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by

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ABSTRACT

The Lebowa Granite Suite of the Bushveld Complex is a large 2054 Ma old, A-type batholith, characterized by several, relatively small magmato-hydrothermal, polymetallic ore deposits. The mineralization is represented by a three-stage paragenetic sequence; early magmatic Sn-W-Mo-F ores (600°C>T>400°C), followed by a Cu-Pb-Zn-As-Ag-Au paragenesis (400°C>T>200°C) and then late-stage Fe-F-U mineralization (<200°C). The first stage of mineralization (typified by the endogranitic Zaaiplaats tin deposit) is related to incompatible trace element concentration during crystal fractionation and subsequent fluid saturation of the magma. Evolution of the late magmatic fluids, as they were channelled along fractures, as well as mingling with externally derived connate or meteoric fluids, resulted in the deposition of the second stage of mineralization. This mineralization is typified by the fracture-related endogranitic Spoedwel and Albert deposits, and also by the exogranitic, sediment-hosted Rooiberg Mine, which is dominated by polymetallic sulphide ores. As the externally derived fluid component became progressively more dominant, oxidation of the polymetallic sulphide assemblage and precipitation of hematite, pitchblende and fluorite occurred generally along the same fracture systems that hosted the earlier sulphide paragenesis.

Small hydrothermal zircons trapped along quartz growth zones from the Spoedwel deposit yielded a U-Pb concordia age of 1965±30 Ma. Whole rock Rb-Sr age determinations from the Lebowa Granite Suite fall in the range 1790±114 Ma to 1604±70 Ma and are interpreted to reflect alkali element mobility and isotopic resetting during exhumation of the Bushveld granite. In contrast to thermal modelling, which indicates that hydrothermal activity should have ceased within 4 my of emplacement, isotopic evidence suggests that mineralization was long-lived, but episodic, and that fluid-flow events were linked to major periods of Palaeo- and Mesoproterozoic orogenic activity along the margins of the Kaapvaal Craton. During these orogenic episodes fluid flow was enhanced by tectonically induced, fluid over-pressuring and/or exhumation of the Bushveld Complex.

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INTRODUCTION

The Bushveld Complex, emplaced into the Kaapvaal Craton during the Palaeoproterozoic, is host to an extraordinary wealth of magmatic and hydrothermal ore deposits. The Bushveld Complex is a large igneous province, covering an area of some 66 000 km^2, which comprises an earlier mafic intrusion, the Rustenburg Layered Suite (RLS), emplaced penecontemporaneously with an acid extrusive event called the Rooiberg Group (Fig. 1). The sill-like, 7000m-thick RLS is characterized by pronounced *in situ* differentiation and episodic magma injection, during which time crystal-fractionation and sulphide immiscibility processes resulted in the concentration of enormous chromite, platinum group element-copper-nickel, and vanadium-rich magnetite ore deposits. Emplacement of the RLS and Rooiberg Group was followed by intrusion of a shallow level, 1500m thick, sheet-like granitoid batholith, with typical A-type characteristics, known as the Lebowa Granite Suite (LGS). The LGS is composed of the Nebo Granite, the Klipkloof Granite and the Makhutso Granite. However, local terms for the Nebo Granite varieties, such as the Bobbejaankop Granite and Lease Granite at the Zaaiplaats Tin Mine, have been informally adopted in other areas of the Bushveld (Pollard *et al.*, 1991).

The composite LGS is pervasively mineralized (Fig. 1), although no world-class deposits are currently known. Widespread polymetallic, magmato-hydrothermal mineralization, typified by a Sn-W-Mo-Cu-Pb-Zn-Ag-Au-Fe-U-F paragenesis, occurs as small- to medium-sized deposits that may be either endo- or exogranitic.

The Bushveld Complex, in general, and the LGS in particular, have proven extremely difficult to date and it is only with the advent of single-zircon techniques that accurate constraints on the emplacement age of the Complex have been obtained. The best current estimate for the age of the RLS is 2061±27 Ma obtained from a Rb-Sr whole rock isochron (Walraven *et al.*, 1990); this age is identical to that of 2061±2 Ma obtained for Rooiberg Group rhyolite by single zircon Pb-Pb evaporation analysis (Walraven, 1997). The age of the LGS is presently believed to be 2054±2 Ma, also obtained by single zircon Pb-Pb evaporation analysis (Walraven and Hattingh, 1993). These data suggest that emplacement of the entire Bushveld Complex was achieved within a time span of some 6 my, although petrogenetic considerations suggest that this interval should be considerably shorter.

Although the RLS-hosted magmatic deposits have been extensively studied, the granitoid-hosted magmato-hydrothermal ores are less well understood. In particular, the processes that gave rise to such an extended paragenetic sequence, as well as the nature and origin of both fluids and metals in these complex, polymetallic deposits, requires more work. Fluid inclusion studies (Ollila, 1981; Pollard *et al.*, 1991; Robb *et al.*, 1994; Freeman, 1998) have recognized a spectrum of fluids that present a range from orthomagmatic through meso- and epithermal entrapment temperatures. Ore genesis studies (Robb *et al.*, 1994; Freeman, 1998) have shown that ore fluids were derived from at least two sources, one originally magmatic and the other meteoric and/or connate, and suggested that metal concentrations are linked to mingling of these fluids. The extended paragenetic sequence has previously been thought of as representing a continuum of ore deposition persisting over an extended period of geological time and isotopic studies have suggested that the hydrothermal system was active for some 1000 my, between 2060 Ma and circa 1100 Ma (McNaughton *et al.*, 1993; Robb *et al.*, 1994). The extended

