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STABLE ISOTOPIC SYSTEMATICS OF THE BUSHVELD COMPLEX: I. CONSTRAINTS OF MAGMATIC PROCESSES IN LAYERED INTRUSIONS

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ABSTRACT. The Bushveld Complex is the world's largest layered igneous intrusion and the greatest repository of magmatic ore deposits ever discovered. This paper reports oxygen isotopic data that impose new constraints on the magmatic evolution of this extraordinary intrusion. The $\delta^{18}O$ values of unaltered plagioclase and pyroxene are uniform in the Lower, Critical, and Main Zones of the intrusion. The average values are: $\delta^{18}O_{plag} = 7.1$, $\delta^{18}O_{opx} = 6.7$, and $\delta^{18}O_{magma} = 6.8$. By contrast, the Upper Zone $\delta^{18}O_{plag}$ values are heavier and more variable (7.6 ± 0.3), and the $\Delta^{18}O_{plag\text{-}opx}$ values are greater than 1.0. Early Proterozoic country rocks belonging to the Transvaal Supergroup have $\delta^{18}O$ values similar to those of Phanerozic examples of the same rock types.

Four aspects of the oxygen isotopic data indicate that the Bushveld magmas were contaminated by a significant amount of crustal material: (1) the $\delta^{18}O$ values of the Bushveld Complex are heavier than the $\delta^{18}O$ values of "normal" mantle-derived magmas; (2) most of the $\Delta^{18}O_{plag\text{-}opx}$ values are typical of magmatic values; (3) the sedimentary country rocks and most of the other potential crustal contaminants have heavier $\delta^{18}O$ values than those of the Bushveld Complex; and (4) the $\delta^{18}O$ values of volcanic country rocks from the Transvaal Supergroup are typical of "normal" mantle-derived magmas. The systematics of the stable isotopic data indicate that subsolidus exchange with hydrothermal fluids was not responsible for the heavy $\delta^{18}O$ values that characterize the Bushveld Complex. Previous Rb-Sr and Re-Os isotopic studies have demonstrated that the Bushveld magmas were strongly enriched in radiogenic Sr and Os relative to the contemporaneous bulk mantle, and these results are consistent with the proposition that the magmas were contaminated by a significant amount of crustal material.

Mass balance calculations indicate that at least 10 percent of the oxygen in the Bushveld Complex was derived from crustal sources. The fact that the $\delta^{18}O_{magma}$ values do not increase with stratigraphic height within the major zones of the intrusion indicates that most of the crustal contamination occurred before the magmas were emplaced in the magma chamber. The different isotopic signatures of the

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parental magmas may reflect differences in the amount and nature of the material they assimilated during their ascent through the continental crust. The assimilation of siliceous material by a mafic magma can alter the crystallization sequence of the cumulus minerals; the precipitation of orthopyroxene before clinopyroxene in the Bushveld Complex may be a manifestation of crustal contamination. The isotopic data indicate that different processes may be responsible for the origin of igneous layering in different parts of the Bushveld Complex. Magma mixing may be responsible for igneous layering in isotopically heterogeneous stratigraphic units, but internal mechanisms of magmatic differentiation appear to be responsible for igneous layering in isotopically homogeneous stratigraphic units.

INTRODUCTION

The study of layered intrusions has had a profound influence on our understanding of the processes responsible for the diversity of igneous rocks. There is a growing consensus that the conventional cumulus theory of crystal sorting by gravity settling (Wager and Deer, 1939; Wager, Brown, and Wadsworth, 1960; Hess, 1960; Jackson, 1961; Wager and Brown, 1968) does not satisfy some of the basic observational constraints on the petrogenesis of layered intrusions (Campbell, 1978). Criticisms of the cumulus theory have inspired an unprecedented proliferation of literature about layered intrusions, and a wide variety of innovative models and processes have been proposed to complement or supersede the cumulus theory (McBirney and Noyes, 1979; Chen and Turner, 1980; Huppert and Sparks, 1980; Irvine, 1980; Irvine, Keith, and Todd, 1983). Some models emphasize the petrological consequences of double diffusive convection and other fluid dynamical processes that operate within a magma chamber. Other models emphasize the role of crustal contamination, magma mixing, or heterogeneity in the mantle source region.

Fundamental questions regarding the petrogenesis of layered intrusions and the evolution of the sub-continental mantle cannot be resolved adequately until the effects of magmatic differentiation can be distinguished from the effects of crustal contamination, magma mixing, source rock heterogeneity, and subsolidus alteration. Stable isotopic data provide a powerful tool for monitoring a variety of magmatic and post-magmatic processes in igneous rocks. This paper reports oxygen isotopic data for unaltered samples from the Bushveld Complex and its country rocks. These data impose new constraints on the magmatic evolution of this extraordinary layered intrusion. A companion study (Schiffries and Rye, 1988) reports stable isotopic data for hydrothermal veins that crosscut the intrusion, and these data impose new constraints on the subsolidus evolution of the Bushveld Complex.

GEOLOGICAL SETTING

The Bushveld Complex is the world's largest layered igneous intrusion and the greatest repository of magmatic mineral resources

ever discovered. It has a stratigraphic thickness ranging between 7 and 9 km and is exposed as three major lobes that enclose an area of 65,000 km^2 (fig. 1). It is 100 to 1000 times larger than the Skaergaard intrusion, the Stillwater Complex, and the Cuillin Complex (Willemse, 1969). Size alone draws attention to the Bushveld Complex, but it is also famous for the remarkable persistence and regularity of its igneous layering. Individual layers and cyclic units have been traced along strike for more than 100 km in both the eastern and western lobes of the intrusion. Some of the layers contain immensely valuable mineral resources, and these deposits produce a major fraction of the world's output of chromium, vanadium, and the platinum-group elements.

The age of the Bushveld Complex is 2050 ± 25 Ma (λ_{Rb} = 0.0142 Ga^{-1}; Hamilton, 1977; Sharpe, 1985). It intruded a 12 km thick pile of early Proterozoic sedimentary and volcanic rocks that belong to the Transvaal Supergroup. The Transvaal Supergroup unconformably overlies Archean basement rocks of the Kaapvaal craton. A portion of the Potgietersrus lobe of the Bushveld Complex is in contact with granitic basement rocks, but the larger eastern and western lobes of the intrusion are separated from the basement by several kilometers of Transvaal Supergroup sedimentary and volcanic rocks. The emplacement of the Bushveld Complex was preceded by an extensive period of volcanism as recorded in the Rooiberg Felsite and Dullstroom Volcanics of the Transvaal Supergroup. Intrusion of the mafic rocks of the Bushveld Complex was followed by a period of granitic plutonism recorded in the Bushveld Granite. A schematic cross section of the contact between the eastern Bushveld Complex and its country rocks is shown in figure 1B.

The Bushveld Complex contains a broad spectrum of plutonic rock types. The magmatic stratigraphy is divided into four major zones (fig. 1). The Lower Zone consists of ultramafic rocks, including dunite, orthopyroxenite, and hartzburgite. Above the Lower Zone lies the Critical Zone, which contains abundant norite, anorthosite, and orthopyroxenite, as well as approx 20 chromite layers that individually are up to 2 m thick. The Main Zone is dominated by gabbro and contains subordinate amounts of anorthosite. The Upper Zone is composed of magnetite gabbro, ferrodiorite, anorthosite, and approx 25 magnetite layers.

The first appearance of cumulus magnetite has been traditionally regarded as the boundary between the Main Zone and the Upper Zone. The recognition of sharp variations in a variety of petrologic indicators at the level of the Pyroxenite Marker has led to the suggestion that this layer, rather than the first appearance of cumulus magnetite, should be regarded as the boundary between the Main and Upper Zones (Sharpe, 1985; Kruger, Cawthorn, and Walsh, 1987). In this paper we consider the Pyroxenite Marker to be the boundary between these zones. This layer was formerly regarded as the boundary between subzones B and C of the Main Zone.

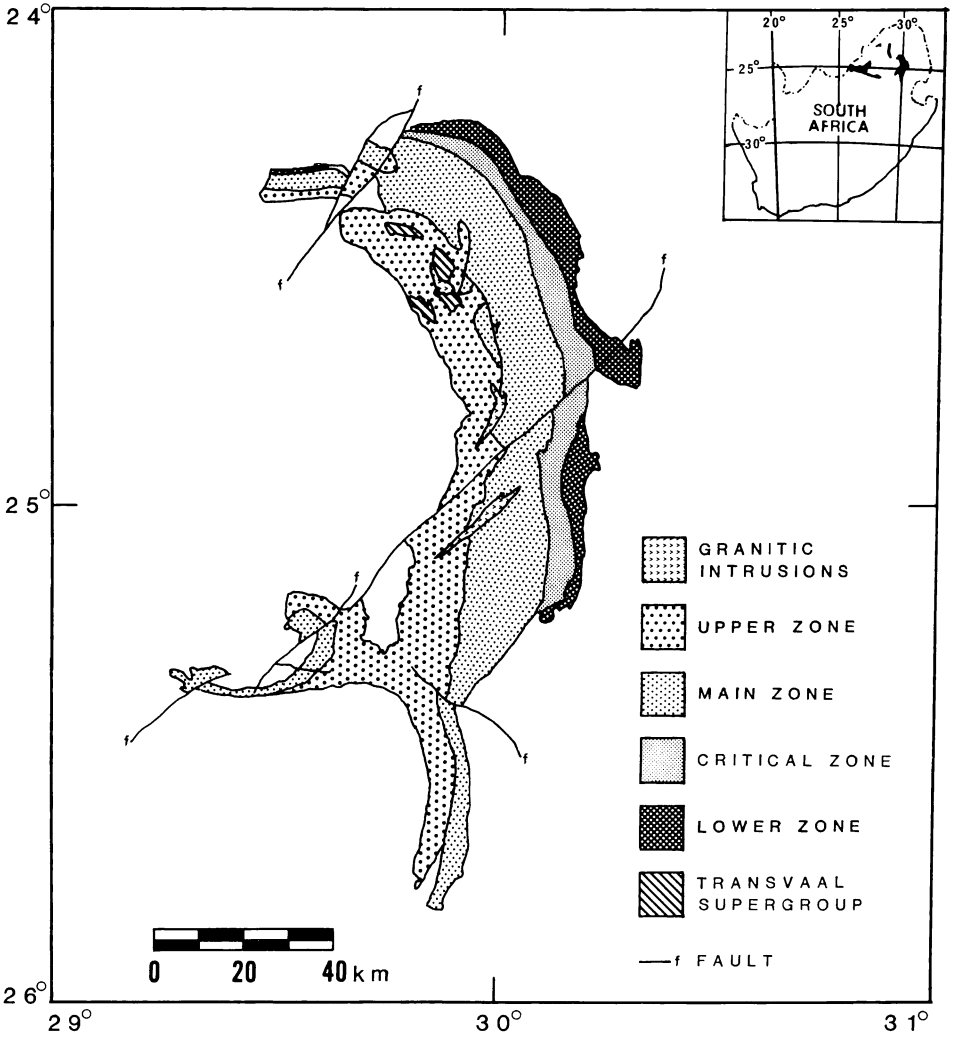
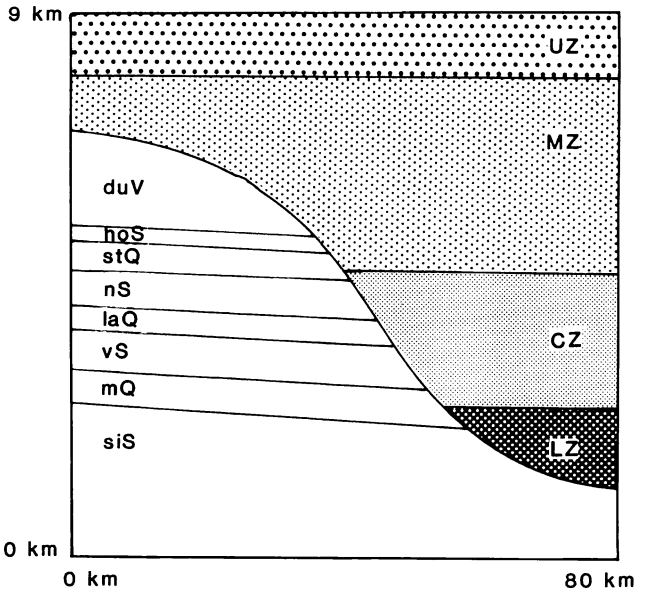


Fig. 1(A) Generalized geologic map of the eastern lobe of the Bushveld Complex.

