The use of phosphorus contents in yielding estimates of the proportion of trapped liquid in cumulates of the Upper Zone of the Bushveld Complex

R. GRANT CAWTHORN AND KEVIN L. WALSH

Department of Geology, University of the Witwatersrand, 1 Jan Smuts Avenue, Johannesburg, 2001, South Africa

Abstract

Phosphorus contents in cumulus rocks occurring close to the level of apatite appearance in the basic rocks of the Bushveld Complex, South Africa, provide a method of calculating the proportion of intercumulus component in these rocks. Previous experimental studies have accurately constrained the phosphorus content of magmas when apatite becomes stable. The ratio of the phosphorus content in the cumulates immediately below the appearance of apatite to this liquid composition defines the proportion of trapped liquid.

Application of this method to rocks from the uppermost mafic rocks of the Bushveld Complex leads to the conclusion that there is from 1 to 6 per cent intercumulus component. Many of these rocks are multiphase cumulates and in such rocks estimation of intercumulus component from textural criteria is difficult.

If crystals grow *in situ* on the floor of the magma chamber such small proportions of interstitial component can be produced without appealing to excessive diffusion and circulation of magma through an unconsolidated crystal pile. The geometry of the intrusion as well as its size might have a major influence on the proportion of the liquid ultimately solidifying within a cumulus rock.

KEYWORDS: layered complexes, intercumulus, apatite, bottom growth.

Introduction

THE concept of physical accumulation of early-formed minerals in some porphyritic lavas and coarse-grained rocks became geologically acceptable with the work of Bowen (1927), although others had previously alluded to such a process. However, it was Wager and Deer (1939) who made the major contribution in quantifying the proportion of accumulated crystals and liquid crystallized *in situ* in such rocks. Inherent in the terminology of 'adcumulus' and 'orthocumulus' of Wager *et al.* (1960) is the dual role of the liquid during crystallization; firstly, in producing overgrowths on minerals with the same composition as the cumulus phase and, secondly, in solidifying as trapped material during the latter stages of crystallization. Geochemical modelling has shown that petrographic criteria may underestimate the former role of the magma (Henderson, 1970 and 1975; Brooks and Nielsen, 1978).

The geochemical method of Henderson (1970) is based on the analysis of two adjacent samples in a layered intrusion, with totally different cumulus mineralogy, for an element which has a large partition coefficient into minerals present in only one of the cumulus assemblages. The major strength of this calculation is that the only unknowns in the equations are the proportion and the concentration of the specified element in that trapped liquid, and both of these are determined by solution of simultaneous equations. Its disadvantages, recognized by Henderson (1970), are that it works best for monomineralic cumulates and its accuracy decreases as the proportion of trapped liquid decreases. As will be shown below, both these constraints apply to the present study.

A more direct technique is based on the concentration of a totally incompatible element in the cumulate and in the liquid from which it crystallized. The simplicity of this principle is

Mineralogical Magazine, March 1988, Vol. 52, pp. 81–89

complicated by the fact that there is no unambiguous way of constraining the liquid composition. A reasonable estimate is possible during the early stages of crystallization if fine-grained marginal rocks can be analysed and if they can unequivocally be shown to represent parental magma (Henderson, 1975). Unfortunately, this is rarely the case. Furthermore, this estimate becomes less reliable as crystallization proceeds, as the degree of fractionation is difficult to determine. If there is addition of magma during crystallization then a knowledge of the compositions of residual and added magma, their relative volumes, and the extent of mixing must all be obtained before these calculations are possible. If magma chambers become chemically stratified due to diffusive processes of unmixing as a result of differentiation (McBirney, 1980; Sparks *et al.*, 1984), predicting the trace element concentration of the magma at the crystal–liquid interface becomes further complicated. Uncertainties in all these parameters compound the difficulties of such calculations for the later-formed or more evolved cumulates. Again, this is the situation in the present study.

The present study involves the uppermost cumulates of the layered basic rocks of the Bushveld Complex. Post-cumulus processes in this intrusion have received considerable attention in the case of chromite layers (Cameron, 1975; Hatton and von Gruenewaldt, 1985), but little attention elsewhere in the complex.

Geological setting

The geology of the mafic rocks of the Bushveld Complex, referred to as the Rustenburg Layered Suite, has been reviewed by von Gruenewaldt *et al.* (1985). The uppermost 2 km of this suite consists of layered rocks containing any of the following cumulus minerals (in order of relative abundance): plagioclase, clinopyroxene, magnetite, olivine, inverted pigeonite, apatite, and ilmenite. The appearance of magnetite in these rocks has been used as a stratigraphic marker, and all overlying rocks grouped into the Upper Zone. The sequence in the eastern lobe of the Complex has been documented by von Gruenewaldt (1973) and Molyneux (1974). In contrast, the sequence in the western sector is less well known due to poor outcrop, but has been studied by Coertze (1970) and Markgraaff (1976). The Geological Survey of South Africa drilled three deep boreholes north of Rustenburg to provide a

