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DIFFUSION OF TRACE ELEMENTS DURING BOTTOM CRYSTALLIZATION OF DOUBLE-DIFFUSIVE CONVECTION SYSTEMS: THE MAGNETITITE LAYERS OF THE BUSHVELD COMPLEX

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Abstract

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Magmas that crystallize to produce layered intrusions may behave as double-diffusive convective systems. In such an intrusion, the magma would be stratified as a stacked series of thick convecting layers separated by thin, static "diffusive" boundary layers. Crystallization is often inferred to occur at the base of the chamber, and chemical changes due to fractional crystallization of the lowermost convecting layer are transmitted by diffusion across the thin "diffusive" boundary layers separating the thick stacked layers of convecting magma.

In this work the Rayleigh distillation law is extended to describe changes in the concentration of a compatible trace element due to fractional crystallization of the convecting magma layer at the base of the stack as well as diffusion of the trace element into the layer across its top diffusive boundary. General equations are presented defining predicted geochemical profiles in terms of partition coefficient, rate of accumulation of the crystal pile, diffusion rates and thickness of discrete magma layers.

Extraction of a single dense phase from the lowermost convecting layer progressively decreases the density of the residual liquid in the layer. Ultimately, the densities of the basal and immediately overlying layers of magma become equal, the diffusive boundary layer breaks down, and the two basal layers mix. This results in a reversal in the trace-element concentration of the crystals being precipitated. A "sawtooth" trace-element concentration profile results.

The Bushveld Complex provides a good test for the double-diffusive convection model envisaged here. Available data on chromium concentration with respect to height in the main magnetitite layer of the Bushveld Complex are well described by these equations.

Introduction

It has been argued that layered intrusions are stratified in the liquid state by the process of double-diffusive convection (d.d.c.) (e.g. McBirney and Noyes, 1979; Irvine, 1980), and that in many cases crystallization occurs only at the base of the lowermost convecting layer. Owing to the strong convective mixing, each liquid layer is homogeneous, but there are chemical, density and thermal differences between layers (McBirney and Noyes, 1979; Irvine, 1980; Huppert and Sparks, 1980; Chen and Turner, 1980; Huppert and Turner, 1981). The

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diffusive boundaries between the convecting layers therefore represent compositional and thermal discontinuities within the liquid. Chemical and thermal changes brought about by bottom crystallization of the basal convecting layer are transmitted by diffusion across these diffusive boundaries. The rate of diffusion versus the rate of crystallization will thus influence the compositional variation of the crystal products at the base of the crystallizing layer (Fig. 1). If the chemical and mineralogical variations seen in cyclic units are the results of bottom crystallization during d.d.c., the rate of chemical diffusion across the boundary layers is slow but not insignificant compared with the rate of heat transfer and crystallization (Huppert and Turner, 1981).

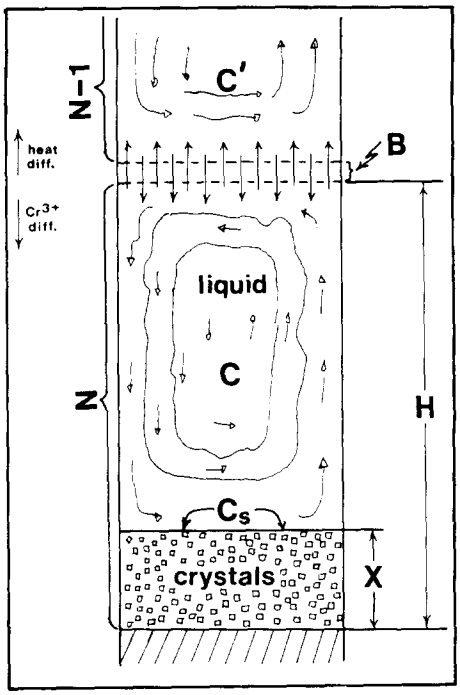

Fig. 1. Schematic diagram (not to scale) illustrating the basal crystallizing layer N, with part of the next higher layer. These are separated by a diffusive boundary of thickness B. The compatible element (Cr) is diffusing down into the basal layer and heat diffuses upward. The concentrations referred to in the text are C, C′ and C_s. H is the initial quantity of liquid in the layer, and X is the quantity of crystals fractionated from the liquid and deposited as a layer.