There is a broad consensus that several major batches of magma were added to the Bushveld magma chamber as it cooled and crystallized, but there is considerable controversy about the chronology of the magma influxes, the number of parental magmas involved, and the origin of the parental magmas. The first major influx of magma formed the Lower Zone and lower Critical Zone. This "U-type" magma had ultramafic affinities (Irvine, Keith, and Todd, 1983). Additions of "A-type" anorthositic magma occurred during the crystallization of the



(B) Schematic cross section of the contact between the Bushveld Complex and the Transvaal Supergroup (after von Gruenewaldt, Sharpe, and Hatton, 1985). Abbreviations are: UZ = Upper Zone, MZ = Main Zone, CZ = Critical Zone, LZ = Lower Zone; duV = Dullstroom volcanics, hoS = Houtenbek shale, stQ = Steenkampsberg quartzite, nS = Nederhorst shale, laQ = Lakenvalei quartzite, vS = Vermont shale, mQ = Magaliesberg quartzite, siS = Silverton shale.

upper Critical Zone. The Main Zone formed from a very large influx of gabbroic or anorthositic magma, and the origin of the Merensky Reef is probably related to the emplacement of this magma. Most workers believe that the Upper Zone formed from a major influx of a new parental magma emplaced at the level of the Pyroxenite Marker. An alternative hypothesis is that the Main Zone represents a liquid layer intruded into the density-stratified magma pile at a level consistent with its density (Sharpe, 1985).

SAMPLES AND ANALYTICAL PROCEDURES

Sample Selection

Careful consideration must be given to the sampling strategy in order to characterize the oxygen isotopic systematics of a large intrusion. The samples analyzed in this study are, with two exceptions, from the eastern Bushveld Complex (table 1). The eastern lobe of the Bushveld Complex was chosen because it is the best exposed and the most intensively studied of the three major lobes of the intrusion. The samples span the entire 9 km stratigraphic thickness of the eastern Bushveld Complex. The sampling density is especially high near the

TABLE 1

Sample descriptions

Sample number	Strat. Height (km)	Locality (farm or mine)	Lithology	Remarks
			Upper Zone	
BDS-104	8.97	Onverwacht 148 JS	Magnetite diorite	Contains ol, ap, bi, qtz, and hbl; moderately altered
BDS-109	8.75	Onverwacht 148 JS	Magnetite diorite	Contains ol, and ap; slightly altered; crosscut by minor veinlet
BDS-82	7.30	Luipershoek 149 JS	Magnetite gabbro	Contains ol and smyplectite
BH40-1-1	6.85	Mapochsgronde 500 JS	Anorthosite	Immediately below Main Magnetite Layer
			Main Zone	
BDS-66	6.53	Mapochsgronde 500 JS	Feldspathic pyroxenite	Pyroxenite Marker; incipient alteration; contains trace of cc
BDS-67	6.51	Mapochsgronde 500 JS	Gabbro	
BDS-68	6.51	Mapochsgronde 500 JS	Gabbro	
BDS-41	5.75	Mapochsgronde 500 JS	Gabbro	Contains 1% calcic myrmekite
85-78-1a	5.00	Kwaggskop 359 JS	Gabbro	3 cm from intermed. temp. vein
PB1-383	4.32	Pietersburg 44 JT	Gabbro	
PB1-3146	3.82	Pietersburg 44 JT	Gabbro	
PB1-4231	3.29	Pietersburg 44 JT	Mottled anorthosite	
PB1-4352	3.27	Pietersburg 44 JT	Leuconorite	
			Critical Zone	
79-156	3.26	Atok Mine	Feldspathic pyroxenite	Merensky Reef
86-19	3.26	Rustenburg Mine	Feldspathic pyroxenite	Merensky Reef; abundant sulfides
R-11	3.26	Rustenburg Mine	Feldspathic pyroxenite	Merensky Reef pothole; abundant sulfides
PB1-4398	3.25	Pietersburg 44 JT	Spotted anorthosite	
86-141c	3.02	Hackney 116 KT	Feldspathic pyroxenite	Pegmatoid below UG-2 chromitite layer
MDH5-202	2.74	Maandagshoek 254 KT	Leuconorite	
MDH22-270	2.40	Maandagshoek 254 KT	Leuconorite	
MDH22-370	2.29	Maandagshoek 254 KT	Feldspathic pyroxenite	
JAG19-180	2.18	Jaglust 418 KS	Orthopyroxenite	
JAG19-346	2.02	Jaglust 418 KS	Orthopyroxenite	Highly fractured
			Lower Zone	
86-152	1.00	Jaglust 418 KS	Orthopyroxenite	Contains olivine

Abbreviations: ol = olivine; ap = apatite; bi = biotite; qtz = quartz; hbl = hornblende; cc = calcite.

stratigraphic boundary between the Critical and Main Zones (approximately at the level of the Merensky Reef) and between the Main and Upper Zones (approximately at the level of the Pyroxenite Marker). Many of the samples were obtained from M.R. Sharpe, and Rb-Sr data for some of these samples are reported by Sharpe (1985).

We have attempted to obtain fresh samples from areas that are relatively free from structural disturbances or obvious networks of hydrothermal veins. Samples from diamond drill cores were used to obtain coverage of stratigraphic intervals for which fresh material is not readily available from surface exposures. Ten of the samples are from diamond drill cores, and they have sample numbers with the prefixes PB1, MDH, or JAG. The remaining samples are from fresh outcrops.

Samples from the Lower Zone and lower Critical Zone are from the Oliphants River trough in the northern sector of the eastern Bushveld Complex, an area studied in detail by Cameron (1978). Additional samples from the Critical Zone are from the central sector of the eastern Bushveld Complex, and the geology of this region is described by Cameron (1980, 1982). Samples from the Main and Upper Zones are from the vicinity of Roossenekal, north of the Laersdrif fault and south of the Steelpoort fault, and the stratigraphy and petrology of this region have been studied by von Gruenewaldt (1973). Two samples of the Merensky Reef were obtained from the Rustenburg Platinum Mine in the western Bushveld Complex, and a third sample is from the Atok Platinum Mine in the northern sector of the eastern Bushveld Complex. One sample from the Main Zone was collected in a dimension-stone quarry south of the Laersdrif fault. This sample, which is 3 cm from an intermediate temperature vein, was analyzed to evaluate the effects of fluid-rock interactions.

Most samples from the Main Zone are gabbros, and most samples from the Critical Zone are norites. However, to test for possible modal effects, several samples of anorthosite and orthopyroxenite have also been analyzed. The interpretation of isotopic data for the Critical and Main Zone samples is greatly simplified, because the samples are composed almost entirely of plagioclase and pyroxene (orthopyroxene and clinopyroxene have extremely similar isotopic properties and can be treated as one mineral for the purposes of this discussion). The Lower and Upper Zones cannot be characterized as easily. Magnetite is a cumulus phase in most Upper Zone rocks, and olivine is present throughout much of the Lower Zone. These minerals pose analytical difficulties, and their presence also complicates the interpretation of the isotopic data. We have determined the oxygen isotopic composition of plagioclase, orthopyroxene, clinopyroxene, olivine, magnetite, chromite, and biotite. However, we have focused on plagioclase and orthopyroxene, because they are the most abundant minerals in the intrusion, and because their isotopic behavior is well understood.

Samples from the Transvaal Supergroup were collected from outcrops of the Dullstroom Volcanics, the Steenkamsberg Quartzite,

the Nederhorst Shale (hornfels), the Lakenvalei Quartzite, the Vermont Shale (hornfels), the Magliesberg Quartzite, and the Malmani Dolomite. The $\delta^{18}O$ values were measured on whole rocks for the hornfels and volcanics, on quartz for the quartzites, and on dolomite for the Malmani Dolomite.

Sample Preparation

If individual grains of a given mineral are isotopically zoned, then the interpretation of the data depends on the textural relations of the material analyzed. For example, the interpretation of mineral-pair geothermometers is dependent on whether the compositions reflect core-core, rim-rim, or core-rim relations. For concentric isotopic zoning or grain-boundary alteration, rim-rim data are most likely to reflect the effects of subsolidus processes, and core-core data are most likely to provide information about the initial crystallization temperature. Results based on indiscriminate mixtures of core and rim material, or of altered and unaltered material, may be highly scattered and are likely to be difficult to interpret. In this study we have attempted to minimize the effects of subsolidus processes by analyzing unaltered material from the cores of cumulus grains.

In order to assure high quality mineral separates, the mineral separates analyzed in this study were prepared by hand picking individual grains (approx 50 mesh) with tweezers under a binocular microscope. Every grain was individually examined for mineral inclusions, alteration, adhering particles, and other contaminants. In most cases the mineral separates were composed of exceptionally clear grains with no visible alteration. It was generally possible to pick tabular grains of plagioclase bounded by two freshly broken cleavage surfaces and equidimensional grains of orthopyroxene bounded by up to four cleavage surfaces. The clarity and morphology of the individual grains indicate that the mineral separates were predominantly composed of unaltered material from the cores of cumulus grains. The mineral separates are believed to be at least 99 percent pure.

Extraction Procedure and Mass Spectrometry

The isotopic ratios reported in this study were measured in the stable isotope laboratory at Yale University. Quantitative separation of oxygen from silicate minerals was accomplished using a reconditioned and modified BrF_5 extraction line (Clayton and Mayeda, 1963). Analytical procedures developed to minimize contamination and increase precision are discussed in greater detail by Schiffries (ms). Samples were loaded into Ni reaction vessels in a glove box under an atmosphere of dry N_2. Plagioclase, pyroxene, biotite, and quartz were reacted with BrF_5 for approx 12 hrs at 550°C. Magnetite and chromite were reacted for 15 hrs at 650°C. The O_2 was converted to CO_2 by reaction with a hot graphite rod in the presence of platinum, and the volume of CO_2 was

measured in a manometer. The average oxygen yield for silicates minerals is 98 ± 2 mole percent. The yields for oxide minerals were less than 90 percent and highly variable.

