

# Magnetite layer formation in the Bushveld Complex of South Africa

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## Article

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# 1 Magnetite layer formation in the Bushveld Complex of South Africa

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## 5 Abstract

6 The great economic significance of layered mafic-ultramafic intrusions like the Bushveld  
7 Complex of South Africa results from the existence within them of some layers highly  
8 concentrated in valuable elements. Here we address the origins of the Main Magnetite Layer,  
9 a globally important resource of Fe-Ti-V-rich magnetite. Previous models of *in situ*  
10 fractional magnetite crystallization require frequent *ad hoc* adjustments to the boundary  
11 conditions. An alternative model of rapid deposition of loose piles of magnetite crystals  
12 followed by compositional convection near the top of the pile and infiltration of the pile from  
13 beneath by migrating intercumulus melt fits observations without any adjustments. The data  
14 admit both explanations, but the latter model, with the fewest unconstrained interventions, is  
15 preferable. The choice of models has pivotal ramifications for understanding of the  
16 fundamental processes by which crystals accumulate and layers form in layered intrusions.

## 18 Introduction

19 Because it is impossible to observe directly the processes that produce layered plutonic mafic  
20 rocks, the nature of these processes must be inferred from the record of composition, mineral  
21 mode, and texture of the final products of emplacement, crystallization, and cooling of the  
22 magma<sup>1</sup>. Even the most basic of questions, such as whether or not crystals form within the  
23 melt and settle to the floor<sup>2-3</sup> or are nucleated and grow *in situ* (i.e., by heterogeneous  
24 nucleation in direct physical contact with the solid base)<sup>4-5</sup>, remain open to controversy after  
25 a century of dedicated research effort. Progress will be difficult without resolution of such

26 fundamental questions. A common approach to attempt to resolve these matters is to produce  
27 a forward geochemical or physical model and use it to demonstrate the feasibility of a  
28 particular hypothetical process; however, there are many examples of cases where totally  
29 contradictory hypotheses can be framed in forward models that successfully match  
30 observations<sup>6-7</sup>. The resolution of these questions is of more than purely academic interest  
31 because some of the layers in question contain mineral deposits of global economic  
32 significance<sup>8</sup>. It is therefore essential to address the larger framework within which individual  
33 hypotheses are framed before we can make reasoned judgements about which ones to favor.

34

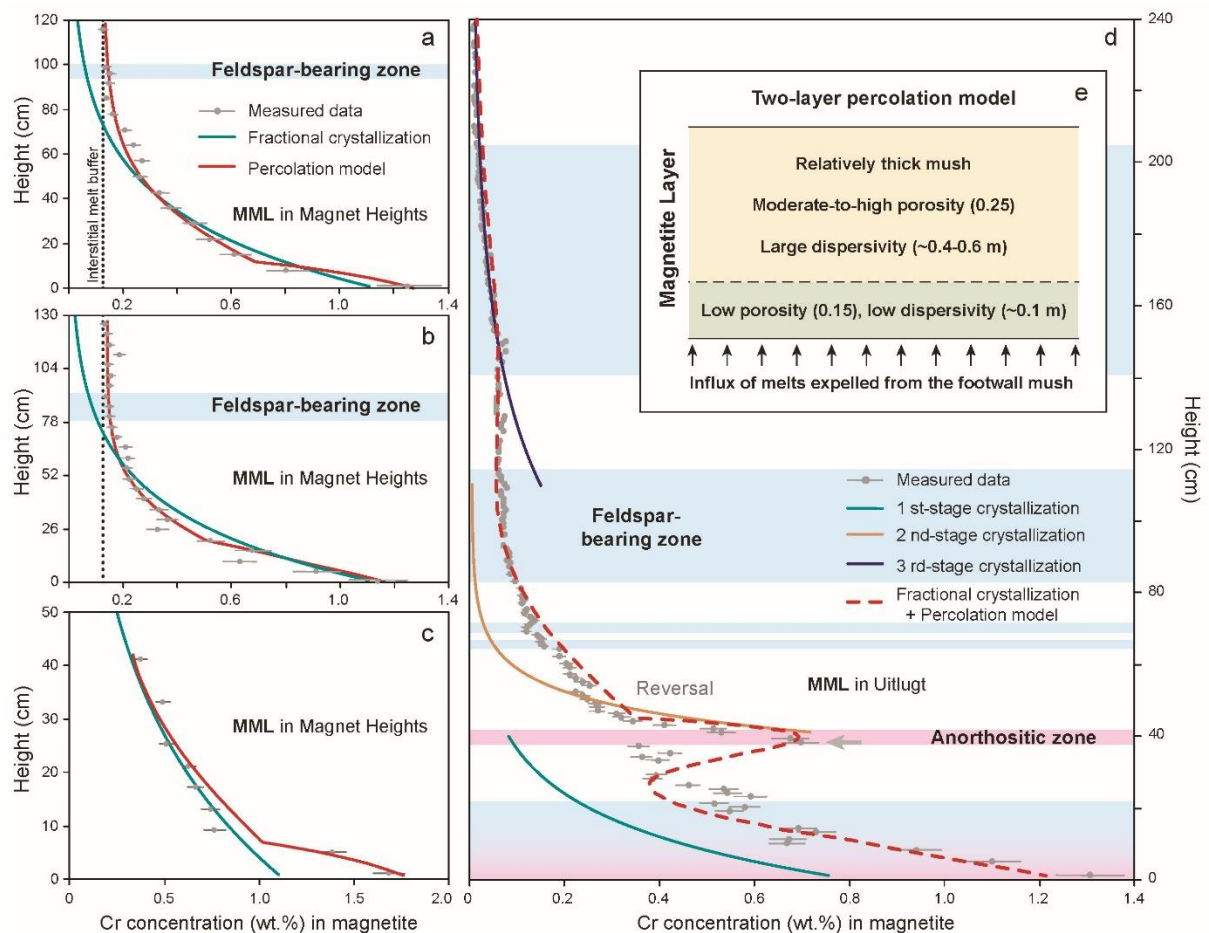
35 One deposit that has long been used as a laboratory to address the controls on layer formation  
36 in layered intrusions is the Main Magnetite Layer (MML) in the Upper Zone of the  
37 Rustenburg Layered Suite of the Bushveld Complex of South Africa<sup>9-19</sup>. Here we compare  
38 observations of the MML with some forward thermodynamic and fluid dynamic models,  
39 appealing to various combinations of fractional crystallization, compositional convection  
40 during adcumulus crystal growth, and reactive transport of upward-migrating Cr-rich melt  
41 from beneath the MML. By comparison of our models with previously published work we  
42 show that multiple model approaches can achieve similar results, with the result that no  
43 model can be used definitively to exclude any other; however, some models are intrinsically  
44 more appealing than others because they require fewer arbitrary adjustments to achieve a  
45 successful fit to the data. In the immediate context of mechanisms of layer formation in  
46 layered intrusions, we show that purportedly incontrovertible evidence for *in situ*  
47 crystallization of magnetite can better be used to support a role for melt infiltration from  
48 below an initially homogeneous magnetite crystal mush, but the choice of model remains a  
49 matter of preference.