The concentrations of compatible trace elements in a magma are strongly influenced by the phases that are crystallizing. These elements can be used as petrogenetic indicators to evaluate the thickness of the double-diffusive layers from which cyclic units in layered rocks crystallized, as well as the influence of diffusion across the boundary layers on the chemistry of the rocks.

The Upper Zone of the Bushveld Complex is a strongly layered sheetlike body of iron-rich cumulates. The mineralogy is dominated by plagioclase, magnetite, Fs-rich orthopyroxene and Fa-rich olivine (Molyneux, 1974). In general, the magma is becoming less dense with differentiation as the molten equivalent of the solid assemblages are more dense than the liquid.

Detailed vertical Cr-concentration profiles of the Main Magnetitite layer in the Upper Zone have been obtained by Cawthorn and McCarthy (1980, 1981). These data can be used to model fractionation processes in the magma chamber as the distribution coefficient of Cr between magnetite and liquid is between 100 and 620 (Irving, 1978).

Derivation of the equations

If bottom crystallization and double-diffusive stratification of magmas operate, it would seem reasonable that as a compatible trace element is extracted by fractional crystallization from the basal magma layer in a d.d.c. system, the concentration difference between that layer and the one above it would increase if the diffusive boundary between them acts as a partial seal (Fig. 1). The amount of interlayer diffusion would be proportional to the concentration difference, and the thickness and diffusion coefficient of this boundary layer. Therefore, a stage could be reached where there is an approximate balance between the extraction of the element by the fractionating crystals and the addition of the element by diffusion from above (Fig. 1). As diffusion is also time dependent,

the rate of extraction (i.e. the rate of crystallization) also has to be considered.

In this derivation we *assume* that the magma is behaving as a d.d.c. system, and that crystallization is occurring at the base of the magma. The distribution coefficient (K) of an element between the crystals and coexisting liquid is held constant. Further, after any amount of crystals had precipitated, the trace-element concentration in that increment of crystals (C_s) reflects the concentration in the coexisting liquid (C) within the basal convecting layer from which the crystals formed, i.e. $C_s = KC$ (see Fig. 1). Changes in the concentration of the element in the liquid of the basal crystallizing layer allowing for boundary layer diffusion can be modelled by the following equation:

$$(H-X)\frac{dC}{dX} = C - KC + D'(C'-C) \qquad (1)$$

Where H is the equivalent solid height of the basal layer before any crystallization takes place, X is the height already crystallized and D' is a dimensionless coefficient taking into account the diffusion coefficient and thickness of the diffusive boundary layer separating the basal from the overlying convecting layer, as well as the rate of crystallization and hence time. The concentration of the trace element in the crystallizing basal layer is C and that of the overlying convecting layer is C'.

The three terms on the right of eqn. (1) describe the change in concentration of the element in the liquid due to decreasing quantity of liquid, extraction by the crystals and addition by diffusion from the layer above respectively. Equation (1) can be written more conveniently:

$$\frac{dC}{dX} = -\frac{(K'-1)}{(H-X)}(C-C_c) \qquad (2)$$

where $K' = K + D'$ and $C_c = C'D'/(K'-1)$.

Because diffusion across the boundary layer is a slow process and the distribution coefficient and properties of the boundary layer are assumed to be constant, C' may be assumed constant as a first approximation, in which case the differential equation has an exact solution:

$$\frac{C-C_c}{C_o-C_c} = \left(\frac{H-X}{H}\right)^{(K'-1)} \qquad (3)$$

where C_o is the initial concentration in the liquid layer.

For $K>1$ (i.e. for compatible trace elements) this represents a concentration that tends to a limit C_c. The case $D'=0$ also implies that $C_c=0$ and eqn. (3) reduces to the well known Rayleigh formula. In Appendix 1, an alternative extended derivation of the equations is presented, and it is shown that $D' = \rho_m D/BR\rho_s$ where ρ_m is the density of the liquid, and ρ_s that of the solid. D is the diffusion coefficient, B is the thickness of the boundary layer and R is the rate of crystallization.

