

**ECONOMIC GEOLOGY  
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University of the Witwatersrand  
Johannesburg

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**METAMORPHISM IN THE WITWATERSRAND BASIN:  
A REVIEW OF RECENT ADVANCES AND SOME  
PREDICTIONS ON GOLD REMOBILIZATION  
BY METAMORPHIC FLUID FLOW**

**G. STEVENS, R.H. BOER and R.L. GIBSON**

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METAMORPHIC FLUID FLOW**

by

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**ABSTRACT**

Recent advances in the understanding of metamorphism of the Witwatersrand Basin, particularly in the previously deeply buried central portions now exposed in the Vredefort Dome, allow for the generation of a Basin-wide metamorphic model. Low-pressure, high-temperature peak metamorphic conditions were attained during the crustal thermal perturbation associated with the Bushveld Event. This study focuses on metamorphic fluid production from West Rand Group shales during this event. Phase proportions of three typical West Rand Group shale bulk compositions were calculated between 250° and 600°C. The post-diagenetic assemblage appears to be characterised by a low clay-mineral content. Thus, the first major metamorphic fluid producing reaction will be the breakdown of muscovite and chlorite to produce biotite. In an average West Rand Group shale composition, where peak metamorphic temperatures were high enough for this model reaction to have run to completion (~450°C), 70 litres of hydrous fluid would have been produced per cubic metre of rock. Further breakdown of hydrous minerals in shales of this composition, to produce the peak metamorphic assemblage recorded in the collar of the Vredefort Dome (~600°C), would have liberated an additional 69 litres of hydrous metamorphic fluid per cubic metre of rock. These calculations, coupled to the peak metamorphic thermal profile through the Basin and the known suboutcrop limits of the West Rand Group suggest that the Witwatersrand Basin lost at least  $7.58 \times 10^{14}$  litres of metamorphic fluid. Fluid inclusion studies in gold-bearing reef horizons of the Central Rand Group near the Basin margins indicate that these horizons experienced fluid flow close to the ~350°C metamorphic peak. The fluids were predominantly aqueous and were likely derived from the underlying West Rand Group as the dominantly quartzitic Central Rand Group has a low metamorphic fluid generation potential. This indicates substantial stratabound flow of metamorphic fluids from the central portions of the depository towards the margins. Calculated gold solubility in the measured fluid inclusion compositions is between 1 and 10 parts per billion. Thus, metamorphic fluids derived from within the lower portions of the Witwatersrand Supergroup had the potential to mobilise at least 7600 tons of gold. In this paper it is proposed that the metamorphic fluid scavenged gold from deep-seated Central Rand Group reefs where peak metamorphic temperatures were highest. This gold was transported towards the cooler Basin margins where deposition in response

to declining temperatures enriched existing gold grades. Ultimately, the fluid must have migrated out of the Basin. The magnitude of the fluid volume, and its calculated gold mobilizing potential, indicate that this fluid-flow event would have had the potential to generate major gold deposits on the Kaapvaal Craton outside the boundaries of the Witwatersrand Basin.

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# METAMORPHISM IN THE WITWATERSRAND BASIN: A REVIEW OF RECENT ADVANCES AND SOME PREDICTIONS ON GOLD REMOBILIZATION BY METAMORPHIC FLUID FLOW

## INTRODUCTION

The increasing body of petrographic evidence that has accumulated in recent years in support of a non-detrital origin for a significant proportion of the gold in the Witwatersrand Basin has led to considerable interest in the postdepositional thermal/metamorphic and fluid history of the basin (e.g., Frimmel, 1994; Phillips & Law, 1994). This interest has, of necessity, been concentrated within the ore-bearing units in the goldfields located around the margin of the Basin and has led to reasonably good constraints being placed on the local conditions of metamorphism, and to some knowledge of fluid compositions (e.g. Frimmel, 1994; Phillips & Law, 1994). However, there remains considerable debate about the ultimate timing and causes of both the metamorphic and fluid-flow events. Recently, Gibson & Wallmach (1995) and Stevens *et al.* (1996) argued that the low-grade metamorphism in the goldfields was the consequence of a more widespread thermal event that effected much of the Kaapvaal Craton. This understanding of metamorphism in the Witwatersrand Basin stems from new data on the metamorphism of the deepest level Witwatersrand rocks and their associated basement exposed within the Vredefort Dome. The amphibolite to granulite facies metamorphic assemblages preserved in these rocks have previously been ascribed to local heat sources, inferred to be associated with the formation of the Dome. However, the recent work referred to above has documented evidence for the peak metamorphic assemblages, as well as the retrogression of these assemblages predating the formation of the Dome. These peak metamorphic assemblages are characterised by high geothermal gradients and are correlated with the Bushveld magmatic event.

This paper reviews data on the metamorphism of the Witwatersrand Basin, with an emphasis on the new findings from rocks exposed in the Vredefort Dome, and then describes a simple modelling exercise aimed at placing some constraints on the volume of metamorphic fluid evolved from within the deep levels of the Witwatersrand Basin during the peak metamorphic event. Lastly, estimates are made to establish the potential of this fluid, largely derived from within the West Rand Group, to interact with the auriferous reefs of the Central Rand Group, and to remobilize gold derived from these reef horizons.

## GEOLOGICAL SETTING

The rocks of the Witwatersrand Supergroup form one of a succession of late Archaean to Proterozoic supracrustal sequences deposited on the Archaean granite-greenstone basement of the Kaapvaal Craton. Deposition commenced with the extrusion of the subaerial mafic volcanic rocks of the Dominion Group at 3.07 Ga (Armstrong *et al.*, 1991). This was followed by the accumulation of up to 8 km of siliciclastic sediments belonging to the Witwatersrand Supergroup (Fig. 1) - a lower argillaceous-arenaceous sub-tidal marine sequence (West Rand Group), and an upper, arenaceous-rudaceous sequence (Central Rand Group) of fluvial/alluvial fan origin. These rocks are overlain by up to 3 km of subaerial mafic lavas and subsidiary rift-related sediments of the ca. 2.7 Ga (Armstrong *et al.*, 1991) Ventersdorp Supergroup, followed, in turn, by the 2.6-2.2 Ga (Walraven & Martini, 1995) Transvaal Supergroup, comprising a 1-2 km thick lower dolomitic sequence (Chuniespoort Group) and an upper 3-4 km thick argillaceous-arenaceous sequence (Pretoria Group).

Following the termination of Transvaal Supergroup sedimentation and an erosional hiatus, the craton experienced widespread magmatism (the Bushveld Event) which led to the eruption of a several kilometre thick sequence of predominantly felsic volcanic rocks (the Rooiberg Group, at 2.06 Ga - Walraven, in press) that broadly coincided with the intrusion of up to 6 km of mafic-ultramafic magmas into the upper levels of the Transvaal Supergroup. The final stages of this event, at 2.054 Ga (Walraven *et al.*, 1990; Walraven & Hattingh, 1993), resulted in voluminous granites of the Lebowa Granite Suite intruding the mafic-ultramafic rocks. The magmatic rocks are exposed mainly within the Bushveld Complex to the north of the Witwatersrand Basin, but correlatives are found in the Losberg Complex (Coetzee & Kruger, 1989), and in the Molopo Farms Complex. Coeval magmatism also occurred in the Phalaborwa and Schiel complexes, indicating that this event influenced much of the craton. Small peralkaline granitic and dioritic intrusions into the Witwatersrand, Ventersdorp and Transvaal Supergroups, found in the central parts of the Witwatersrand Basin in and around the Vredefort Dome, also appear to be close to Bushveld in age (Moser, 1996). Later, Phanerozoic sediments, lavas and intrusions of the Karoo Supergroup were unconformably deposited over the Bushveld Complex and the Transvaal Supergroup in the south and east of the Kaapvaal Craton.

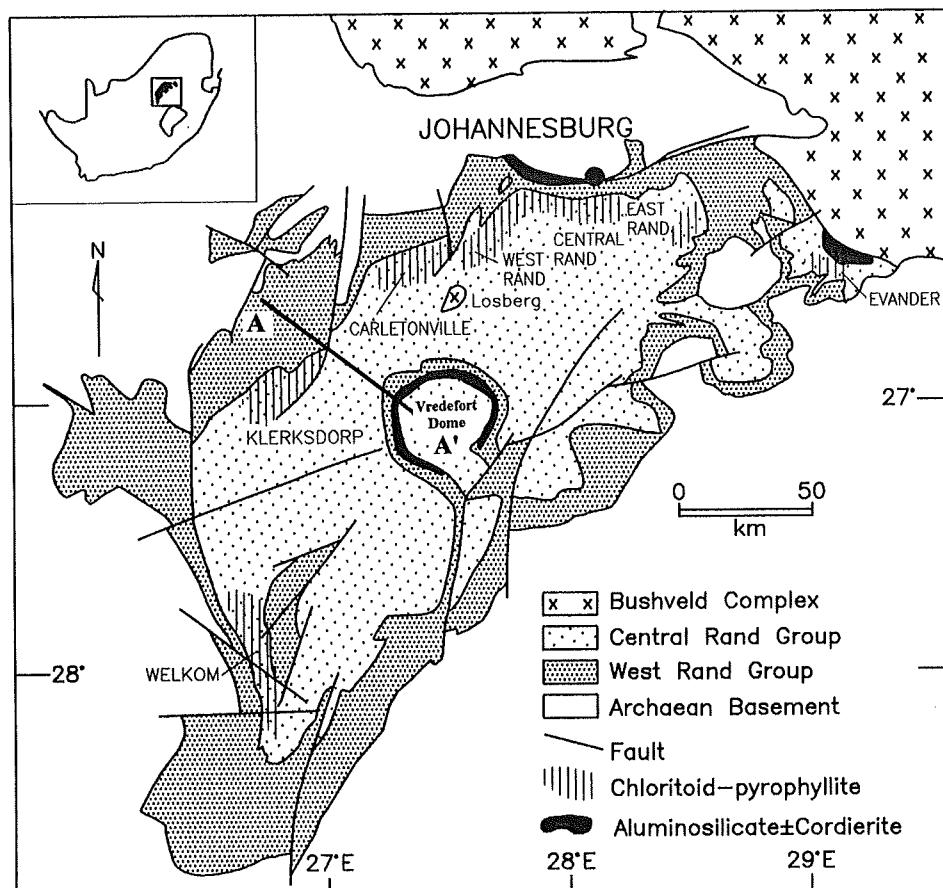


Figure 1: Simplified geology of the Witwatersrand Basin showing the suboutcrop distribution of the West Rand and Central Rand Groups. An indication of metamorphic grades is also included. Line A to A' indicates the position of the section presented in Figure 3.