The isotopic composition of CO_2 gas was measured with a new Finnegan MAT 251E mass spectrometer equipped with seven collectors and an automatic gas handling system. The internal precision of the instrument was typically 6×10^{-6}, corresponding to 0.006 permil ($2\sigma_m$). Backgrounds at masses 44, 45, and 46 were measured before each data block, and the baseline for each amplifier was determined by peakstepping to mass 43.5 on the mass 44 collector cup. The purity of the CO_2 gas was checked by peakstepping and measuring the intensities of masses 18 (H_2O), 14 (N), and 40 (Ar) relative to the intensity of mass 44 after each run. The relative intensities of the potential contaminants were negligible in every case.

Representation of the Isotopic Data

All isotopic data are presented in the standard delta notation:

$$\delta^{18}O_{spl} = 1000\left[\frac{(^{18}O/^{16}O)_{spl}}{(^{18}O/^{16}O)_{std}} - 1\right]$$

where the sample (spl) is either a mineral separate or a whole rock, and the standard (std) is V-SMOW.

The isotopic fractionation between two coexisting phases (A and B) is expressed as:

$$\Delta^{18}O_{A-B} \equiv \delta^{18}O_A - \delta^{18}O_B$$
$$\approx 1000[\ln(\alpha_{A-B})]$$

where

$$\alpha_{A-B} = \frac{(^{18}O/^{16}O)_A}{(^{18}O/^{16}O)_B}$$

Precision of the Isotopic Data

During the course of the Bushveld study we extracted and analyzed the oxygen from 16 aliquots of standard NBS-28 (African glass standard). The mean and standard deviation of the 16 analyses is 9.70 ± 0.20 permil (2σ). In some cases the NBS standard was run in a reaction vessel immediately after a sample having a low yield or unusual isotopic composition (for example, magnetite), and therefore the standard deviation of the 16 analyses is somewhat poorer than the reproducibility achieved under routine conditions that characterize the majority of the data. The last six aliquots of standard NBS-28 were run under routine conditions, and the standard deviation of this group of analyses is 0.03 permil (2σ).

RESULTS

Oxygen Isotope Analyses

Intrusive rocks.—The $\delta^{18}O$ values of a representative suite of samples from the Bushveld Complex are presented in table 2, and the average values ($\pm 1\sigma$) for the major stratigraphic zones and for the entire intrusion are given in table 3. Stratigraphic profiles of $\delta^{18}O_{plag}$, $\delta^{18}O_{opx}$, and $\Delta^{18}O_{plag\text{-}opx}$ are shown in figure 2.

The $\delta^{18}O_{plag}$, $\delta^{18}O_{opx}$, and $\Delta^{18}O_{plag\text{-}opx}$ values from the composite section of the eastern Bushveld Complex are nearly uniform across the entire 6.5 km stratigraphic thickness that comprises the Lower, Critical, and Main Zones (Schiffries and Rye, 1987). This remarkable uniformity of the $\delta^{18}O$ values appears to extend laterally as well as vertically. There appears to be little or no difference between the oxygen isotopic values in different parts of the eastern lobe of the Bushveld Complex. In addition, the few samples from the western Bushveld Complex and the sample collected 3 cm from a hydrothermal vein have $\delta^{18}O$ values that are indistinguishable from values for fresh samples from the eastern lobe of the Complex. The $\delta^{18}O_{opx}$ value for a sample from the Lower Zone is 6.9. The average values ($\pm 1\sigma$) for samples from the Critical Zone are: $\delta^{18}O_{plag} = 7.1 \pm 0.2$; $\delta^{18}O_{opx} = 6.7 \pm 0.2$; and $\Delta^{18}O_{plag\text{-}opx} = 0.5 \pm 0.1$. The average values for samples from the Main Zone are: $\delta^{18}O_{plag} = 7.0 \pm 0.2$; $\delta^{18}O_{opx} = 6.6 \pm 0.2$; and $\Delta^{18}O_{plag\text{-}opx} = 0.4 \pm 0.2$. The average $\delta^{18}O$ and $\Delta^{18}O$ values for the Main and Critical Zones are indistinguishable at the 1σ level. Variations in $\delta^{18}O_{plag}$ and $\delta^{18}O_{pyx}$ are strongly correlated, and the standard deviation of the mean $\Delta^{18}O_{plag\text{-}opx}$ values for the Critical and Main Zones are smaller than the standard deviations of the mean $\delta^{18}O_{plag}$ and $\delta^{18}O_{opx}$ values. The difference between the maximum and minimum values of $\delta^{18}O_{plag}$, $\delta^{18}O_{opx}$, and $\Delta^{18}O$ for all samples from the Critical and Main Zones is approx 0.5 permil, and in each case the standard deviation of the mean (0.1–0.2 permil) is comparable to the analytical uncertainty for replicate analyses of a single sample. By contrast, the Upper Zone $\delta^{18}O_{plag}$ values are heavier and more variable (7.6 ± 0.3), and the $\Delta^{18}O$ values are larger than 1.0 (fig. 3).

Country rocks.—The $\delta^{18}O$ values of a representative suite of Bushveld Complex country rocks are listed in table 4 and illustrated in figure 4. The $\delta^{18}O$ values reported here for Transvaal Supergroup rocks are typical of values reported for Phanerozoic examples of the same rock types (Hoefs, 1987). The sedimentary country rocks have $\delta^{18}O$ values of 9 to 11 for quartzites, 11 to 15 for pelites, and 19 to 21 for carbonates. The mafic to intermediate volcanic country rocks have a $\delta^{18}O$ value of 5.5. Oxygen isotope analyses of basement gneisses beneath the Transvaal Supergroup are not available. Heuber, Kyser, and Nisbet (1986) report $\delta^{18}O$ values of 9 to 10 for high grade gneisses in the Limpopo belt, and these values are typical of lower crustal gneisses from other areas (Longstaff and Schwartz, 1977).

TABLE 2

Oxygen isotope analyses of intrusive rocks

Sample number	Strat. Height (km)	$\delta^{18}O_{plag}$ (‰)	$\delta^{18}O_{opx}$ (‰)	$\Delta^{18}O_{plag\text{-}opx}$ (‰)	$\delta^{18}O_{min}$ (‰)	T (°C)	$\delta^{18}O_{melt}$ (‰)
			Upper Zone				
BDS-104	8.97	7.71					
BDS-109	8.75	7.64	6.12	1.52		960	7.25
BDS-82	7.30	7.65	6.49	1.16	4.11 (ol)	1040	7.28
BDS-82	7.30				1.84 (mt)		
BH40-1-1	6.85	7.36					
			Main Zone				
BDS-66	6.53	6.86	6.44	0.42		1220	6.62
BDS-67	6.51	6.94	6.32	0.62		1170	6.66
BDS-68	6.51	7.00			6.28 (cpx)		
BDS-41	5.75	7.07	6.67	0.40		1220	6.84
85-78-1a	5.00	6.97	6.61	0.36		1230	6.75
PB1-383	4.32	6.77	6.46	0.31		1250	6.56
PB1-3146	3.82	7.18	6.97	0.21		1270	7.00
PB1-4231	3.29	7.17					
PB1-4352	3.27	7.17	6.42	0.75		1140	6.86
			Critical Zone				
79-156	3.26	7.20	6.79	0.41	4.29 (chr)	1220	6.97
86-19	3.26	7.48	6.86	0.62	6.84 (cpx)	1170	7.20
86-19	3.26				6.07 (bi)		
R-11	3.26	7.03	6.61	0.42	5.78 (bi)	1220	6.80
PB1-4398	3.25	6.88	6.45	0.43		1220	6.64
86-141c	3.02	7.24	6.80	0.44		1220	7.00
MDH5-202	2.74	6.85	6.40	0.45		1210	6.61
MDH22-270	2.40	7.02	6.44	0.58		1180	6.75
MDH22-370	2.29	7.07	6.59	0.48		1200	6.82
JAG19-180	2.18		6.93				
JAG19-346	2.02		6.77				
			Lower Zone				
86-152	1.00		6.94				

TABLE 3

Average $\delta^{18}O$ values ($\pm 1\sigma$) of the intrusive rocks

	$\delta^{18}O_{plag}$ (‰)	$\delta^{18}O_{opx}$ (‰)	$\Delta^{18}O_{plag\text{-}opx}$ (‰)	$\delta^{18}O_{melt}$ (‰)	T (°C)
Upper Zone	7.6 ± 0.4	6.3 ± 0.3	1.3 ± 0.4	7.3 ± 0.0	1000 ± 40
Main Zone	7.0 ± 0.2	6.6 ± 0.2	0.4 ± 0.2	6.8 ± 0.2	1220 ± 40
Critical Zone	7.1 ± 0.1	6.7 ± 0.2	0.5 ± 0.1	6.8 ± 0.2	1210 ± 20
Lower Zone		6.9			
Average	7.2 ± 0.4	6.6 ± 0.2	0.6 ± 0.3	6.9 ± 0.2	1190 ± 80

INTERPRETATION

Subsolidus Processes

In order to estimate the $\delta^{18}O$ value of the Bushveld magmas it is necessary first to evaluate the effects of both mineralogical alteration and subsolidus isotopic exchange on the $\delta^{18}O$ values reported in table 2. The Bushveld Complex is crosscut by extensive networks of hydrothermal veins produced by the fracture-controlled flow of aqueous fluids during the subsolidus cooling history of the intrusion (Schiffries and Skinner, 1987). Petrological studies of the veins indicate that fluid-rock interactions occurred over a wide range of physical and chemical conditions.

Mineralogical alteration.—Although the Bushveld Complex is crosscut by extensive networks of hydrothermal veins, the sampling proce-

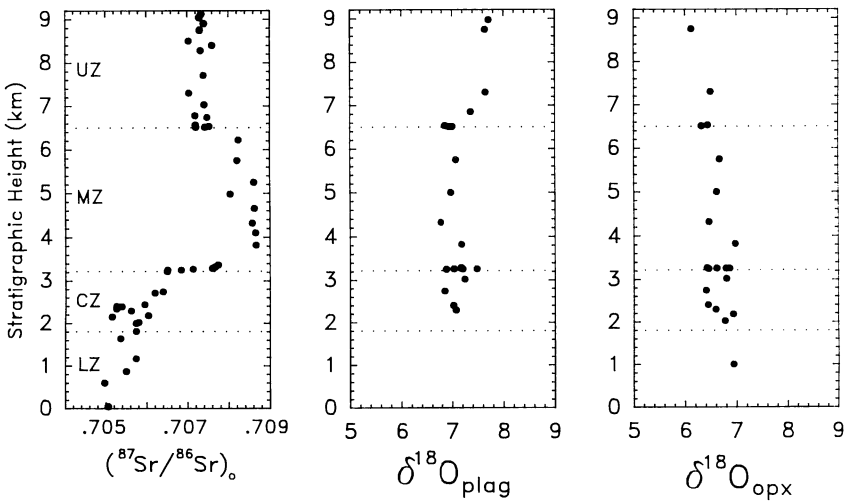


Fig. 2. Stratigraphic variations of $(^{87}Sr/^{86}Sr)_0$, $\delta^{18}O_{plag}$, and $\delta^{18}O_{opx}$. The $(^{87}Sr/^{86}Sr)_0$ values are from Sharpe (1985, 1986).

