The entire 10 m of pyroxenite has the Sr isotopic ratio of the type 1 magma, but the overlying norite shows increasing values with height due to the crystallization of interstitial plagioclase from type 2 magma.

The grains of chromite and orthopyroxene in the Merensky package formed a framework, initially with considerable porosity. Compaction due to annealing resulted in a decrease in porosity with increasing depth in the crystal pile (Figure 9). The actual permeability, rather than porosity, depends upon grain size as well proportion of pore space. Thus, the permeability in the footwall succession shows a minimum at the chromitite layer because of its fine-grain size. Cawthorn (1999a) suggested that the settling sulphide liquid had low viscosity and high density and so it filtered downward through the uppermost pyroxenite, as shown by the modeling of Mungall and Su (2005). It was partially arrested as it tried to penetrate the chromitite layer, but some did filter through. The rapidly decreasing permeability downward, and the small volume of sulphide available, meant that it did not penetrate more than about 1 m into this crystal mush. Thus, the sulphide is now found slightly below the level it which it first separated. This relationship was first discussed by Wagner (1929; p. 113) who recognized this process in a very

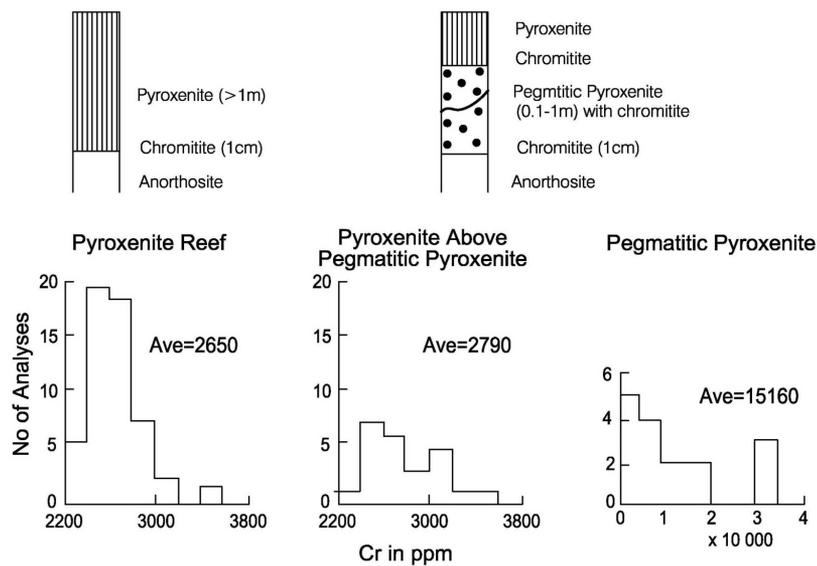


Figure 7. Histogram showing the Cr content of Merensky pyroxenite in different reef facies. Where there is no pegmatitic pyroxenite facies (in thin or contact reef) the average Cr content is 2650 ppm, close to the typical value of Cr in primary pyroxene. At the other extreme where there is a well-developed pegmatitic pyroxenite the average value increases by a factor of 6, attributed here to the incongruent melting of orthopyroxene to leave abundant chromite in the pegmatitic rocks. Data from Boerst (2001)