50

51 The Upper Zone of the Bushveld Complex crystallized from an iron-rich basaltic andesite  
52 magma at the top of the Rustenburg Layered Suite in what appears to have been a melt-  
53 dominated sill hundreds of km wide and at least several hundred m deep<sup>17</sup>. The evolution of  
54 this magma and its cumulates can be modeled broadly as the product of a protracted process  
55 of fractional crystallization at the scale of the entire Upper Zone<sup>3,17,20</sup>, however the Upper  
56 Zone contains at least 26 magnetite layers that are ~0.1-10 m thick and concordant with the  
57 igneous layering. The formation of these magnetite layers cannot be accommodated by a  
58 simple process of fractional crystallization from a single homogeneous body of liquid the size  
59 of the entire Upper Zone. Instead, it seems probable that a repeated cyclic process of  
60 spontaneous double diffusive convection occurred wherein the rejected liquid complementary  
61 to the gabbroic cumulates was denser than the main body of magma, causing the system to  
62 become stratified with a dense Fe-rich residual melt separating the growing cumulate pile  
63 from the rest of the magma<sup>17,21</sup>. It is thought that when magnetite became saturated and a  
64 layer of magnetite was crystallized from this dense Fe-rich layer the liquid density rapidly  
65 diminished, causing convective overturn and re-homogenization of the magnetite-depleted  
66 basal layer with the rest of the magma and terminating magnetite crystallization until the  
67 closing stage of the next such cycle. Magnetite layers generally have sharp and smooth or  
68 undulating boundaries with underlying anorthosites, but grade upwards into magnetite-rich  
69 gabbro due to smoothly increasing plagioclase modal abundance<sup>13,17</sup>. Along with abundant  
70 anorthosite xenoliths, narrow plagioclase-rich layers are widely developed within magnetite  
71 layers (Fig. 1). The other portions are composed almost entirely of magnetite with minor  
72 interstitial plagioclase and pyroxene, effectively constituting monomineralic adcumulates.  
73 The economically important Main Magnetite Layer (MML), near the bottom of the Upper  
74 Zone, has ~1-2.5 m thickness and crops out intermittently over hundreds of km of strike  
75 extent<sup>10-14,22</sup>. It has been the focus of efforts to explain magnetite formation.

76

77 Although the origin of magnetite layers has spurred an ongoing debate between various  
78 hypotheses<sup>22</sup>, there has been general agreement that the magnetite we observe now is an  
79 adcumulate evolved from a magnetite-dominated mush with a high initial amount of  
80 interstitial melt<sup>13,23</sup>. However, Kruger and Latypov<sup>18</sup> recently proposed that this mushy zone  
81 had negligible thickness, being deposited directly onto a solid substrate *in situ* to form  
82 magnetite adcumulates with little or no initial interstitial melt. The highly compatible  
83 behavior of Cr in magnetite<sup>24</sup> makes it sensitive to magmatic processes (e.g., crystallization  
84 and reactive melt infiltration), and the quasi-exponential vertically decreasing Cr gradients in  
85 monomineralic magnetite layers have been widely attributed to fractional crystallization at  
86 or near the magma chamber floor<sup>10-12,14-15,18</sup>. Failure of a simple one-stage Rayleigh  
87 fractionation model to match observed compositional profiles (e.g., [Fig. 1a-d](#)) led to  
88 propositions that the supply of Cr was modulated by double-diffusive convection in overlying  
89 melt domains<sup>10-12,14-15</sup>. By repeatedly changing the Cr concentration in the melt, model trends  
90 could be produced that matched the data. Models of this sort can be shown to be consistent  
91 with observations but are not persuasive means to compare hypotheses when the boundary  
92 conditions are endlessly changeable, thus ensuring that a fit to the data can always be  
93 achieved.



94

95 **Fig. 1 (a-d)** Measured and modeled vertical profiles of Cr contents in magnetite from the  
 96 MML. Grey arrow in (d) reflects a major reversal in Cr content where a new crystallizing  
 97 sequence began. (e) Diagram for the two-layer percolation model. All data collected from  
 98 multiple sources cited in the text, e.g., profiles in (a-b)<sup>22</sup>, (c)<sup>18</sup> and (d)<sup>12</sup>.

99

100 **Results**

101 **Fractional crystallization models.** Vertical profiles of Cr distribution in the MML, shown  
 102 from Magnet Heights and Uitlugt in Figure 1, commonly show a curvilinear upward  
 103 depletion from the bottom, which broadly matches expectations based on the compatibility of  
 104 Cr during fractional crystallization of magnetite<sup>10-12</sup>, but in each case the predictions do not  
 105 match the data exactly. The Cr content decreases markedly more steeply at the base of every  
 106 section, it locally shows extreme reversals (Fig. 1d), and it becomes constant at higher levels,

107 none of which it could do during fractional crystallization unless the melt composition or  
108 basic physical parameters changed repeatedly during the process<sup>14-15</sup>. A further complicating  
109 matter is the common occurrence of exceptionally Cr-rich nodes decorating the basal contact  
110 of the MML, showing strong lateral as well as vertical compositional gradients<sup>16,18-19</sup>. These  
111 features have been taken as evidence that magnetite initially nucleated and crystallized  
112 around discrete nodes which coalesced as they grew outwards and upwards<sup>16,18</sup>.

113

114 We begin by addressing the profiles at Magnet Heights, shown in Figure 1a-c. The more  
115 complex Uitlugt profile will be addressed afterward. Here we assume that the MML is  
116 crystallized from a thick magma sheet, and that magnetite is the sole crystalline phase during  
117 the formation of the crystal mush of the MML (Methods). In Figure 1 we show models of  
118 fractional crystallization of magnetite from a homogeneous body of melt containing 57 ppm  
119 Cr<sup>3,25</sup>, with  $f_{O_2}$  between zero and one log unit below the quartz-fayalite-magnetite oxygen  
120 buffer (i.e.,  $-1 < \Delta QFM < 0$ )<sup>20,26-27</sup> using a magnetite-melt partition coefficient of 200 at  
121 1150 °C<sup>14,24</sup>. If the initial thickness of the overlying magma is set to 120 m as shown (Table  
122 1), on the order of the vertical distance to the next magnetite layer up section<sup>17,20,28</sup>, then the  
123 exponential decrease in Cr between 10 and 40 cm is reproduced by the model but in the upper  
124 portions of all of the profiles the model predicts much greater depletion of Cr than is  
125 observed. Furthermore, there is a short interval at the base of the MML in which Cr decreases  
126 much faster than predicted by the model (Fig. 1a-c). A simple model of fractional  
127 crystallization alone therefore only matches a short interval of the observed profile and is  
128 inadequate. Like our predecessors<sup>14-15</sup>, we conclude that more complex processes have been  
129 at play.

130

131 We propose two mechanisms to account for the departures of the Magnet Hill Cr profiles  
132 from fractional crystallization, both contingent on the assumption that the magnetite  
133 cumulate was initially solidified as a mush that retained some porosity occupied by interstitial  
134 melt<sup>13,23</sup>. This could be consistent either with *in situ* crystallization or settling of magnetite  
135 crystals formed by homogeneous nucleation. First, convection of intercumulus melt at the top  
136 of the pile<sup>29-30</sup> might have promoted thorough re-equilibration of magnetite grains with the  
137 overlying melt body (i.e., *compositional convection*) to homogenize the upper portions of the  
138 pile and generate the flat profiles above ~60-80 cm in all profiles. Second, expulsion of Cr-  
139 rich intercumulus melt from underlying anorthositic cumulates (i.e., *reactive melt infiltration*)  
140 might have allowed upgrading of Cr contents in the basal parts of the layer. Both processes  
141 depend on the rapid re-equilibration of magnetite crystals with percolating melt, which we  
142 address below after considering the effects of compaction of the mush.