Shortly after the emplacement of the Bushveld Complex, the Witwatersrand Basin and overlying sequences were subjected to widespread deformation associated with the formation of the Vredefort Dome (McCarthy *et al.*, 1986). The presence, in the rocks of the Dome, of unusual shock deformation features including shatter cones, microdeformation features, coesite and stishovite (see review in Reimold & Gibson, 1996) has led to widespread acceptance that the Dome represents the deeply eroded roots of a large meteorite impact crater, recently dated at 2.02 Ga (Kamo *et al.*, 1996). The geographic coincidence between the Dome and the Witwatersrand Basin is attributed to the effects of syn-doming downwarping of the Witwatersrand strata in the Potchefstroom Synclinorium (McCarthy *et al.*, 1990). In other words the Witwatersrand Basin, as seen today, is a structural basin. McCarthy *et al.* (1990) further suggested that regional tilting of the craton occurred at some time

after the formation of the Dome, leading to variable amounts of exhumation of the rocks in the Witwatersrand Basin (up to 7 km in the vicinity of the Vredefort Dome, but decreasing northwards).

The Vredefort Dome exposes within its core the pre-Dominion Archaean basement. Surrounding this core is a collar of steeply dipping to overturned strata exhibiting the full stratigraphic sequence observed in the region (Dominion Group and Witwatersrand, Ventersdorp and Transvaal Supergroups) (Fig. 2). The collar is between 15 and 20 km wide, but estimates of the thickness of the supracrustal sequence suggest that deformation, probably related to the formation of the Vredefort Dome, thickened the stratigraphic sequence. Estimated thicknesses for the different units are: 5-6 km for the Witwatersrand Supergroup, < 2 km for the Ventersdorp Supergroup (P. Linton, pers. comm., 1995) and < 4.5 km for the Transvaal Supergroup (P. Erikson, pers. comm., 1995). The rocks of the core and collar are intruded by numerous mafic sills and dykes, the bulk of which are of Ventersdorp age (Pybus *et al.*, 1995) and the remainder of Bushveld Complex age (Bisschoff, 1972).

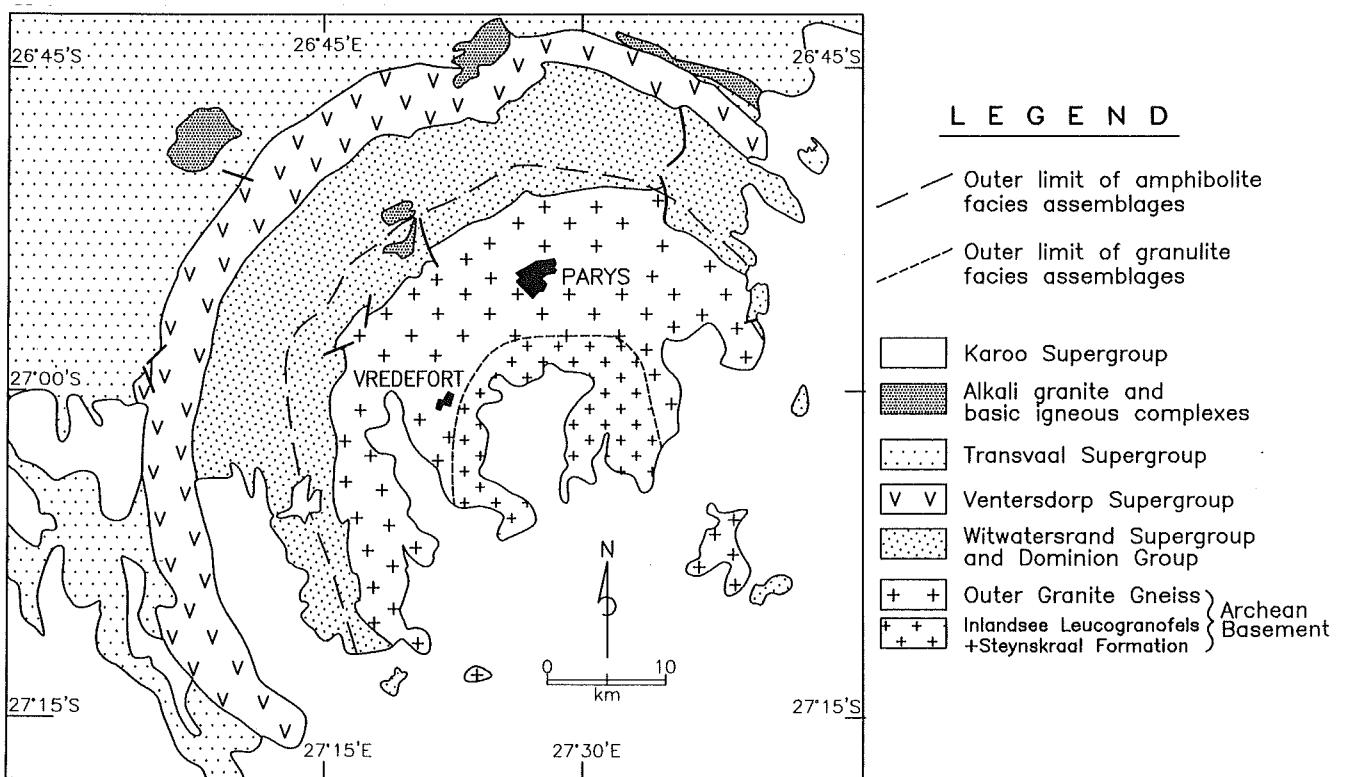


Figure 2: The geology of the Vredefort Dome (after Gibson and Walmach, 1995).

## METAMORPHISM IN THE WITWATERSRAND BASIN - A REVIEW

Strata of the Witwatersrand Supergroup are currently accessible in five main geographic areas in the Witwatersrand Basin. The most extensive surface and underground exposures occur along the northern and northwestern margins of the Basin around Springs, Johannesburg and Carletonville (East Rand, Central Rand, West Rand and Carletonville Goldfields and Klerksdorp Goldfield) (Fig. 1). In the Evander and Welkom Goldfields only underground exposures are available. A large area of surface exposure occurs in the centre of the Basin due to upturning of the strata in the Vredefort Dome. Reports of metamorphic mineral assemblages in the largely conglomerate "reef packages" in the goldfields date from as early as Young (1917), and in the Vredefort Dome from even earlier (Molengraaf, 1903). However, it was only following the work of Phillips (1987) that a systematic approach was adopted to address the question of metamorphism in all the goldfields. Phillips showed that all the goldfields experienced a similar, lower greenschist facies, grade of metamorphism. In the Vredefort Dome, metamorphic grade increases radially inwards from lower greenschist facies to mid-amphibolite facies in the collar rocks, up to granulite facies in the core. Phillips & Law (1994) stated that no conclusive links could be established between the metamorphism in the goldfields and that observed in the Vredefort Dome. Furthermore, they concluded that the relationship between the greenschist, amphibolite and granulite facies metamorphism in the Vredefort Dome was also not well understood. Since then, however, significant strides have been made towards understanding the metamorphic features of all rocks in the Witwatersrand Basin and towards developing a cogent metamorphic evolutionary model.

### **Metamorphism in and Adjacent to the Goldfields**

Phillips (1987) established the existence of a metamorphic assemblage involving pyrophyllite, chloritoid, muscovite, chlorite and quartz in pelitic horizons in all the goldfields. From this, he concluded that the rocks in the goldfields were affected by a regional-scale lower greenschist facies metamorphic event. Subsequent studies have confirmed Phillips' estimates of pressure-temperature conditions of  $\sim 350 \pm 50$  °C, 2-3 kbar for the metamorphic peak (e.g. Wallmach & Meyer, 1990; Frimmel, 1994; Zhou *et al.*, 1994). In a few areas around the goldfields, slightly higher-grade assemblages involving kyanite, andalusite, cordierite and/or biotite have been reported in the Witwatersrand Supergroup (e.g., Schreyer & Bisschoff, 1982; Tweedie, 1986; Phillips, 1987). Only

a few occurrences of biotite have been recorded in the goldfields (Phillips & Law, 1994). In the Evander goldfield, Tweedie (1986) observed a progressive increase in metamorphic grade towards the northeast and interpreted it as a contact effect adjacent to a Bushveld-age pluton. Schreyer & Bisschoff (1982) speculated that the kyanite/andalusite metapelites observed along the northern margin of the Basin are also related to heating effects emanating from the Bushveld Complex. McCarthy *et al.* (1986) noted what they interpreted to be andalusite in shales from both the Ventersdorp and lower Transvaal Supergroups overlying the goldfields north of Johannesburg. In the upper parts of the Transvaal Supergroup (Pretoria Group), the metamorphic sequence is inverted and metamorphic grade increases to mid- to upper amphibolite facies beneath the contact with the Bushveld Complex.