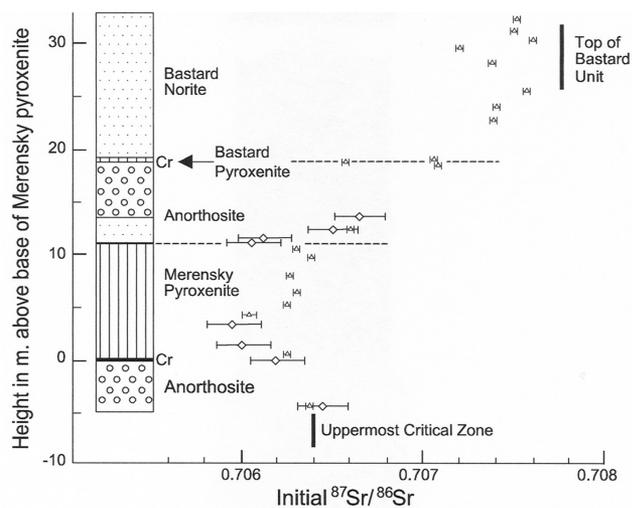


Figure 8. Plot of initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of whole-rocks from a section through the thick Merensky package on Western Platinum Mines shown by the triangle symbols (from Shelembe, 2006). Note that the entire pyroxenite sequence has ratios identical to the footwall succession formed from type 1 magma. The increase in ratios, indicating the presence of a new magma (type 2) occurs only at the level of the norite above the pyroxenites. For comparison, the typical values obtained for thin Merensky Reef package from Kruger and Marsh (1982) are included as the diamond symbols. However, their positions are vertically exaggerated to match the lithological succession shown here. Also the typical Upper Critical Zone and Main Zone ratios, from Kruger (1994), are included as thick vertical bars

prescient section of his book entitled, 'Effect of movements during and subsequent to consolidation'. It is an issue that has been almost totally neglected in most subsequent publications in which the timing of the formation of sulphides is related to the exact layer in which they are now found. I suggest that this vertical migration of probably about 1 m is crucial in understanding the genesis of the succession. This model explains the fact that in thick reef

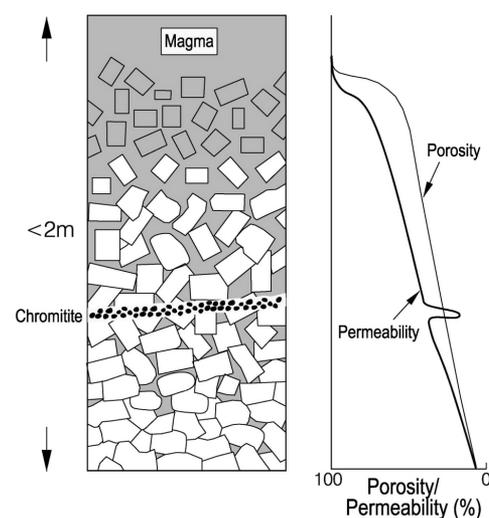


Figure 9. Schematic porosity and permeability diagram as a function of height through the uppermost cumulate succession that includes the chromitite layer of the Merensky package (taken from Cawthorn, 1999b). The grains shown stippled are pyroxene, and those shown blank are plagioclase

the mineralization is always top loaded and that in thin reef the mineralization may descend into the leucocratic footwall succession (Figure 1). Its coexistence with pegmatitic pyroxenite in some areas of some mines is coincidental and does not reflect their simultaneous formation or genetic association.

#### PGE grades and ratios

There is one aspect of the grades and ratios of Pt/Pd that is relevant to the genesis of the mineralization. The detailed study by Barnes and Maier (2002) of thin pegmatitic reef on Impala is shown in Figure 2a. Both the Pt + Pd grade and Pt/Pd ratio are shown. It can be seen that the maximum grades coincide with the very thin chromitite layers, and

also that the ratio of Pt/Pd is very much higher in these layers than in the intervening silicate rocks. Note that the chromitite layers are too thin to be analysable separately, and the length of sample cut for analysis included some of the associated silicate rocks. Hence, the grade in the pure chromitite might have been even higher. These relations led Barnes and Maier (2002) to suggest that there were two collection mechanisms operative, one associated with chromite and the other with sulphide. This dual process is shown in Figure 2b–e. This dual collection mechanism was ignored in the calculations made by Naldrett *et al.* (2009) who assumed that all PGE had been sequestered by sulphides. This issue will be addressed once the mineralization in the UG2 has been discussed.