143

144 ***Compaction of magnetite mush.*** Isolated plagioclase crystals surrounded by magnetite  
145 record weaker deformation than clustered grains, suggesting that magnetite mush contained  
146 enough interstitial melt during early compaction to let the isolated crystals move or rotate  
147 freely<sup>23</sup>. Poikilitic pyroxene in magnetite commonly encloses many separate and loosely  
148 packed subhedral-euhedral magnetite grains<sup>13</sup>; therefore, whether magnetite crystallized in-  
149 situ or settled into place, there was initially substantial intercumulus melt. The sizes of the  
150 primary grains (~0.5-2 mm) are similar with that of disseminated magnetite in associated  
151 cumulate rocks, but far less than the crystals in silicate-free regions (~5-20 mm)<sup>13</sup>, indicating  
152 extensive overgrowth, compaction, and annealing to form the adcumulates. If it formed by a  
153 process of fractional crystallization as a loose pile of settled crystals<sup>3-4</sup> or as a loose  
154 arrangement of chains of magnetite crystals nucleated against others *in situ*<sup>16,18-19</sup> and  
155 accumulated faster than the rate of compaction, the initial magnetite cumulate may have had



156 a high porosity ( $\sim 0.52$ )<sup>31</sup>. Compaction is driven by the large density contrast between solid  
157 ( $\sim 4955 \text{ kg}\cdot\text{m}^{-3}$ ) and liquid ( $\sim 2683 \text{ kg}\cdot\text{m}^{-3}$ ) (Methods). Generally, this process initiates with a  
158 mechanical reorganization to achieve optimum packing and continues with pressure  
159 solution/precipitation and possible viscous deformation of cumulate minerals<sup>29,31-33</sup>. The  
160 thickness of the MML ( $< \sim 2.4 \text{ m}$ ) may not have provided sufficient gravitational load and  
161 effective stress to drive plastic deformation of magnetite<sup>23</sup>, and hence we argue that the  
162 compaction process was likely to have been achieved mainly through pressure solution and  
163 reprecipitation. On the basis of centrifuge experiments, Manoochehri and Schmidt<sup>31</sup>  
164 quantified the compaction rate for a chromite cumulate in basaltic magma via dissolution-  
165 precipitation compaction. Given the affinity between chromite and magnetite, here we  
166 assume that the rate of chemical compaction for a magnetite mush layer is similar to that for  
167 chromite cumulate. The required time to reduce porosity from the initial value ( $\sim 0.52$ , the  
168 average porosity after crystal settling) to about 0.25 in a magnetite cumulate is in the range  
169 from 10 to 40 years, or even less if crystals are smaller (Fig. 2a; see [Methods for details of](#)  
170 [this calculation](#)).

171

172 ***Compositional convection and re-equilibration.*** While compaction is under way, continued  
173 crystallization of magnetite from intercumulus melt locally decreases the density of  
174 interstitial melt to a value lower than that of overlying magma, possibly causing it to rise as  
175 buoyant plumes into the overlying melt that must be convectively replaced by melt from the  
176 main magma body above the cumulate in the process of compositional convection<sup>29-30</sup>.

177

178 During this process, the interstitial melt is continuously expelled and replaced by the  
179 overlying melt from the main magma body, maintaining a near-constant composition for the  
180 pore melt<sup>29,34</sup>. The occurrence of significant compositional convection requires that the

181 dimensionless local solutal Rayleigh number ( $R_a$ ) exceeds a critical value of about 25<sup>30,35</sup> and  
182 that the characteristic convective velocity is higher than the rate of crystal accumulation<sup>29</sup>.  
183 The latter requirement is easily achieved in deep and large intrusions (e.g., the Upper Zone of  
184 the Bushveld Complex) due to the small accumulation rate ( $\sim 0.1\text{-}1\text{ m}\cdot\text{year}^{-1}$ )<sup>29</sup>. Therefore, the  
185 minimum thickness of the magnetite cumulate pile to allow compositional convection to be  
186 operative can be estimated when the  $R_a$  value of porous medium equals the critical value  
187 (Methods). Figure 2b shows the minimum mush layer thickness to achieve the critical  
188 Rayleigh number permitting the onset of compositional convection for a range of grain sizes  
189 if the decrease of melt density is driven by the crystallization of a further 2.5 wt.% magnetite  
190 from the intercumulus melt. For example, convection begins when the thickness of an initial  
191 1 mm-grained crystal mush exceeds  $\sim 0.44\text{ m}$  at 25% porosity (black dot in Fig. 2b), while a  
192 lower porosity (15%) will require a much thicker magnetite cumulate ( $\sim 2\text{ m}$ , grey dot in Fig.  
193 2b) for compositional convection. Compositional convection is therefore expected to operate  
194 in the loosely packed upper portion of the mush layer but will cease once compaction has  
195 reduced porosity.

196

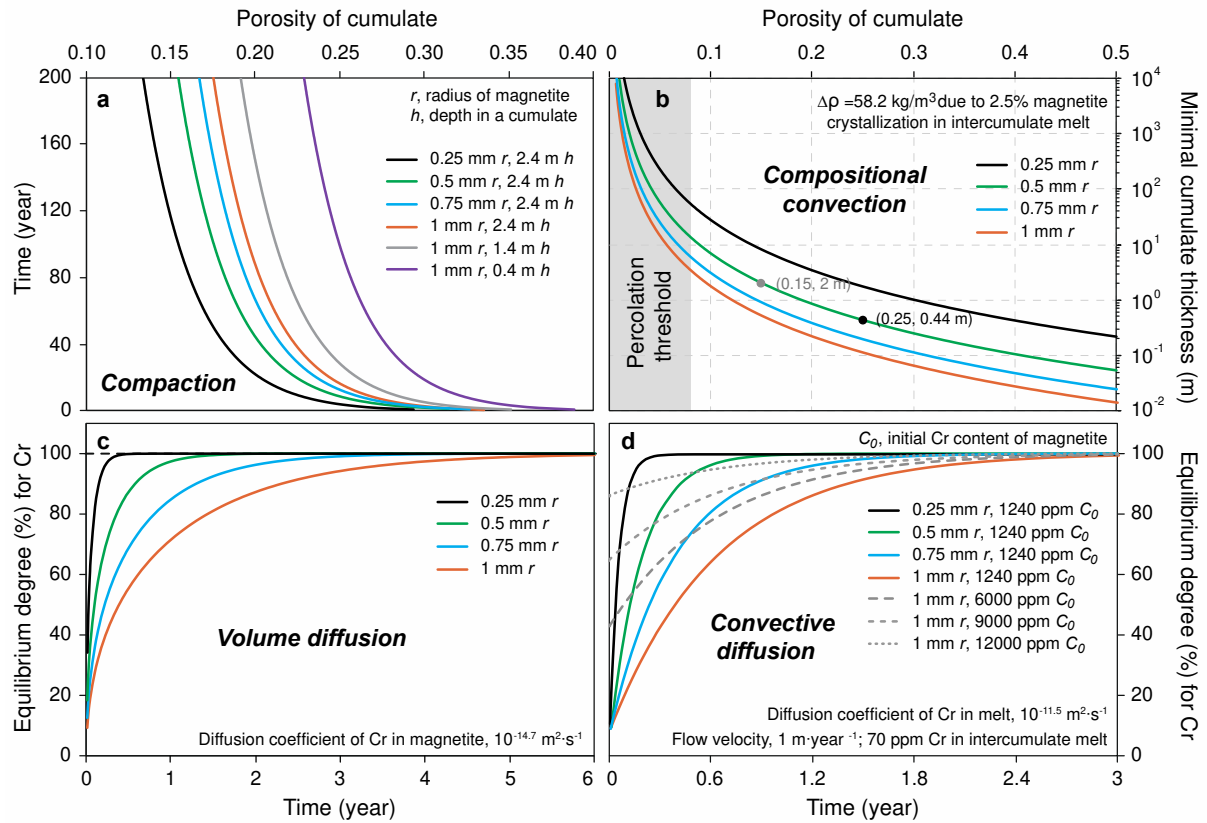
197 During compositional convection and percolation, the interstitial melt will continuously  
198 modify the compositions of packed magnetite grains. Previous investigations have identified  
199 occurrences of compositionally heterogenous magnetite grains, but the observed systematic  
200 zonations of Al and Mg are solely related to the exsolution of spinel phases in the sub-solidus  
201 stage<sup>36</sup>. Some fluctuations of Cr occur in the rims of magnetite grains in contact with ilmenite  
202 or plagioclase and are also attributed only to the local sub-solidus re-equilibrium<sup>36</sup>. Observed  
203 lack of other types of zonation therefore indicates that any compositional modifications of  
204 magnetite grains driven by the convection and percolation are likely to have been completed  
205 in the early stage of post-cumulate processes. Re-equilibration between convecting interstitial