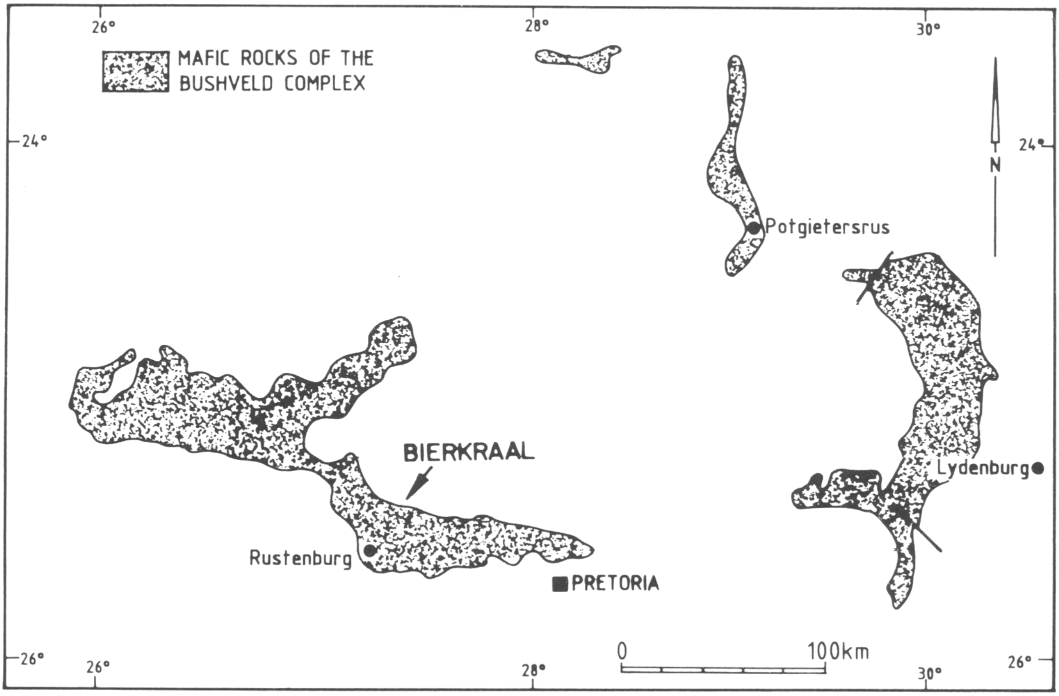

FIG. 1. Location of the farm Bierkraal on which boreholes were drilled, in relation to the outcrop of the mafic rocks of the Bushveld Complex.

continuous profile through the entire Upper Zone (Fig. 1). Aspects of the mineralogy and geochemistry of these rocks have been documented by Cawthorn and McCarthy (1985) and Reynolds (1985). Further samples from these boreholes have been taken for the geochemical study reported here.

Borehole BK1 was collared in Bushveld granite overlying the layered mafic rocks, the uppermost of which was intersected at a depth of 380 m. BK3 was drilled to overlap the lowest section of BK1. Owing to the absence of reliable marker horizons, the extent of overlap was estimated to be 600 m from a comparison of the stratigraphy and the vanadium content of magnetite in boreholes BK1 and BK3 (unpublished data). The average dip in this area is 24° (Walraven and Wolmarans, 1979). However, all depths reported in the diagrams are vertical depths in the boreholes. Thus all distances exceed true stratigraphic thicknesses by about 10 per cent.

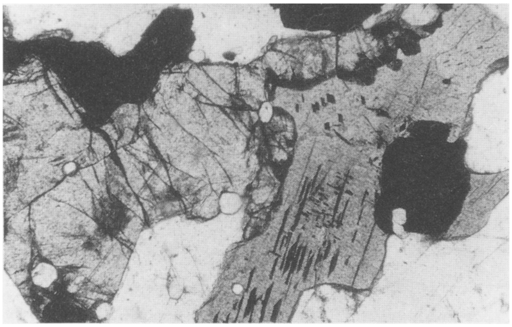

FIG. 2. Photomicrograph of a polyphase cumulate with subhedral crystals of olivine (left), augite (right of centre), plagioclase (at edges), apatite (enclosed near edges of all other minerals) and magnetite. Plane-polarised light; field of view 0.8 × 0.5 cm.

Petrography

Rock types range from almost monomineralic anorthosites and magnetites to polyphase samples with up to five cumulus minerals: plagioclase, olivine, clinopyroxene, magnetite, and apatite. However, as noted by Wager *et al.* (1960), the recognition of definitive cumulus textures becomes more difficult with increasing number of cumulus phases. The distinction between adcumulus growth onto early-formed minerals and crystallization from trapped liquid in the presence of several cumulus phases is almost impossible.