### **Metamorphism in the Vredefort Dome**

In the central parts of the Witwatersrand Basin, metamorphic grade increases broadly with stratigraphic age towards the centre of the Vredefort Dome and metamorphic grades are significantly higher than in the corresponding stratigraphic horizons in the goldfields: biotite occurs in association with chlorite and muscovite in the Booysens Formation shales, and kyanite and andalusite have also been reported from Central Rand Group units. In the lower West Rand Group, porphyroblastic cordierite, andalusite, biotite, garnet and/or staurolite metapelites indicate the attainment of mid-amphibolite facies conditions (Hall & Molengraaff, 1925; Nel, 1927; Bisschoff, 1982; Gibson & Wallmach, 1995). Gibson & Wallmach (1995) estimated peak metamorphic temperature of 570-600 °C at 4.0-4.5 kbar. In the core of the Dome, garnet-, cordierite- and/or orthopyroxene-bearing metapelites, with or without migmatitic leucosomes, indicate the attainment of granulite facies conditions (850 °C to >920 °C, ca. 5.0 kbar; Stevens *et al.*, 1996). Until recently this metamorphism was interpreted as a localised effect associated with the formation of the Dome, and its relationship to the metamorphism observed in the wider Basin has proved problematic (see, e.g. Phillips & Law, 1994; Frimmel, 1994). However, field evidence indicates that the peak metamorphic assemblages in both the collar and the core predate the shock deformation linked to the formation of the Dome (Bisschoff, 1982; Schreyer, 1983; Gibson & Wallmach, 1995; Stevens *et al.*, 1996). Some authors have favoured different ages for the metamorphism in the collar supracrustals and the basement rocks (e.g., Bisschoff, 1982; Hart *et al.*, 1991). However, studies by Gibson & Wallmach (1995) and Stevens *et al.* (1996) have shown (a) similar, anticlockwise, pressure-temperature path geometries

for the collar and core metamorphism, and (b) strong circumstantial evidence that both the medium- and high-grade metamorphism occurred within only a few tens of millions of years of the 2.02 Ga Vredefort Event. Single-zircon U-Pb SHRIMP data from an anatetic melt body in the granulite facies core terrane (Gibson *et al.*, in press) corroborate this timing. The extreme geothermal gradients necessary to effect the amphibolite and granulite facies metamorphism (40-50 °C/km) in the Vredefort rocks, together with the pressure-temperature path and geochronological data, are most consistent with a Bushveld Complex timing for the metamorphism. The medium- to high-grade Vredefort metamorphism is, thus, interpreted as a consequence of intraplating of mantle-derived mafic-ultramafic magmas into lower to mid- crustal levels in the Kaapvaal Craton broadly contemporaneously with the intrusion of similar magmas at shallow levels to form the Bushveld Complex. This intraplating also resulted in the voluminous silicic crustal melts that constitute the Rooiberg Group and the Lebowa Granite Suite.

### **Origin and Timing of the Metamorphism in the Goldfields**

Given the above scenario for the medium- to high- grade metamorphism in the mid-crustal levels exposed in the Vredefort Dome, it is logical to expect that shallower crustal levels would have experienced lower-grade metamorphism characterised by an abnormally high geothermal gradient during the Bushveld Event. Layer *et al.* (1988) concluded that the main metamorphic event in the Witwatersrand goldfields occurred at this time based on the resetting of the remnant magnetism in shales throughout the Witwatersrand Basin to a ca. 2.0 Ga palaeopole orientation, and on K-Ar ages of ca. 1.95 Ga from Witwatersrand shales. Although Phillips & Law (1994) recognised that the most likely cause of the metamorphism in the goldfields was the emplacement of a large mafic body 'near the base of the crust' (p. 26), they favoured a timing for the peak event at ca. 2.3 Ga, based on geochronological data. Frimmel (1994) favoured an even older age, ca. 2.5 Ga, for the metamorphic peak, also based on geochronological data, although he recognised a significant retrogressive event associated with the Bushveld Event. In contrast, Robb & Meyer (1995) argued that the Basin was characterised by temperatures less than 250°C until at least 2.3 Ga, based on hydrocarbon maturation studies. While we recognise that other thermal events may have affected the Witwatersrand Basin prior to the Bushveld Event, the fact remains that the Bushveld Event is the single most important magmato-thermal event to affect the Kaapvaal Craton since deposition of the Witwatersrand Supergroup. This is borne out primarily by the evidence of voluminous crustally-derived felsic melts

associated with the mafic rocks, which indicate that a significant proportion of the lower to middle crust experienced metamorphic conditions conducive to high temperature "dry" anatexis. Figures 3 and 4 summarize our view of the thermal structure of the Kaapvaal crust immediately prior to and during the Bushveld Event, as well as the current distribution of metamorphic facies, following the formation of the Vredefort Dome and erosion to the present level. The remainder of this paper builds on this new perspective on metamorphism in the Witwatersrand Basin in an effort to calculate how much fluid was produced as the result of prograde metamorphism, and what the potential of this fluid was to remobilise existing gold concentrations in the Basin.

## **FLUID FLOW IN THE WITWATERSRAND BASIN**

### **Diagenetic Fluids**

Diagenesis of the Witwatersrand sediments would have involved substantial amounts of fluid flow upwards and towards the basin margins. These fluids were derived from interstitial pore waters liberated by mechanical compaction, as well as from water bound within the crystal structure of clay minerals and liberated during low temperature structural transformation of the clays. The likely characteristics of this fluid have been comprehensively reviewed by Phillips *et al.* (1990), who proposed that the entire depository lost an amount of interstitial water equivalent to 20% of the rock volume during diagenesis. After this, the only water available in the Basin was bound in mineral structures in the clay-, chlorite- and mica-bearing horizons. Phillips *et al.* (1990) also proposed that the shale horizons of the Basin lost approximately 5 vol% of this bound water during diagenesis.

### **Metamorphic Fluids**

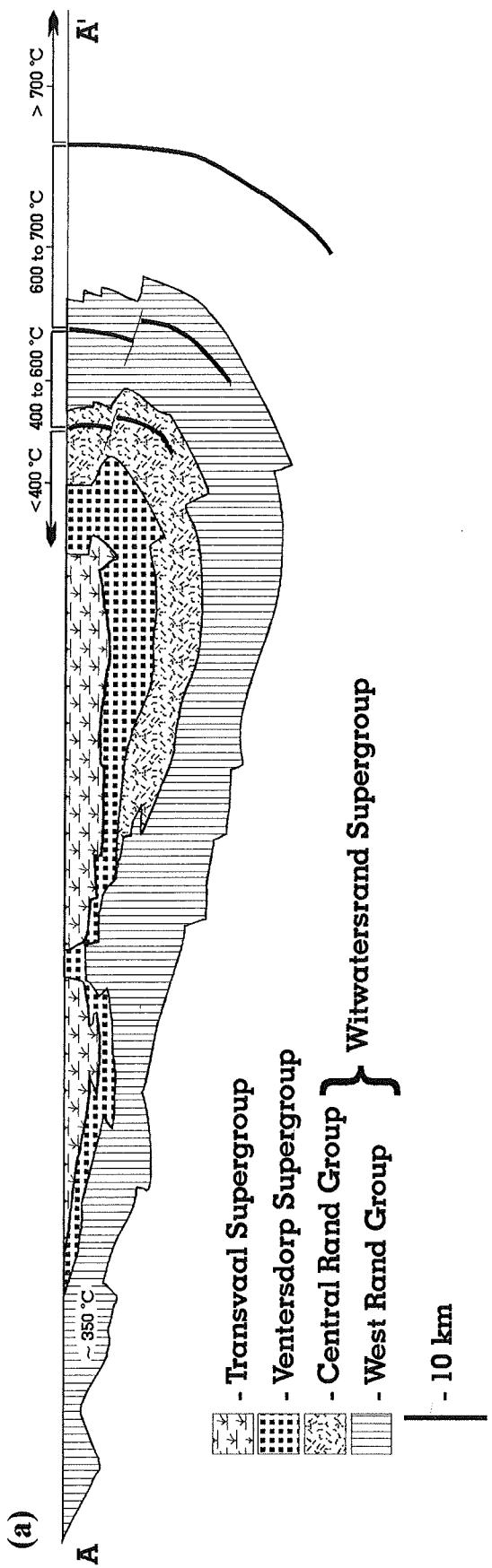
Prograde metamorphism of sedimentary protoliths drives devolatilization reactions. Thus, at low to medium grades of metamorphism fluid-phases should be present in such rocks when devolatilization reactions occur. Average metapelitic and shale compositions (Shaw, 1956) suggest that the total amount of H<sub>2</sub>O available to be released during the metamorphism of an average pelite will correspond to 12 vol% of the rock (Walther & Orville, 1982). The lower density of this fluid phase will ensure that any accumulation of a fluid volume will fracture the rock, allowing the fluid to escape upwards. The potential for volatile production is a function of the volatile content of the