206 melt and magnetite grains involves two kinetic processes: the volume diffusion in single  
207 magnetite grains and the diffusion of elements through a viscous and diffusive boundary  
208 layer between the surrounding melt and magnetite (i.e., convective diffusion)<sup>37</sup>. The volume  
209 diffusion of Cr in a magnetite grain is simplified as the penetration diffusion of Cr into a  
210 spherical magnetite from an infinite source (i.e., interstitial melt)<sup>38-39</sup> (Methods). Due to the  
211 relative motion between flowing interstitial melt and magnetite, the supply of nutrient for  
212 magnetite growth is maintained fast enough to support our assumption of a constant boundary  
213 composition on the surfaces of magnetite crystals for the volume diffusion<sup>40</sup>. Our models for  
214 magnetite growth controlled by diffusion through a narrow boundary layer against a  
215 convecting melt<sup>40</sup> and volume diffusion within solid magnetite<sup>38-39</sup> confirm that Cr-poor  
216 grains (size < 1 cm) rapidly approach equilibrium with passing melt, and the required time is  
217 reduced with increasing initial Cr contents of the grains (Fig. 2c-d). Most magnetite grains  
218 would attain local equilibrium with the percolating melt in the post-cumulus stage, except for  
219 the retention of Cr gradients in rare coarse crystals (> 1 cm)<sup>36,41</sup>. We therefore assume that Cr  
220 content of magnetite in the dominant fine-grained population will be equilibrated with  
221 percolating melts.

222

223 Based on the Cr content of clinopyroxene in the closest hanging wall of the MML<sup>27</sup>, the  
224 overlying magma is proposed to have contained ~6.2 ppm Cr after the formation of the  
225 magnetite mush. Continuous loss of buoyant interstitial melt and its replacement by Cr-  
226 depleted melt containing ~6.2 ppm Cr would maintain the upper magnetite grains at a  
227 constant Cr content (~1240 ppm), which fits the profiles in Fig. 1a-b. Plagioclase can appear  
228 in the sequence without any correlation to the Cr trend (Fig. 1), because in this situation Cr  
229 content of magnetite has become decoupled from its initial value, whereas plagioclase would  
230 resist diffusive re-equilibration because solid state diffusion is so slow in feldspar<sup>42</sup>. We

231 conclude that flattening of the profiles could have resulted from compositional convection  
 232 within the magnetite mush rather than requiring changes in the composition or depth of the  
 233 magma column from which it initially crystallized (cf. McCarthy and Cawthorn<sup>12</sup>).



234  
 235 **Fig. 2** Modeling results for (a) variation of magnetite mush porosity with time due to  
 236 compaction; (b) Minimum thickness allowing compositional convection for mushes with  
 237 indicated porosities. More thoroughly compacted lower portions of the mush may not  
 238 spontaneously initiate compositional convection whereas more open upper portions could; (c)  
 239 Approach of the bulk composition of magnetite grains (integrated over the entire diffusion  
 240 profile) during volume diffusion of Cr due to re-equilibration of magnetite with migrating  
 241 intercumulus melt; (d) Approach of bulk composition to equilibrium via convective diffusion  
 242 for various sizes and initial Cr contents of magnetite. Coloured lines are for crystal sizes as in  
 243 (a, c), and dashed lines represent magnetite grains with different initial Cr contents. Time  
 244 scales of diffusive re-equilibration are much shorter than the time scale of compaction,

245 supporting the assumption of instantaneous magnetite-melt equilibrium in the percolation  
 246 model.

247 **Table 1** Parameters for the fractional crystallization and reactive melt infiltration models

Vertical profile	Fractional crystallization		Reactive melt infiltration model		
	Initial Cr in magma (ppm)	Thickness of magma sheet (m)	Cr in upwelling melt (ppm)	Initial Cr in magnetite (ppm)	Duration time (year)
Section-a	57	120	73.5	1240	70
Section-b	57	120	73.5	1240	58
Section-c	57	120	108	1240	51
Section-d	1-st	40	70	*	24
	2-nd	40			
	3-rd	7.5			

248 \* The initial Cr content of magnetite in this profile follows the variation driven by the first,  
 249 second, and third fractional crystallization events, and is shown as the coloured lines in  
 250 [Figure 1d](#).

251

252 **Reactive melt infiltration.** The footwall of the MML is proposed to have been permeated by  
 253 interstitial melt that carried a slightly more primitive signature with it from lower parts of the  
 254 UUMZ crystal pile, causing reverse zoning of cumulus plagioclase<sup>21</sup> and crystallization of  
 255 more anorthitic interstitial grains<sup>23</sup>. This slightly more primitive interstitial melt would retain  
 256 high Cr content because it had not yet equilibrated with magnetite in the footwall of the  
 257 MML, further either enhanced by crystallization of Cr-free plagioclase in the footwall  
 258 anorthositic mush or minimally diminished by the onset of crystallization of trace amounts of  
 259 intercumulus magnetite. This Cr-rich melt would be discharged upward by the compaction of  
 260 the footwall mush<sup>28</sup>. Hence, we presume an upward percolation of Cr-rich melt into the base  
 261 of the recently formed magnetite mush, causing continuous modification of the Cr  
 262 concentrations of magnetite grains.