Extraction of the trace element from the convecting layers stacked above the crystallizing layer owing to the downward diffusion across the boundary layers, can be modelled by an extension of eqn. (1). In this case $C = C_N$ is the concentration in the basal (N^{th}) layer, and $C' = C_{N-1}$ is not assumed constant, but is governed by an equation describing diffusion into layer $N-1$ from the overlying layer $N-2$ as well as down to layer N (Fig. 1). The extended model which is derived in Appendix 2 does not have an exact solution, and predictions have to be made using a computer program, which is available from the second author on request.

This model differs fundamentally from that proposed by Cawthorn and his co-workers (Cawthorn and McCarthy, 1980, 1981; Cawthorn et al., 1983; Wright et al., 1983), as their model does not consider the possibility of a *discretely* stratified magma as assumed here. The model also differs from that considered by McCarthy et al. (1985) where they consider the possible effects of lateral accretion proposed by Irvine et al. (1983).

Application to the Bushveld Complex

In the case of the Upper Zone of the Bushveld Complex, the system involves an iron-rich liq-

uid near the magnetite/plagioclase cotectic. The magnetite/liquid system represents a large density contrast, as the magnetite is almost twice as dence as the liquid. Hence, the compositional changes brought about by fractional crystallization of magnetite might produce instability leading to d.d.c.

The Upper Zone of the Bushveld Complex is about 3 km thick and evolved from a single, well mixed magma (Kruger et al., 1986). The present boundaries of the Bushveld Complex indicate that the Upper Zone magma layer had a lateral to vertical aspect ratio of more than 100. The sidewall cooling effects that were important in the evolution of the small Skaergaard Intrusion (McBirney et al., 1985) are probably negligible in this case. Thermal modelling of intrusive sheets (Gray, 1978) indicates that the thick cumulate floor to the Upper Zone (ca. 5 km) would prevent downward heat loss, and probably actually supply heat to the liquid part, thus aiding convection. Heat flux was thus vertically upward, through the roof rocks. Thus both the density changes due to changes in composition and heat flux serve to produce instabilities that could cause d.d.c. in the Upper Zone of the Bushveld Complex. This is a different situation to that considered by McBirney (1985) which deals with tholeiitic magmas that have not yet reached the stage of maximum iron enrichment and are still becoming more dense as a result of fractional crystallization.

Vertical concentration profiles for chromium in the Main Magnetitite layer (MML) in the Upper Zone of the Bushveld Complex have been determined by Cawthorn and McCarthy (1980, 1981). These stratigraphic concentration profiles can be used to test our model for the crystallization of d.d.c. layers. In the MML, the profiles show a "sawtooth" variation starting with a high concentration which rapidly decreases to a constant concentration significantly above zero. A number of abrupt reversals occur which give the "sawtooth" pattern, in which each section shows a similar depletion profile and successive sections with progressively lower concentrations at the reversals. The possibility that these reversals are caused by magma replenishments from outside the magma chamber is excluded by recent Sr-isotopic data on the Upper Zone (Kruger et al., 1986).

The magnetitite is assumed to crystallize at the base of the lowermost layer of the d.d.c. system and that there was *no* significant redistribution of the chromium after crystallization by vertical diffusion within the crystal pile. The latter assumption is supported by the presence of abrupt reversals and very steep concentration gradients (Cawthorn and McCarthy, 1980, 1981; Cawthorn et al., 1983). Further, work by Reynolds (1985) on the magnetite layers suggests that sintering of the magnetite occurs concurrently with or very soon after precipitation, causing the layers to become impermeable. Thus, postcumulus processes such as that proposed by Kerr and Tait (1985) may be excluded in this case. Therefore, at any height within the magnetitite layers, the concentration of Cr reflects that of the coexisting liquid from which that magnetite crystallized. Since a monomineralic layer (pure magnetite) is under consideration, and loss of heat of crystallization is the rate controlling factor, the rate of crystal accumulation is assumed constant as a first approximation.

During crystallization, diffusion of Cr occurred across the diffusive boundary separating the basal crystallizing layer from the convecting layers above (Fig. 1). This is due to marked changes in concentration of Cr brought about by fractional crystallization of the basal magma layer. As the Cr concentration contrast across the diffusive boundary layer becomes greater, diffusion increases, until a stage is reached where diffusion *into* the layer matches the extraction by magnetite crystallization, and the concentration tends to the constant C_c.