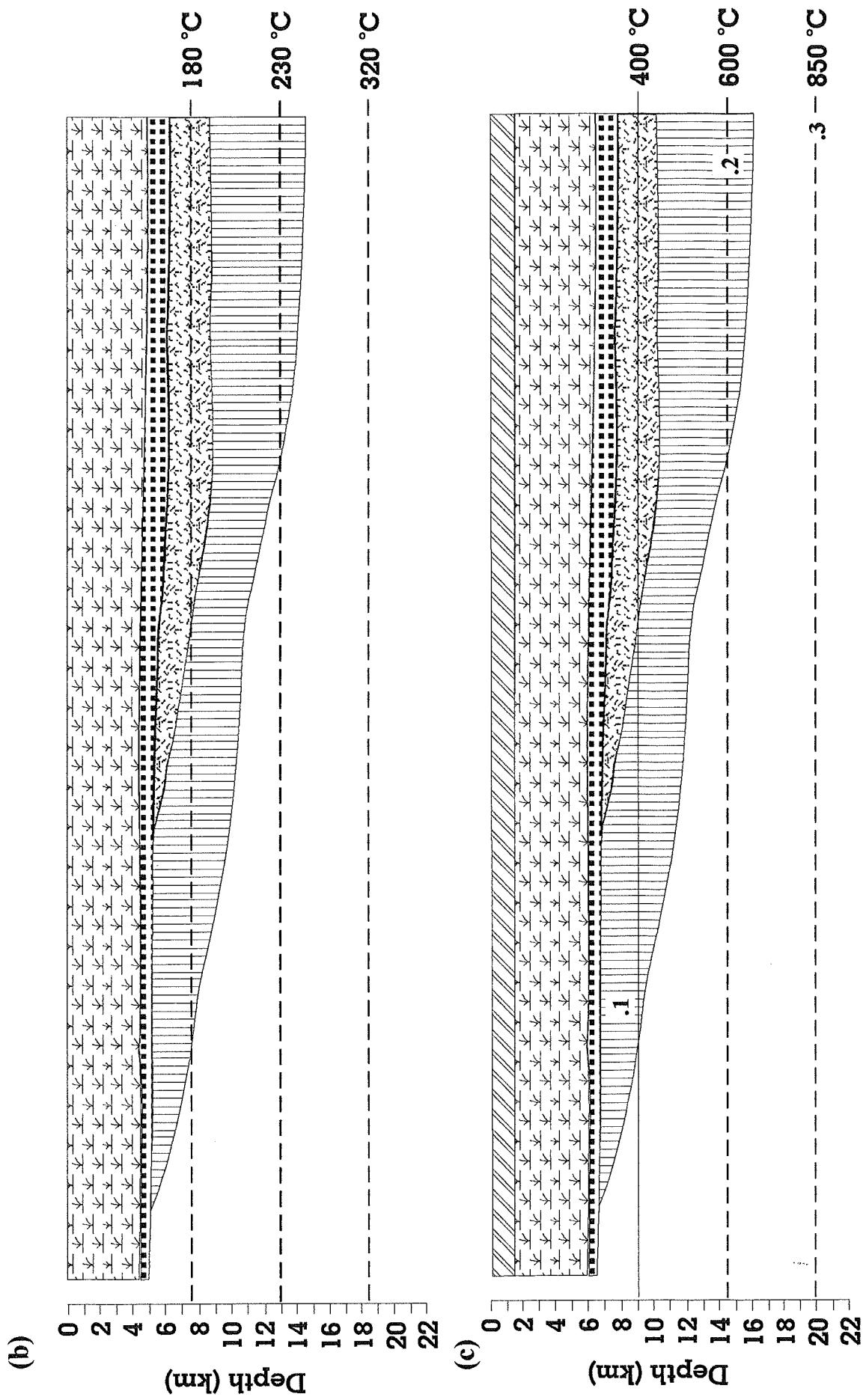
post-diagenetic sediment and the metamorphic grade attained. In clastic sedimentary successions, those dominated by argillite will have higher initial volatile contents and the higher the grade of metamorphism attained, the more anhydrous the peak metamorphic assemblage, and the higher the volume of volatiles liberated. In this regard, the argillaceous-arenaceous West Rand Group of the Witwatersrand Supergroup is a vastly richer volatile source than the predominantly arenaceous-rudaceous Central Rand Group. It is at the bottom of the sequence and therefore, attained higher temperatures during regional metamorphism (Fig. 3). It also contains thick shale units, compared to the Central Rand Group which contains only the thin "reef package" shales described by Phillips (1988) and the ~150m thick Booysens Shale and is dominated by conglomerate reef horizons and thick quartzite units.

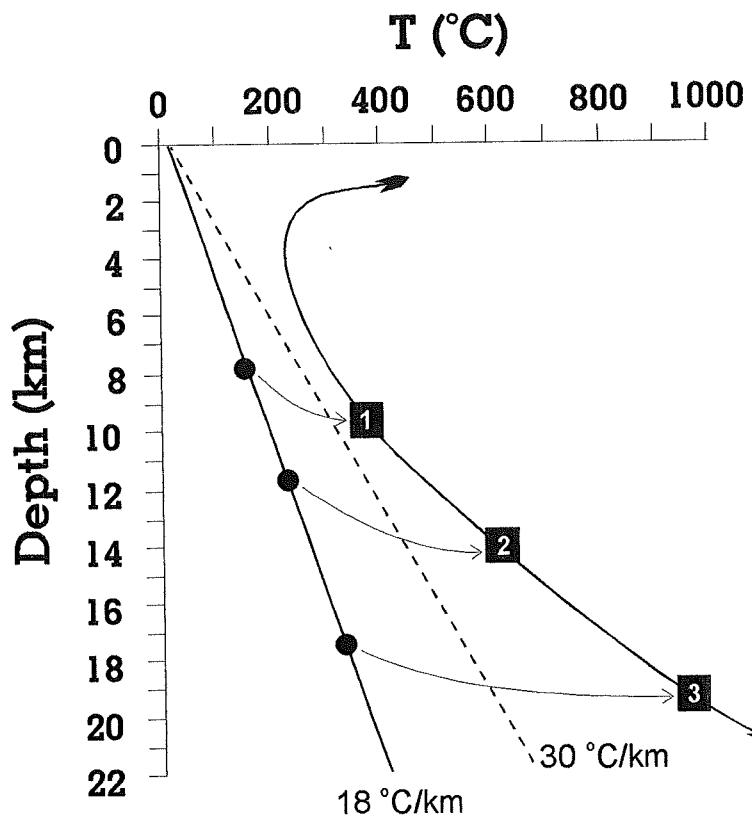
The study by Phillips *et al.* (1990) is the only past work to consider the volume of metamorphic fluid produced in the Witwatersrand Basin. These authors proposed that the shale horizons lost 2.5 vol% metamorphic water. The model for the thermal evolution of the crust during the Bushveld Event, summarized in Figure 3, allows for a first order calculation of metamorphic fluid production in the Witwatersrand Basin that is substantially more realistic and accurate than this previous estimate because it takes into account changes in metamorphic grade with depth, as well as the bulk rock compositions likely to best represent the most significant fluid source in the Basin.

*Figure 3 (Pages 10 & 11): Thermal and structural crustal profiles through the "Potchefstroom Gap" in the Witwatersrand Basin. The line of the section (A to A') is indicated in Figure 1.*

- (a) *Illustrates the geophysically constrained present day section (after Friese *et al.*, 1995). Much of the deformation displayed in this section is believed to be due to the formation of the Potchefstroom synclinorium, and is associated with the formation of the Vredefort Dome. Surface metamorphic grades and the inferred orientation of the metamorphic isograds in the Vredefort Structure are also indicated.*
- (b) *Illustrates the proposed pre-doming crustal structure and crustal thermal profile shortly before the beginning of Bushveld-related magmatism, assuming a stable crustal geotherm of 18 °C/km.*
- (c) *Illustrates the pre-doming crustal structure and crustal thermal profile shortly after the intrusion of the Bushveld Complex. Points 1, 2 and 3 refer to Figure 4.*







*Figure 4: An illustration of the depth-temperature evolution of points 1, 2 and 3 from a stable cratonic geotherm of 18 °C/km (Fig. 3a) to the low pressure, high temperature Bushveld-related, geotherm (Fig. 3c) that existed at ~2.06 Ga.*

### Bulk Compositions Studied

The following discussion deals with the West Rand Group argillites as the bulk composition and structure of the Witwatersrand rocks dictate that they formed the dominant metamorphic fluid reservoir in the Basin. However, the general principles are equally applicable to the minor shale horizons within the Central Rand Group. Bulk composition data has been collected in this study from the Hospital Hill and Government Subgroups exposed in the collar of the Vredefort Dome. The average West Rand Group shale composition, based on a comprehensive study of shale geochemistry in the Heidelberg area (Camden-Smith, 1980) is also included (Table 1). The data scatter widely on the standard AFM projection from muscovite (Fig. 5). Despite the scatter two broad groups can be identified. The first includes both Hospital Hill and Government Subgroup samples and generally plots at A values above 0.2 and is relatively magnesian. The second group is dominated by Hospital Hill Subgroup samples and is relatively ferruginous and plots at A values ranging from 0.25 to less

**Table 1: Bulk compositions of the Hospital Hill and Government Subgroups from the collar of the Vredefort Dome. The average West Rand Group shale (WGS) of Camden-Smith (1980) is also listed**

	<b>HOSPITAL HILL</b>												
	<b>WGS</b>	<b>13a</b>	<b>13b</b>	<b>13c</b>	<b>13d</b>	<b>S10</b>	<b>S6</b>	<b>V31</b>	<b>VH32</b>	<b>VH33</b>	<b>VH34</b>	<b>VH35</b>	<b>VH36</b>
<b>SiO<sub>2</sub></b>	64.71	70.09	62.71	59.77	58.27	59.91	61.18	56.01	60.73	57.94	61.93	66.44	67.07
<b>TiO<sub>2</sub></b>	0.63	0.42	0.56	0.49	0.49	0.77	0.62	0.50	0.50	0.53	0.60	0.55	0.54
<b>Al<sub>2</sub>O<sub>3</sub></b>	14.90	11.97	16.86	16.17	15.55	20.70	19.38	16.60	16.85	18.22	19.52	15.96	15.50
<b>Fe<sub>2</sub>O<sub>3</sub></b>	12.49	8.73	9.72	13.28	15.31	7.18	7.17	15.72	11.64	11.58	7.30	6.75	9.55
<b>MnO</b>	0.00	0.08	0.10	0.12	0.15	0.09	0.08	0.14	0.13	0.12	0.08	0.09	0.10
<b>MgO</b>	4.76	3.06	3.82	4.42	4.49	4.84	4.62	5.49	4.96	5.21	4.78	5.15	3.73
<b>CaO</b>	0.45	0.52	0.53	0.23	0.63	0.23	0.07	0.73	0.00	0.41	0.02	0.21	0.15
<b>Na<sub>2</sub>O</b>	0.00	1.29	1.46	1.05	1.23	1.49	0.87	0.26	0.23	0.79	0.76	1.05	0.61
<b>K<sub>2</sub>O</b>	2.10	2.35	2.24	2.96	2.42	2.17	3.01	2.47	2.21	2.86	3.00	2.23	1.93
<b>P<sub>2</sub>O<sub>5</sub></b>	0.00	0.13	0.13	0.16	0.18	0.11	0.10	0.14	0.11	0.09	0.08	0.12	0.12
<b>LOI</b>	NA	1.20	0.88	0.99	0.49	1.42	1.75	2.26	1.50	1.15	1.10	1.16	0.90
<b>Total</b>	100.00	99.83	98.99	99.65	99.21	98.90	98.84	100.31	98.84	98.90	99.15	99.71	100.22