263

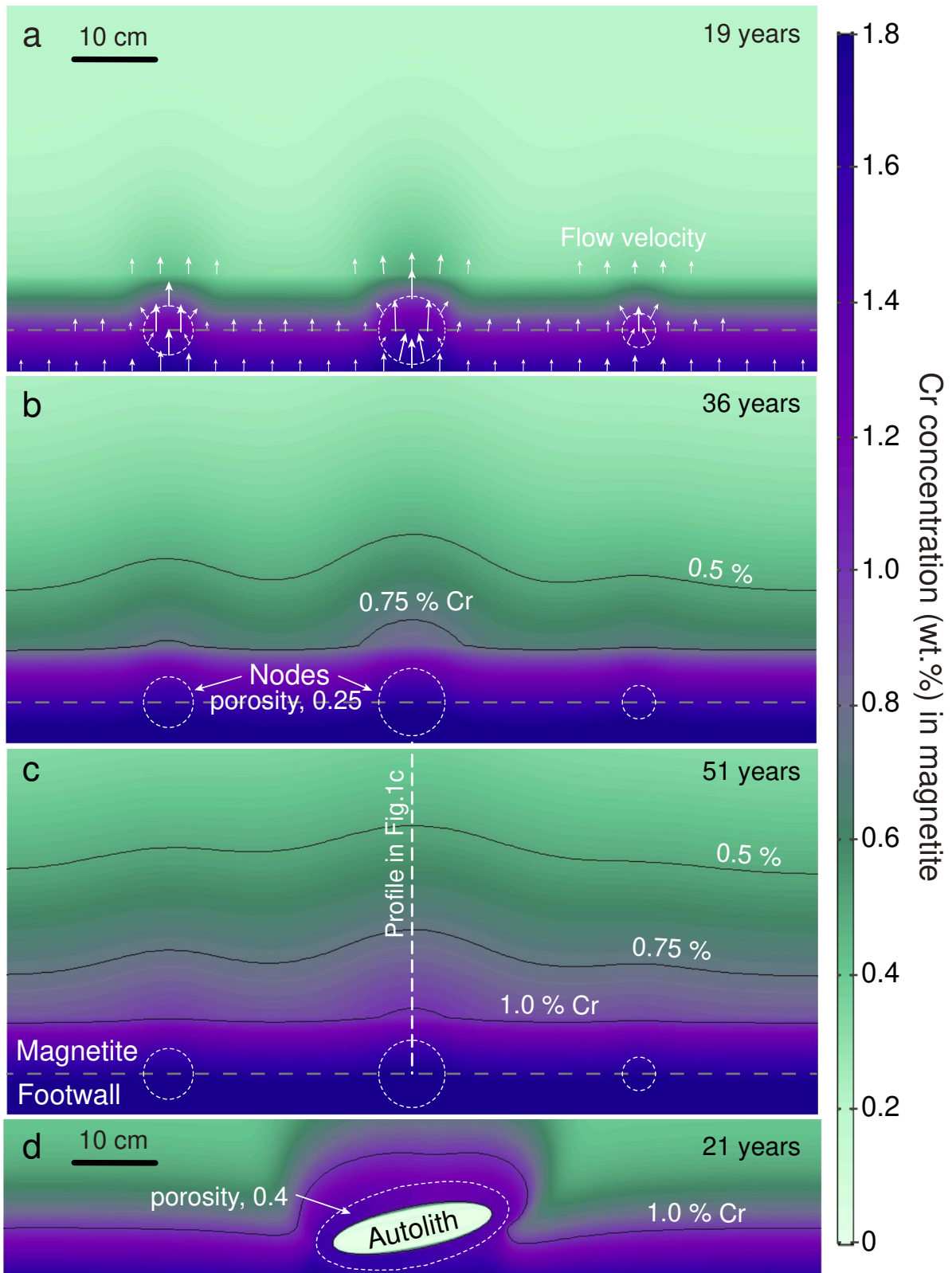
264 We solved this reactive transport problem using the Brinkman equations coupled with  
265 consideration of the dispersion around magnetite crystals via COMSOL Multiphysics for a  
266 two-dimensional system with a horizontal magnetite layer sitting above an anorthositic layer  
267 ([Methods](#)). The variation of porosity in the compacting magnetite cumulate as shown in  
268 [Figure 2a](#) is simplified as two-layer model: a thin bottom zone (10 cm) with low porosity is  
269 overlain by thick mush with a higher porosity ([Fig. 1e](#)). In our model, a slightly Cr-enriched  
270 interstitial melt (~70-74 ppm Cr for the profiles in [Figs. 1a-b, d; Table 1](#)) is expelled from  
271 underlying anorthositic mush at a compaction-driven velocity of  $\sim 1 \text{ m}\cdot\text{year}^{-1}$  <sup>29,32-33</sup>. The  
272 basal magnetite from the section in [Figure 1c](#) contains a larger Cr content ( $\sim 1.7 \text{ wt.}\%$ ) and,  
273 accordingly, the magnetite-saturated melt expelled from footwall mush is assumed to have  
274 had a higher Cr content ( $\sim 108 \text{ ppm}$ , [Table 1](#)), which is equal to the Cr amount of the  
275 intruding parental magma above the Pyroxenite Marker<sup>43</sup>.

276

277 As a simple end-member scenario, the red lines in [Figure 1a-c](#) show the results of the  
278 percolation model imposed on a pile of magnetite crystals along a one-dimensional profile  
279 through the top of one of the nodes in the two-dimensional model described below ([Fig. 2](#)).  
280 To identify better the influence of the percolation process, the magnetite grains in the whole  
281 cumulate pile are assumed initially to be equilibrium with a Cr-depleted, evolved melt ( $\sim 6.2$   
282 ppm Cr, [Table 1](#)), i.e., they initially contain a constant Cr content ( $\sim 1240 \text{ ppm}$ ) in the whole  
283 profile due to the effects of the compositional convection described above. Our percolation  
284 model trends account well for the steepening of Cr compositional gradients at the bottoms of  
285 all four one-dimensional sequences documented in [Figure 1a-d](#), and the flat upper portions  
286 remain as imposed by reaction with the overlying melt due to compositional convection  
287 within the mush.

288

289 ***Lateral variations in Cr concentration.*** Others have inferred that concentration contours  
290 concentric around node-like structures at the base of the MML were caused by *in situ* growth  
291 that was initiated at discrete points on the floor<sup>16,18-19</sup>. Instead, we used our two-dimensional  
292 model of percolative reactive transport to address the alternative hypothesis that local  
293 variations in porosity in the underlying anorthosite mush may allow some sites to become  
294 preferred pathways for underlying melt during upward percolation. If Cr-rich melts flow  
295 through these nodes with a higher velocity and greater net mass flux, the grains here are able  
296 to extract more Cr than surrounding crystals, inducing lateral variations in Cr content along  
297 the base (Fig. 3a). Along with melt dispersion and upward flow in mush, this effect extends  
298 outwards to form undulations in Cr-concentration contours which gradually weaken over  
299 ~20-30 cm height (Fig. 3b-c), coinciding with the geochemical mapping on field outcrops of  
300 the MML<sup>18</sup>. We included an anorthosite autolith in the basal mush and considered that a solid  
301 block may impede early compaction of magnetite, leaving a high-porosity domain around the  
302 block which would enhance percolation. The resulting perturbation in the flow field led to  
303 concentric Cr enrichment around this autolith (Fig. 3d), producing spatial variations very  
304 similar to those shown by Kruger and Latypov<sup>18-19</sup>. This also partly accounts for the  
305 confusing phenomenon that Cr-rich growth nodes are found in the nooks beneath the  
306 autoliths but not directly on the upper surfaces of autoliths<sup>18-19</sup>.



307

308

309

310

**Fig. 3 (a-c)** Snapshots of modeled Cr compositional mapping for the percolation of basal magnetite mush with three high-porosity spheric nodes (0.25, white dashed lines). White arrows in (a) plot for melt flow vectors. (d) Cr contours drape over the anorthosite autolith



311 due its high-porosity contact zone (0.4, white dashed line) with magnetite mush. Black lines  
312 represent Cr isopleths.