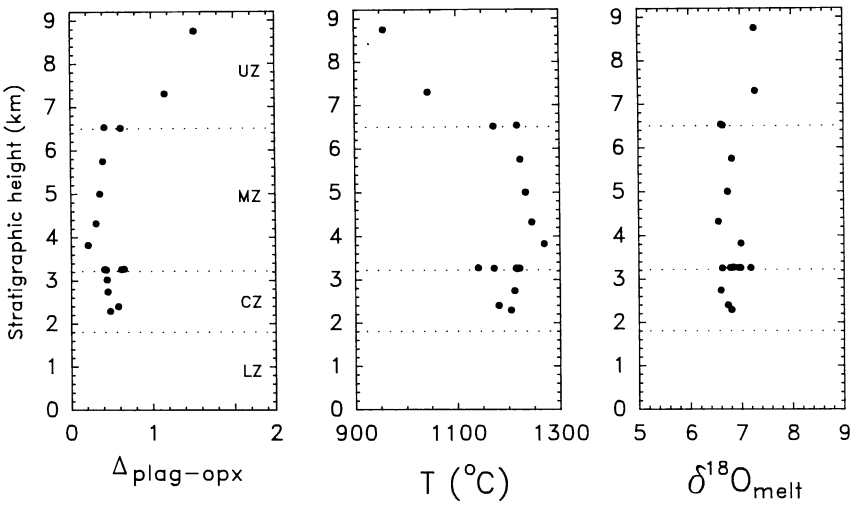


Fig. 3. Stratigraphic variations of $\Delta^{18}O_{plag\text{-}opx}$, minimum initial crystallization temperature, and $\delta^{18}O_{magma}$.

dure used for the preparation of the mineral separates analyzed in this study virtually assures that none of the mineral separates was contaminated with alteration minerals. Thus, any subsolidus changes in the $\delta^{18}O$ values of the minerals are the result of isotopic exchange reactions rather than net-transfer reactions that involve the formation of secondary hydrothermal minerals. A detailed discussion of the oxygen isotopic systematics of hydrothermally altered samples is given by Schiffries (ms). It should be noted that hydrothermally altered plagioclase is generally enriched in ^{18}O relative to unaltered samples, whereas hydrothermally altered pyroxene (amphibole present) is generally depleted in ^{18}O relative to fresh material. The magnitude of these shifts is up to 1 permil.

Subsolidus isotopic exchange.—The coordinated use of $\delta^{18}O_{plag}$ and $\delta^{18}O_{pyx}$ data provides a sensitive tool for monitoring the effects of subsolidus isotopic exchange in gabbroic rocks (Taylor and Forester, 1979; Gregory and Taylor, 1981; Criss, Gregory, and Taylor, 1987; Gregory and Criss, 1986). The systematics of a $\delta^{18}O_{pyx}$ versus $\delta^{18}O_{plag}$ diagram are illustrated in figure 5. The equilibrium isotopic fractionation between plagioclase and pyroxene is 0.5 ± 0.1 permil at magmatic temperatures. For a "normal" mantle-derived mafic magma, the value of $\delta^{18}O_{pyx}$ is 5.5, and the value of $\delta^{18}O_{plag}$ is 6.0 (point B in fig. 5). If pyroxene and plagioclase are the dominant minerals in the rock, then closed-system equilibrium exchange would produce complementary depletions and enrichments in the $\delta^{18}O$ values of pyroxene and plagioclase, respectively, and the resulting samples would plot within triangle ABC (fig. 5). Plagioclase-pyroxene rocks that crystallized from melts

TABLE 4

Oxygen isotope analyses of sedimentary and volcanic country rocks from the Transvaal Supergroup

Sample number	Dist. from Contact (km)	Sample	$\delta^{18}O$ (‰)	Comments
		Dullstroom Volcanics		
85-175	0.02	whole rock	6.82	Migmatitic zone of contact aureole
85-147	0.9	whole rock	5.53	Hydrothermal veins spaced 1-10 cm apart
85-182	2.5	whole rock	5.53	Hydrothermal veins spaced 1-10 cm apart
		Steenkamsberg Quartzite		
86-187	0.1	quartz	9.20	Recrystallized; coarse grained
86-197	0.5	quartz	9.66	Recrystallized; coarse grained
86-198	1.2	quartz	10.37	Recrystallized; coarse grained
86-199	1.9	quartz	9.94	Medium grained
86-200	3.3	quartz	9.99	Medium grained
86-204	10.1	quartz	10.11	Medium grained
		Nederhorst Shale		
86-207	3.5	whole rock	11.06	Hornfels
		Lakenvalei Quartzite		
86-208	5.5	quartz	11.06	Medium grained
		Vermont Shale		
86-210	6.5	whole rock	15.05	Hornfels
86-211	11.5	whole rock	15.12	Hornfels
		Magaliesberg Quartzite		
85-336	0.3	quartz	10.10	Oliphants River trough
86-1	1.5	quartz	11.00	Rustenberg area; coarse grained
		Malmani Dolomite		
Bar-1	0-1	dolomite	20 ± 1	Potgietersrust lobe; 6 samples

with different $\delta^{18}O$ values and that attained isotopic equilibrium at a given temperature would define a line with a slope of 1.0 and an intercept equal to the equilibrium fractionation factor on a $\delta^{18}O_{plag}$ versus $\delta^{18}O_{pyx}$ diagram. Open-system equilibrium exchange at submagmatic temperatures can generate compositions that lie between the lines represented by $\Delta^{18}O = 1.5$ and $\Delta^{18}O = 0.5$, which correspond to closure temperatures of approx 550° and 1200°C, respectively.

Gabbroic rocks affected by fluid-rock interactions generally plot along a steeply sloping disequilibrium trend on a $\delta^{18}O_{plag}$ versus $\delta^{18}O_{pyx}$ diagram (line EBF in fig. 5). This trend is generated because the reaction rate for isotopic exchange between plagioclase and water is much faster than the reaction rate for exchange between pyroxene and water (Taylor and Forester, 1971, 1979; Gregory and Taylor, 1981). Disequilibrium exchange with isotopically light fluids is characterized by reverse fractionations between plagioclase and pyroxene ($\Delta^{18}O < 0$) and by samples that plot near line segment BE in figure 5. This behavior is

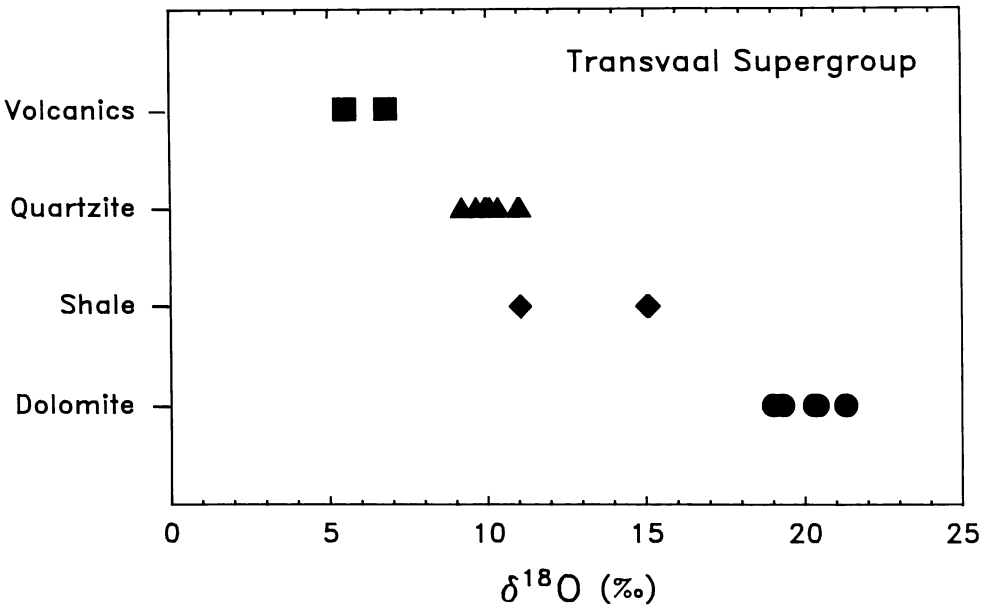


Fig. 4. Oxygen isotope analyses of sedimentary and volcanic country rocks belonging to the Transvaal Supergroup. Assimilation or isotopic exchange between the magma and the sedimentary rocks would increase $\delta^{18}O_{magma}$. Assimilation or isotopic exchange between the magma and the volcanic rocks would have little effect on $\delta^{18}O_{magma}$.

exemplified by data for the Skaergaard intrusion (Taylor and Forester, 1979). Disequilibrium exchange with isotopically heavy fluids is characterized by samples that plot near line segment BF in figure 5, and some samples may have abnormally large isotopic fractionation between plagioclase and pyroxene ($\Delta^{18}O > 1.5$). This behavior is exemplified by data for the Samail ophiolite (Gregory and Taylor, 1981).

The $\delta^{18}O_{plag}$ and $\delta^{18}O_{opx}$ data for the Critical and Main Zones of the Bushveld Complex do not exhibit any of the patterns that are diagnostic of appreciable subsolidus exchange under either equilibrium or disequilibrium conditions (fig. 5). The $\Delta^{18}O$ values are indicative of magmatic temperatures, and we conclude that the isotopic compositions of pyroxene and plagioclase are very close to their magmatic values.