	<b>HOSPITAL HILL (contd.)</b>								<b>GOVERNMENT</b>				
	<b>VH37</b>	<b>VH38</b>	<b>VH39</b>	<b>VH40</b>	<b>VH41</b>	<b>VH42</b>	<b>VH43</b>	<b>VH44</b>	<b>K12d</b>	<b>S1</b>	<b>S2</b>	<b>V17</b>	<b>V40</b>
<b>SiO<sub>2</sub></b>	54.37	59.72	57.46	60.06	65.16	59.32	60.85	56.52	53.20	62.74	68.75	60.45	57.31
<b>TiO<sub>2</sub></b>	0.51	0.48	0.44	0.46	0.43	0.45	0.57	0.44	0.80	0.56	0.52	0.57	0.54
<b>Al<sub>2</sub>O<sub>3</sub></b>	18.48	21.44	15.14	15.45	13.31	17.97	20.99	16.27	24.59	18.42	14.29	19.21	18.92
<b>Fe<sub>2</sub>O<sub>3</sub></b>	16.15	6.49	18.27	14.23	12.82	11.93	7.47	17.18	9.28	6.75	6.79	6.73	14.09
<b>MnO</b>	0.13	0.08	0.23	0.18	0.21	0.35	0.07	0.56	0.12	0.08	0.09	0.09	0.14
<b>MgO</b>	5.67	4.66	4.75	5.01	4.49	5.26	4.68	4.23	6.15	5.27	4.63	4.85	4.97
<b>CaO</b>	0.29	0.40	0.48	0.12	0.90	0.30	0.03	0.96	0.03	0.37	0.07	1.47	0.06
<b>Na<sub>2</sub>O</b>	0.23	0.91	0.54	0.21	0.15	0.37	0.55	0.54	0.28	0.61	0.65	2.14	0.14
<b>K<sub>2</sub>O</b>	2.70	2.64	1.91	3.06	2.37	3.22	2.89	1.82	2.66	2.63	2.59	2.27	2.82
<b>P<sub>2</sub>O<sub>5</sub></b>	0.20	0.13	0.12	0.15	0.10	0.14	0.08	0.14	0.12	0.10	0.10	0.45	0.18
<b>LOI</b>	1.50	1.98	1.16	1.34	1.08	1.17	2.20	0.28	1.34	2.12	1.24	1.25	1.12
<b>Total</b>	100.20	98.95	100.49	100.26	100.19	100.48	100.38	98.93	98.58	99.65	99.70	99.47	99.99

than 0.1. The average West Rand Group shale (WGS) plots almost centrally within this second group. A representative sample from each group (S6 and V40 from groups 1 and 2 respectively) as well as the WGS have been chosen for further analysis in this study. These compositions should

provide a representative estimate of metamorphic fluid production from the Hospital Hill and Government Subgroups, as well as from the bulk West Rand Group.

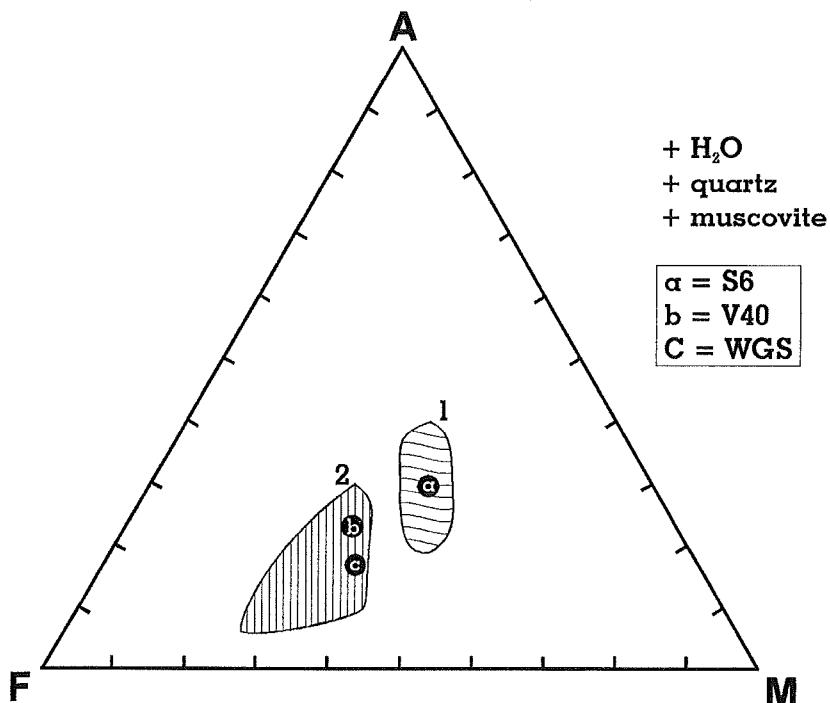


Figure 5: AFM projection from muscovite, quartz and water for the bulk compositions determined in this study.

### Metamorphic Assemblages, Phase Proportions and Bulk Rock Water Contents

#### *Post-diagenetic assemblage*

This study is not concerned with diagenetic fluids as their role in the evolution of the Witwatersrand Basin was comprehensively analysed by Phillips *et al.* (1990). Accordingly, the starting point needs to be the mineral assemblage that existed in composition S6, V40 and WGS at the end of diagenesis. A temperature of 250°C has been chosen for this poorly defined transition from diagenesis to metamorphism. Hoffman & Hower (1979) suggested that at this temperature a shale should consist of a mixture of quartz, chlorite, white mica and clay minerals. The following normative calculation has been used to calculate the phase proportions in this assemblage:

- 1) all Fe and Mg are in chlorite;
- 2) all K and Na are in muscovite;
- 3) excess Al, above that used to make chlorite and muscovite, is in kaolinite; and
- 4) excess Si, above that used in steps (1) to (2), is in quartz.

The chlorite compositions used in each bulk composition are identical in terms of Al substitution i.e.  $(\text{Mg}, \text{Fe})_9\text{Al}_3[\text{Si}_{6.39}\text{Al}_{1.61}\text{O}_{20}](\text{OH})_{16}$ , Fe/Mg ratios are those of the bulk rocks, as chlorite is the only ferromagnesian mineral. Muscovite is assumed to be ideal  $(\text{K}, \text{Na})_2\text{Al}_4(\text{Si}_6\text{Al}_2\text{O}_{20})(\text{OH})_4$ , and kaolinite and quartz are assumed to be pure  $\text{Al}_4(\text{Si}_4\text{O}_{10})(\text{OH})_8$  and  $\text{SiO}_2$ , respectively. The resultant phase proportions in wt%, phase  $\text{H}_2\text{O}$  contents and bulk rock  $\text{H}_2\text{O}$  contents are listed in Table 2.

**Table 2: Calculated phase proportions, phase  $\text{H}_2\text{O}$  contents and bulk rock  $\text{H}_2\text{O}$  contents in the 250°C, post diagenetic assemblage. All values are in wt%**

	WGS				V40				S6			
	Qtz	Chl	Musc	Kaol	Qtz	Chl	Musc	Kaol	Qtz	Chl	Musc	Kaol
Prop.	41.17	37.72	19.11	1.90	27.58	40.47	26.94	3.02	34.42	26.12	39.06	0.39
$\text{H}_2\text{O}$ phase	0.00	11.18	4.04	13.95	0.00	11.18	4.04	13.95	0.00	11.14	4.04	13.95
$\text{H}_2\text{O}$ total	5.26				6.02				4.54			

### *Metamorphism to 400 °C*

In many shale compositions an important low temperature reaction involves the disappearance of kaolinite and the appearance of pyrophyllite at ~300°C (Frey, 1987), probably as a result of the reaction  $\text{Kln} + \text{Qtz} = \text{Prl} + \text{H}_2\text{O}$  (mineral symbols are those recommended by Kretz, 1983). The low kaolinite contents in the three bulk compositions used in this study suggest that this reaction contributed very little fluid to the Witwatersrand Basin. Even in V40, the most kaolinite-rich rock, the breakdown of kaolinite would have produced less than 0.3 wt%  $\text{H}_2\text{O}$  in the bulk rock. This is a function both of the low kaolinite content and the fact that the reaction produces pyrophyllite with 5 wt%  $\text{H}_2\text{O}$ . Similarly, as a result of the fact that pyrophyllite results from kaolinite breakdown, the pyrophyllite contents of the rocks will also be very low (approximately  $1.5 \times$  the kaolinite content in the 250°C assemblage). Pyrophyllite breakdown by the reaction  $\text{Prl} = \text{Ky} + \text{Qtz} + \text{H}_2\text{O}$ , at ~400 C (Franceschelli *et al.*, 1986) will liberate a maximum of 0.25 wt%  $\text{H}_2\text{O}$  in the bulk rock in sample V40. These calculations suggest that metamorphism of the West Rand Group shales to 400°C should proceed with only minor fluid production. This is largely a function of the fact that the rocks contain little alumina in excess of that needed to stabilise muscovite and chlorite. The average residual of  $\text{Al}^{3+}$  after subtracting the Al incorporated into muscovite and chlorite, for all the analyses listed in Table 1, is 2.07. This translates into an average kaolinite clay content of approximately 2.3 wt% in the bulk rock. The low temperature peak metamorphic assemblages should thus be characterised by low clay-

mineral contents or, alternatively, by less aluminous K<sub>2</sub>O-, FeO- and MgO-bearing phases than those used in this modelling.