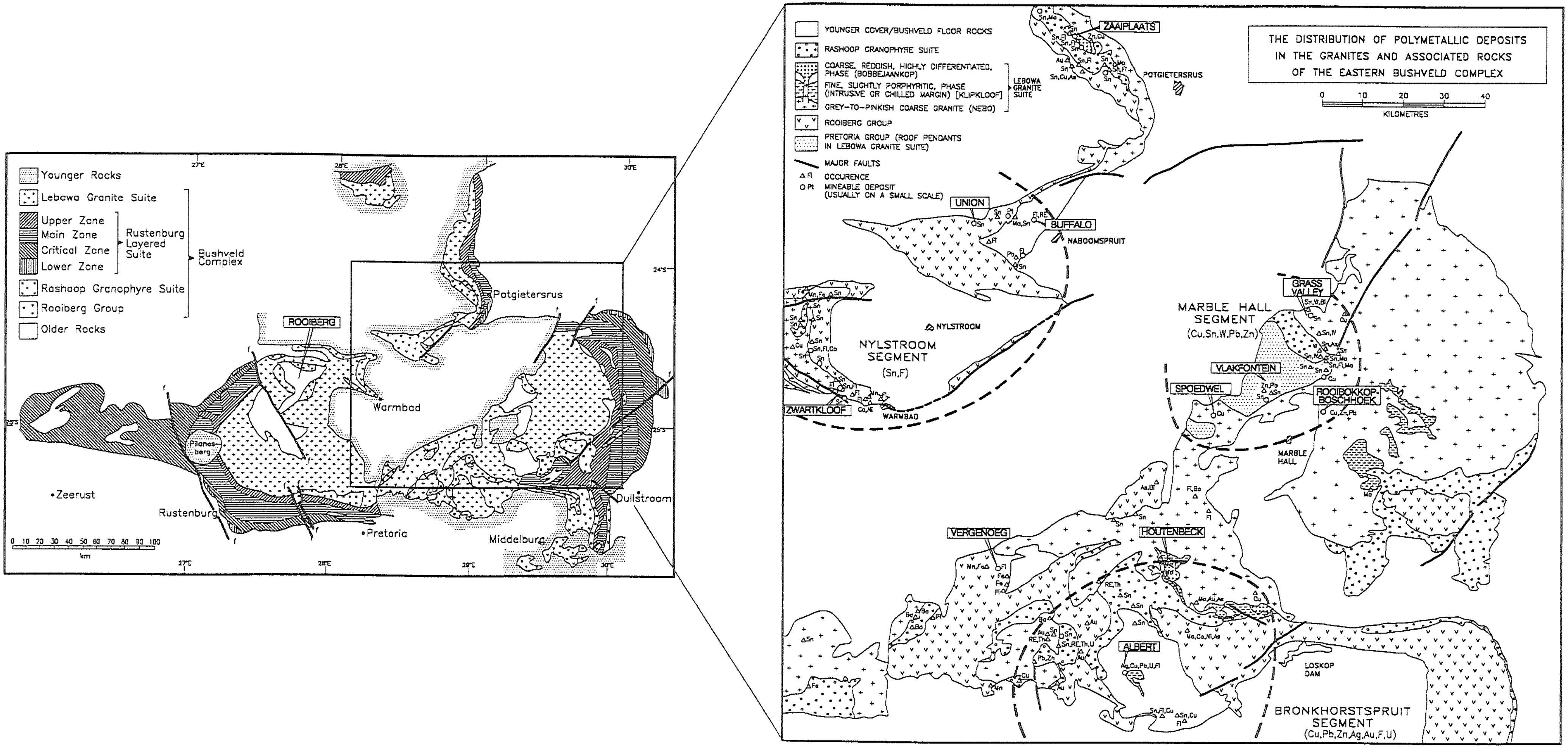


Figure 1: *Simplified map of the Bushveld Complex with an enlarged plan showing the exposed Lebowa Granite Suite (LGS) and Rooiberg Group in the eastern section of the Complex and the widespread distribution of granite-related polymetallic ore deposits.*

duration of this mineralizing system was attributed to the high radiothermal heat productive capacity of the host granitoids. The extended duration of this hydrothermal system is, however, contradicted by recent studies (e.g. Cathles *et al.*, 1997) which indicate that magmato-hydrothermal fluid flow tends to be short-lived, typically lasting for about 1 my.

The present paper considers the duration of a granite-related hydrothermal fluid flow scenario for the LGS in terms of thermal modelling as well as fluid inclusion and isotopic data for the various styles of mineralization. New, precise U-Pb age data for authigenic zircons from mineralized portions of the LGS, coupled with published whole-rock and mineral age data, are presented to show that mineralization in the Bushveld granites was indeed long-lived, but episodic. An attempt is made to relate the complex paragenetic sequence to multiple fluid and metal sources and the episodes of fluid flow to major craton-margin orogenic events, coupled with exhumation of the Bushveld Complex.

NATURE OF POLYMETALLIC MINERALIZATION IN THE LGS

Ore deposits in the Lebowa Granite Suite may be broadly divided into two categories: (1) orthomagmatic (formed during crystallisation of the granite); and (2) structurally controlled (formed during migration of mineralizing fluids and precipitation of ore minerals along regional and local fracture systems). In addition, mineralization tends to be concentrated in the upper portion of the LGS and may be either endogranitic or exogranitic. Freeman (1998) examined the nature of fluids associated with mineralization in five fracture-related deposits of the eastern lobe of the Bushveld Complex and comparisons were made with the orthomagmatic tin deposit at Zaaiplaats. The deposits, namely Grass Valley, Spoedwel, Dronkfontein, Albert, and Houtenbek, all occur within a graben, flanked by the Wonderkop and Zebediela fault systems (Fig. 1).

Robb *et al.* (1994) and Freeman (1998) showed that the diverse occurrences of polymetallic mineralization in the LGS can be described in terms of at least three episodes of ore formation. The first episode of mineralization (typified by the Zaaiplaats tin deposit) occurred at relatively high temperatures (600-400°C) and resulted in the formation of a cassiterite-scheelite-wolframite-fluorite assemblage. The mineralization is disseminated and formed by processes related to fractional crystallisation and subsequent fluid saturation of the magma. Evolution of the late magmatic fluids, as they interacted with the wallrocks and mingled with externally derived connate and/or meteoric fluids, resulted in the deposition of a second episode of mineralization at lower temperatures ($200°C<T<400°C$). This latter mineralization is represented by a polymetallic sulphide paragenesis comprising pyrite-galena-tetrahedrite/tennantite-sphalerite-chalcopyrite (typified by the Spoedwel, Boschhoek and Albert deposits). As the external fluid component became progressively more dominant, the third episode of mineralization took place with deposition of hematite, pitchblende and significant fluorite. The introduction of the latter assemblage (which is prominent at the Albert and to a lesser extent at Spoedwel deposits) along the *same* fracture systems which localised the sulphide assemblage, resulted in alteration of the sulphides to secondary minerals such as bornite, chalcocite, digenite, covellite and limonite.