### UG2 chromitite

The UG2 chromitite reef is generally considered to be a single layer of chromitite. However, Lee (1996) pointed out that there was a fundamental textural break in the middle of the layer, the lower part being a massive chromitite and the upper part containing abundant pyroxene oikocrysts. This boundary also coincided with changes in composition, and Lee (1996) suggested that the UG2 is a composite layer. Unlike the Merensky where there is a tail of PGE (albeit quite short) into the hangingwall pyroxenite to norite, above the UG2 there are no reports of any mineralization in the overlying silicate rocks. Below the UG2 there is usually a pegmatitic pyroxenite, although on some mines it is an anorthosite. The presence of PGE in the pegmatitic footwall has been reported, but it appears that it is confined to schlieren of chromitite within the pegmatitic pyroxenite rather in the silicate host itself. So it can be inferred that the PGE is totally hosted in chromitite. It is also important to note that the amount of sulphide is extremely low, as evidenced by Cu and Ni contents typically less than 0.05% (McLaren and de Villiers, 1982; Hiemstra, 1985; 1986; Lee, 1996).

The PGE grades most frequently reported are for 20 cm lengths through the layer, which is the standard practice for mining and exploration companies. However, I consider the most definitive data were obtained in two studies by Hiemstra (1985; 1986) who sampled every two cm through sections of UG2 from Lonplats mines. Typical mining company data show a basal enrichment of PGE and also a secondary enrichment either nearer the middle of towards the top. However, when analysed in great detail it is seen that there may be three upward depletion trends as shown in Figure 10 (from Hiemstra, 1985; 1986).

### Relationship of UG2 and UG3 chromitite layers

Several lines of evidence suggest that the UG3 chromitite that is discrete in the northeastern part of the Bushveld has merged with the top of the UG2 chromitite in all other regions. The existence of a UG3 cyclic unit in the northeastern limb of the Bushveld was first discussed by Gain (1985). In fact, he noted that there was also a UG3A cyclic unit in which there were two discrete thin (15 and 10 mm) chromitite layers and which can be seen on the farm Maandagshoek (Figure 11). I have denoted the two layers as 3A and 3B for clarity. Gain also suggested that these uppermost cycles were not laterally continuous but merged with the underlying cycle, as shown in Figure 11 between layers 3A and 3B. I suggest that the UG3 also merged with the underlying UG2 in most sections of the Bushveld, only being recognizable as a discrete entity in the northeast.

These geometrical relations are shown in Figure 12. The data of Gain (1985) showed that the UG3 had a distinctively high Pt/Pd ratio, a feature also revealed by Hiemstra's detailed profiles at the top of the UG2

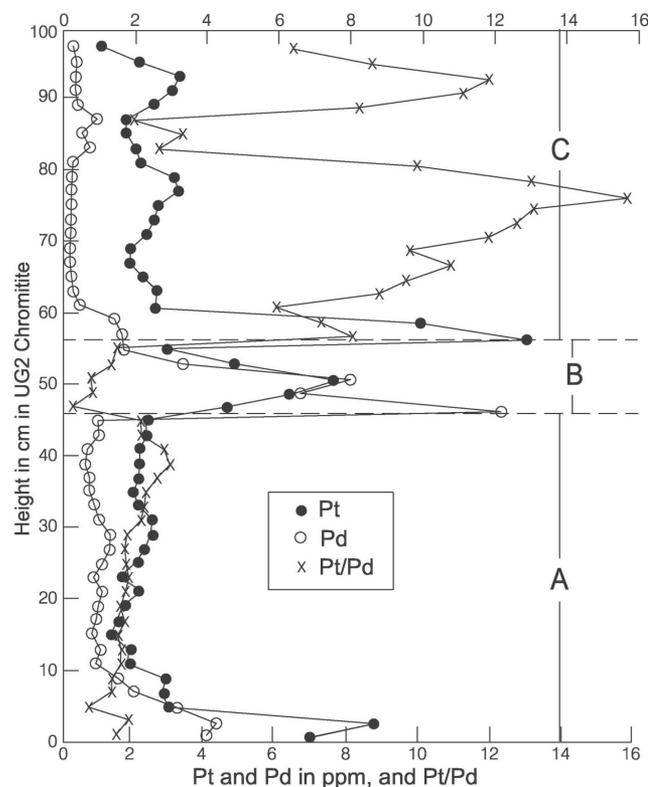


Figure 10. Plot of Pt, Pd and Pt/Pd ratio in 2 cm-thick samples of the UG2 chromitite from Lonplats Mines (from Hiemstra, 1985), showing the three distinct cycles each with decreasing grade, but constant and distinct Pt/Pd ratio