#### ***Assemblage above muscovite - chlorite - quartz stability***

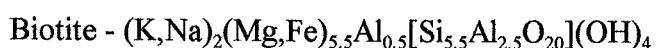
The first appearance of biotite by the reaction Mu + Chl = Bt + And + Qtz + H<sub>2</sub>O, at a temperature of approximately 400°C (Ferry, 1984), has the potential to produce substantial volumes of fluid in the West Rand Group shales because of the muscovite- and chlorite-dominated nature of the lower temperature assemblages. Unlike the reactions discussed above, which involve little or no solid solution and which will approach univariance and occur at a single discrete temperature, the biotite-producing reaction involves Fe Mg<sup>-1</sup> exchange between biotite and chlorite, as well as possible Al<sub>2</sub>O<sub>3</sub>, Fe<sup>3+</sup>, Cl and F exchange. Thus, the reaction will only run to completion, with the consumption of chlorite or muscovite, at some temperature above 400°C. For the purposes of this study a temperature of 450°C has been chosen as the upper stability limit of the quartz-chlorite-muscovite assemblage. In calculating the phase proportions in this assemblage the ratio of Fe+Mg/K+Na in biotite is modelled as 2.75. In rocks with a higher ratio of these components than 2.75, excess chlorite will result and the following rules apply:

- 1) all K and Na are in biotite;
- 2) excess Fe and Mg, above that incorporated into biotite, is in chlorite;
- 3) Al not used in biotite and chlorite forms andalusite; and
- 4) excess Si is in quartz.

In rocks with a Fe+Mg/K+Na ratio less than 2.75 excess muscovite will be produced and the following rules apply:

- 1) all Fe and Mg are in biotite;
- 2) excess K and Na are in muscovite;
- 3) Al not used in biotite and muscovite is in anandalusite; and
- 4) excess Si is in quartz.

In order to avoid having to make decisions on Fe-Mg partitioning between chlorite and biotite these components were grouped together, the biotite and the chlorite effectively have the Fe:Mg ratios of the bulk rocks. As in the modelling of the 250°C assemblage, identical Al contents are used in all samples in phases where this parameter is variable (biotite and chlorite). The resultant mineral formulae used are:



Chlorite -  $(\text{Mg},\text{Fe})_9\text{Al}_3[\text{Si}_{6.39}\text{Al}_{1.61}\text{O}_{20}](\text{OH})_{16}$

Muscovite -  $(\text{K})_2\text{Al}_4(\text{Si}_6\text{Al}_2\text{O}_{20})(\text{OH})_4$

Andalusite -  $\text{Al}_2\text{SiO}_5$

Quartz -  $\text{SiO}_2$

The resultant phase proportions in wt%, phase H<sub>2</sub>O contents and bulk rock H<sub>2</sub>O contents are listed in Table 3.

**Table 3: Calculated phase proportions, phase H<sub>2</sub>O contents and bulk rock H<sub>2</sub>O contents in the 450°C assemblage. All values are wt%**

	WGS				V40				S6			
	Qtz	Chl	Biot	And	Qtz	Chl	Biot	And	Qtz	Biot	Musc	And
Prop.	47.04	20.08	20.03	12.13	35.61	17.34	29.64	17.42	34.42	26.12	39.06	0.39
H <sub>2</sub> O phase	0.00	11.18	3.85	0.00	0.00	11.18	3.84	0.00	0.00	3.95	4.04	0.00
H <sub>2</sub> O total	3.10				3.07				1.59			

The large discrepancy between the water content of S6 and the other two samples results from the fact that chlorite is exhausted by the reaction in this sample and the hydrous phases in the final assemblage are muscovite and biotite.

#### ***Highest temperature peak metamorphic assemblages***

The peak metamorphic assemblages developed in the collar of the Vredefort Dome suggest temperatures of 600°C (Gibson & Wallmach, 1995). The good preservation of these highest-grade West Rand Group shales has allowed for the identification of the peak metamorphic assemblage and determination of the mineral compositions involved in the two samples from this study. The peak assemblage in S6 consists of cordierite, biotite, andalusite and quartz; the corresponding assemblage in V40 is biotite, garnet, cordierite and quartz. The mineral compositions in these two samples have been determined by electron microprobe analyses on the OXFORD microprobe at the University of Pretoria and are presented in Table 4. Comparison of the positions of V40 and WGS plotted on the AFM projection in Figure 5 suggests that the hypothetical sample represented by the average composition WGS should have the same garnet-biotite-cordierite peak metamorphic assemblage as V40. Therefore, at the same metamorphic grades, these two samples should be characterised by the same mineral compositions. Thus, the compositions presented for V40 in Table 5 can also be used

to model the peak metamorphic phase proportions in WGS. The well-constrained mineral composition data available for these samples allows the phase proportions to be calculated by a simple least squares mixing routine. These results as well as the H<sub>2</sub>O contents of the phases and bulk rocks are presented in Table 5.

**Table 4: Mineral compositions in the peak metamorphic assemblages of S6 and V40**

	V40			S6	
	Garn	Biot	Cord	Biot	Cord
<b>SiO<sub>2</sub></b>	38.35	34.51	46.24	36.69	45.10
<b>TiO<sub>2</sub></b>	0.00	1.72	0.00	1.99	0.00
<b>Al<sub>2</sub>O<sub>3</sub></b>	21.87	19.54	32.27	19.97	39.85
<b>FeO</b>	33.40	24.36	11.33	21.21	7.94
<b>MnO</b>	4.48	0.00	0.16	0.00	0.00
<b>MgO</b>	1.66	8.05	7.37	8.15	5.06
<b>CaO</b>	0.16	0.00	0.00	0.00	0.00
<b>Na<sub>2</sub>O</b>	0.00	0.00	0.00	0.00	0.00
<b>K<sub>2</sub>O</b>	0.00	8.05	1.64	8.89	1.70
<b>Total</b>	99.92	96.23	99.01	96.90	99.65

**Table 5: Calculated phase proportions, phase H<sub>2</sub>O contents and bulk rock H<sub>2</sub>O contents in the 600 °C, peak metamorphic assemblage. All values are wt%**

	WGS (ssr <sup>1</sup> = 0.7)				V40 (ssr = 0.2)				S6 (ssr = 0.9)			
	Qtz	Garn	Biot	Cord <sup>2</sup>	Qtz	Garn	Biot	Cord	Qtz	Biot	Cord	And
Prop.	38.51	6.40	32.19	22.73	25.67	6.84	34.09	33.53	34.42	26.12	39.06	0.39
H <sub>2</sub> O phase	0.00	0.00	3.85	1.5	0.00	0.00	3.85	1.5	0.00	3.85	1.5	0.00
H <sub>2</sub> O total	1.58				1.82				1.44			

<sup>1</sup>The sum of squared residual values (ssr) arise through squaring the residuals of the bulk rock chemical components remaining after the compositions of the phases have been subtracted according to their modelled proportions. These values are an indication of the accuracy of the calculations and a value of 0 indicates that the mixing routine models the bulk composition perfectly.

<sup>2</sup>The cordierite H<sub>2</sub>O content is based on the value published for a cordierite-garnet-biotite hornfels from Angus, Scotland (Deer *et al.*, 1977).

## Discussion

The approach used in this study for calculating bulk rock water contents during the peak of metamorphism has advantages over laboratory determinations of this parameter in Witwatersrand metasediments. This study follows on several others (e.g. Slawson, 1978; Hart *et al.*, 1981) in contending that the collar of the Vredefort Dome provides an approximate vertical section through the Witwatersrand strata. Gibson & Wallmach (1995) and Stevens *et al.* (1996) have demonstrated that in this area the Witwatersrand shales and the underlying basement are characterised by relatively high-temperature, low-pressure retrogression. Thus, the actual bulk rock H<sub>2</sub>O contents do not record the peak metamorphic H<sub>2</sub>O content and cannot be used to calculate the amount of fluid liberated during prograde metamorphism. The approach used in this study provides a reliable estimation of metamorphic fluid production from the West Rand Group shales. The amount of fluid produced between 250°C and either the 450°C or peak metamorphic assemblage can be calculated by subtracting the relevant bulk rock H<sub>2</sub>O content from that of the post-diagenetic assemblage (Table 6). However, it should be remembered that fluid production will not occur as a linear function of increasing temperature. Rather, fluid will be produced in discrete pulses during the occurrence of prograde dehydration reactions. The pattern of fluid production only emerges if the calculated fluid production values from Table 6 are integrated with the likely temperature intervals of the dehydration reactions discussed earlier. This is illustrated for the average West Rand Group shale (WGS) in Figure 6. The suggested diagenetic fluid production values and intervals suggested by Phillips *et al.* (1990) are included.