The extended nature of this three stage paragenetic sequence is considered to reflect widespread mingling between an early fluid derived by H_2O-saturation of the granitic magma and an external meteoric/connate fluid. The descriptions that follow are summaries intended to provide a framework upon which an assessment of the nature and longevity of hydrothermal activity and mineralization in and around the LGS can be made.

Orthomagmatic Deposits

Endogranitic, orthomagmatic Sn, with minor W and Mo mineralization, is represented at the Zaaiplaats Tin Mine (Fig. 1). At Zaaiplaats, *in situ* crystal fractionation within a highly differentiated granite has resulted in a centripetally concentrated zone of disseminated, low-grade, cassiterite mineralization (Groves and McCarthy, 1978). In addition, higher-grade, pipe-like tin ore bodies cut through the granite and represent tubular conduits for hydrothermal solutions that precipitated an assemblage comprising cassiterite-feldspar-sericite-calcite-tourmaline with minor sulphide and rare earth minerals (Crocker, 1986). A photomicrograph showing disseminated cassiterite within altered and silicified granite from the Zaaiplaats Mine is shown in Figure 2a.

The orthomagmatic Sn-dominated deposits associated with the Lebowa Granite Suite are characterised by high-temperature magmatic fluids. Ollila (1981) measured homogenisation temperatures for fluid inclusions from a variety of granites associated with the Zaaiplaats deposit and found evidence for high temperature fluids (Fig. 3). The Bobbejaankop Granite yields T_h values of between 480 °C and 719 °C, while those in the Lease Granite yield values between 455 °C and 554 °C. Fluid inclusions in the contact pegmatite have homogenisation temperatures between 365 °C and 525 °C, and CO_2-rich fluid inclusions in vuggy quartz associated with cassiterite between 371 °C and 438°C. Salinities in these fluids are typically high and range between 35 - 50 wt% eNaCl although vein-related fluorite, calcite and quartz contained fluids of lower salinity between 5 - 35 wt% eNaCl. Ollila (1981) also identified a later low-temperature fluid in secondary inclusion trails with T_h in the range 150 - 200°C (Fig. 3). In a later study at Zaaiplaats Pollard *et al.* (1991) identified high salinity, halite-sylvite-Fe chloride-bearing primary inclusions in granite-hosted quartz, cassiterite and fluorite, with T_h ranging up to 600°C. Lower salinity and lower temperature L+V aqueous inclusions (T_h 88-240°C) were also found in paragenetically later minerals such as quartz, fluorite and calcite. In addition, the presence of H_2O-NaCl-CO_2 inclusions was also identified in secondary inclusion trails and in the paragenetically late minerals, and attributed to immiscibility on cooling of the primary magmatic fluid.

Pollard *et al.* (1991) concluded that early fluid inclusions contain solid phases (halite, sylvite and Fe chloride) and circulated at temperatures up to 600°C and lithostatic pressure of around 1250 bars. They noticed a progressive decrease in temperature from 600°C to 200°C, which is accompanied by a decrease in salinity from 68 to 15 equivalent weight per cent NaCl and a decrease in $\delta^{18}O_{fluid}$ values from 6.4 ‰ to 1.4 ‰. They suggested that fluid evolution was dominated by the interaction between granites and a magmatically derived fluid phase and that the compositional range and homogenisation behaviour of the inclusions suggests immiscibility around 350°C and 1000 bars.

Evidence of magmatic fluids associated with mineralization in the Rooiberg volcanics was also found in a study of the Vergenoeg fluorite deposit (Borrok *et al.*, 1998). High-temperature (T_h>500°C) high-salinity (up to 67 wt% eNaCl) fluids were found to be associated with the early fluorite-fayalite-ilmenite-apatite-allanite-pyrrhotite assemblage and these co-existed with a CO_2 vapour phase. A later paragenesis, comprising magnetite-hematite-ferroactinalite-siderite-sulphides, was found to be associated with lower temperature and lower salinity fluids which, on the basis of stable isotope compositions, were deemed to be of mixed magmatic and meteoric origin.

Figure 2: *Photomicrographs showing ore minerals and paragenetic relationships in polymetallic ore deposits of the Lebowa Granite Suite..*

A. *Disseminated cassiterite mineralization in altered granite from the Zaaiplaats Mine.*

B. *Early cassiterite followed by later precipitation of pyrite and chalcopyrite in a vein cutting altered sediments from the Rooiberg Mine.*