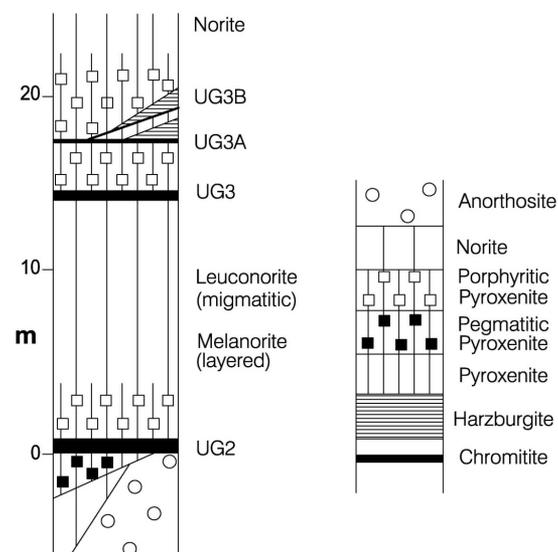
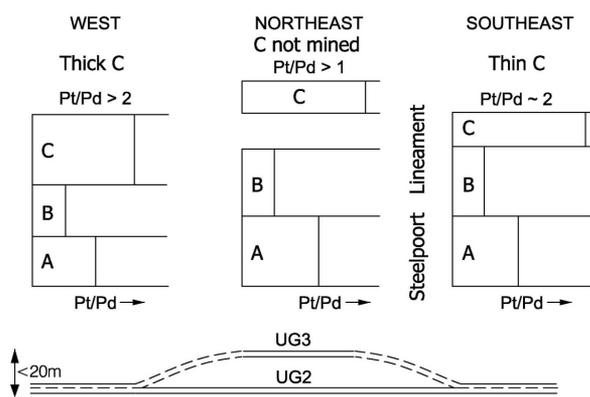


Figure 11. Section showing the relationship between the UG2 and UG3 chromitite layers. Relationships taken from the work of Gain (1986) on the farm Maandagshoek, eastern limb, showing the merging of two thin chromitites above the UG3, based on field relationships and borecore data



**Figure 12.** Larger scale relations showing the preservation of a discrete UG3 in the northeastern limb, and its merging into the UG2 elsewhere. The UG3 has a distinctly higher Pt/Pd ratio than the UG2 producing the high Pt section at the top of UG2 as shown in Figure 10

(Figure 10). A pyroxenite overlies the UG2 chromitite layers in the northeastern sections. Above the UG3A chromitite is an olivine-bearing pyroxenite (Gain, 1985). In the western Bushveld the pyroxenite overlying what is taken to be the UG2 is often olivine-bearing (Cawthorn and Barry, 1992). These assorted lines of evidence suggest that in the western Bushveld the UG2 chromitite could be comprised of three distinct layers of chromitite, between which there may be occasional thin partings of silicate material (Lee, 1996). Gain (1985) also showed that the top of the UG2 in this northeastern area did not have the high Pt content found elsewhere in the UG2. Thus, the lower Pt/Pd ratio of the UG2 in the northeastern region compared to the rest of the sections becomes explicable.

There is significant PGE mineralization above the UG2 in the extremely thin chromitite layers that have been called the leaders or stringers, ranging from 0.5 to 1 m above the UG2. The grades of these layers have not been reported because they are so thin, but such information might assist in elucidating the correlations through this interval.