**Table 6: H<sub>2</sub>O production between 250 and 400°C, between 450 and 600°C and in total, in the three bulk compositions studied . The values are in wt%**

	250 to 450 °C	450°C to peak T	Total
<b>WGS</b>	1.89	1.52	3.41
<b>V40</b>	2.95	1.25	4.20
<b>S6</b>	2.95	0.15	3.1

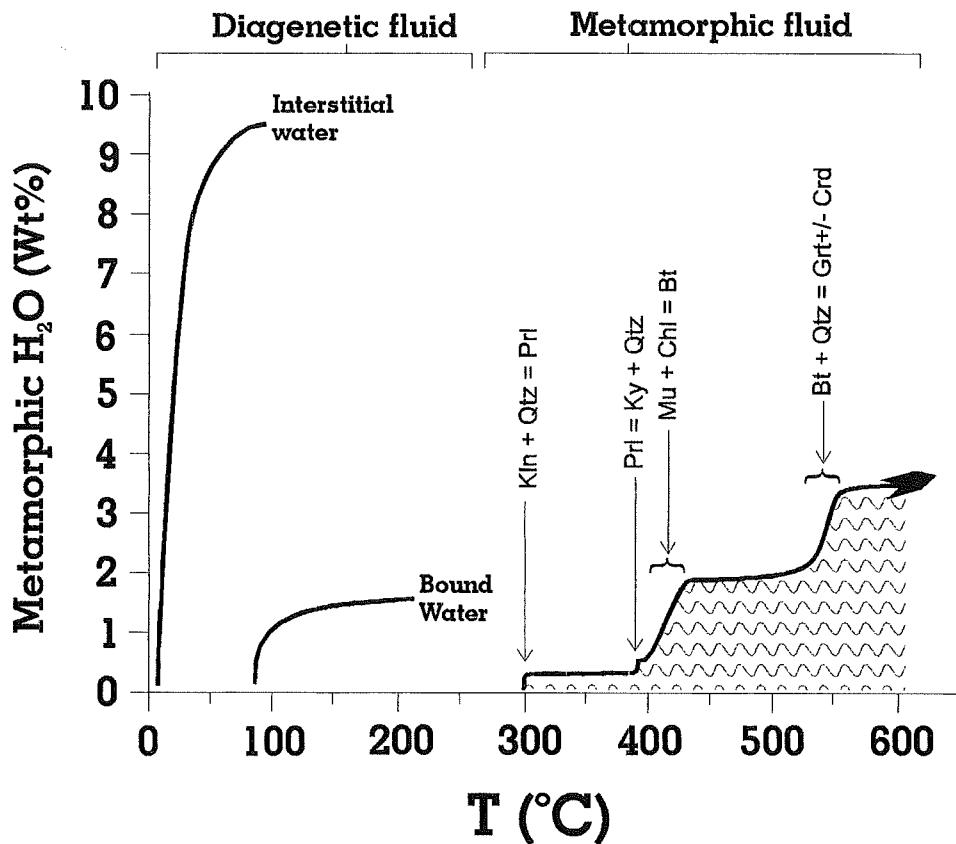


Figure 6: Hydrous fluid volume produced from the average West Rand Group shale since the onset of sedimentation to the peak of metamorphism in the deepest portions of the Witwatersrand Basin. Note that substantial metamorphic fluid production appears to be restricted to rocks that were heated to above 400 °C.

The data clearly indicate that substantial volumes of hydrous metamorphic fluid were produced from the portions of the West Rand Group that evolved to temperatures above the onset of chlorite breakdown. At temperatures below this very little fluid appears to have been produced, primarily due to the low kaolinite content of the rocks. The above calculations suggest that an average West Rand Group shale, metamorphosed to above 450 °C, lost 70 litres of water per cubic meter of rock. The same composition and volume metamorphosed to 600 °C would have lost 139 litres of water (high pressure-temperature fluid density from Burnham *et al.* (1969), metasediment density estimated to be 3.0 at 450 °C and 3.1 at 600 °C). A comparison of the pre-Vredefort Dome profile in Figure 3 and the established suboutcrop limits of the West Rand Group (Fig. 1) suggest that the West Rand Group

evolved to a temperature appropriate for the formation of biotite over an area of at least 5000 km<sup>2</sup>. Similarly, it appears that an area of at least 1200 km<sup>2</sup> evolved to above 600°C. Thus, the profile in Figure 3c suggests that ~9896 km<sup>3</sup> evolved to above 450°C, and ~471 km<sup>3</sup> of West Rand Group attained a temperature in excess of 600°C. Published sedimentary profiles (e.g. Holland *et al.*, 1990) suggest that a shale to quartzite ratio of 1:1 is accurate for the West Rand Group. These values indicate that the West Rand Group shales lost at least  $7.58 \times 10^{14}$  litres of hydrous metamorphic fluid. This fluid would have mobilized upwards and out of the Basin and is most likely to have passed through the overlying Central Rand Group. The remainder of this paper considers the potential of this fluid to remobilize gold during interaction with the overlying Central Rand Group.

## **GOLD REMOBILIZATION POTENTIAL OF THE METAMORPHIC FLUID**

The fluids produced during prograde metamorphism should be delivered in pulses, corresponding to the thermal evolution of the key fluid reservoirs through metamorphic reaction intervals (Fig. 6). The significant questions when considering the capacity of these fluids to remobilise gold contained within the Central Rand Group are: 1) is there a significant component of strata-bound fluid-flow along auriferous horizons; and 2) does the fluid have the ability to carry gold in solution? These questions can largely be answered by fluid inclusion studies of Central Rand Group reef horizons. To some degree such studies also provide a test of the metamorphic model presented above. If the modelling is accurate, and if there was appreciable stratabound fluid-flow within the Basin, then fluid inclusions in the goldfields should record the presence of aqueous inclusions in authigenic phases trapped at a temperature of approximately 350°C. The precise composition of these inclusions will determine their capacity to have transported gold.

### **Fluid Inclusion Microthermometry - A Review**

#### ***Inclusions inherited from the provenance regions***

Surprisingly little work has been published on the fluid inclusions occurring in authigenic minerals in the Witwatersrand Basin. Fluid inclusion studies in the Witwatersrand Basin have generally examined the nature of fluids hosted within the detrital quartz pebbles (for an overview see Phillips *et al.*, 1988). However, a knowledge of the composition of these fluids does not contribute towards the understanding of fluids circulating in the Witwatersrand Basin, but rather

describes the nature of the fluids in the enigmatic source region/s. Shepherd (1977) distinguished five principal types of pre-depositional fluid inclusions in the pebbles of the Witwatersrand conglomerates. These types differed in the amount of CO<sub>2</sub> and collectively indicate a moderate-to high-pressure, high-temperature environment of original presumed vein-quartz formation. Systematic variation in the relative abundance of these inclusion assemblages for different sections of the ore field demonstrated the presence of several different, well-defined provenance areas or multiple entry points into the Basin. A marked sympathetic relationship between uraniferous blanket ores and the presence of vein quartz containing inclusions rich in liquid CO<sub>2</sub>, together with a corresponding antipathetic relationship with Au, strongly suggested separate sources for the two metals (Shepherd, 1977). Post-depositional inclusions are subordinate and offer no support for the alternative epigenetic model, showing only later interaction of relatively cool circulating ground waters. A series of papers by Hallbauer (1982a,b;1983) and Hallbauer & Kable (1979; 1982) indicated that data from fluid inclusions in quartz pebbles have permitted the recognition of another distinct class of "blue opalescent quartz pebbles". These authors have identified numerous solid phases in the inclusions in both quartz and pyrite by SEM, including orthoclase, muscovite, calcite, apatite, iron-rich phyllosilicate, barium feldspar, chlorite, rutile, corundum, anhydrite, cassiterite and various mixed chlorides, including CaCl<sub>2</sub>. Although called "daughter crystals", some of the crystals in their photomicrographs actually look more like solid inclusions. Whichever they are, these crystals provide valuable insight into the original environments of formation of the quartz pebbles.

### *Authigenic fluid inclusions*

Killick *et al.* (1988) focussed on pseudotachylite associated with the bedding-parallel fault zone between the Ventersdorp and Witwatersrand Supergroups in the Western Areas Gold Mine. Weak brines with up to 6 wt% eNaCl were identified in pseudotachylite-associated discordant quartz veins. Clathrate melting temperatures of 6° to 13°C are indicative of CO<sub>2</sub>. The authors suggested that the pseudotachylites may have formed at similar depths to the associated mylonites as the result of intermittent reactivation of fault zones and, furthermore, suggested a genetic link between the pseudotachylites in the Vredefort area and those on the bedding-parallel fault zones. This suggests a close temporal link between the entrapment of these fluids and the Bushveld-related prograde metamorphic fluid-flow suggested by this study.

Fluid inclusion studies on authigenic material from sedimentary reef horizons have generally yielded low- to moderate salinity aqueous inclusions showing low homogenization temperatures. Although the data on the post-depositional fluids are far from comprehensive, the composition of the fluid inclusions indicate complex mixed fluids, with divalent ions  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , apart from Na and K, as well as Cl and S being the most common anions (*e.g.* Klemd *et al.*, 1989, 1994; Meyer *et al.*, 1991; Boer *et al.*, 1995). Homogenization temperatures, *i.e.* minimum entrapment temperatures of reef-hosted fluid inclusions, centre around  $150^\circ \pm 40^\circ\text{C}$ , with a certain population of high temperature fluids showing homogenization temperatures in excess of  $250^\circ\text{C}$ : Basal Reef in the Welkom goldfield: -  $130^\circ$  to  $140^\circ\text{C}$  (Frimmel & Minter, 1991; Frimmel *et al.*, 1993); shear zones in the Kimberley Reef and Ventersdorp Contact Reef: -  $85^\circ$  to  $279^\circ\text{C}$  (Klemd *et al.*, 1994); and the matrix quartz in the Ventersdorp Contact Reef: -  $115^\circ$  to  $210^\circ\text{C}$  (Boer *et al.*, 1995). Frimmel (1994) categorized the fluid evolution in the Witwatersrand Basin into three major stages, *viz.* (1) diagenesis; (2) burial associated with infiltration of fluids that interacted with the sediments of the Transvaal Supergroup; and (3) hydrothermal activity related to the Bushveld/Vredefort events. No distinction was made on chemical grounds between formation waters and post-depositional alteration fluids, as both these fluids are characterized by low pH, low  $f\text{O}_2$  and relative high  $f\text{S}_2$  conditions.