Curves predicted by eqn. (3) are a good fit (see the procedure below) for the observed vertical concentration profiles of chromium in the MML (Fig. 2), recorded by Cawthorn and McCarthy (1981). Convergence to C_c is rapid,

and is observable even though the process terminates in abrupt reversals to higher concentration while X is only a *small* fraction of H. This is probably due to the mixing of the basal layer with that above when the densities become equal, as a result of the extraction of dense magnetite from the lowermost convecting layer. This process would result in a thicker convecting layer at the base of the chamber with an intermediate chromium concentration, which would crystallize magnetite with a higher chromium concentration. This aspect is examined in greater detail below.

Fitting the parameters

In the application of eqn. (3) to chromium in magnetite, X/H is always small and $K'-1$ is large. For large values of the coefficient K' the right-hand side of eqn. (3) is approximately exp $(-\lambda X)$ where $\lambda=(K'-1)/H$. For $K'-1>300$ (a likely value for chromium deposition in magnetite) the difference between the right-hand sides of eqns. (3) and (4) is either less than 1% (for small values of λX) or less than 0.001 (for larger values of λX), or both.

The exponential approximation leads to the simpler model:

$$C-C_c/C_o-C_c=\exp(-\lambda X) \qquad (4)$$

Concentrations predicted by eqns. (3) and (4) were fitted to data selected from Cawthorn and McCarthy (1981) by choosing parameters to minimize the root mean square relative error (RMS) over the data points.

The exponential model (eqn. 4) was fitted first to give starting values for fitting eqn. (3). It became apparent that there was a range of parameters giving almost equally good fits even apart from the relationship between K' and H. The method adopted to locate this range was to plot the RMS values (to the nearest percent) for given C_c (which could be estimated from the graph of the data, see Fig. 2) and K' and a range of values for C_o and H. Clearly then, an accurate estimate of either the distribution coefficient K or the initial concentration of Cr in the magma

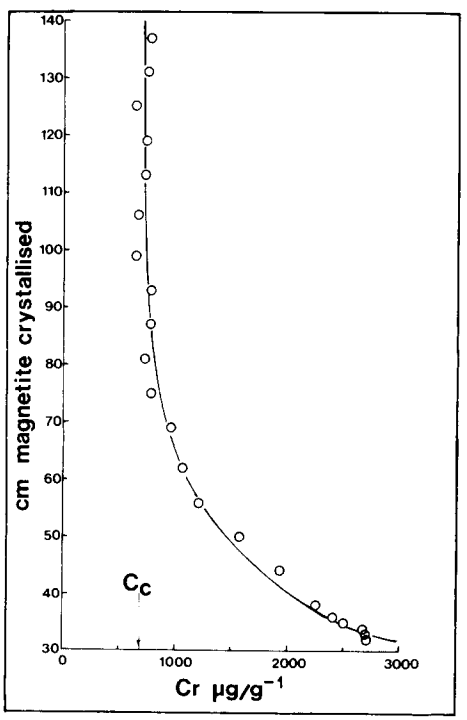

Fig. 2. Actual chromium concentrations in a pure magnetitite layer (Section C from Cawthorn and McCarthy, 1981) show the depletion predicted by eqn. (3). In the *solid* C_c is 700 $\mu g\ g^{-1}$, and the initial concentration is predicted to be 16,000 $\mu g\ g^{-1}$ in agreement with the data of Cawthorn et al. (1983). The curve plotted used $K'-1=300$ and $H=5000$ cm as suggested by the error contour plot (Fig. 3).