C. *The early pyrite-chalcopyrite-arsenopyrite paragenesis from the Albert Mine.*

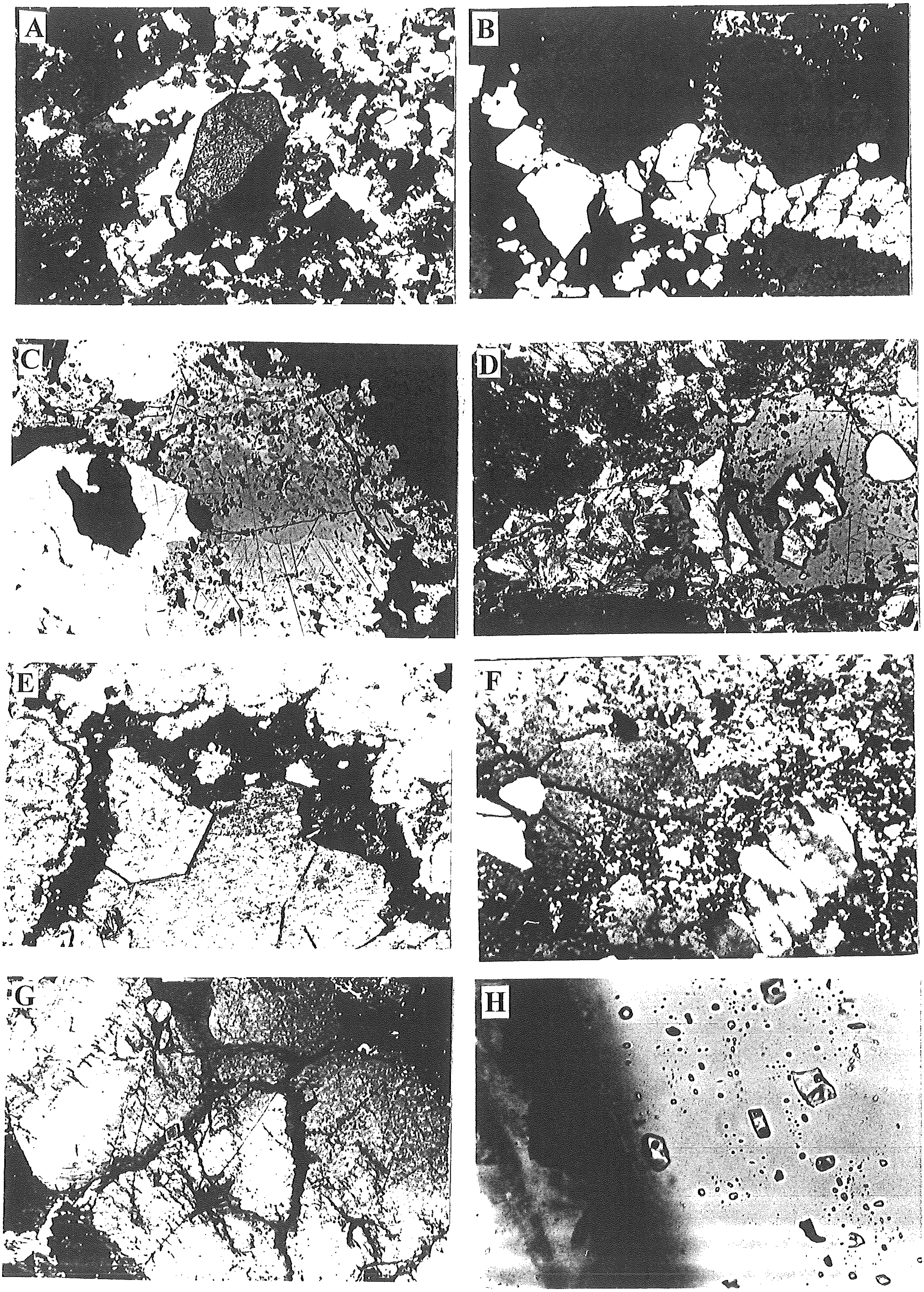
D. *Oxidation of the early sulphide mineralization at the Spoedwel Mine, exhibited by covellite-muscovite-chlorite overprinting chalcopyrite.*

E. *The late vug-filling hematite-chlorite-fluorite paragenesis from the Albert Mine.*

F. *Pitchblende intimately associated with hematite from the late Fe-F-U assemblage at the Albert Mine.*

G. *Cathodoluminescence image of small authigenic zircons associated with a late stage of hydrothermal quartz growth at the Spoedwel Mine.*

H. *Primary fluid inclusions trapped along a quartz growth zone, some of which contain small hematite crystals; sample from late stage quartz at the Spoedwel Mine.*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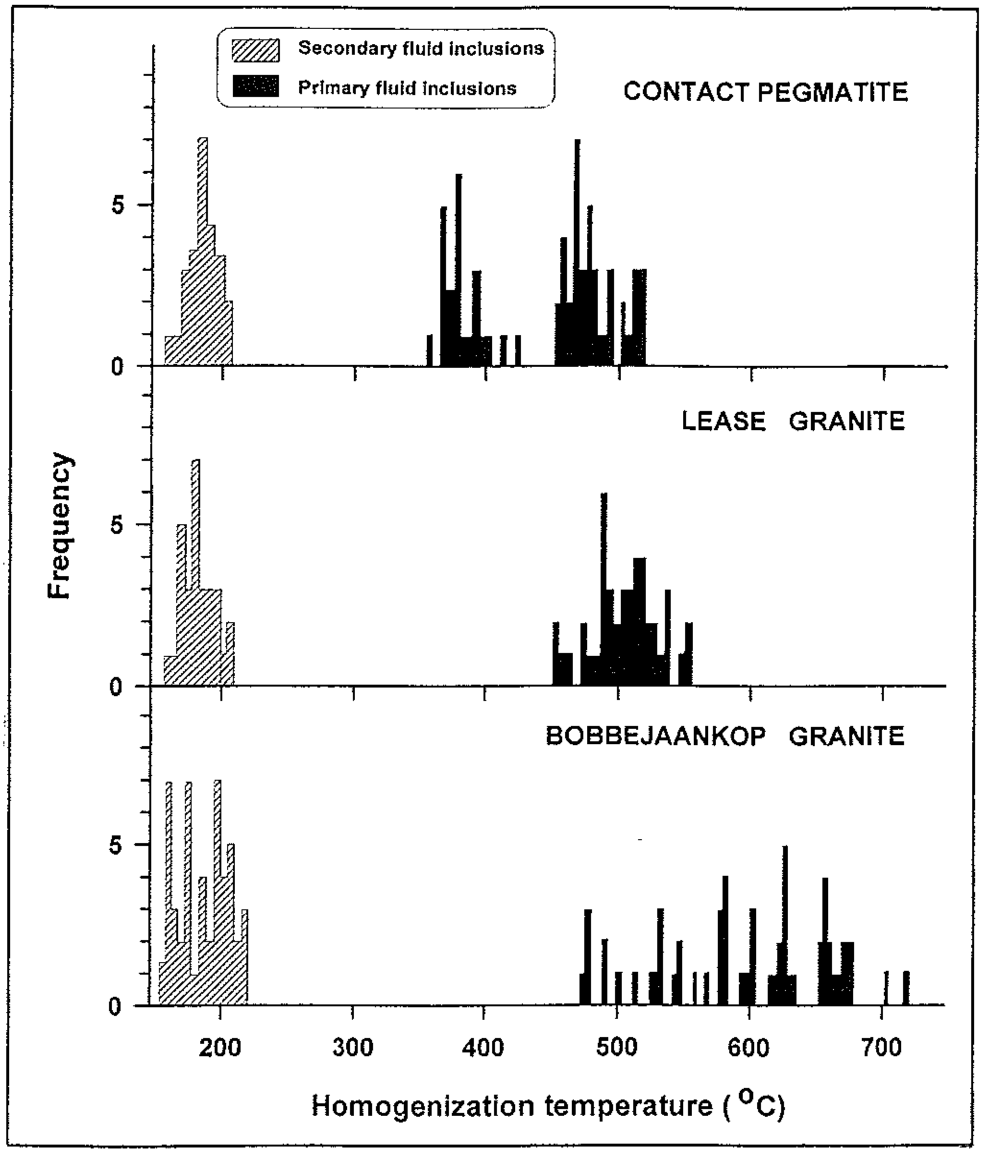

Figure 3: Frequency histogram showing the ranges of fluid inclusion homogenization temperatures obtained by Ollila (1981) for granitic host rocks to the endogranitic tin mineralization at the Zaaiplaats Mine. P = primary fluid inclusions in a variety of gangue and ore minerals; S = secondary fluid inclusions.