The homogenization temperatures presented in previous studies are an indication of the minimum entrapment temperatures for these fluid inclusions. In order to determine the actual temperature of entrapment it is necessary to apply a pressure correction which relies on an external geobarometer for the Upper Witwatersrand rocks. Fault-associated kyanite occurrences close to the Kimberley and Monarg reefs indicate pressures in the range of 2 to 2.5 kbar (Wallmach & Meyer, 1990). This indicates that the kyanite-bearing faults were activated or reactivated after deposition of more than 8 to 9 km of rock on top of the Witwatersrand Supergroup (during late or post-Transvaal times). Recently, sericite pseudomorphs after post-tectonic kyanite were found in a shear zone near the top of the Central Rand Group (Frimmel, 1992), which, by assuming a pressure in the order of 2 to 2.5 kbar, places the kyanite-forming event clearly into post-Transvaal time. The shales of the Orange Grove Quartzite Formation also contain kyanite which, according to Schreyer & Bischoff (1982), occurs on a regional scale. Subsequent work (McCarthy *et al.*, 1986; Wallmach, pers. comm., 1996) on the overlying Black Reef and Ventersdorp Supergroup have indicated the presence of andalusite, with no kyanite. These observations indicate that

conditions during metamorphism of the Upper Witwatersrand sequence were close to this alumino-silicate phase transition, and that a pressure assumption of 2 to 2.5 kbar for the Ventersdorp Contact Reef, for example, is reasonable. Consequently, entrapment temperatures for the aqueous - carbonic fluids in these rocks centre around  $300^{\circ}\pm50^{\circ}\text{C}$ . Late-stage, low salinity fluids show lower homogenization temperatures and have probably been emplaced at lower pressures. These pressures are difficult to evaluate since there is no evidence relating to the absolute timing of their entrapment and, thus, the tectonic conditions influencing the reef horizon. The available data indicate that a temperature of  $300^{\circ}\text{C}$  and a pressure of 2 kbar is a reasonable estimate for the thermodynamic modelling that follows.

### **Gold Solubility in Witwatersrand Metamorphic Fluids**

A detailed chemical analysis of fluid compositions, using quadrupole mass spectroscopy, was conducted by Boer *et al.* (1995; and in prep.) on the Ventersdorp Contact Reef and associated lithologies, at the Elandsrand and Vaal Reefs (No. 10 Shaft) gold mines. In almost all analyses of the Ventersdorp Contact Reef (VCR) samples  $\text{H}_2\text{O}$ ,  $\text{CO}_2$  and  $\text{CH}_4$  make up more than 99 mol% of the total gas composition, although the relative abundances of  $\text{CH}_4$  and  $\text{CO}_2$  vary greatly among samples from Elandsrand and Vaal Reefs. On a  $\text{CO}_2$ - $\text{CH}_4$ - $\text{C}_2\text{H}_6$  ternary diagram the Elandsrand and Vaal Reefs samples plot in distinctly separate fields, with  $\text{CO}_2$  not exceeding 0.60 mol% in the Elandsrand samples and  $\text{CH}_4$  not surpassing 0.59 mol% in the Vaal Reefs samples. The fluids are thus  $\text{H}_2\text{O}$  dominated (>95 mol%). A well-defined positive correlation exists between  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in the Vaal Reef samples, whereas  $\text{H}_2\text{O}$  and  $\text{CH}_4$  show a distinct correlation in samples from the Elandsrand Mine. These sympathetic correlations suggest that the nature of the carbonic species is a function of the oxygen fugacity. This finding is corroborated by the sulphide mineralogy of the two localities (Boer *et al.*, in prep.). Alkanes show enhanced concentrations compared to hydrothermal deposits of the mesothermal type with values ranging up to 0.14 mol%, but do not correlate consistently with other gaseous compounds. The average total sulphur gas content ( $\sum S_{\text{Total}} = \text{H}_2\text{S} + \text{SO}_2$ ) for Elandsrand and Vaal Reefs is 0.0202 and 0.0109 mol%, respectively. However, the  $\text{SO}_2/\text{H}_2\text{S}$  ratio varies significantly in the range 0.01 to 0.39, irrespective of locality or rock type. HCl values are significantly enriched in all of the VCR samples. Moderate  $\text{N}_2$  and Ar values confirm that no atmospheric contamination took place during sample preparation (*e.g.* by air entrapment along cleavage planes).

Mean  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  concentrations in the Elandsrand fluids were 0.27 and 1.03, corresponding to activities of  $a\text{CO}_2 = 0.34$  and  $a\text{CH}_4 = 0.46$ , respectively. Mean concentrations for Vaal Reefs samples were  $X_{\text{CO}_2} = 1.95$  and  $X_{\text{CH}_4} = 0.33$ , corresponding to  $a\text{CO}_2 = 2.44$  and  $a\text{CH}_4 = 0.15$ . Fluid salinities were moderate around 2.6 molar with a mean Na:K ratio of 4. In terms of the reaction  $\text{CO}_{2(\text{aq})} + \text{H}_2\text{O} \leftrightarrow \text{HCO}_3^- + \text{H}^+$  ( $\log K = -7.658$ ; calculated using SUPCRT92, Johnson *et al.*, 1992;  $\log aK = -1.11$ ; ionic strength = 2.46) an acidic pH of approximately 3 (neutral pH at 350°C is 5.1) was calculated for the VCR fluids ( $\gamma_{\text{CO}_2} = 2.26$  [calculated from Patterson *et al.*, 1981]). QMS gas data were utilized to determine redox conditions:  $f\text{O}_2$  was calculated using the reactions  $\text{CH}_4 + 2\text{H}_2\text{O} \leftrightarrow 4\text{H}_2 + \text{CO}_2$  and  $\text{H}_2\text{O} \leftrightarrow \text{H}_2 + \frac{1}{2}\text{O}_2$ , which yielded values of  $10^{-29.02}$  and  $10^{-26.05}$  for Elandsrand and Vaal Reefs samples, respectively. QMS data yielded sulphur fugacities for Elandsrand and Vaal Reefs of  $f\text{H}_2\text{S}_g = 1.099$  & 0.598, assuming  $\sum S_{\text{Total}} = X_{\text{H}_2\text{S}} + X_{\text{SO}_2}$ , which corresponds to  $a\text{H}_2\text{S}_{,\text{aq}}$  of  $10^{-1.5}$  to  $10^{-2.6}$ , respectively.

Using these fluid characteristics, inferences on the gold speciation and precipitation mechanism are possible. Solubility calculations indicate that VCR gold in the Elandsrand area was an order of magnitude more soluble as a gold chloride complex than as a gold bisulphide complex. In the Vaal Reefs area, gold was equally soluble as a chloride complex or as a bisulphide complex. The calculated result is confirmed by the higher abundance of HCl in the Vaal Reef samples, compared to the Elandsrand samples. It appears that the post-depositional fluids were certainly capable of taking appreciable volumes of gold into solution, in the order of 1 to 10 parts per billion. These values, coupled to the previous calculation of metamorphic fluid production, indicate that metamorphic fluid flow in the Witwatersrand Basin had the potential to mobilise up to 7600 tons of gold. This is in excess of 12% of all the gold contained in the depository prior to the beginning of mining (43000 tons of gold mined to date plus 20000 tons estimated reserves). This figure is substantially lower than the >300000 tons of potentially mobilised gold suggested by Phillips *et al.* (1990). This extravagant value largely results from their assumption of a gold concentration in the fluid of 1 part per million, some three orders of magnitude greater than appears realistic from our calculations. Precipitation mechanisms for gold dissolved in the metamorphic fluid are not well understood. However, it was suggested by Frimmel *et al.* (1996) and Boer *et al.* (1995) that changes in sulphur- and oxygen fugacity, as well as temperature and pH may all play a dominant role.

## CONCLUSIONS

With the realisation during the past two decades that large-scale fluid migration is a normal predictable phenomena in sedimentary and metamorphic environments, it becomes possible to make advances towards unravelling the long-standing controversy regarding the origin of gold and uranium in the Witwatersrand Basin. Numerous bedding-parallel shear zones occur within some of the auriferous and uraniferous conglomerates, particularly in the Ventersdorp Contact Reef. These shear zones have served as channelways for the migration of hydrothermal fluids. It is suggested that the Witwatersrand Basin lost at least  $7.58 \times 10^{14}$  litres of aqueous metamorphic fluid during the Bushveld Event at  $\sim 2.06$  Ga. Some of this fluid may have mobilized directly upwards. However, fluid inclusion populations from authigenic phases in reef horizons suggest that an aqueous fluid, most likely derived from the argillaceous portions of the West Rand Group, migrated through the reef horizons in the goldfields at close to the peak of metamorphism. This suggests a significant proportion of stratabound metamorphic fluid flow. Gold, dissolved from reefs in the hotter, deeper portions of the Basin, was mobilised towards the cooler Basin margins, thereby enriching existing reefs where conditions were appropriate for gold precipitation. The authors propose that this mechanism accounts for the demonstrably low gold grades recorded from reefs occurring near the central portion of the Basin - particularly those exposed around the Vredefort Dome - and the extremely high gold grades in some of the reefs of the goldfields. Finally, a most important finding from an exploration viewpoint, is that the estimates of the gold mobilizing potential of the fluid, and the availability of a vast source of gold in the Central Rand Group through which the fluids had to migrate, predicts the existence of significant gold deposits outside the confines of the Witwatersrand Basin.

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