#### Models for the formation of chromitite layers

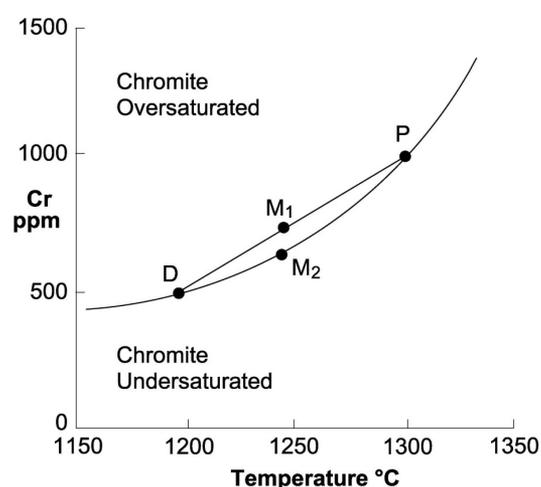
##### Magma mixing

Most models for the genesis of chromitite layers appeal to magma addition and mixing (as originally proposed by Irvine, 1977). He suggested that a slightly differentiated resident magma, equivalent to type 1 when applying Irvine's model to the Bushveld Complex, mixed with a new injection of primitive type 1 magma. This process is illustrated on a Cr-saturation diagram from Barnes (1986) and Murck and Campbell (1986), shown in Figure 13. The primitive type 1 magma would have had a composition given by P. As a result of its cooling and differentiation it would change composition to D. The chamber was then replenished by more of magma of composition P. P and D mixed. The resultant mixture ( $M_1$ ) was oversaturated in Cr and so formed chromite grains until its composition had fallen to  $M_2$ . It can be seen in this diagram that a maximum of 100 ppm Cr could have been extracted from  $M_1$ . Chromitite layers contain about 45%  $\text{Cr}_2\text{O}_3$  or 300 000 ppm Cr. Thus to produce a layer 1 m thick (with a density of  $4.5 \text{ g/cm}^3$ ) from magma (density 2.8) by the loss of 100 ppm would require the efficient mixing of a total 4 to 5 km of

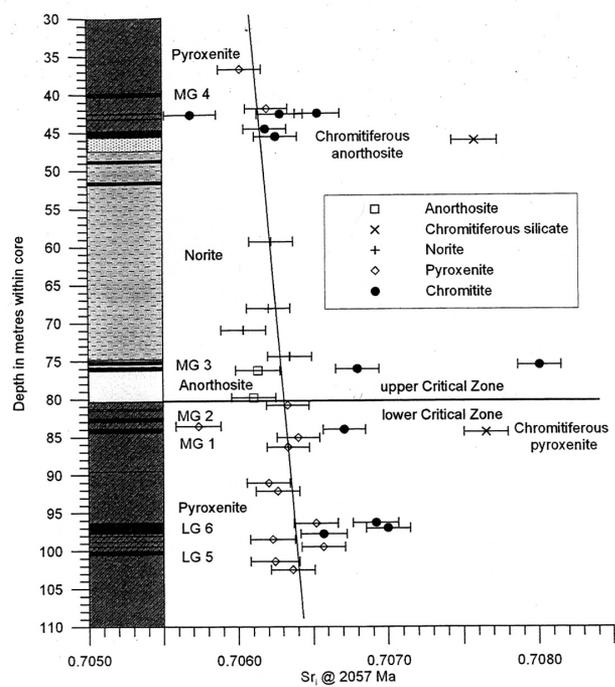
magma (Campbell and Murck, 1993). If mixing was not in equal proportions then a smaller oversaturation in Cr would have resulted, requiring even greater thicknesses of magma. Inspection of the basal contact of the UG2, and almost every other chromitite layer, indicates an extremely sharp contact implying a very abrupt process. Again, this must have happened on a scale approaching 30 000  $\text{km}^2$ . Instantaneous mixing on such a lateral and vertical scale is considered very unlikely.

It should be possible to test the concept of magma addition by comparing the  $100 \cdot \text{Mg}/\text{Mg}+\text{Fe}$  (mg#) ratio of orthopyroxene above and below a chromitite layer. Magma compositions P and D would be expected to crystallize orthopyroxene with mg# of close to 90 and nearer 80 respectively, and so composition M ought to produce a composition of about mg# 85. Thus, primitive magma addition ought to produce a reversal of several units in the mg# in the pyroxene above a chromitite layer. In his very detailed studies of the eastern limb, Cameron (1980, 1982) did not report any reversals.