313

314 ***Multiple episodes of fractional crystallization combined with reactive transport.*** More  
315 complex profiles than those at Magnet Heights (Fig. 1a-c) require more complex adjustments  
316 to boundary conditions. We have combined the fractional crystallization model with the  
317 reactive transport model to address the complexity of the Uitlugt profile, which was sampled  
318 at 1 cm spacings. The large excursion in Cr content at 40 cm shown in Figure 1d, which  
319 contains about 65% plagioclase, was reported not to be present in flanking boreholes by  
320 McCarthy and Cawthorn (1983), who attributed it to unspecified local convective  
321 fluctuations in magma composition during magnetite crystallization. To account for the  
322 evident presence of three segments of the profile, each decaying from progressively lower  
323 starting points, we accept the possibility that convective overturn after deposition of the first  
324 ~ 40 cm of magnetite led to the start of a new episode of fractional crystallization from fresh  
325 Cr-rich melt, and a third such event occurred after deposition of about 110 cm of magnetite,  
326 however we do not resort to a change in magma composition every time the profile departs  
327 from a perfect exponential decay. In Figure 1d we show the three initial profiles of Cr due  
328 purely to fractional crystallization, as well as the trend resulting from 24 years of continued  
329 evolution of Cr due to percolative reactive transport of Cr-rich intercumulus melt from below  
330 (Table 1). The combined fractional crystallization and reactive transport model duplicates all  
331 the observed peaks and inflections in slope of Cr variation, using only three discrete  
332 crystallization sequences, for each of which the only adjustable parameter was the initial Cr  
333 content of the new magma.

334

335 **Discussion and comparison with previous work**

336 The difference between models of *in situ* magnetite crystallization and collection of loose  
337 crystals of magnetite may seem like a moot point, but it drives at the difference between two  
338 fundamentally different mechanisms of layer formation. If crystals in layered intrusions form  
339 within the melt and are transported to their eventual resting places, the implications for  
340 petrogenesis are dramatically different than they are if crystals form *in situ* on a hard  
341 substrate. A very wide range of possibilities is consistent with deposition of a mush, but only  
342 one is permitted if crystallization proceeded *in situ*. It should be evident at this point that the  
343 match between observations and models afforded both by our models and by the previously  
344 published models places one in the uncomfortable position of having to choose between two  
345 very different sets of mechanisms to explain exactly the same observations with equal  
346 fidelity. In summary, all existing models including ours rely fundamentally on the idea that  
347 fractional crystallization of magnetite has generated the rapid and approximately exponential  
348 decay of Cr concentration in vertical profiles. The models differ in their explanations of  
349 inflections or reversals in the Cr profiles (Fig. 1) and of perturbations of Cr concentration  
350 contours due to lateral variations (Fig. 3).

351

352 Previous workers have inferred that the deposition of magnetite occurred *in situ* along the  
353 base of the magma chamber<sup>10-12,16,18</sup>. In these models there was no significant role for trapped  
354 liquid between magnetite crystals as they formed; lateral variations in Cr content were  
355 accounted for as resulting from initial magnetite nucleation at isolated growth centres that  
356 grew concentrically until they merged, and inflections in slope of Cr concentration in vertical  
357 profiles were accounted for by numerous adjustments to the boundary conditions<sup>16,18</sup>. These  
358 adjustments varied from one model to the next, but all were designed to control the supply of  
359 Cr to the fractionating magnetite. In our models, magnetite collected as discrete crystals to  
360 form an orthocumulate-textured mush during fractional crystallization; reversals of Cr

361 gradients in more complex profiles are accounted for by as many as three episodes of  
362 convective overturn in the overlying magma (Fig. 1d). Percolation of melt through the mush  
363 could both have erased early compositional gradients in the upper portions of thick mushes  
364 and might also have allowed upward-migrating intercumulus liquid from below to steepen  
365 the Cr concentration gradients near the base. Lateral variations in Cr concentration are  
366 accounted for by postulating that the porosity of the underlying anorthosite mush was not  
367 uniform; randomly distributed local enhancements in anorthosite permeability drove  
368 increased percolation at discrete locations in the base of the mush, generating concentrically-  
369 zoned domical patches of Cr enrichment (Fig. 3).

370

371 Geological observations can impose the primary criteria for testing these forward models.  
372 First, we consider whether it is more likely that magnetite arrived in its present location by *in*  
373 *situ* growth due to heterogeneous nucleation on a hard floor or by homogeneous nucleation in  
374 the melt followed by crystal settling to form a mush. The presence of plagioclase with  
375 magnetite in continuously variable proportions along with the abundance of autoliths is  
376 suggestive of co-accumulation of discrete grains due to crystal settling. The preservation of  
377 considerable relict intercumulus volume in pyroxene-poikilitic domains<sup>13</sup> indicates that  
378 whether it occurred heterogeneously or homogeneously, magnetite crystallization initially led  
379 to formation of an open framework through which intercumulus melt would have been free to  
380 migrate before compaction and/or adcumulus growth brought porosity down below the  
381 percolation threshold (Fig. 2b)<sup>29,44</sup>. In-situ crystallization experiments at low undercooling  
382 document the homogeneous nucleation of magnetite in basaltic melt even after initial  
383 heterogeneous nucleation on a solid substrate<sup>45</sup>. During diffusion-controlled growth, the  
384 preferred sites of incipient nucleation and rapid growth of magnetite would be expected to lie  
385 at the highest points on the surface rather than in deep reentrants under autoliths, regardless

386 of their origins as relicts of eroded floor rocks<sup>19</sup> or as xenoliths. Taken all together, these  
387 observations and interpretations alone do not allow a firm choice between homogeneous  
388 nucleation followed by crystal settling, or heterogeneous nucleation *in situ* at the magma  
389 chamber floor, but we lean towards a process dominated by homogeneous nucleation from  
390 the melt followed by settling of magnetite to form a highly permeable initial cumulate  
391 framework.

392

393 The choices of model parameters and boundary conditions may reflect the robustness and  
394 reliability of these forward models. Previous models resorted to diffusive addition of Cr from  
395 convecting layers above<sup>10-12,14-15,46</sup>. Considering diffusive supply from a reservoir of fixed  
396 size to the crystallizing magnetite Wright et al.<sup>46</sup> and McCarthy et al.<sup>14</sup> found that, to fit the  
397 measured profiles for a fixed value of the Cr diffusion coefficient  $D$  in the melt, the rate of  
398 crystal accumulation had to be arbitrarily constrained by the observed compositions. The rate  
399 of crystal accumulation implied by the model would require as much as 3 Myr to crystallize  
400 the 1 m thick MML<sup>15</sup>. Alternatively, Kruger and Smart<sup>15</sup> showed that the profile could be  
401 fitted very well by assuming a continuous influx of Cr into the crystallizing layer of magma  
402 from an upper layer, above a stable diffusive boundary layer, in a stratified magma  
403 undergoing double diffusive convection driven by crystallization at the base and heat loss out  
404 the top. However, their model could only be made to fit the observed compositional profiles  
405 by adopting extremely high and unrealistic values of  $D_{Cr}$  of about  $10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$  whereas  
406 measured values in comparable andesitic melts are about 300 times smaller ( $3.2 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$   
407 at  $1150 \text{ }^\circ\text{C}$ )<sup>47</sup>. It is possible that by changing other model parameters their curves could be  
408 brought into line with measured data. The parameter least well constrained in their model is  
409 the thickness of the diffusive boundary layer separating the crystallizing lower layer from the  
410 layer above it. Since they never specified the value that they chose for the boundary layer

411 thickness it is impossible to judge whether or not it could accommodate a change sufficient to  
412 match the data using correct values for  $D_{Cr}$ . In any case, both of these previous models work  
413 only if a key model parameter such as the thickness of a hypothetical diffusive boundary  
414 layer can be adjusted at will. Evidently, without independent evidence to guide the choices of  
415 such parameters, they can be dialed in to match any profile and the modeling process  
416 becomes an arbitrary exercise in curve fitting.