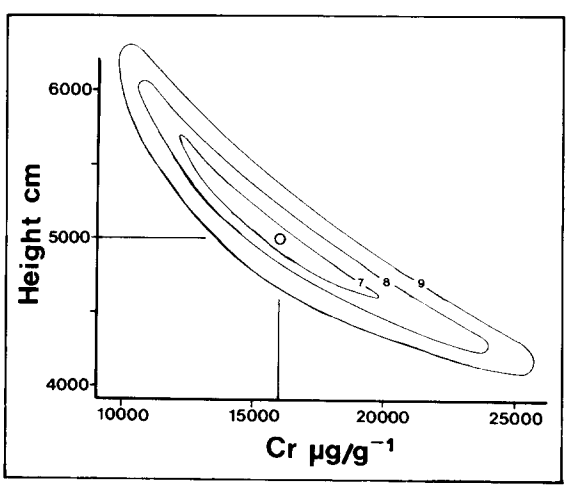

Fig. 3. Error contour plot used to estimate the best fit parameters for eqn. (3). The best fit was 6.5% RMS relative error for $H=5000$ cm and $C_o=16{,}000$ $\mu g\ g^{-1}$ in the solid. This technique can also be used to fit other parameters.

is required before accurate predictions about H can be made.

Figure 3 shows a typical error contour plot for data from the smooth sequence from 32 cm to 137 cm of section C of Cawthorn and McCarthy (1981). The RMS relative error has been plotted for 9% error and less. Taking $K'-1=300$ and $C_c=700\ \mu g\ g^{-1}$ Cr in the solid, the lowest RMS was about 6.5% for $H=50$ m and $C_0=16{,}000\ \mu g\ g^{-1}$ Cr in the magnetite. However, RMS lies between 6.5% and 7.5% for a range of values between $H=57$ m, $C_0=12{,}000\ \mu g\ g^{-1}$ and $H=45$ m, $C_0=21{,}000\ \mu g\ g^{-1}$ Cr.

In addition to this flexibility in fitting the physical parameters, values of $K'-1$ (dominated by the distribution coefficient K in this case) and H can be varied widely while keeping a constant ratio as discussed above. For example, $K'-1=1500$ produces the same error contour plot as Fig. 3 if the H-scale is revised to run from 200 m to 300 m. The best fit in this case would be obtained with $H=250$ m and the same $C_0=16{,}000\ \mu g\ g^{-1}$ Cr. The values for K available in the literature range between 100 and 620 (Irving, 1978), and thus the value of 1500 for $K'-1$ is an extreme used for illustration. But for most of the calculations K is about 275 (in the published range) and thus 50 m is not an unreasonable value for H which is definitely less than 250 m. Clearly, the equivalent solid heights quoted here have to be multiplied by ρ_s/ρ_m (about 5/3 for magnetite/liquid) to give the actual height of the magma layer.

The precipitated magnetite is a very small fraction of the magma layer (between 0.4 and 2% in the examples above), and thus the model does not demand unlikely phase relationships such as lengthy single phase saturation. In fact thin plagioclase partings in the magnetitite layers (Cawthorn and McCarthy, 1981; Reynolds, 1985) suggests that the magma was close to the magnetite/plagioclase phase boundary.

Several other smooth sequences from the different sections in Cawthorn and McCarthy (1981) were fitted in the same way. Despite the parameter flexibility, it appeared that if K' was assumed constant (i.e. the distribution coefficient and the characteristics of the diffusive boundary were similar in each case), the best fit C_0 and H were different for the different sections, and thus the basal crystallizing layers were not always the same thickness.

We have also used the extended model (Appendix 2) to fit the same data, but as diffusion is slow, the results were not significantly improved. The number of layers above the base can be adequately modelled using 4 to 5 layers of equal thickness in the application considered; higher layers showed almost no change in the concentration of chromium.

The parameter D' can be calculated from the relationships given in eqn. (2) and for section C is approximately 13.6, but clearly it is also subject to error. An alternative approach would be to calculate D' independently using the equation $D'=\rho_m D/BR\rho_s$ derived in Appendix 1, but the parameters necessary (B, D and R as well as K) are not well known as yet and may be in error by an order of magnitude, hence the present approach.