A variant on the magma mixing model was proposed by Kinnaird *et al.* (2002), based on the original model of Irvine (1975), using Sr isotope ratios (Figure 14). They showed that every chromitite layer was associated with an increase in this ratio, which they attributed to crustal assimilation. This model also raises the question as to why it should be such an abrupt event, producing such a sharp basal contact over such a large area. Mondal and Mathez (2007) challenged this model on the grounds that the succession overlying chromitite layers ought to show higher Sr isotope ratios, which they do not. However, in the model of Kinnaird *et al.* (2002) the assimilation involved reaction between magma and a partial melt from the roof of the intrusion. The chromite crystals then cascaded down from the roof carrying a selvage of the liquid with the high Sr isotope ratio. The Sr isotope ratio of the magma towards the base that was the source of the subsequent pyroxenite was not affected.



**Figure 13.** Relationship of Cr solubility in magma as a function of temperature. The parental magmas to the Bushveld may have had a composition P. On fractionation of chromite, olivine and orthopyroxene it may have differentiated to D. Addition of more magma P and mixing with D could have produced a magma  $M_1$  which would have crystallized chromite until the composition has changed to  $M_2$  (from Murck and Campbell, 1986)



**Figure 14. Plot of the initial Sr isotope ratio versus height through the transition from the Lower Critical Zone to Upper Critical Zone including 6 chromitite layers. Almost all samples enriched in chromite (denoted by solid dots) have anomalously high isotopic ratios. In contrast, the overall trend for the intervening silicate-rich rocks show a slightly decreasing trend (from Kinnaird *et al.*, 2002)**

#### ***Injection of a chromite slurry***

Eales (2000) argued that there was an imbalance in the total of amount of Cr (both as chromite and in the Cr-rich orthopyroxenes) in the Lower and Critical Zones of the Bushveld Complex. He therefore appealed to the injection of a chromite slurry that had been formed in some deeper magma chamber. The first issue here is the mass balance of Eales (2000). He suggested that there is an imbalance in Cr in the lower half of the Bushveld, but by inference, no imbalance was implied for any other element. The fact is that there is a difficult mass balance problem associated with the entire lower half of the Bushveld. Using the proposed composition for type 1 magma, Cawthorn and Davies (1983) showed that the magma would crystallize the sequence olivine, orthopyroxene, plagioclase as observed in the intrusion. This evolution was modelled quantitatively, constrained by the mg# of the pyroxene, by Li *et al.* (2001), who showed that only 20% of the magma had crystallized at the level of the Merensky unit. Thus, a mass balance approach demands the question as to what happened to the remaining 80% of this magma. Sourcing the chromite from another chamber does not resolve the much bigger issue of this remaining 80%. The Cr imbalance is no bigger a problem than the rest of the evolving type 1 magma.

The problem with an injection of a chromite-rich slurry is its lateral distribution once in the Bushveld chamber. A dense slurry of chromite would be dumped quite close to the site of entry, and any layer that formed would thin dramatically with distance away from the entry point. The layers of chromite, especially the UG2, show remarkable uniformity of thickness over their entire occurrence with no evidence for thinning. Thus this model must be questioned.

An even more extreme version of this model of emplacement of a chromite-rich slurry has been proposed by Maier and Barnes (2008) and Voordouw and Beukes (2009). They have proposed that the chromite slurry was intruded into a pre-existing succession of rocks. In the case of the UG2 (to which Maier and Barnes specifically referred) this model asks the question as to why it intruded exactly along the contact between two different rock types. It is also challenging to ask whether a crystal mush could intrude over an area of 30 000 km<sup>2</sup> with such uniform thickness. Furthermore, these two publications do not address the question as to whether they consider all chromitite layers to be intrusive or only the UG2 discussed in their publications. If they consider only this one layer to be intrusive it is necessary to identify criteria that distinguish layers that are intrusive from layers that formed by downward accumulation of chromite.