Apatite is present as a cumulus mineral (Fig. 2) in almost all the samples from borehole BK1 except near the base, but is only sporadically present in the upper half of BK3. It first appears as a cumulus phase in an isolated sample of anorthosite at 600 m depth, where it is a minor cumulus phase. Above this sample for 100 m, it is absent. At 500 m depth, it reappears as a minor cumulus phase and, in decreasing abundance, is present in samples over the next 20 m before disappearing again. Apatite is present in nearly all samples from 400 m upwards, although the proportion is very variable. Intercumulus apatite is very scarce, and is absent in the samples immediately below the first appearance of cumulus apatite, unlike the situation documented in the Skaergaard Complex (Wager and Brown, 1968), where intercumulus apatite gradually increases in abundance upwards until it attains cumulus status. In several intrusions the appearance of a cumulus mineral is heralded in the underlying rocks by an increasing proportion of that phase as an intercumulus component: for example, plagioclase in La Perouse layered gabbro (Himmelberg and Loney, 1981) and magnetite in the Bushveld Complex (Molyneux, 1974).

Chemistry

Analysis for P was undertaken in 378 samples from 2500 m of borehole core by X-ray fluorescence spectrometry. Phosphorus is commonly analysed along with other major elements on a fusion disk (Norrish and Hutton, 1969). However, at concentrations below 0.1% the precision is poor, and hence pressed powder pellets were utilized as described in the Appendix. Calibration was by standards used for major element analysis and synthetic blanks.

The data for P are plotted as a function of depth in the boreholes in Figs. 3 and 4. (Analyses are available from the senior author.) It is evident that there is no increase in the P content in the zone immediately underlying the appearance of apatite at 600 m in borehole BK3, although the sample immediately below the re-entry of apatite at 500 m shows a threefold increase in P above the normal background values.

Calculation of trapped liquid content

Wager (1960) modelled the fractional crystallization of the Skaergaard Complex, using the behaviour of phosphorus and calculations on the proportion of cumulus to intercumulus material (Fig. 5). An estimate of the proportion of trapped liquid can be made if the values for C_S and C_L in Fig. 5 are known. The concentration of P in the cumulates (C_S) is determined directly by analysis.