Clearly, if these parameters can be determined more accurately (e.g. using other fluid dynamic, thermal modelling and geophysical data), the model can be tested further. For example, the average rate of accumulation during crystallization of the Bushveld Complex was of the order of 2.3 cm per year (7.3×10^{-8} cm s^{-1}) calculated using a cooling time of ca. 350,000 years and a thickness of 8 km (Walraven, 1981). Substituting this value for R, setting $D'=13.6$ and $B=10$ and 100 cm in the above equation, results in diffusion coefficients D of 2×10^{-5} to 2×10^{-6} cm^2 s^{-1}. This is greater than ionic diffusion in a static magma (ca. 10^{-6} — 10^{-8}, see Hofmann, 1980), which may indicate that the diffusive boundary may have processes which aid diffusion occurring within it, or that some movement of the boundary into the overlying Cr-rich layer occurs during crystallization. These suggestions can be tested in tank experiments using the equations derived in this work. If the same R is used in the work of Wright et

al. (1983) a very high D is required (0.1 to 660), which seems unreasonable. The rates of accumulation quoted in that work are also unreasonably small (10^{-12} cm s^{-1}) as the 100-cm-thick MML would require ca. 3 Ma to accumulate, which is in excess of the time required for the whole of the layered sequence.

Modelling reversals in the data profiles

As briefly discussed above, extraction of a dense phase (magnetite) from a less dense liquid will result in the residual liquid becoming less dense. Thus magnetite fractionation of the basal layer will result in the residual liquid in the layer becoming less dense until it equals that of the overlying layer. When the two layers become equal in density, the diffusive boundary will break down, and the two layers will mix. This results in a thicker basal layer which will then continue to crystallize when its liquidus temperature is reached. Alternatively, this new, thicker mixed basal layer may split into two or more individual layers the lowest of which continues to crystallize. This alternative scenario appears unlikely, at least in the initial stages, but may become more likely if the basal layer becomes very thick due to repetitions of the mixing process, or a different phase (plagioclase) starts to crystallize and produces a different convective regime.

In the modelling presented in Fig. 4, the profiles produced by both processes are shown for illustration. In this modelling, parameters similar to those derived in the previous section were used, and were assumed constant throughout. The stacked layers were assumed to be of constant thickness, and the same amount of crystals were precipitated after each cycle. Clearly this is a simplification, but it does serve to illustrate the principle, and the form of the resulting depletion profiles. Because more than one layer is modelled, the depletion of the overlying layers due to diffusion may be significant when the mixing events occur and thus the extended model (Appendix 2) was used to compute the profiles.

Comparison of the shape of the profiles with those of Cawthorn and McCarthy (1981) indicates that for most of the profiles the first model fits best, but that the second model may also apply to some reversals. It is self evident that for any sequence of reversals the initial profile is the same for both models.

A possible process whereby layers may split (process 2), is if magnetite crystallization resulting in less dense residual liquid is followed by plagioclase crystallization which results in a more dense residual liquid (Campbell et al., 1983). This may have been the case in the Bushveld Complex. Plagioclase crystallization may result in ponding of the dense residual liquid enriched in mafic components at the base of the crystallizing layer which may then form a separate convecting layer. Alternate magnetite/plagioclase crystallization could thus possibly modulate the formation and coalescence of d.d.c. layers in the liquid.

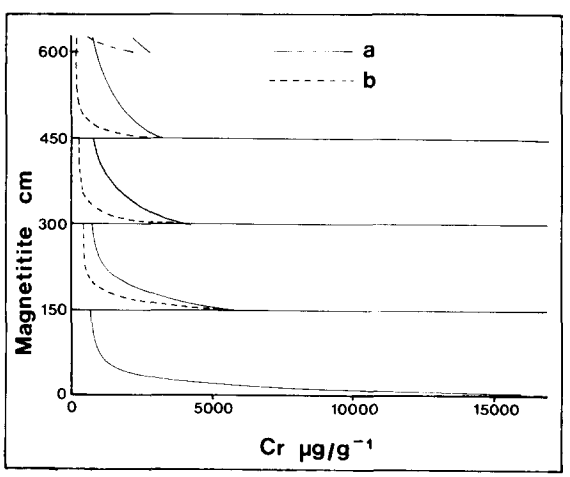

Fig. 4. The form of the reversals if the basal layer mixed with the one above and then (a) the double thickness layer continued to crystallize (solid line) and (b) the mixed layer split into two equal layers and continued to crystallize (dashed line). Repetition of process (a) eventually results in a single very thick convecting layer, whereas process (b) results in the same number of layers being maintained. Parameters used were: $D' = 20$, $K' - 1 = 300$ and a reversal was produced every 150 cm. Initially, $H = 5000$ cm and $C_0 = 57$ $\mu g\ g^{-1}$ for all the layers.