Orthomagmatic cassiterite from the Zaaiplaats Mine has yielded a relatively precise, but inaccurate, U-Pb isotope age of 2099±3 Ma (Gulson and Jones, 1992). The significance of this data is unclear and the fact that the age is considerably older than the emplacement age of the host granite might indicate the presence of inherited lead from an older source.

Structurally Controlled Deposits

A host of fracture-related deposits with broadly similar characteristics are known to be associated with the LGS. Early studies on the Grass Valley Tin Mine (Wagner, 1921) recognized a three-stage paragenetic sequence similar to that which has recently been determined for the Spoedwel and Albert deposits, and introduced the concept of meteoric fluid ingress along fractures as being responsible for the formation of the later stages of the paragenetic sequence. Smits (1980) also recognized a three stage paragenetic sequence at the

Rooibokkop-Boschhoek copper deposit and incorporated fluid channeling along fracture zones and the subsequent mixing of magmatic and meteoric fluids in a model for the mineralization.

Endogranitic Fracture-related Deposits

Polymetallic sulphide deposits in the LGS commonly occur in the apical portions of highly differentiated granites and mineralization is typically associated with sub-vertical, quartz-filled fractures. In certain cases there is evidence to suggest that fluids have accumulated beneath, or within, the fine-grained roof-zone facies of the granite sheet. A schematic diagram depicting the geological characteristics of these deposit types, which are typified by the Spoedwel and Albert mines, is presented in Figure 4.

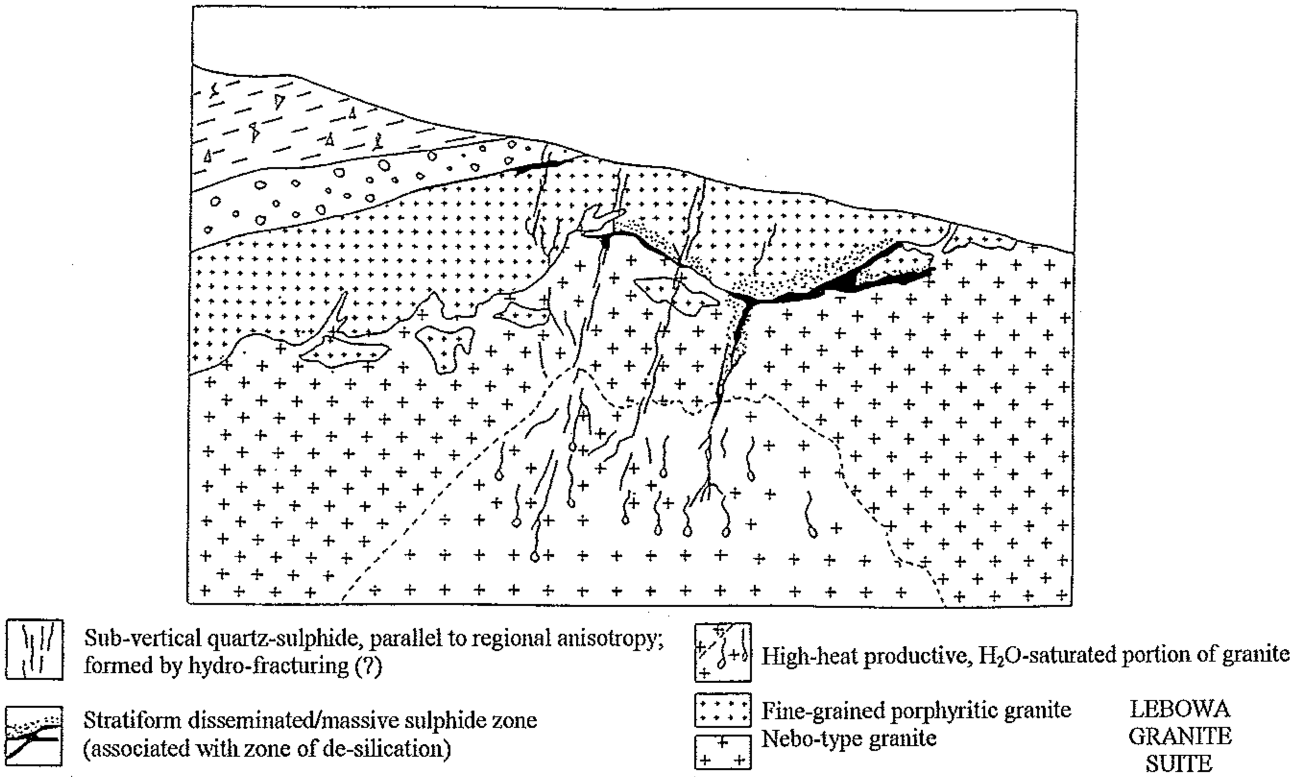

Figure 4: *Schematic cross section showing the characteristics of polymetallic Cu-Pb-Zn-Ag-Fe-F-U mineralization in Albert/Spoedwel type deposits.*

At Spoedwel mineralization occurs at the contact between coarse-grained granite and fine-grained granite in the form of a tabular massive sulphide body dipping gently to the north. In addition, vertical, stockwork-like mineralization occurs to the south of the tabular ore and is interpreted as being a feeder to the flat-lying body. The ore body reaches a maximum width of approximately 60m in the central portion and a thickness of up to 5m. Mineralization and de-silication of the host Nebo Granite occurred simultaneously, with excess silica being expelled into the immediate hanging-wall, where it manifests as quartz veins. The northeast-trending quartz veins crop out at surface approximately 1 km south of the Spoedwel Mine, and are interpreted to be the result of leakage from a tabular "ponded" body trapped below the fine-grained granite cap-rock.