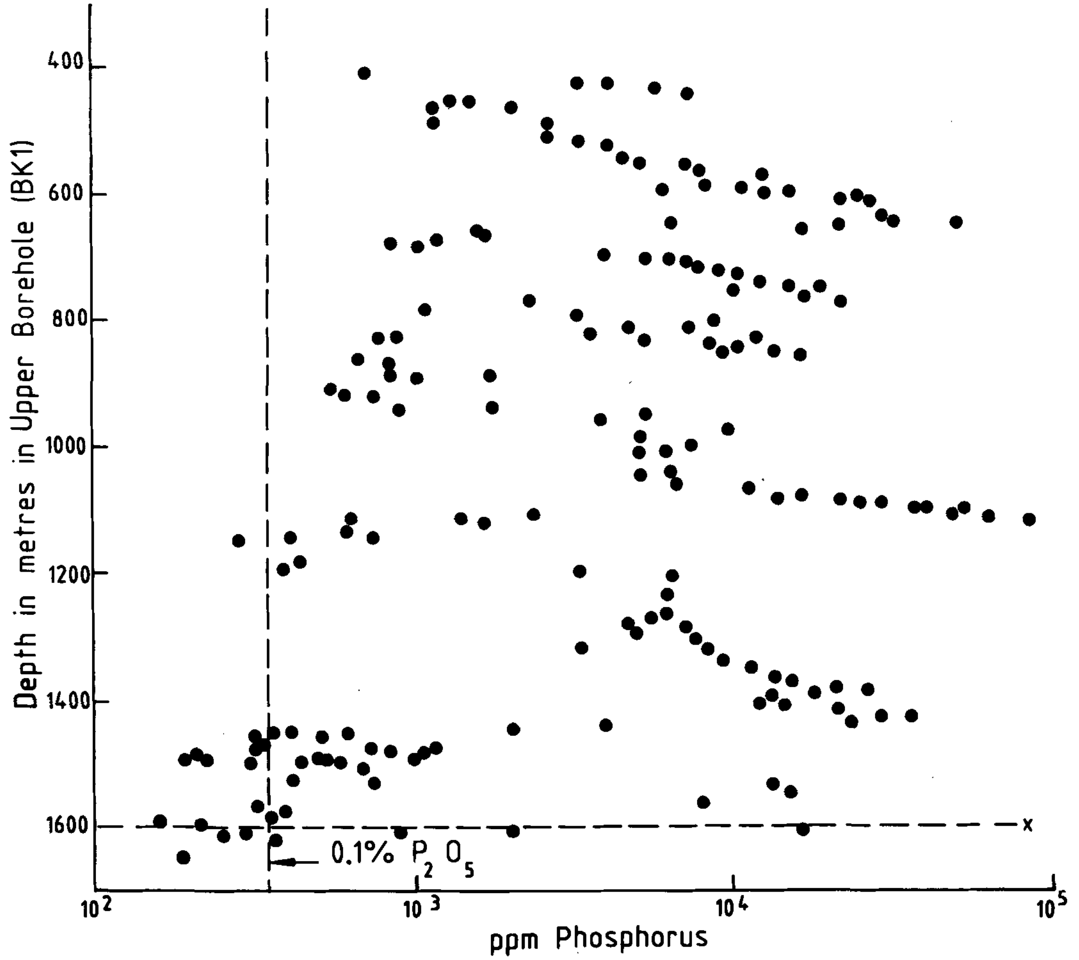

FIG. 3. P content in whole rocks from the magnetite gabbro samples in upper borehole (BK1). Borehole depths are uncorrected for dip. Horizontal dashed line (×) at 1600 m refers to inferred correlation between boreholes BK1 and BK3 (Fig. 4) at 590 m. Vertical dashed line at 0.1% P_2O_5 separates samples with cumulus apatite from those devoid of apatite.

The value of C_L is more elusive in layered complexes, but, with the appearance of apatite, may be constrained by experimental studies.

Watson and coworkers (Watson and Capobianco, 1981; Watson and Green, 1981; Green and Watson, 1982; Harrison and Watson, 1984) have undertaken experimental studies which have made the quantitative evaluation of apatite stability possible. Using a variety of starting compositions and methods of reaching apatite saturation they were able to determine the P content of liquids at which apatite began to crystallize as a function of temperature, pressure, and water and SiO_2 contents of the liquid. The pressure effect, with and without water, is minimal, and the effect of the principal variables is indicated in Fig. 6.

The temperatures at the end stages of differentiation of a body like the Skaergaard intrusion or the Upper Zone of the Bushveld Complex have been inferred from pyroxene compositions (Lindsley *et al.*, 1969) to be approximately 985 °C. The appearance of apatite somewhat before these extreme differentiates suggests that a slightly higher temperature might be appropriate for its nucleation. An upper limit on temperatures of crystallization near the end-stages of differentiation may be obtained from the experimental study of Thompson (1972). He showed that the cotectic temperature

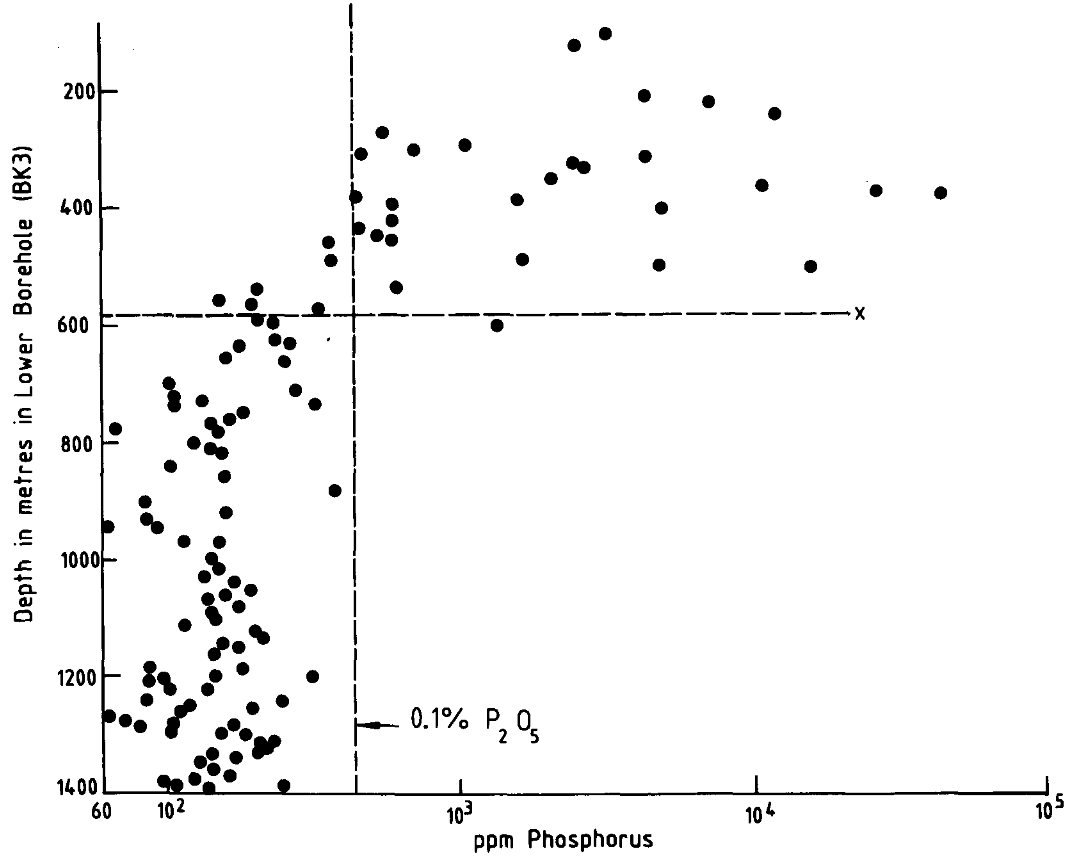

FIG. 4. P content in samples from lower borehole (BK3).