The large $\Delta^{18}O$ values for samples from the Upper Zone (table 2) clearly indicate that these rocks have undergone some subsolidus exchange. However, the nature of the exchange is uncertain, because the rocks contain cumulus magnetite as well as some hydrothermal alteration. The $\delta^{18}O_{plag}$ values for the Upper Zone are heavier by about 0.5 permil than the values for other parts of the Bushveld Complex, whereas the $\delta^{18}O$ value for unaltered pyroxene is indistinguishable from the values for other parts of the intrusion. The rocks in the Upper Zone contain an appreciable amount of magnetite, and therefore it is possible

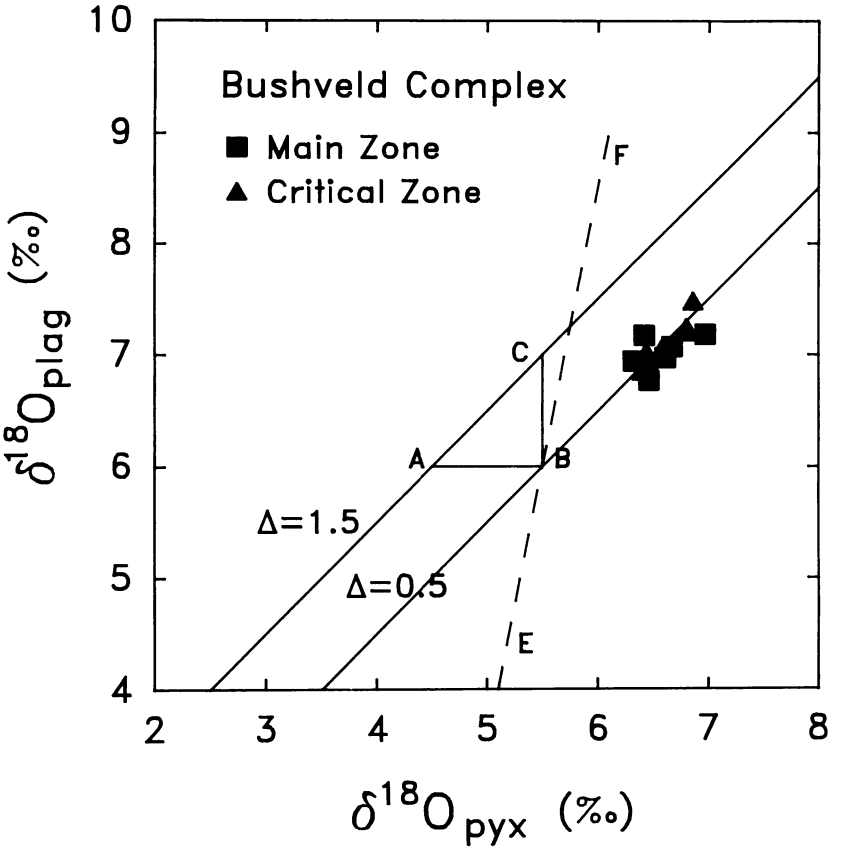


Fig. 5. Plot of $\delta^{18}O_{opx}$ versus $\delta^{18}O_{plag}$. At magmatic temperatures, plagioclase and pyroxene in equilibrium with a "normal" mafic magma have $\delta^{18}O$ values of 6.0 and 5.5, respectively (point B). If pyroxene and plagioclase are the dominant minerals in the rock, then closed-system, equilibrium cooling will result in compositions that lie in triangle ABC. Subsolidus exchange between a "normal" basalt and externally derived aqueous fluids produces a steeply sloping disequilibrium trend (dashed line). The lines defined by $\Delta^{18}O$ = 1.5 and $\Delta^{18}O$ = 0.5 correspond to temperatures of approx 550° and 1200°C, respectively.

that the $\delta^{18}O_{plag}$ values were affected by subsolidus exchange with magnetite, but pyroxene was not significantly affected by exchange with either mineral. Alternatively, these rocks may have exchanged with a high-$\delta^{18}O$ fluid. In either case we do not have sufficient information to determine unambiguously the original $\delta^{18}O$ value of the Upper Zone magma.

Estimation of the Initial Crystallization Temperature

The fractionation of oxygen isotopes between coexisting plagioclase and orthopyroxene can be used to place a lower bound on the

initial crystallization temperature of the Bushveld magmas. The temperature dependence of the plagioclase-pyroxene fractionation factor has been calibrated by several experimental and empirical techniques under a variety of chemical and physical conditions. A regression of the available data (Muehlenbachs and Kushiro, 1974; Bottinga and Javoy, 1975; Friedman and O'Neil, 1977; Matthews, Goldsmith, and Clayton, 1983) for magmatic temperatures gives (Dunn, 1986):

$$T(°C) = 1320 - 239(\Delta^{18}O_{plag\text{-}opx})$$

The uncertainty of the temperature calibration is approx ±60°C.

The calculated isotopic temperatures are listed in tables 2 and 3, and a stratigraphic temperature profile is shown in figure 3. Minimum initial crystallization temperatures for the Critical and Main Zones are 1200°C. The results of the isotopic thermometers are in excellent agreement with independent estimates of the crystallization temperatures summarized by Sharpe and Hulbert (1985). On the other hand, the calculated temperatures for the Upper Zone are 960° to 1040°C and reflect the large $\Delta^{18}O_{plag\text{-}opx}$ values produced by subsolidus processes.

Isotopic Composition of the Magmas

Oxygen isotopic composition of the melts.—The $\delta^{18}O$ value of a silicate melt that is in isotopic equilibrium with a mineral can be calculated from:

$$\delta^{18}O_{melt} = \delta^{18}O_{min} - \Delta^{18}O_{min\text{-}melt}(T)$$

where $\delta^{18}O_{min}$ is the isotopic composition of a mineral in equilibrium with the melt, T is determined from the isotopic fractionation between plagioclase and pyroxene (see above), and $\Delta^{18}O_{min\text{-}melt}(T)$ is calculated from a regression of the available data (Dunn, 1986). The calculated values of $\delta^{18}O_{melt}$ are presented in tables 2 and 3, and a stratigraphic profile of $\delta^{18}O_{melt}$ is shown in figure 3. The calculated $\delta^{18}O_{melt}$ values for the Bushveld Complex range from 6.6 to 7.2. The calculated average $\delta^{18}O_{melt}$ values are 6.8 ± 0.2 for the Critical Zone and 6.8 ± 0.1 for the Main Zone. Assuming that the unaltered pyroxene in the Upper Zone is representative of the magmatic value, the calculated $\delta^{18}O_{melt}$ value is 6.8 for a temperature of 1200°C. These values are remarkably uniform and considerably heavier than the global average of 5.7 for mantle-derived mafic magmas.

Previous Rb-Sr results.—Several Rb-Sr studies have demonstrated that the Bushveld magmas ($^{87}Sr/^{86}Sr$ = 0.705 to 0.709) were strongly enriched in radiogenic strontium relative to the contemporaneous $^{87}Sr/^{86}Sr$ ratio of the bulk mantle (0.7022 at 2050 Ma), and that there are large stratigraphic variations (fig. 2) in the initial $^{87}Sr/^{86}Sr$ ratio (Hamilton, 1977; Kruger and Marsh, 1982; Sharpe, 1985, 1986; Harmer and Sharpe, 1985; Kruger, Cawthorn, and Walsh, 1987). Two alternative interpretations of the data have been proposed. On the one

hand, the high and variable $(^{87}Sr/^{86}Sr)_0$ ratios have been attributed to derivation of the Bushveld magmas from a heterogeneous mantle source region that was variably enriched in incompatible elements (Hamilton, 1977; Harmer and Sharpe, 1985). On the other hand, the high and variable $(^{87}Sr/^{86}Sr)_0$ ratios have been attributed to crustal contamination of the magmas (Davies and others, 1980; Sparks, 1986). Evidence from radiogenic isotopes is generally not sufficient to discriminate between crustal contamination and heterogeneity of the sub-continental mantle (James, 1981). The oxygen isotopic data can help discriminate between the two possibilities, but we must first evaluate the effects of fractional crystallization on the isotopic ratios.

Fractional Crystallization

Fractional crystallization has very little effect on the oxygen isotopic composition of a gabbroic magma. Plagioclase and pyroxene fractionation have nearly offsetting effects on the value of $\delta^{18}O_{melt}$, and therefore simultaneous fractionation of these minerals tends to buffer the oxygen isotopic composition of a gabbroic magma. Variations in $\delta^{18}O_{melt}$ values that result from changes in the mineralogy of the fractionating assemblage are generally both small and gradual. Data from the Kiglapait intrusion have been used to model variations in the isotopic composition of the melt during closed-system fractional crystallization, and the results indicate that the maximum variation in $\delta^{18}O_{melt}$ values is approx 0.3 (Kalamarides, 1984). An investigation of the effects of fractional crystallization on the oxygen isotopic composition of three idealized mafic and ultramafic magmas in the system olivine–plagioclase–quartz indicates that the maximum variation in $\delta^{18}O_{melt}$ value is on the order of 0.2 (Dunn, 1986).

Fractional crystallization has no measurable effect on the $^{87}Sr/^{86}Sr$ ratio of a magma. Closed-system fractional crystallization would therefore be reflected by cumulates with uniform values of $(^{87}Sr/^{86}Sr)_0$. Stratigraphic variations in the initial $^{87}Sr/^{86}Sr$ ratio of a layered intrusion can be produced by crustal contamination or by influxes of new magma.

The $(^{87}Sr/^{86}Sr)_0$ profile of the Bushveld Complex (fig. 2) has sharp discontinuities between the Critical and Main Zones and between the Main and Upper Zones. The $\delta^{18}O_{melt}$ profile (fig. 3) has an apparent discontinuity between the Critical Zone and the Main Zone. In addition, the $\delta^{18}O_{melt}$ values decrease slightly from 7.0 to 6.6 from the bottom to the top of the Main Zone.

The discontinuities in the isotopic profiles cannot be explained by fractional crystallization or by other internal mechanisms of magmatic differentiation. However, the declining values of $\delta^{18}O_{melt}$ within the Main Zone may reflect the effects of fractional crystallization. Most of the rocks in the Main Zone are essentially bi-mineralic, consisting primarily of plagioclase and pyroxene. Fractional crystallization would

produce a small decrease in $\delta^{18}O_{melt}$ with increasing stratigraphic height, because plagioclase is more abundant than pyroxene.

Magma Replenishment and Parental Magmas

There is a broad consensus that several major batches of magma were added to the Bushveld magma chamber as it cooled and crystallized, but there is considerable controversy about the origin of the magmas, the chronology of the magma influxes, and the number of parental magmas involved. The stratigraphic variations in the initial $^{87}Sr/^{86}Sr$ ratio of the Bushveld Complex have been exploited as a tool for monitoring additions of new magma to a crystallizing intrusion. Oxygen isotopic data provide additional constraints on the magmatic evolution of the Bushveld Complex.

The first major influx of magma formed the Lower and lower Critical Zones. This magma had ultramafic affinities and was probably picritic in composition. The middle portion of the Critical Zone is characterized by large and irregular variations in $(^{87}Sr/^{86}Sr)_0$ values, and these data along with discontinuities in indices of magmatic differentiation are interpreted as indications of modest influxes of anorthositic or gabbroic magma (von Gruenewaldt and others, 1985).

The Main Zone formed from a very large influx of gabbroic or anorthositic magma, and the origin of the Merensky Reef is probably related to the emplacement of this magma. The replenishment of the magma chamber with an isotopically distinct parental magma is clearly manifested by a large and sharp increase in the $(^{87}Sr/^{86}Sr)_0$ value at the boundary between the Critical and Main Zones (fig. 2) and is further corroborated by a sharp increase in the $\delta^{18}O_{melt}$ value at the same stratigraphic level (fig. 3). The fact that the $\delta^{18}O_{melt}$ values within the Main Zone can be understood in terms of fractional crystallization indicates that there was little or no influx of other magmas during the crystallization of the Main Zone.