#### ***Pressure change***

The model proposed by Lipin (1993) invoked pressure changes that would occur throughout the entire magma chamber instantaneously. Thus the entire magma chamber could be processed to produce chromite, minimizing the mass balance problems. However, it is a very difficult model to test because it leaves no effect in the magmatic system except the change in the order of crystallization of minerals (as reviewed by Naslund and McBirney, 1996). Naslund and McBirney also referred to the possibility of seismically induced nucleation. As pointed out by Wager and Brown (1968, p.548) the sequence of mineral nucleation would likely be in order of structural complexity of the minerals, namely, oxide, pyroxene then plagioclase. They noted that this order also matches that expected from density sorting and normal crystallization from the type 1 magma, thus a unique solution to the process that causes the layering and especially the chromitite layers remains elusive.

#### ***PGE mineralization in the UG2***

Whatever the mechanism that produced the chromitite layers, the concentration mechanism of the PGE into the chromitite requires examination. It should be emphasized that all chromitites in the Bushveld Complex contain significant concentrations of PGE (Scoons and Teigler, 1994), and they argued that the concentrations observed in the UG2 chromitite are part of a continuum of a pattern throughout the entire Critical Zone.

#### ***Sulphide association***

It has been suggested that the chromitite layers are associated with sulphide formation that actually concentrated the PGE (Naldrett and Lehmann, 1987). In this model, the sulphide subsequently broke down by reaction with chromite, the iron entering the oxide phase and the sulphur being lost as a vapour. However, if sulphide had been present, the chromitites ought to have contained some Cu and Ni, possibly comparable in grade to the Merensky Reef. The UG2 chromitite contains typically 0.03 and 0.06% Cu and Ni respectively (Hiemstra, 1985, 1986; McLaren and de Villiers, 1982; Lee, 1996). The Anglo Platinum Annual report quotes 0.01 to 0.04% Cu and 0.07 to 0.16% Ni for the UG2, which are much less than the grades in the Merensky Reef of about 0.1 and 0.25%. Although the report does not specify whether the analysis is total Ni in whole rock or on acid soluble leachate, it is probable that approximately 0.1% of that Ni in

the UG2 resides in the oxide phase (de Waal; 1975), and is not in the sulphide fraction, and so the difference between sulphide Cu and Ni in the two reefs is very significant. Thus, this model requires that much of the Cu and Ni were also lost from the sulphide during the proposed desulphidation process. It would be far more likely that the Ni would enter the oxide phase rather than being released into a vapour. Hence, the suggestion that there was a higher original sulphide content in these chromitite layers must be challenged.

#### ***Solid solution in chromite***

It has been suggested that the PGE can partition into chromite in solid solution (obeying Henry's law), and that on cooling the PGM exsolved from the chromite grains. However, experimental evidence suggests that significant solution of PGE into chromite is not likely (as reviewed by Finnigan *et al.*, 2008).

#### ***The role of micronuggets, nanonuggets and clusters of PGM***

Hiemstra (1979; 1985; 1986) suggested that micronuggets of PGE phases existed, suspended in the magma. Ertel *et al.* (1999) concluded that they were probably no greater than 0.05  $\mu\text{m}$ , and so would be far too small to sink independently, but remained suspended due to Brownian movement. Hiemstra suggested that once chromite grains formed the PGE micronuggets were attracted to these grains, and thereby carried to the base of the magma chamber with the chromite. Evidence for or against this model is hard to obtain because such micronuggets would anneal into the larger PGMs now identified associated with chromite. Extremely small grains of Pt species are being identified (Ballhaus and Sylvester, 2000; Godel *et al.* 2007) and determining their original form remains elusive. Ertel *et al.* (2008) have reviewed the problems of identifying what they call nanonuggets, and predicting their behaviour.