for the crystallization of olivine-clinopyroxene-plagioclase decreased with increasing Fe/(Fe+Mg). When this ratio reaches a value of 0.8, which would correspond to the composition when magnetite appears (according to Osborn, 1979), the temperature would be 1050-1100 °C. As apatite begins to crystallize after magnetite, slightly lower temperatures than this can be envisaged. The temperature considered likely for the appearance of apatite is 1000-1050 °C.

Estimates of SiO_2 content of the *liquid* which produced apatite saturation in these layered complexes is more tenuous. A major injection of tholeiitic magma with 49% SiO_2 occurred some distance below the base of the Upper Zone (Davies and Cawthorn, 1984) and its differentiation initially was by iron enrichment rather than silica enrichment (cf. Cox, 1980; Osborn, 1979, Fig. 5-9). In the Bushveld Complex quartz is limited to the uppermost 400 m of the intrusion and, except for the top 50 m, does not exceed 1 per cent by mode (Molyneux, 1974). Cawthorn (1977) explained the modal data by showing that the abundant precipitation of orthopyroxene rather than olivine restricted the trend towards silica-enrichment. A sudden change in the relative trends for iron and silica occurs with magnetite precipitation. Magnetite crystallized below the appearance of apatite. Thus the magma had begun the silica-enrichment trend prior to apatite crystallization. Using the curves of Osborn (1979), SiO_2 contents of 50-55% seem reasonable for the magma at the onset of apatite crystallization. This is higher than the value of 48% calculated by Wager (1960) for the Skaergaard Complex, but makes little difference to the following calculations.

The boxed area in Fig. 6 defines the likely conditions prevailing in these two magmas at the time that apatite began to crystallize. This implies that the P content of the magma at this stage was between 1.0 and 1.6% P_2O_5. The upper value is close to that (1.75% P_2O_5) calculated by Wager

(1960) based on an assumed parental magma composition and the relative proportions of fractionated minerals. These two methods are different yet the similarity in results is remarkable. The similarity in mineralogy of the Bushveld magnetite gabbros and those of the Skaergaard suggests that a similar estimate for P would be reasonable. In the following calculations a figure of 1.3% P_2O_5 will be used and an uncertainty of ±30% will be assumed to cover the possible range of temperature and silica content.

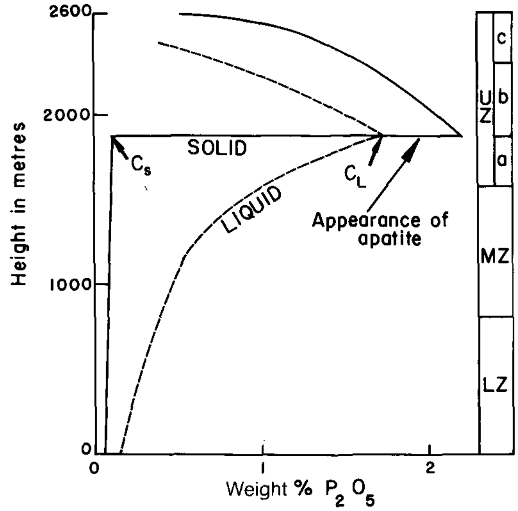

FIG. 5. Illustration of the behaviour of P_2O_5 in a differentiating liquid and the cumulates forming therefrom. The diagram is adapted from Wager and Brown (1968) using data from the Skaergaard Complex. Data for the cumulates have been smoothed out.

The geometry of the curves in Fig. 6 shows that with differentiation leading to SiO_2-enrichment, and decreasing temperature, the P content at saturation will decrease. This results in a greater proportion of apatite crystallizing than is present in the normative composition of the liquid. 1.3% P_2O_5 corresponds to 3.1% normative apatite. The calculations made by Wager (1960) suggested a decrease from 1.75 to 0.4% P_2O_5 or 4.2 to 1% normative apatite at the end-stages of differentiation (Wager and Brown, 1968, Table 10), which is in qualitative agreement with Fig. 6. In the Bushveld Complex modal data on apatite have been published by von Gruenewaldt (1973) and Molyneux (1974), and are reproduced in Fig. 7. These data specifically exclude the magnetite-olivine-apatite-rich horizons which have been described by Reynolds (1985). Despite the wide scatter in the proportions, there appears to be a slight initial upward enrichment in apatite before the expected decrease. The average modal apatite content ranges from 2.2% (von Gruenewaldt, 1973) to 2.4% (Molyneux, 1974). Allowing for the apatite-olivine-magnetite-rich bands and the high density of apatite compared to the silicate phases, the weight proportion of apatite, in the section from where it first occurs to the top of the intrusion, would be closer to 3%. This is in good agreement with the estimate of 1.3% P_2O_5 in the liquid at apatite saturation.