At the Albert Silver Mine geological mapping by Robb *et al.* (1994) indicated that a fine-grained granitic roof zone, or possibly a chilled apical phase, overlies and is intruded by a coarsergranite. The region of mineralization is defined on surface by at least three major sub-vertical vein systems trending approximately east-west. The paragenetic sequence at Albert has been divided into two stages: (1) an early sulphide assemblage comprising pyrite, chalcopyrite, sphalerite, galena, arsenopyrite and argentiferous tennantite; and (2) a later assemblage of thuringitic chlorite, hematite, pitchblende and fluorite.

Exogranitic Fracture-related Deposits.

Lower temperature, exogranitic Sn-base metal mineralization, is best represented by the Rooiberg and Union Mines (Fig. 1). At Rooiberg, cassiterite mineralization is hosted in both bedding-parallel and cross-cutting lodes within strongly metasomatized shaley arkose of the Leeuwpoort Formation, a largely arenaceous sequence of the upper Transvaal Supergroup, into which the LGS has intruded (Rozendaal *et al.*, 1986). The paragenetic sequence begins with orthoclase-quartz, followed by tourmaline-apatite-cassiterite, and finally, carbonate-chalcopyrite-pyrite (associated with minor galena, sphalerite bismuthinite and gersdorffite) (Leube and Stumpfl, 1963). A photomicrograph showing a typical vein-fill assemblage comprising prismatic cassiterite growing from the vein walls, and later precipitation of pyrite and chalcopyrite in the middle of the vein, is shown in Figure 2b.

Rooiberg mineralization is believed to have derived from an original magmatic fluid that migrated some distance from its granitic source. Similar characteristics pertain to the Union Mine, except that the cassiterite-copper sulphide ore assemblage is hosted within volcaniclastic sediments of the Rooiberg Group, which unconformably overlie the Transvaal Supergroup (Pringle, 1986a).

Fluid Characteristics of Fracture-related Deposits.

Studies carried out on deposits in the LGS clearly point to a complex fluid evolution in the Bushveld granites that is commensurate with the long-lived paragenetic sequence of polymetallic ore formation (Wagner, 1921; Smits, 1980; Ollila, 1981; Pollard *et al.*, 1991; Robb *et al.*, 1994; Borrok *et al.*, 1998; Freeman, 1998). In contrast with the orthomagmatic Sn-dominated deposits, however, the polymetallic sulphide-dominated ores representative of the later stages of the paragenetic sequence show little or no preservation of the high-temperature, high-salinity fluids that characterize the former. Recent fluid inclusion studies (Freeman, 1998) have shown that multiple pulses of fluid flow were associated with mineralization at deposits such as Grass Valley, Albert and Spoedwel. At least three generations of quartz precipitation were recognized by cathodoluminescence imagery (Fig. 2g) in the ores at each of the deposits, and fluids associated with each were examined using various analytical techniques. The results of microthermometry and stable isotopic studies for these deposits are presented here to provide supporting evidence for the model of fluid mixing that is being developed for mineralization in the Bushveld granites.

The homogenisation and final melting temperatures of fluids associated with mineralization at several fracture-related, sulphide ore deposits in the LGS are plotted in Figure 5. These fluid populations are dominated by low-to-intermediate temperatures (100-300°C) and intermediate salinities, and the detection of clathrate melting indicates that they occasionally contain small amounts of CO_2. Raman microspectrometry and quadrupole mass spectrometry of fluid inclusions also revealed the existence of small molar proportions of other volatiles such as CH_4, N_2, H_2S and SO_2. In general, the most saline fluids among these data are associated with the mineralization at Spoedwel, whereas fluids associated with late-stage hematite-pitchblende-

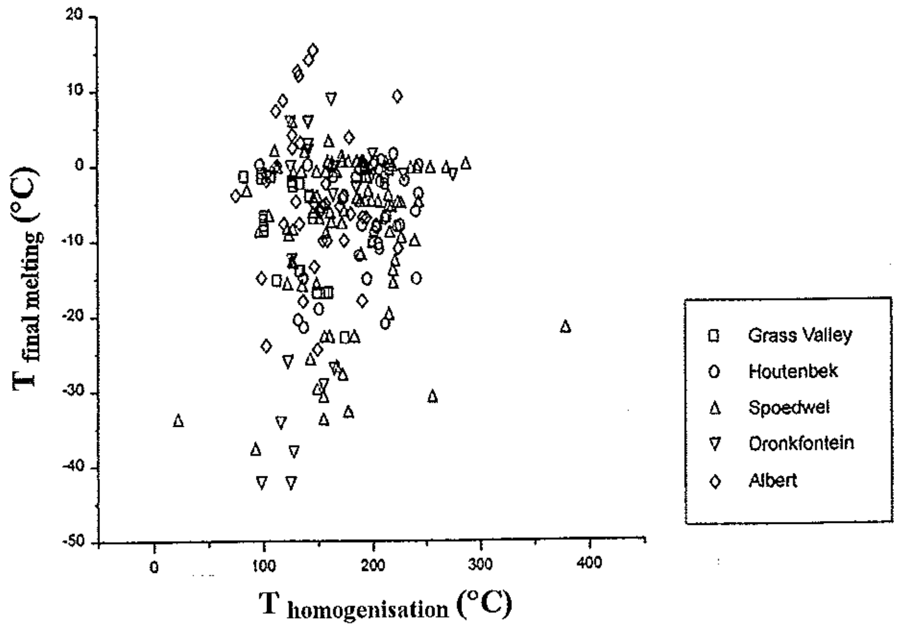

Figure 5: Plot of final melting versus homogenization temperatures for fluid inclusions from a variety of the polymetallic deposit types (i.e. Grass Valley, Houtenbek, Spoedwel, Dronkfontein and Albert) which represent the intermediate- to late-stages of the paragenetic sequence in the LGS.