The boundary between the Main and Upper Zones is marked by a sharp decrease in $(^{87}Sr/^{86}Sr)_0$ and by reversals in indices of magmatic differentiation. One interpretation of the data is that the Upper Zone formed from a major influx of a third isotopically distinct magma (Krueger and others, 1987). An alternative hypothesis is that the Main Zone represents a liquid layer intruded into the density-stratified magma pile at a level consistent with its density, and that the Upper Zone crystallized from the pre-existing magma that was elevated above the liquid layer that formed the Main Zone (Sharpe, 1985). According to the latter hypothesis, the entire Bushveld Complex formed from just two isotopically distinct parental magmas, and the Upper Zone is petrogenetically related to the upper Critical Zone. Unfortunately, the present oxygen isotopic data cannot distinguish between these possibilities, because we do not have a reliable value of $\delta^{18}O_{melt}$ for the Upper Zone.

Whether there were two or three isotopically distinct parental magmas, any model that explains the origin of the Bushveld magmas must account for the high and variable $(^{87}Sr/^{86}Sr)_0$ values and the very heavy $\delta^{18}O_{melt}$ values. Two possibilities are that the Bushveld magmas were derived from a highly anomalous region of the mantle or that the magmas were contaminated by crustal rocks. Below we consider both possibilities and conclude that the likely process was crustal contamination.

Mantle Heterogeneity

The Bushveld magmas were strongly enriched in radiogenic Sr relative to the contemporaneous bulk mantle. Hamilton (1977) and Harmer and Sharpe (1985) interpret the Sr isotopic data to represent derivation of the Bushveld magmas from a heterogeneous mantle source region variably enriched in incompatible elements. This hypothesis involves selective melting of metasomatically enriched material which constitutes a very small proportion of the mantle source region. The major element characteristics of the magmas would be controlled by the large reservoir of "normal" mantle material, whereas the trace element characteristics (concentration and isotopic composition) would be strongly influenced by the volumetrically insignificant amount of metasomatically enriched material. This process would result in a decoupling between the chemistry of major and trace elements, and it should produce $\delta^{18}O_{melt}$ values typical of mantle-derived mafic magmas.

The oxygen isotopic data indicate that the Bushveld magmas were strongly enriched in ^{18}O relative to the bulk mantle. Mass balance considerations indicate that the high $\delta^{18}O_{melt}$ values of the Bushveld magmas could not be produced by the selective incorporation of a relatively small reservoir of metasomatically enriched mantle material. Thus, either the magmas did not acquire their $\delta^{18}O_{melt}$ signatures in the mantle or the sub-Kaapvaal mantle had anomalous values.

If mantle heterogeneity was a major factor in controlling the oxygen isotopic ratios of the Bushveld magmas, then the bulk mantle beneath a large area of the Kaapvaal craton must be anomalously enriched in ^{18}O. At present there is no independent evidence for large-scale anomalies in the $\delta^{18}O$ value of the sub-Kapvaal mantle. Mantle-derived volcanic rocks in the Transvaal Supergroup (table 4) have oxygen isotope ratios consistent with "normal" $\delta^{18}O_{melt}$ values. Mantle-derived igneous rocks from other parts of the Kaapvaal craton also have "normal" $\delta^{18}O_{melt}$ values (Taylor and Magaritz, 1975; Taylor, 1977; Smith, O'Neil, and Erlank, 1984). For example, the $\delta^{18}O_{melt}$ value of Archaean komatiites from the Barberton Mountain Land is approx 5.7, which is indistinguishable from the average $\delta^{18}O$ value of modern ocean floor basalts. Given the widespread occurrence in the Kaapvaal craton of mantle-derived igneous rocks with "normal" $\delta^{18}O_{melt}$ values, we consider it highly unlikely that the Bushveld magmas inherited their

unusual isotopic signature from an anomalous region of the sub-Kaapvaal mantle. It appears likely, therefore, that the heavy $\delta^{18}O_{melt}$ values that characterize the Bushveld Complex are the result of crustal contamination. The Sr isotopic data can also be explained by crustal contamination, and there is no need to invoke arguments that require unusual characteristics of the mantle to explain the Sr isotopic results.

Evidence for Crustal Contamination

Introduction.—Our understanding of both the petrogenesis of layered intrusions and the nature of the sub-continental mantle are critically dependent upon our knowledge of the extent to which mantle-derived magmas have been affected by crustal contamination. In addition to altering the chemical and isotopic composition of a magma, crustal contamination has been proposed as a mechanism for producing modal layering (Irvine, 1975) and for altering the crystallization sequence of the cumulus phases (Irvine, 1970; Longhi, 1982; Sparks, 1986). In the following sections of this paper we summarize the arguments for crustal contamination of the Bushveld magmas based on oxygen isotopic data.

The following interpretation of the oxygen isotopic data is discussed in detail, because there is considerable controversy concerning the role of crustal contamination of the Bushveld Complex. The interpretation is largely based on four aspects of the data: (1) the $\delta^{18}O$ values of the Bushveld Complex are heavier than "normal" mantle-derived magma values; (2) most of the $\Delta^{18}O_{plag\text{-}opx}$ values are typical of magmatic values; (3) the $\delta^{18}O$ values of the volcanic rocks of the Transvaal Supergroup are typical of "normal" mantle magmas; and (4) sedimentary country rocks and most of the other potential crustal contaminants are enriched in ^{18}O relative to the Bushveld Complex. Considered individually, none of these observations is sufficient to justify the conclusion. However, collectively the four observations provide strong evidence that the Bushveld magmas were contaminated by a significant amount of crustal material. In addition to the stable isotopic evidence, several Rb-Sr studies have demonstrated that the Bushveld magmas were strongly enriched in ^{87}Sr relative to the contemporaneous bulk mantle (Hamilton, 1977; Kruger and Marsh, 1982; Sharpe, 1985; Harmer and Sharpe, 1985; Kruger, Cawthorn, and Walsh, 1987), and this result is consistent with the proposition that the magmas assimilated a significant amount of crustal material.

Heavy $\delta^{18}O$ values of the intrusive rocks.—Numerous oxygen isotope studies have established that the $\delta^{18}O$ values of uncontaminated, mantle-derived basaltic and gabbroic rocks are: $\delta^{18}O_{plag} = 6.0$; $\delta^{18}O_{pyx} = 5.5$; and $\delta^{18}O_{rock} = 5.7$ (Taylor, 1968). The average $\delta^{18}O$ values of the Bushveld Complex ($\delta^{18}O_{plag} = 7.1$; $\delta^{18}O_{opx} = 6.6$; and $\delta^{18}O_{magma} = 6.8$) are appreciably heavier than the values for "normal" mafic igneous rocks (tables 2 and 3). Heavy $\delta^{18}O$ values can be produced by subsolidius exchange with

an external fluid reservoir, by partial melting of a region of the mantle that had anomalously high $\delta^{18}O$ values, or by crustal contamination of the magma.

Small $\Delta^{18}O_{plag\text{-}opx}$ values.—As discussed earlier, the systematics of the oxygen isotopic data for the Lower, Critical, and Main zones do not exhibit any of the characteristics that are diagnostic of subsolidus exchange with hydrothermal fluids under either equilibrium or disequilibrium conditions. Most fresh samples from the Bushveld Complex have $\Delta^{18}O_{plag\text{-}opx}$ values (tables 2 and 3) typical of magmatic values ($\approx$0.5 permil). We interpret this observation to indicate that the $\delta^{18}O_{plag}$ and $\delta^{18}O_{opx}$ values were set at magmatic temperatures (approx 1200°C) and that the oxygen isotopic compositions of plagioclase and pyroxene were not appreciably modified by subsequent subsolidus processes.

Volcanic rocks with 'normal' $\delta^{18}O$ values.—Volcanic rocks from the Transvaal Supergroup have $\delta^{18}O$ values of 5.5 (table 4), typical of "normal" mantle-derived magmas. The occurrence of volcanic rocks with "normal" $\delta^{18}O$ values indicates that the mantle beneath the Kaapvaal craton was not anomalously enriched in ^{18}O. The volcanic rocks with normal $\delta^{18}O$ values are older than the Bushveld Complex, and one could appeal to large temporal or spatial variations in the $\delta^{18}O$ values of the sub-Kaapvaal mantle in order to account for the high $\delta^{18}O$ values of the intrusive rocks. However, these possibilities remain unconvincing due to a lack of independent evidence for large-scale enrichment of the sub-Kaapvaal mantle in ^{18}O. It is, therefore, unlikely that the high $\delta^{18}O_{plag}$ and $\delta^{18}O_{opx}$ values that characterize the Bushveld Complex were inherited from a high-$\delta^{18}O$ source region within the sub-Kaapvaal mantle.

Crustal rocks with heavy $\delta^{18}O$ values.—In order to build a strong case for crustal contamination it is necessary to identify potential contaminants that have heavy $\delta^{18}O$ values. The Bushveld Complex is intruded into a 12 km thick pile of early Proterozoic sedimentary and volcanic rocks which belong to the Transvaal Supergroup. The $\delta^{18}O$ values of the principal sedimentary rock types are 9 to 11 for quartzites, 11 to 15 for pelites, and 19 to 21 for carbonates (table 4; fig. 4). These values are 3 to 15 permil heavier than the $\delta^{18}O$ values of typical mantle-derived mafic magmas (5.7 permil), and therefore assimilation or isotopic exchange with these rock types would increase the $\delta^{18}O$ value of the magma.

The Bushveld magmas may have been affected by crustal contamination during their ascent through the lower and middle crust. Direct measurements of the isotopic composition of the rocks present at lower structural levels than the Transvaal Supergroup are unavailable. However, the $\delta^{18}O$ values of the quartzites may provide an indirect indication of the ^{18}O values of the crystalline basement rocks. Quartz is very resistant to isotopic exchange at low temperatures, and the $\delta^{18}O$ value of a quartzite can be used to infer the $\delta^{18}O$ value of its source rock. The laterally extensive Transvaal quartzites were probably derived from

weathering of granitic basement rocks. The $\delta^{18}O$ values of most quartzites are 10 to 11. These values are typical of quartz derived from granitic source rocks with $\delta^{18}O$ values between 8 and 10.

Timing of Crustal Contamination

Introduction.—The preceding discussion indicates that the high $\delta^{18}O$ values were neither inherited from an anomalous source region in the sub-continental mantle nor superimposed on the intrusion during its post-magmatic history. We conclude that the Bushveld magmas became enriched in ^{18}O as a result of contamination with crustal material. This section evaluates the relative roles of contamination that occurred during magma ascent through the crust and contamination that occurred during the crystallization of the magma chamber.

Local contamination.—The presence in the Bushveld Complex of partially digested xenoliths of local country rocks (Hall, 1932; Willemse, 1969) provides compelling evidence that the magmas were affected by at least a small amount of local contamination as the intrusion cooled and crystallized. The Bushveld magmas were also contaminated with a small amount of anatectic melt derived from migmatized country rocks that are locally present in the contact aureole immediately adjacent to the intrusion (Schiffries and Skinner, 1987). However, there is little field, petrologic, or geochemical evidence indicating that assimilation of local country rocks was a major process in the eastern and western lobes of the Bushveld Complex. Calculations based on AFC models of magma evolution (DePaolo, 1985, eq 6) indicate that the ratio of the assimilation rate to the crystallization rate was approx 10^{-2} during the crystallization of stratigraphic intervals that have a constant $\delta^{18}O_{magma}$ value (Schiffries, ms).