417

418 Echoing the suggestion of Cawthorn<sup>16</sup>, Kruger and Latypov<sup>18</sup> modeled solidification of the  
419 MML by *in situ* crystallization of magnetite directly attached to a solid substrate in a process  
420 of fractional crystallization from a well-mixed convecting reservoir of melt. As in other  
421 examples, their model would predict a continuous exponential decrease in Cr concentration  
422 and would fail to match the observed flattening of compositional profiles after the initial  
423 rapid decline (Fig. 1). To deal with this issue they added batches of fresh melt to their magma  
424 reservoir in amounts and with Cr contents arbitrarily chosen to force the model Cr trend to  
425 follow the observed trend. For example, Kruger and Latypov<sup>18</sup> required successive injections  
426 of four discrete magma pulses with variable volumes to account for only the lowest 9  
427 measurements in a 40-cm-thick profile (Fig. 1c). When a new pulse of magma is required in a  
428 model to account for every second data point, the model again becomes an arbitrary exercise  
429 in curve-fitting that loses predictive explanatory power. Furthermore, the two-dimensional  
430 model of magnetite crystallization had to use measured Cr concentration contours to guide  
431 their modeled crystallization sequence, rather than having the crystallization pattern governed  
432 by any physical model of diffusion-mediated crystal growth in a diffusive boundary layer.

433

434 There are weaknesses in our models as well. We have chosen boundary conditions and values  
435 for parameters like diffusivities and partition coefficients to match external constraints as

436 well as we can; however, to match the complex Cr concentration profile at Uitlugt we must  
437 resort to three separate magma recharge events timed to generate the observed inflections and  
438 we must perforce select a Cr content of the incoming melt to match the overall pattern. The  
439 principal differences between our models and previous ones are that we minimized the  
440 number of adjustable parameters (e.g., at most three magma influxes rather than one for every  
441 two data points, model melt compositions fixed by equilibrium with measured mineral  
442 compositions, use of experimentally measured compaction rates and diffusion coefficients)  
443 and we accounted for processes widely thought to have occurred in the Upper Zone such as  
444 post-cumulus melt migration and infiltration<sup>23,28,48</sup> which were overlooked by previous  
445 approaches.

446

#### 447 **Conclusion and implications**

448 Forward models of magmatic processes that cannot be directly observed can be valuable as  
449 descriptions of what could plausibly have happened, but they cannot be regarded uncritically  
450 as demonstrations of what really happened. When we are faced with multiple contradictory  
451 models that can adequately generate an acceptable quantitative match to observations, we  
452 must make our final choice based on the law of parsimony, also called Occam's Razor - we  
453 favor the simplest hypothesis that requires the smallest number of artificially imposed  
454 external constraints while simultaneously acknowledging that we cannot be sure of the  
455 answer. A modeling scenario that can be set up to match the observations by setting nothing  
456 more than a plausible set of initial conditions and acceptable parameter choices is greatly  
457 preferable to another that requires continuous and arbitrary fine-tuning of model parameters  
458 to achieve a match to observations, even more so when the arbitrary models can only succeed  
459 through the adoption of physical parameters that are far outside of the ranges dictated by  
460 experimental observations.

461

462 In the balance, although both sets of models can be shown to work to explain the data, we  
463 prefer our model of melt migration and reaction with a magnetite crystal mush that was itself  
464 generated in some localities by multiple pulses of deposition from a magma sheet undergoing  
465 episodic replenishments due to convective overturn. The significance of the conclusion is that  
466 it speaks to a fundamental bifurcation in our understanding of the genesis of layered  
467 intrusions - do crystals form *in situ* on solid substrates or are they formed on one place and  
468 deposited in another by transport and settling? These two concepts have dramatically  
469 important effects in controlling how we understand the formation of layered intrusions in  
470 general and, more specifically, the genesis of critically important mineral deposits of Fe, Ti,  
471 V, Ni, Cu, Co, Pt, Pd etc. The importance of *in situ* crystal growth is not unequivocally  
472 established by observations of the MML. Evidently our current knowledge of the Main  
473 Magnetite Layer does not, as yet, carry the information we need to make a definitive choice,  
474 and it is important for us to remember that as we continue to search for more definitive  
475 answers to a perplexing problem at the heart of igneous petrology.

476

## 477 **Method**

478 **Equations for fractional crystallization model.** In each increment of the fractional  
479 crystallization, the conservation of Cr mass in a closed system requires that:

$$480 \quad C_{i-1}^{melt} \cdot H_{i-1} \cdot \rho_{i-1}^{melt} = C_i^{melt} \cdot k_D^{mt} \cdot (H_{i-1} - H_i) \cdot \rho_{mt} + C_i^{mt} \cdot H_i \cdot \rho_i^{melt} \quad (1)$$

481 where  $C_i^{melt}$ ,  $H_i$  and  $\rho_i^{melt}$  are the Cr content, thickness and density of melt at  $i$ -th increment,  
482 respectively, and the  $k_D^{mt}$  is the partition coefficient of Cr between magnetite and melt, and  
483 here is assumed as 200 based on recent experimental measurements<sup>24</sup>. The density of  
484 magnetite ( $\rho_{mt}$ ) is estimated as  $\sim 4955 \text{ kg} \cdot \text{m}^{-3}$ <sup>49</sup>. The melt density keeps decreasing due to  
485 the crystallization of denser magnetite phase, and if we make the simplifying assumption that

486 the partial molar volume of the Fe<sub>3</sub>O<sub>4</sub> component in the melt is the same as that of magnetite,  
487 its value at the *i*-th increment can be calculated as:

$$488 \quad \rho_i^{melt} = \frac{\rho_0 \cdot H_0 - \rho_{mt} \cdot (H_0 - H_i)}{H_i} \quad (2)$$

489 where  $\rho_0$  is the density of initial melt ( $\sim 2683 \text{ kg}\cdot\text{m}^{-3}$ ), and  $H_0$  is the initial thickness of melt.

490 The thickness of the magnetite layer at the *i*-th increment is expressed as  $(H_0 - H_i)$ , and the Cr  
491 concentration of crystallized magnetite within this increment ( $C_i^{mt}$ ) is calculated as:

$$492 \quad C_i^{mt} = C_i^{melt} \cdot k_D^{mt} \quad (3)$$

493 **Compaction time calculation.** The compaction time (*t*) is the required time to reduce  
494 porosity from the initial porosity ( $\sim 0.52$ ) to a given lower value in the basal layer of a  
495 magnetite cumulate, and can be calculated as<sup>31</sup>:

$$496 \quad t = \frac{5.471 \times 10^{18}}{10^{(14.35 \cdot \varphi)}} \times \frac{d}{h \cdot \Delta\rho} \quad (4)$$

497 where  $\varphi$  is the porosity of cumulate pile, *d* is the diameter of magnetite grains (m), *h* is the  
498 depth in crystal mush (m), and  $\Delta\rho$  is the density difference between the magnetite and  
499 interstitial melt ( $\text{kg}\cdot\text{m}^{-3}$ ). On the basis of our fractional crystallization model<sup>3</sup>, the density of  
500 the evolved melt that corresponds to the position of the MML in the Upper Zone can be  
501 estimated as  $\sim 2683 \text{ kg}\cdot\text{m}^{-3}$  via the parameterized equation from Lesher & Spera<sup>50</sup>.