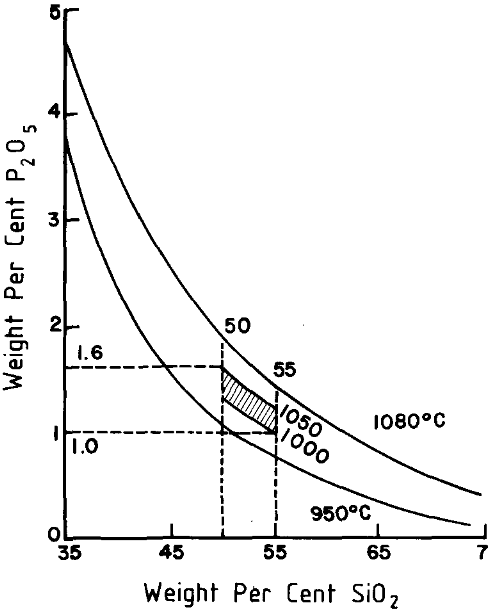

FIG. 6. Experimentally determined compositions of liquid in equilibrium with apatite as a function of temperature (from Green and Watson, 1982). The estimated parameters in the Bushveld Complex when apatite began to crystallize are shown as the hatched area and are 50 to 55% SiO_2 in the liquid, and 1000 to 1050 °C.

An alternative way to estimate the P content of the magma when apatite becomes stable is to calculate the weighted average of the P in all the samples from the level where apatite appears to the top of the intrusion. The concentration in each sample was multiplied by the half-distance from the overlying to underlying sample, and the final summation divided by the total thickness. Such a weighted average is considered more reliable than a simple average of all concentrations. The weighted average for all these data is 6524 ppm P, which corresponds to 3.5% of apatite.

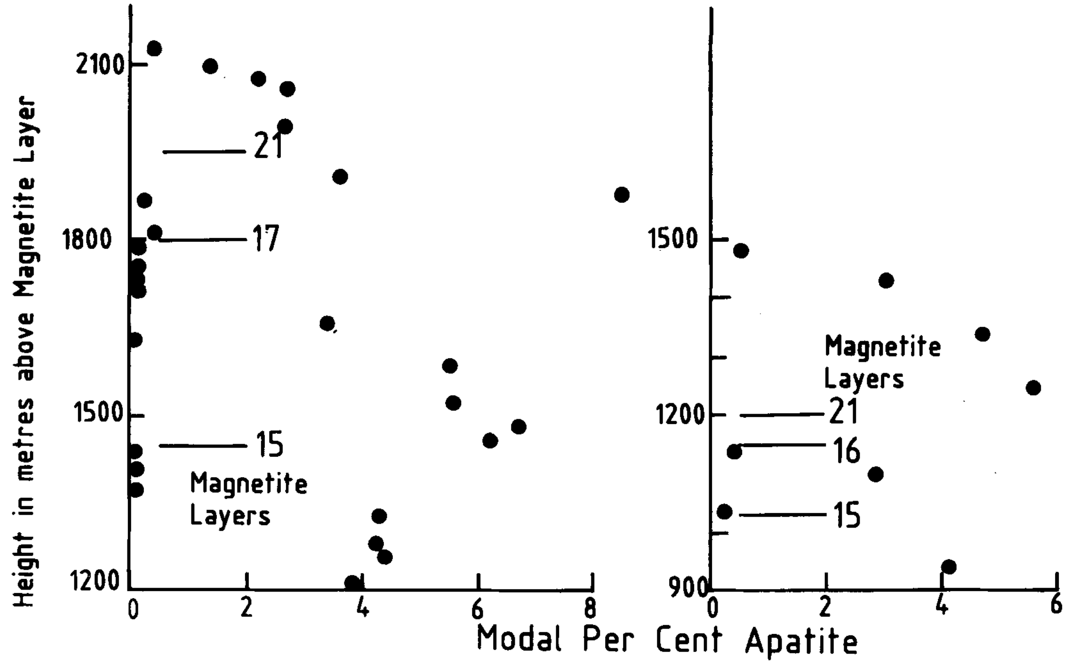

FIG. 7. Modal apatite content in the uppermost rocks of the Bushveld Complex, based on profiles by von Gruenewaldt (1973) on left and Molyneux (1974) on right. The positions and numbers of the uppermost magnetite layers are included for reference. Positions are given relative to the Main Magnetite Layer.

The estimates of P in the liquid at the onset of apatite saturation, based on experimental studies and observed concentrations through the layered sequence, agree and constrain the value of C_L to 1.3% P_2O_5 of 5670 ppm P in Fig. 5 and in the following calculations.