Conclusion

We have extended the well known Rayleigh distillation law to include one type of partly open system in which diffusion occurs. This extended equation may lead to a better understanding of the behavior of trace elements during d.d.c. The equations also serve as an alternative model to that presented by Wright et al. (1983) for the magnetitite layers of the Bushveld Complex, and the lateral accretion model of Irvine et al. (1983) for layered intrusions as applied by McCarthy et al. (1985), to the magnetitite layers.

Reversals in the compositional trends of the cumulates can result from the breakdown of the diffusive boundaries between layers and may not necessarily be the result of influxes of magma. The processes envisaged may result in the eventual homogenization of the whole residual magma, or the d.d.c. layers may be maintained.

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Appendix 1: Derivation of the diffusion model.

The total mass per unit area of the trace element in the magma layer is increased in each time interval by diffusion from the layer above, and decreased by deposition of the element in the crystal fraction.

The rate of diffusion across the diffusive boundary layer is $D\rho_m(C'-C)/B$ (μg cm^{-2} s^{-1}) where C'(μg g^{-1}) is the concentration in the overlying convecting layer, B cm is the thickness of the boundary layer, ρ_m and ρ_s (g cm^{-3}) are the densities of the magma and solid respectively (see Fig. 1) and D (cm^2 s^{-1}) is the diffusion coefficient. The magma layer is assumed to be equivalent to a *solid* layer H cm thick, and thus the *actual* thickness is $H\rho_s/\rho_m$ (cm).

Consider a situation when the crystal fraction (cumulate pile) has a height of X cm (Fig. 1). The residual magma will then be $(H-X)\rho_s/\rho_m$ (cm) and will contain $(H-X)$ $C(X)\rho_s$ (μg cm^{-2}) of the trace element. Deposition of the next ΔX cm of crystals causes $C_s(X)\rho_s\Delta X = KC(X)\ \Delta X\rho_s$ (μg cm^{-2}) of the trace element to be deposited, as $C_s = KC$. The time taken is $\Delta X/R$ (s), where R (cm s^{-1}) is the rate of accumulation, during which time $D\rho_m(C'-C)\ \Delta X/BR$ (μg cm^{-2}) of the trace element diffuses through from the layer above.

Combining these effects will give the total quantity of the trace element in the liquid when the height of the cumulate pile is $X+\Delta X$ cm. Collecting these terms and dividing by ρ_s results in:

$$(H-X-\Delta X)C(X+\Delta X)$$
$$= (H-X)C(X) - KC\Delta X + D'(C'-C)\Delta X$$

where

$$D' = D\rho_m/BR\rho_s$$

Thus:

$$\frac{d}{dX}\left[(H-X)C(X)\right] = -KC + D'(C'-C)$$

This is equivalent to differential equation (1) in the text.

Note that multiplying the differential equation by K changes the concentrations to μg g^{-1} in the *crystal* fraction making the model predictions directly comparible with the data.

Appendix 2: Multi-layered diffusion model.

For N layers the basal or N^{th} layer will satisfy eqn. (1) in the form:

$$(H-X)\frac{dC_N}{dX}=C_N-KC_N+D'(C_{N-1}-C_N)$$

The layers stacked above the basal crystallizing layer are assumed not to crystallize and to remain constant in thickness, i.e. there are no terms like the first two on the right-hand side of the equation above. However, diffusion will take place to the layer below as well as from the layer above for the intermediate layers. Thus:

$$H\frac{dC_J}{dX}=D'(C_{J-1}-C_J)-D'(C_J-C_{J+1})$$

where $1<J<N$

while the top layer will be subject only to diffusion downwards:

$$H\frac{dC_1}{dX}=-D'(C_1-C_2)$$

The height H of each layer and the boundary layer characteristics D' are assumed to be the same for all the layers in this derivation, but different values could be used if this is felt to be appropriate. These N differential equations provide a model for multi-layer d.d.c. which can be used to test theories about d.d.c. such as the causes of compositional reversals.

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