502 **Compositional convection model.** Based on the critical value of the Rayleigh number ( $R_a$ ,  
503  $\sim 25$ )<sup>30,35</sup>, the minimum thickness ( $h_{min}$ ) of the magnetite cumulate pile allowing  
504 compositional convection to be operative can be estimated as<sup>29</sup>:

$$505 \quad h_{min} = \frac{R_a \cdot \mu_m \cdot D}{k \cdot g \cdot \Delta\rho'} \quad (5)$$

506 where  $\mu_m$  is the melt viscosity ( $\sim 160 \text{ Pa}\cdot\text{s}$ )<sup>3,51</sup>, *D* is the effective chemical diffusivity in the  
507 melt, *k* is the permeability of crystal mush, *g* is the gravitational acceleration, and  $\Delta\rho'$  is the  
508 density difference between the interstitial melt and overlaying magma. The diffusion



509 coefficient of Cr in melt is estimated as  $3.2 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$ <sup>47</sup>, and adopted as the  $D$  here. The  
 510 decrease of melt density driven by the crystallization of 2.5 wt.% magnetite from the pore  
 511 melt is calculated as  $58.2 \text{ kg} \cdot \text{m}^{-3}$ , which can be considered as the  $\Delta\rho'$ . At a high porosity, the  
 512 permeability of crystal mush is simplified as<sup>33,52-53</sup>:

$$513 \quad k = d^2 \cdot \varphi^3 / 300 \quad (6)$$

514 where  $d$  is the diameter of magnetite grains and  $\varphi$  is the porosity of crystal mush.

515 **Equation for the diffusion model.** For the bulk diffusion of Cr in a spherical magnetite  
 516 grain with uniform initial composition and a boundary set to a different fixed composition by  
 517 reaction with an infinite surrounding medium, the equilibrium degree of Cr can be expressed  
 518 as the ratio of  $M_t$  (total amount of diffusing Cr entering or leaving the magnetite at time  $t$ ) to  
 519  $M_\infty$  (corresponds the amount of diffusing Cr after infinite time to reach equilibrium)<sup>38</sup>:

$$520 \quad \frac{M_t}{M_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \cdot e^{-D_{Cr-mt} \cdot n^2 \cdot \pi^2 t / r^2} \quad (7)$$

521 where  $r$  is the radius of magnetite grain,  $t$  is the time and  $D_{Cr-mt}$  is the diffusion coefficient of  
 522 Cr in magnetite ( $\sim 2 \times 10^{-15} \text{ m}^2 \cdot \text{s}^{-1}$ )<sup>24,37</sup>. On the other hand, during the convective diffusion, a  
 523 compositional gradient of Cr persists within a thin diffusive boundary layer around the  
 524 magnetite grain, and the mass transfer ( $dM_{Cr}$ ) of an element Cr via this compositional  
 525 boundary layer within a short time interval ( $dt$ ) can be approximated as<sup>40</sup>:

$$526 \quad \frac{dM_{Cr}}{dt} = 4\pi r^2 \rho_m \cdot D_{Cr-melt} \cdot \frac{C_m - C_0}{\delta_{Cr}} \quad (8)$$

527 where  $r$  is the magnetite radius,  $\rho_m$  is the density of melt ( $\sim 2683 \text{ kg} \cdot \text{m}^{-3}$ ),  $D_{Cr-melt}$  is the  
 528 diffusion coefficient of Cr in melt ( $\sim 3.2 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$ )<sup>47</sup>,  $C_m$  is the Cr content of interstitial  
 529 melt,  $C_0$  is the initial Cr concentration of magnetite, and  $\delta_{Cr}$  is the boundary layer thickness  
 530 for Cr. The Cr flux predicted by this equation is summed for successive time intervals to

531 track the compositional variations of magnetite during this convective diffusion. The  $\delta_{Cr}$  is  
 532 mostly controlled by the strength of convection, and can be expressed as<sup>54</sup>:

$$533 \quad \delta_{Cr} = 2r / \left[ 1 + \left( 1 + \frac{2rV}{D_{Cr-melt}} \right)^{1/3} \right] \quad (9)$$

534 where  $V$  is the flow rate of interstitial melt ( $\sim 1 \text{ m} \cdot \text{year}^{-1}$ )<sup>29,32-33</sup>.

535 **Reactive melt infiltration model.** The Brinkman equations extend Darcy's law to model the  
 536 dissipation of kinetic energy by viscous shear, which is similar to the Navier-Stokes  
 537 equations, and are also connected to a set of reaction-diffusion-convection equations. Here,  
 538 the Brinkman equations for steady state flow are:

$$539 \quad \left\{ -\nabla \cdot \frac{\mu}{\varphi} [\nabla \mathbf{u} + (\nabla \mathbf{u})^T] \right\} - \left( \frac{\mu}{k} \cdot \mathbf{u} + \nabla p - \mathbf{F} \right) = 0 \quad \text{and} \quad \nabla \cdot \mathbf{u} = 0 \quad (10)$$

540 where  $\mu$  is melt viscosity,  $\mathbf{u}$  represents the velocity vector,  $p$  is pressure,  $\varphi$  is the porosity and  
 541  $k$  denotes the permeability of crystal mush. Gravity is also included in this model, and its  
 542 influence is accounted for by the force term,  $\mathbf{F}$ . The grain size of magnetite is assumed as 2  
 543 mm in the model<sup>13</sup>, and the relation between porosity and permeability follows the Kozeny-  
 544 Carman model<sup>55</sup>. The transport of solute Cr in the migrating melt due to liquid dispersion,  
 545 advection and molecular diffusion is modeled via the advection-dispersion equation:

$$546 \quad \varphi \frac{\partial \gamma}{\partial t} + \nabla \cdot (-\varphi \cdot D_L \cdot \nabla \gamma + \mathbf{u} \gamma) = S_C \quad (11)$$

547 where  $\varphi$  is the porosity,  $\gamma$  is the mass concentration of Cr in melt ( $\text{kg} \cdot \text{m}^{-3}$ ),  $\mathbf{u}$  is the flow  
 548 velocity ( $\text{m} \cdot \text{s}^{-1}$ ),  $D_L$  is the hydrodynamic dispersion tensor ( $\text{m}^2 \cdot \text{s}^{-1}$ ), and  $S_C$  represents the  
 549 quantity of Cr added per unit volume of porous medium per unit time at the bottom of  
 550 magnetite cumulate pile ( $\text{kg} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$ ). The hydrodynamic dispersion tensor ( $D_L$ ) describes the  
 551 combined influences of the chemical diffusivity in the melt and the mechanical dispersivity  
 552 via grain-scale flow in porous media. Hence, the diagonal components of the  $D_L$  are  
 553 controlled by the diffusion coefficient of Cr in the andesitic melt ( $\sim 3.2 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$ )<sup>47</sup>, and

554 the dispersivities along the longitudinal and transverse flows. Previous work demonstrated a  
555 systematic increase of the longitudinal dispersivity with the corresponding observational  
556 scale of porous media<sup>56</sup>, and we assume ~0.1 and ~0.4-0.6 m for the two layers in modeled  
557 mush, respectively (Fig. 1e). The transverse dispersivity in experiments is typically an order  
558 of magnitude smaller than that of longitudinal dispersivity<sup>57</sup>, which is also adopted here.

559

#### 560 **Data availability:**

561 All data shown here was obtained from the cited references.

562

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708 J.E.M. and Z.-S.Y. discussed the concepts; Z.-S.Y. performed modeling calculations. J.E.M.  
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711 **Competing interests:**

712 The authors declare no competing interests.

713

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