fluorite assemblages at Albert tend to be of a lower temperature and salinity. In detail, however, the fluids associated with the early stages of sulphide precipitation became progressively more saline, peaking with the deposition of sphalerite, and then decrease in salinity with the precipitation of the late sulphide assemblage. The relatively high salinity of the earlier fluid suggests that at this stage it was not meteoric, but rather a basinal brine or connate fluid, perhaps derived from the Transvaal sediments into which the granites were intruded. The trend towards progressively lower salinities in fluids associated with the later stages of the paragenetic sequence possibly reflect addition of a low salinity meteoric fluid component and the highly variable salinities observed in Figure 5 are certainly consistent with a dilution trend involving mingling between at least two fluids. The magmatic fluid component is apparently not preserved in these deposit types, either because it may have evolved to an extent where it is no longer recognizable as such, or it has been swamped by externally derived fluids. The paragenetically late, polymetallic, fracture-controlled sulphide-dominated deposits, together with the very late hematite-pitchblende-fluorite assemblage, appear to be have been dominated by externally derived fluids of possible connate and meteoric origin. Evidence supporting a fluid mixing model for mineralization in the LGS is also provided by direct oxygen and hydrogen isotope determinations carried out at the Department of Geological Sciences, University of Michigan, on fluid decrepitates from inclusions hosted in non-oxygen bearing minerals such as fluorite and sphalerite. CO_2, CH_4 and H_2O liberated from such samples can be used to directly determine the C, O and H isotopic signatures of ore-bearing fluids using procedures described in Vennemann and O'Neil (1993). This data (Table 1) is plotted on a δD - $\delta^{18}O$ diagram in Figure 6, where it is seen that fluids reflect mixtures between either magmatic and meteoric waters or between magmatic and connate waters, where the latter have equilibrated with organic matter. Carbon isotope values (Table 1) typically fall in the range $\delta^{13}C = -2.6$ to 3.5, which is diagnostic of

Table 1. Oxygen, hydrogen and carbon isotope determinations for fluid inclusion decrepitates from various polymetallic deposits in the Lebowa Granite Suite

SAMPLE	$\delta^{18}O$ (water)	δD (water)	$\delta^{18}O$ (methane)	δD (methane)	$\delta^{13}C$ (carbon dioxide)	$\delta^{18}O$ (carbon dioxide)
Houtenbek1	1.2	-52.7	21.4	n.d.	-7.5	21.4
Houtenbek2	1.6	-93.0	55.6	n.d.	1.4	55.6
GrassValley2	1.0	-96.2	15.5	n.d.	-2.6	15.6
GrassValley8	17.8	n.d.	19.1	-150.6	3.5	17.8
Spoedwel2	n.d.	-60.8	n.d.	-167.3	n.d.	n.d.
Spoedwel13	72.1	n.d.	n.d.	n.d.	2.4	72.1

Notes: 1. All values in per mil. 2. Fluid inclusion decrepitates were obtained from fluorite, except for Houtenbek 2 (sphalerite) and Grass Valley 8 (quartz). 3. Oxygen isotope ratios relative to SMOW; carbon isotope ratios relative to PDB. 4. Oxygen and hydrogen isotope ratios in columns 3 and 4 were determined on water converted from fluid inclusion decepitated methane. Details of analytical procedures in Vennemann and O'Neil (1993).

carbon derived from a sedimentary carbonate (dolomitic?) source. A single lower value of $\delta^{13}C$ = -7.5 from a Houtenbek fluorite suggests contamination by organic carbon, a possibility that is consistent with the δD - $\delta^{18}O$ signatures indicated in Figure 6.

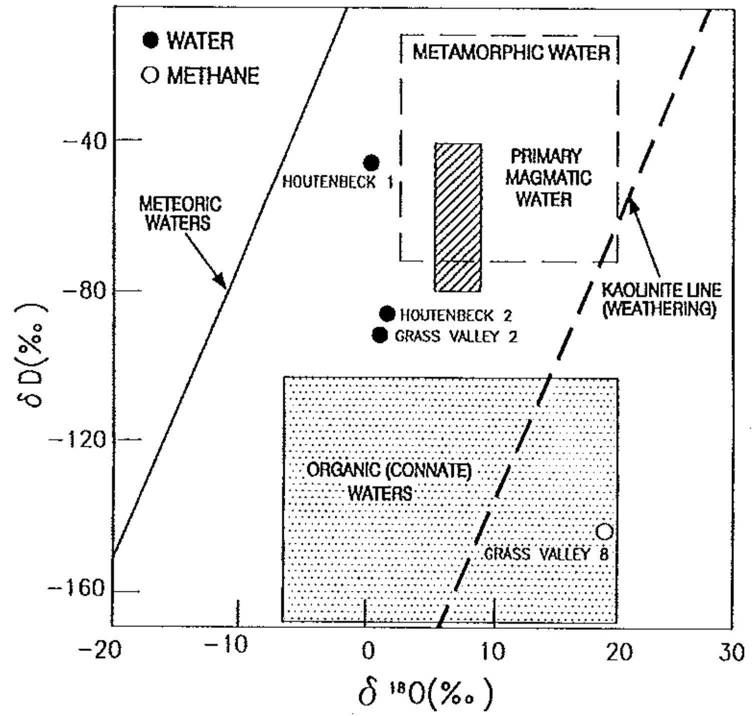

Figure 6: Plot of δD versus $\delta^{18}O$ of fluid inclusion decripitates from Houtenbek and Grass Valley mines (data from Table 1) analysed by quadrupole mass spectrometry.

THERMAL MODELLING

Thermal modelling calculations designed to place constraints on the duration of a hydrothermal event associated with an igneous intrusion show that convective fluid flow is geologically short-lived and closely linked to a specific intrusion event. Cathles *et al.* (1997) demonstrated that for a single, 40km x 2km, ultramafic sill-like intrusion in the mid-crust, hydrothermal fluids at T>200°C would circulate for a maximum of 0.8my in a low permeability (0.1 mD) environment.

In a high-permeability environment (1.0 mD), such as might be expected to pertain in a mineralizing system, the duration of hydrothermal fluid flow at T>200°C would be even less, and would effectively cease some 0.3 my after magma emplacement. The Bushveld Complex is a much larger igneous body and its emplacement history is characterized by multiple intrusions initiated by a voluminous 7km thick composite mafic sill followed by a further 1.5 km thick, composite felsic intrusion. This scenario has been modelled to see if the duration of hydrothermal fluid flow is significantly different from the time scales produced by Cathles *et al.* (1997).