Local contamination played a much greater role in portions of the relatively small Potgietersrus lobe of the Bushveld Complex (Buchanan and others, 1981; Cawthorn, Barton, and Viljoen, 1985). The assimilation of a substantial amount of local siliceous material is indicated by the presence of quartz, biotite, and relatively sodic plagioclase in rocks near the margin of the intrusion. Extensive local contamination is corroborated by Rb-Sr data. The initial $^{87}Sr/^{86}Sr$ ratios are very high and variable (0.705–0.723), and they correlate with the $(^{87}Sr/^{86}Sr)_{2050}$ ratios of the local floor rocks (Barton, Cawthorn, and White, 1986). Sulfur isotopic data indicate a contribution of sedimentary-derived sulfur to the Potgietersrus lobe, and the mineralization of its basal Platreef unit has been largely attributed to local contamination (Buchanan and others, 1981). None of these features is typical of the much larger eastern and western lobes of the Bushveld Complex.

Contamination during magma ascent through continental crust.—Two aspects of the isotopic data indicate that most of the contamination in the eastern and western Bushveld Complex occurred during magma ascent through the continental crust. First, the earliest formed cumu-

lates in each of the major stratigraphic divisions have high $\delta^{18}O$ and $(^{87}Sr/^{86}Sr)_0$ values, indicating that the magmas were already heavily contaminated by the time they were injected into the magma chamber. Secondly, if crystallization of the cumulates was accompanied by large-scale assimilation of local country rocks, then the $\delta^{18}O$ and $(^{87}Sr/^{86}Sr)_0$ values of the cumulates would increase monotonically within the major stratigraphic units of the intrusion. The uniform $(^{87}Sr/^{86}Sr)_0$ values of the Upper Zone (Kruger, Cawthorn, and Walsh, 1987) and the uniform $(^{87}Sr/^{86}Sr)_0$ values (Sharpe, 1985) and uniform or decreasing $\delta^{18}O_{magma}$ values of large portions of the Main Zone are not consistent with appreciable amounts of local contamination during the crystallization of these stratigraphic units.

Numerical simulations of turbulent magma ascent through continental crust indicate that tholeiitic and picritic magmas should become contaminated with 5 to 15 wt percent of crustal material (Huppert and Sparks, 1985). The amount of contamination varies greatly with the rate of magma ascent, the temperature of the magma, and the geothermal gradient. Ultramafic magmas are relatively indiscriminate in the material they assimilate, but basaltic magmas tend to assimilate selectively rocks of low fusion temperature (Huppert and Sparks, 1985). A consequence of this difference is that material from the relatively refractory lower crust probably constitutes a greater proportion of the bulk contaminant of an ultramafic magma than of a mafic magma. The lower crust is commonly depleted in ^{18}O, Rb, and ^{87}Sr in comparison with the upper crust, and therefore the $^{18}O/^{16}O$ and $^{87}Sr/^{86}Sr$ ratios of a highly contaminated ultramafic magma may be lower than the ratios of a less contaminated mafic magma. The parental magmas of the Bushveld Complex may have obtained their distinct isotopic signatures as a result of variations in the nature and amount of the material they assimilated during their ascent through the continental crust.

Constraints on the Amount of Crustal Contamination

Mass balance calculations based on oxygen isotopic data can be used to place constraints on the amount of crustal contamination (James, 1981; Graham and Harmon, 1983). To a first approximation, the $\delta^{18}O$ value of a contaminated magma ($\delta^{18}O_m$) can be expressed as a weighted average of the mean $\delta^{18}O$ value of the uncontaminated magma ($\delta^{18}O_{m_o}$) and the mean $\delta^{18}O$ value of the assimilants ($\delta^{18}O_a$):

$$\delta^{18}O_m = (X_{m_o})\,\delta^{18}O_{m_o} + (X_a)\,\delta^{18}O_a$$

where X_a is the mole fraction of oxygen contributed by the crustal contaminants, and $X_{m_o} = (1 - X_a)$ is the mole fraction of oxygen contributed by the uncontaminated magma. The concentration of oxygen in the assimilated rocks is approximately equal to the concentration of oxygen in the magma, and the mineral-melt distribution coefficient for oxygen is assumed to be 1.0. The $\delta^{18}O$ value of the uncontami-

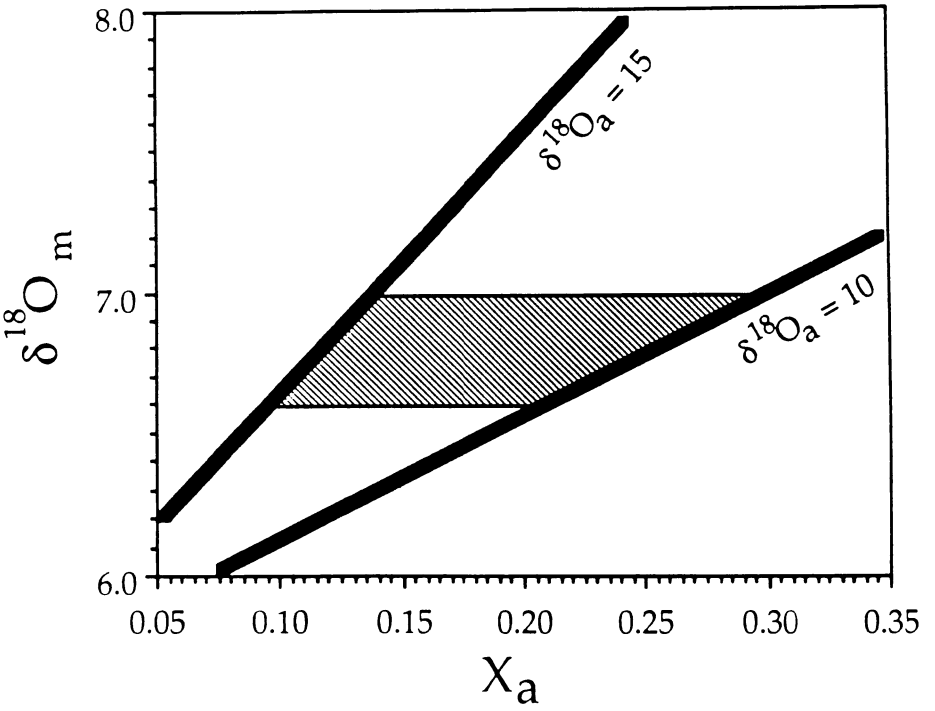


Fig. 6. The two lines in this figure are solutions to the mass balance equation for oxygen isotopes at different values of $\delta^{18}O_a$. The $\delta^{18}O$ value of the uncontaminated magma is assumed to be 5.7, and the calculated average $\delta^{18}O$ value of the contaminated magmas is approx 6.8 ± 0.2 (table 3). The shaded region represents the range of possible combinations of $\delta^{18}O_a$ and $\delta^{18}O_m$ that satisfy the mass balance constraints. If the assimilated material had an average $\delta^{18}O$ value of 15 (a typical value for shales or for a composite of crustal rock types), then 10 to 14 percent of the oxygen in the intrusion was derived from crustal sources. If the assimilated material had an average $\delta^{18}O$ value of 10 then 20 to 29 percent of the oxygen was derived from crustal sources.

nated magma is assumed to be 5.7, and we estimate that the mean $\delta^{18}O$ value of the Bushveld magmas was approx 6.8 (table 3). Figure 6 shows the range of possible combinations of $\delta^{18}O_a$ and $\delta^{18}O_m$ that satisfy the mass balance constraints. If the contaminants had an average $\delta^{18}O$ value of 15 (a typical value for shales or for a composite of crustal rocks from the Transvaal Supergroup and the Kaapvaal craton), then between 10 and 14 percent of the oxygen in the intrusion was derived from crustal sources. These values are certainly within the expected range if the magmas experienced turbulent flow during their ascent through the crust (Huppert and Sparks, 1985). Contamination by granitic rocks with a $\delta^{18}O$ value of 10 would require that between 20 and 29 percent of the oxygen in the intrusion was derived from crustal sources.

CONSEQUENCES OF CONTAMINATION

Crystallization Sequence of the Cumulus Minerals

The contamination of a mafic magma with a modest amount of crustal material can have a profound effect on the crystallization sequence of the cumulus phases (Irvine, 1970, 1975, 1979; Longhi, 1982; Longhi, Wooden, and Coppinger, 1983; Sparks, 1986). Average continental crust is enriched in SiO_2 and alkalis relative to mafic magmas. One consequence of increasing the SiO_2 content of a mafic magma is to advance the crystallization of orthopyroxene relative to clinopyroxene. A simple example is illustrated in figure 7, which is a psuedo-ternary liquidus diagram for the system forsterite–clinopyroxene–plagioclase–quartz projected from plagioclase (after Irvine, 1979). The bulk composition of a typical uncontaminated basaltic magma is represented by point B. The addition of approx 10 wt percent silica causes the bulk composition of the contaminated magma to shift to point C. Although the bulk compositions of the contaminated and uncontaminated magma are not dramatically different, their fractional crystallization paths begin to diverge at an early stage in their evolution. Fractional

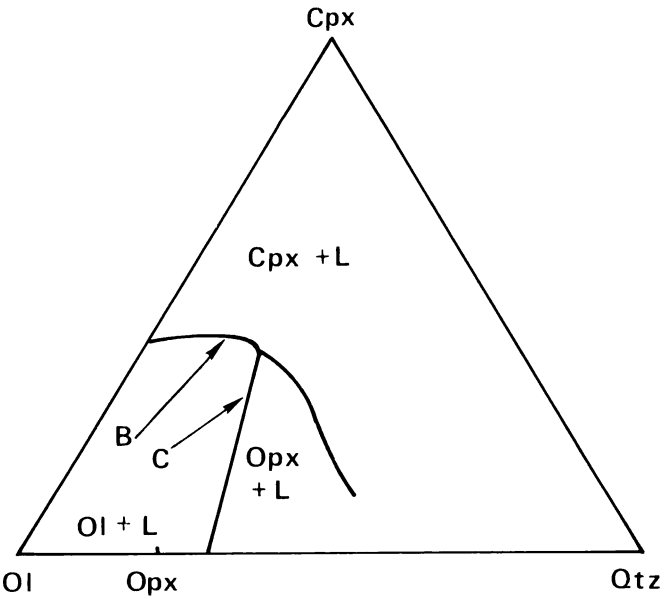


Fig. 7. A portion of the pseudo-ternary liquidus phase diagram for the system olivine–clinopyroxene–plagioclase–quartz projected from plagioclase (after Irvine, 1979). Fractional crystallization of a magma with a composition designated by point B will result in the precipitation of clinopyroxene before orthopyroxene. A reversal of this sequence can be achieved if the bulk composition of the magma shifts toward point C due to the assimilation of quartz-rich crustal material.

crystallization of magma B begins with the precipitation of olivine and then olivine + clinopyroxene, whereas fractional crystallization of magma C begins with the precipitation of olivine and then orthopyroxene. The fact that the latter sequence occurs in the Bushveld Complex is consistent with the contamination model for the intrusion.