Variations on this chromite-PGM association were developed by Scoon and Teigler (1994), Ballhaus *et al.* (2006) and Finnigan *et al.* (2008). The first authors did not specify a process. The second authors suggested that the PGM nucleated on chromite grains, whereas the latter argued that when chromite crystallized it induced a zone of reduced (low oxygen fugacity) liquid composition around the grains, and that at lower oxygen fugacity the PGM would nucleate directly and adhere to the growing chromite. The last model depends upon the relative diffusion of rates of the oxygen versus Cr. Unless Cr can diffuse through the magma towards the growing chromite quicker than oxygen can diffuse in the same direction then a zone of low oxygen fugacity around the growing grains cannot be sustained and the PGM will not form.

Yet another variant on this model of Hiemstra (1979) was developed by Tredoux *et al.* (1995). They noted that the catalytic properties of PGE result from their ability to exist as clusters of atoms, which are highly reactive. They suggested that such clusters would become stabilized by forming surface bonds with either sulphur or iron. (In exactly the same way that catalytic car exhaust converters can be poisoned by sulphur in fuel.) Thus, these clusters may react with, and become incorporated into, either an immiscible sulphide liquid or bonded to oxide (either chromite or magnetite). However, a note of caution needs to be sounded about the intimate association of the PGMs with chromite because the metallurgical processes employed for beneficiation discard a chromite-rich residue very depleted in PGEs.

Any of these chromite-related mechanisms can explain the duality of concentration mechanisms seen in Figure 2, namely that both chromite and sulphide have the chemical or physical properties necessary to concentrate the PGE.

### **Variation in PGE content in Merensky and UG2**

Hiemstra (1985; 1986) recognized the great variation in PGE grade upward and its cyclicity and concluded that the first chromite grains to form would attract the greatest proportion of the postulated micronuggets or clusters. Subsequent settling chromite grains would attract progressively fewer of these grains as the magma was swept clear of these particles. If it is accepted that the UG2 may be a composite of two or even three sub-layers, each new chromite-forming event might scavenge a new crop of these micronuggets or clusters. Similarly, in the Merensky Reef the first sulphide droplets would scavenge a high concentration of the particles, with subsequent sulphide droplets finding fewer such particles to attract. In this way, the upward depletion PGE trends noted by Naldrett *et al.* (2009) for the Merensky Reef become predictable, as postulated by Hiemstra (1986) for the chromitite patterns.

### **Summary**

The PGE are very unevenly distributed vertically throughout the Merensky and UG2 chromitite reefs in the Bushveld Complex. In the Merensky Reef, mineralization may be found in the anorthosite below the chromitite and pyroxenite layers in what is called thin or contact reef. Where the reef is thicker, the mineralization can be seen to be top loaded (i.e. near the upper chromitite). The mineralization need not be associated with pegmatitic rocks. It is only where the reef is thickest that the entire sequence of events may be recognized. In thin reef too much of the temporarily present succession has been removed by syn-magmatic erosion to fully appreciate the processes involved. There may have been up to three injections of magma, each producing a chromitite and pyroxenite, but each subsequent event removed variable amounts of the previous depositional cycle by thermal erosion or reconstitution. The last event produced the most significant proportion of the mineralization, carried down by an immiscible sulphide liquid that percolated through the uppermost metre of crystal mush.

The UG2 chromitite probably did not contain significant sulphide, and the PGE are concentrated mainly by the chromite itself. Various nucleation-triggering processes have been proposed to explain this association of chromite and PGE. The UG2 chromitite is probably not a single event, but represents two or three layers that have been superposed, occasionally with thin, intervening silicate rock lenses. Each layer has its own distinctive upward depletion cycle of PGE content. The uppermost of the three layers may exist as a discrete layer, separated by a significant thickness of silicate rocks from the UG2 in the northeastern Bushveld, where it is termed the UG3 chromitite. It has a high Pt/Pd ratio, but is too thin to mine. This high Pt/Pd signature may be recognized in the uppermost part of the thick UG2 chromitite elsewhere in the Bushveld, and so it is suggested that the UG3 accumulated directly onto the UG2 in most places.

## Acknowledgements

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