To determine a value for C_S the concentration of P in six samples in the 100 m section below the point at which apatite saturation occurs were averaged. The average is 211 ppm with a maximum of 267 and a minimum of 105 ppm. The percentage of trapped liquid in these cumulates is therefore $100 \times 211/5670$ or 3.7%. Using the maximum and minimum values of C_S of 265 and 105 and for C_L of 4360 and 6980 ppm, the most extreme range of trapped liquid is calculated to be 1.5 and 6.1%. Henderson (1968) presented two analyses of 174 and 312 ppm P in samples from the Upper Zone which are consistent with our data. He also gave an analysis of 20 ppm for a plagioclase separate. The previous calculations are based on the assumption that P is totally incompatible in the cumulus phases. If some of the P resides in cumulus minerals then the actual proportion of trapped liquid will be even less than calculated. All of these estimates demonstrate that these rocks are extreme adcumulates.

Formation of adcumulates

McKenzie (1984) showed that residual porosities as low as 3% may be produced by compaction. The efficiency of this process depends mainly on the rate of deformation of the matrix, which is related to the shear and bulk viscosity. Such parameters are inadequately known for a crystal mush in a high-level magma chamber to permit detailed calculations.

The process of compositional convection by which interstitial liquids partially crystallize to give less dense residual magmas which may then migrate upwards to be replenished by denser, less-differentiated magma has been proposed by Sparks *et al.* (1985). This may occur in horizons where there is overgrowth onto magnetite and pyroxene, as this will reduce the density of residual liquids, but, where monomineralic anorthosites are forming, residual liquids will increase in density and retard the circulation process.

The concept of a rock composed of cumulus crystals and a trapped interstitial component equivalent to the liquid composition is a legacy of the model involving crystal settling to produce a pile of unconsolidated crystals. Such accumulation of primocrysts is possible, especially as a result of slumping of unconsolidated material growing on the walls or on steeply dipping surfaces bounding an intrusion (Irvine, 1982). However, in intrusions with very high area-to-height ratios this method of accumulation may be insignificant (Irvine *et al.*, 1983) and *in situ* growth may be the dominant mechanism (Campbell, 1978; McBirney and Noyes, 1979; Cawthorn and McCarthy, 1980). In view of the experimentally determined densities of sodic labradorite feldspars and the iron-enriched magma from which they crystallize in the Upper Zone of the Bushveld Complex (Campbell *et al.*, 1978), *in situ* growth would seem the more plausible way in which feldspar-rich layers can accumulate. Given this premise, there is then no need to postulate an uncompacted pile of euhedral crystals which need to be cemented together by the cooling magma.

The mechanism for adcumulus growth discussed by Sparks *et al.* (1984) is for almost complete solidification of the cumulate while at the crystal-liquid interface. This can occur only if the rate of accretion of the cumulus pile is extremely slow (Hess, 1939). In view of the immense size of the Bushveld Complex this seems highly plausible. Other very large intrusions, such as the Stillwater Complex, also contain cumulates with very small proportions of trapped liquid (Czamanske and Scheidle, 1985). In contrast, smaller intrusions, such as Skaergaard, or those with a large height-to-area ratio are more likely to contain greater proportions of trapped liquid in their cumulates.

Acknowledgements

This project was made possible by the generosity of the Geological Survey of South Africa in giving access to their borehole core. We thank the Chief Director, Dr P. J. Smit for permission to publish, and Dr F. Walraven for much assistance in obtaining core samples. Financial support from the C.S.I.R. is gratefully acknowledged. Drs P. Henderson and S. Sparks are thanked for reviewing an earlier draft of this manuscript. Technical assistance from Mrs S. Hall, P. King, A. Davies and Mr M. Hudson is appreciated.