Origins of Igneous Layering

Introduction.—Numerous models have been proposed to explain the origins of igneous layering. To a first approximation, they can be divided into two groups. The first group relies on open-system processes, such as crustal contamination and magma replenishment. The second group is based on internal processes, such as crystal settling, stagnant bottom layers, and double-diffusive convection. There is little doubt that both types of processes operate in layered intrusions. In the following paragraphs we discuss the possibility that different types of processes may have been responsible for the origin of layering in different parts of the Bushveld Complex.

Open-system processes.—The addition of new material to a crystallizing magma chamber can result in the formation of igneous layering (Irvine, 1975, 1977). New material may be introduced as a result of crustal contamination or magma replenishment. Portions of the Critical Zone are characterized by large and irregular variations in $(^{87}Sr/^{86}Sr)_0$ ratio, and these data along with reversals in cryptic zoning and other indices of differentiation are interpreted as indications of influxes of new magma (von Gruenewaldt and others, 1985). The Critical Zone is strongly layered, containing numerous anorthosite and chromitite layers. It is widely accepted that replenishment of the magma chamber played a key role in the development of this layering, and it is also possible that platinum mineralization is related to influxes of new magma (Irvine, 1977: Campbell, Naldrett, and Barnes, 1983).

Internal processes.—Although the addition of new material remains an attractive mechanism for producing layering in the Critical Zone, open-system processes have less appeal for explaining the origin of layering in stratigraphic units that are isotopically homogeneous. The Upper Zone and large portions of the Main Zone of the eastern Bushveld Complex have uniform $\delta^{18}O_{magma}$ and $(^{87}Sr/^{86}Sr)_0$ values. An appreciable quantity of new material could not have been added to the magma chamber during the crystallization of these cumulates, unless the material added had the same $^{87}Sr/^{86}Sr$ and $^{18}O/^{16}O$ ratios as the resident magma. This coincidence is highly unlikely, because the resident magma was a mixture of several magmas and crustal components, and the $^{87}Sr/^{86}Sr$ and $^{18}O/^{16}O$ ratios of the resident magma were generally different from those of any individual component. An implication of this argument is that there were no major additions of new magma or crustal material to the resident magma during the crystallization of the isotopically homogeneous intervals. If this conclusion is correct, then it follows that the modal and cryptic layerings in these

intervals were probably produced by internal processes of magmatic differentiation, rather than by open-system processes such as magma replenishment and crustal contamination.

Interpretation of $^{187}Os/^{186}Os$ Isotopic Data

The Re-Os isotopic system is a potentially powerful tracer of crust-mantle interactions. The $^{187}Os/^{186}Os$ ratio of a laurite ($Ru(Os)S_2$) sample from the Merensky Reef has been measured by Allègre and Luck (1980). Figure 8 shows that the Bushveld sample is strongly enriched in radiogenic Os ($^{187}Os/^{186}Os \approx 1.45$) relative to the mantle evolution curve ($^{187}Os/^{186}Os \approx 0.91$ at 2050 Ma). Allègre and Luck (1980) suggest

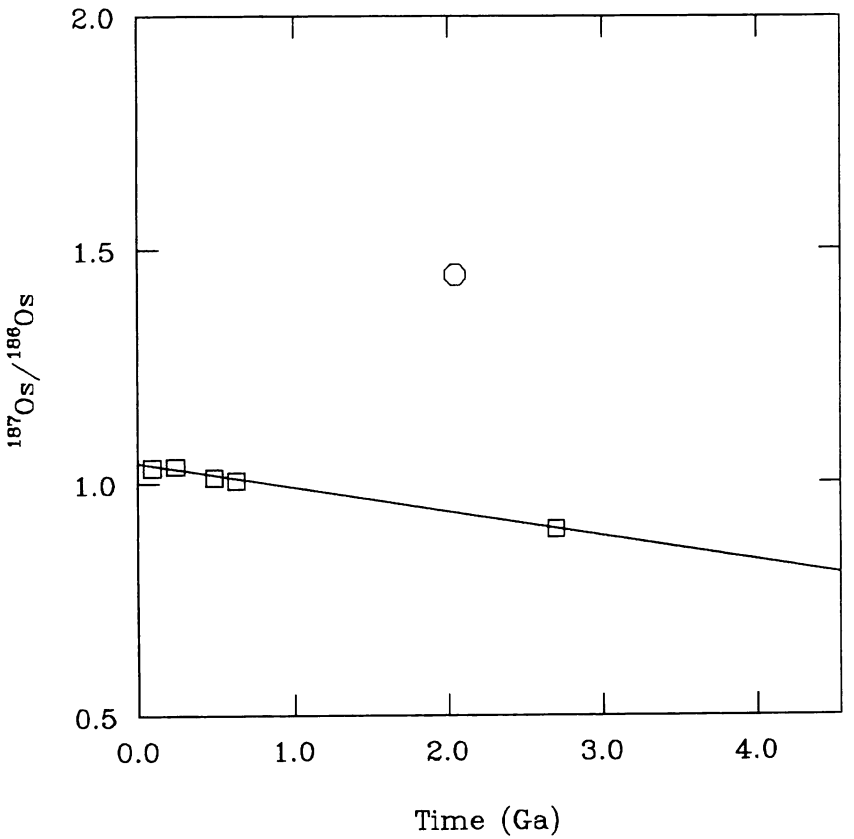


Fig. 8. Mantle evolution curve for $^{187}Os/^{186}Os$ assuming that $\lambda_{Re} = 1.61 \times 10^{-11}$ yr^{-1}, $^{187}Re/^{186}Os = 3.15$, and $(^{187}Os/^{186}Os)_0 = 0.805$. The data points (squares) that plot on the mantle evolution curve are for osmiridium samples from placer deposits of known age. A sample of laurite ($Ru(Os)S_2$) from the Bushveld Complex (octagon) has a much higher $^{187}Os/^{186}Os$ ratio than the contemporaneous bulk mantle. Data from Allègre and Luck (1980).

that the presence of highly radiogenic Os in the Bushveld Complex indicates that the magmas may have been derived from a strongly enriched region of the subcontinental mantle. The possibility that crustal contamination was responsible for the high $^{187}Os/^{186}Os$ ratio can be discounted if the Merensky Reef crystallized from an ultramafic magma that was strongly enriched in osmium relative to the contaminants. However, there is no reason to assume that the Merensky Reef crystallized from an Os-rich ultramafic magma. Petrologic and geochemical studies of marginal rocks indicate that the upper Critical Zone, which contains the Merensky Reef, crystallized from a magma that was not enriched in platinum-group elements relative to average continental flood basalts (Davies and Tredoux, 1985; Naldrett and others, 1987). Average tholeiites have a lower Os concentration and a lower Re/Os ratio than many types of crustal rocks that may have been assimilated by the Bushveld magmas. For example, average granite has a slightly higher Os concentration and a much higher Re/Os ratio than average tholeiite (Allègre and Luck, 1980). Certain black shales have very high Os concentrations and extremely high Re/Os ratios relative to average tholeiites (Ravizza and Turekian, 1987). Contamination of the Bushveld magmas with a modest amount of granite or shale could account for the high $^{187}Os/^{186}Os$ ratio of the Bushveld laurite sample. For example, if the magma had the Re-Os characteristics of a typical continental tholeiite (Os = 0.05 ppb, and $(^{187}Os/^{186}Os)_{2050} = 1.0$) and the bulk contaminant had Re-Os characteristics of a typical granite (Os = 0.06 ppb, and $(^{187}Os/^{186}Os)_{2050} = 10$), then contamination of the magma with only 5 wt percent granite would increase the $^{187}Os/^{186}Os$ ratio of the magma to the measured value of 1.45.

If crustal contamination is responsible for the presence of extremely radiogenic osmium in laurite from the Merensky Reef, then it is possible that an appreciable fraction of the platinum-group elements in the Bushveld Complex were derived from crustal sources.

CONCLUSIONS

We report oxygen isotopic data for unaltered mineral separates from the Bushveld Complex. These data place new constraints on the relative importance of several major petrological processes that controlled the magmatic evolution of the Bushveld Complex. The main conclusions of this study are summarized below.

1. The Bushveld magmas were strongly enriched in ^{18}O, ^{87}Sr, and ^{187}Os relative to the contemporaneous bulk mantle.

2. The high $\delta^{18}O$ values that characterize the Bushveld Complex are not the result of subsolidus exchange with hydrothermal fluids. The systematics of the $\delta^{18}O_{plag}$ and $\delta^{18}O_{pyx}$ data for the Lower, Critical, and Main Zones provide strong evidence that the samples have retained their magmatic isotopic signature, and that they have not been appreciably affected by subsolidus exchange.

3. The fractionation of oxygen isotopes between coexisting plagioclase and pyroxene indicates that the initial crystallization temperature was greater than 1200°C, which is in agreement with the results of independent temperature estimates.

4. The occurrence in the Transvaal Supergroup and elsewhere in the Kaapvaal craton of mantle-derived volcanic rocks with "normal" $\delta^{18}O$ values indicates that the sub-continental mantle was not anomalously enriched in ^{18}O relative to the bulk mantle. It is, therefore, very unlikely that the high $^{18}O/^{16}O$ and $(^{87}Sr/^{86}Sr)_0$ ratios of the Bushveld magmas were inherited from a heterogeneously enriched source region in the sub-continental mantle.

5. Mass balance calculations indicate that at least 10 percent of oxygen in the Bushveld Complex was derived from crustal sources.

6. The parental magmas probably acquired their differing isotopic signatures as a result of variations in the nature and amount of material they assimilated during their ascent through the continental crust.

7. The magmas probably experienced turbulent flow during their ascent through the crust.

8. In addition to its chemical and isotopic effects, crustal contamination may have had a profound influence on the crystallization sequence of cumulus minerals in the Bushveld Complex. The crystallization of orthopyroxene before clinopyroxene may be due to the contamination of a mantle-derived magma with siliceous crustal material.

9. The isotopic homogeneity of certain thick stratigraphic units in the Bushveld Complex indicates that neither assimilation of local country rocks nor replenishment of the magma chamber played a major role during the crystallization of these units. The ratio of the assimilation rate to the crystallization rate was approx 0.01 during the crystallization of isotopically homogeneous units.

10. The origin of igneous layering remains a major problem in geology. It appears that different mechanisms may be responsible for igneous layering in different parts of the Bushveld Complex.

11. Crustal contamination can explain the origin of extremely radiogenic osmium in laurite from the Merensky Reef, and it is possible that an appreciable fraction of the osmium and other platinum-group elements in the Bushveld Complex may be derived from crustal sources.

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