References

Abbey, S. (1983) *Geol. Surv. Canada*, Paper 83-15.
Bowen, N. L. (1927) *Amer. J. Sci.* **14,** 89-108.
Brooks, C. K., and Nielsen, T. F. D. (1978) *Lithos*, **11,** 1-14.
Cameron, E. N. (1975) *Geochim. Cosmochim. Acta*, **39,** 1021-33.
Campbell, I. H. (1978) *Lithos*, **11,** 311-23.
——Roeder, P. L., and Dixon, J. M. (1978) *Contrib. Mineral. Petrol.* **67,** 369-77.
Cawthorn, R. G. (1977) *Trans. Geol. Soc. S. Afr.* **80,** 139-44.
——and McCarthy, T. S. (1980) *Earth Planet. Sci. Lett.* **46,** 335-43.
————(1985) *Econ. Geol.* **80,** 1016-26.
Coertze, F. J. (1970) *Geol. Soc. S. Afr., Spec. Publ.* **1,** 5-22.
Cox, K. G. (1980) *J. Petrol.* **21,** 629-50.
Czamanske, G. K., and Scheidle, D. L. (1985) *Montana Bureau Mines Geol., Spec. Publ.* **92,** 334-45.
Davies, G., and Cawthorn, R. G. (1984) *Mineral. Mag.* **48,** 469-80.
Green, T. H., and Watson, E. B. (1982) *Contrib. Mineral. Petrol.* **79,** 96-105.
Harrison, T. M., and Watson, E. B. (1984) *Geochim. Cosmochim. Acta*, **48,** 1467-77.
Hatton, C. J., and von Gruenewaldt, G. (1985) *Econ. Geol.* **80,** 911-24.
Henderson, P. (1968) *Geochim. Cosmochim. Acta*, **32,** 897-911.
——(1970) *J. Petrol.* **11,** 463-73.
——(1975) *Mineral. Mag.* **40,** 285-91.
Hess, H. H. (1939) *Trans. Am. Geophys. Union*, 430-2.
Himmelberg, G. R., and Loney, R. A. (1981) *U.S. Geol. Surv. Prof. Pap.* 1195, 24 pp.
Irvine, T. N. (1982) *J. Petrol.* **23,** 127-62.
——Keith, D. W., and Todd, S. G. (1983) *Econ. Geol.* **78,** 1287-334.
Lindsley, D. H., Brown, G. M., and Muir, E. D. (1969) *Mineral. Soc. Am. Spec. Pap.* No. 2, 193-201.
Markgraaff, J. (1976) *Trans. Geol. Soc. S. Afr.* **79,** 217-24.
McBirney, A. R. (1980 *J. Volcanol. Geotherm. Res.* **7,** 357-71.
——and Noyes, R. M. (1979) *J. Petrol.* **20,** 487-554.
McKenzie, D. P. (1984) Ibid. **25,** 713-65.
Molyneux, T. G. (1974) *Trans. Geol. Soc. S. Afr.* **77,** 329-38.
Norrish, K. and Hutton, J. T. (1969) *Geochim. Cosmochim. Acta*, **33,** 431-53.
Osborn, E. F. (1979) In *The Evolution of Igneous Rocks* (H. S. Yoder, Jr., ed.) Princeton Univ. Press, Princeton, New Jersey: 133-70.
Reynolds, I. M. (1985) *Econ. Geol.* **80,** 1027-48.
Sparks, R. S. J., Huppert, H. E., and Turner, J. S. (1984) *Phil. Trans. Roy. Soc. London*, **A130,** 511-34.
————Kerr, R. C., McKenzie, D. P., and Tait, S. R. (1985) *Geol. Mag.* **122,** 555-68.
Thompson, R. N. (1972) *Am. J. Sci.* **272,** 901-32.
von Gruenewaldt, G. (1973) *Trans. Geol. Soc. S. Afr.* **76,** 207-27.
——Sharpe, M. R., and Hatton, C. J. (1985) *Econ. Geol.* **80,** 803-12.
Wager, L. R. (1960) *J. Petrol.* **1,** 364-98.
——and Brown, G. M. (1968) *Layered Igneous Rocks*, Oliver and Boyd, Edinburgh, 588 pp.
——and Deer, W. A. (1939) *Medd. Grönland*, **105.**
——Brown, G. M., and Wadsworth, W. J. (1960) *J. Petrol.* **1,** 73-85.
Walraven, F. and Wolmarans, L. G. (1979) *Annals Geol. Surv. S. Afr.* **13,** 109-14.
Watson, E. B. and Capobianco, C. J. (1981) *Geochim. Cosmochim. Acta*, **45,** 2349-58.
——and Green, T. H. (1981) *Earth Planet. Sci. Lett.* **56,** 405-21.

Appendix: Analytical techniques

Analysis for P was by X-ray fluorescence spectrometry on crushed, whole-rock samples using a *Phillips* PW 1410 spectrometer. Operating conditions were 50 kV and 50 mA under vacuum (20 Pa) using a Cr tube. The P-*K*α peak, which is not overlapped by other spectral peaks, was measured using a Ge crystal and a flow-proportional counter. Counting time on the peak was for 40 seconds and background (2° away) for 20 seconds. Mass absorption effects were not taken into consideration, being slight. A set of international standards with P concentrations between 0.01% and 1.05% P_2O_5 was used for calibration.

Analysis of one sample (standard NIM-S) 143 times yielded a range of values between 568 and 589 ppm P. A close correlation between reported (Abbey, 1983) and calculated values for the standards was attained, although a certain imprecision must exist in the former as these are given as percentages, not parts per million.

Values in ppm P which we have obtained in our calibration for standardization are given below.

Standard	Calculated concentration (ppm)
GH	44
FK-N	87
DT-N	393
GA	524
G2′	567
NIM-S	568–589
SY 1	960
DR-N	1091
GSP-1	1222
MICA-FE	1964
AGV 1	2226
BR	4582

[*Manuscript received 11 November 1986; revised 6 April 1987*]