In the present study, the equations for conductive heat loss with time along a crustal profile of given thermal gradient, and perturbed by one or more igneous intrusions, have been incorporated into a computer-driven algorithm named THERMOS (Cawthorn and Walraven, 1996). In the first model a single felsic intrusion, 2km thick, with a liquidus temperature of 800°C, and emplaced at 4km depth into a crust with a thermal gradient of $20^{\circ}C.km^{-1}$, is considered. Heat loss is by conduction alone, but accommodates a significant radiothermal heat production of 30 $\mu W.m^{-3}$ (after calculations by McNaughton *et al.*, 1993). Figure 7a shows that, 50 000 years after emplacement, the crust immediately below and above the intrusion is still hot enough to sustain orthomagmatic fluid flow. However, 0.5my after emplacement, the granite and surrounding crust would have cooled to around 200°C and hydrothermal activity would have effectively ceased (Fig. 7b). This scenario accords well with the constraints provided by Cathles *et al.* (1997).

A more realistic scenario for the Bushveld Complex, however, is provided if consideration is given to the influence of the thick mafic underplate that must have existed beneath the LGS at the time of its emplacement. Although the present age constraints for emplacement of the Bushveld Complex (2060 - 2054 Ma) suggest intrusion over a 6 my period, it is considered likely, for petrogenetic reasons, that the mafic and felsic components were essentially coeval. In this scenario, therefore, the LGS is emplaced 250 000 y after intrusion of the 7km thick mafic sill representing the RLS. At the time of LGS emplacement the RLS still represents a major mid-crustal thermal anomaly that likely kept the base of the granite sheet above its solidus temperature for some time. Conductive heat decay shows, in this case, that the crust in and around the LGS would decrease to around 200°C only 4 my after intrusion of the Bushveld Complex.

By any standards, this represents a situation that likely sustained an extremely long-lived hydrothermal system. This is only feasible because of the enormous volumes of magma present and the extent of the thermal anomaly that they represent. It should be noted that the effects of the high heat productive capacity of the LGS play a minimal role in extending the duration of the thermal anomaly in and around the granite above 200°C. The most important consequence of the model, therefore, is that even with the 7km of hot magma beneath it and its high heat productive capacity, any magmatic hydrothermal fluid originating from the granite itself would have cooled to, or below, 200°C within five million years of emplacement of the Complex. Any hydrothermal fluid activity in the Bushveld Complex that significantly post dates 2050 Ma, is no longer likely to be magmatic in origin and must have been externally derived.

AGE AND RADIOGENIC ISOTOPE CHARACTERISTICS

A compilation of all the age data pertaining to the LGS is provided in Figure 8, where it is seen that apparent ages for the granite suite range from 2100 Ma to 950 Ma (for the sources of this data see Fig. 8). This range is interpreted to reflect widespread and long-lived open-system

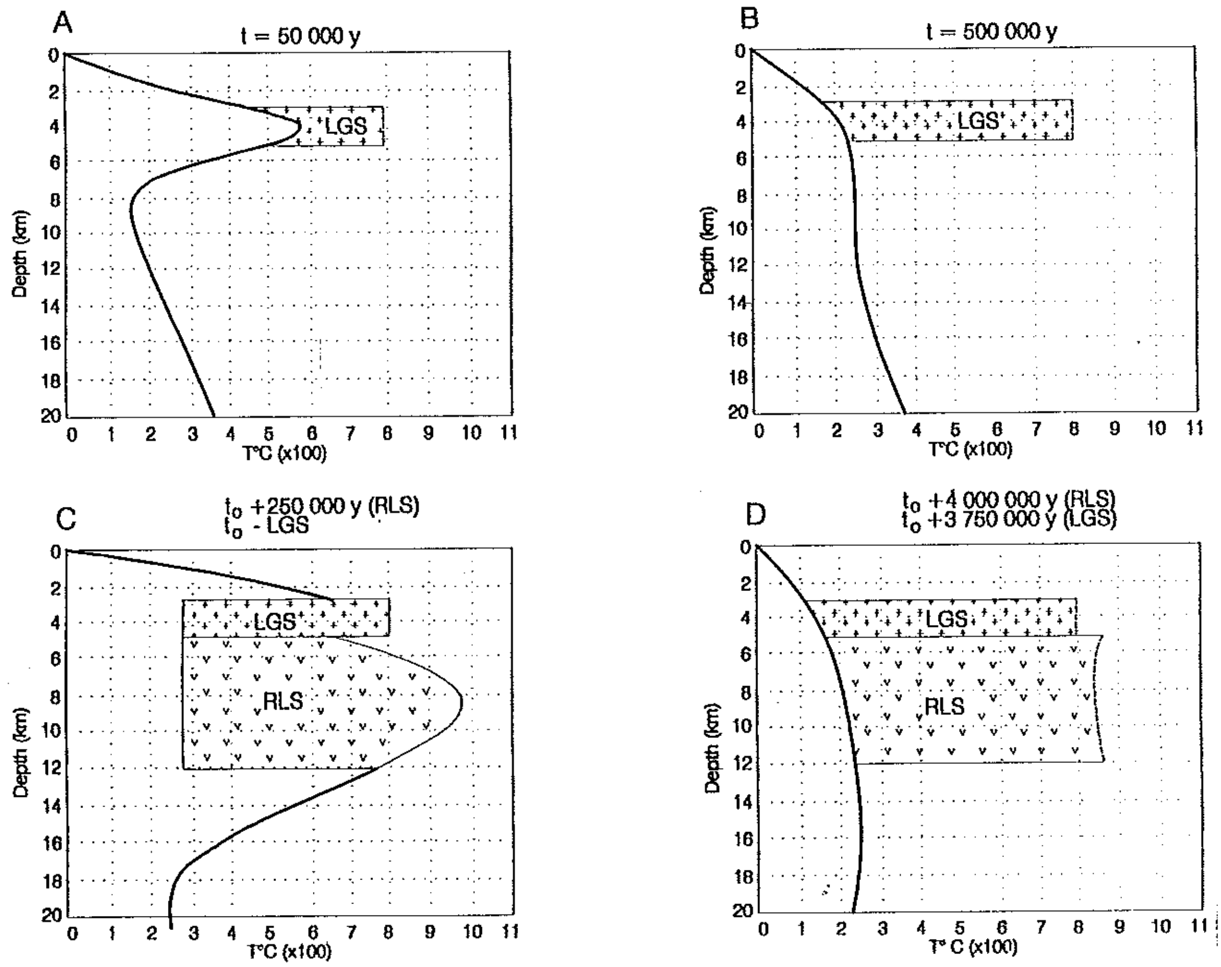

Figure 7: *Modelled thermal profiles of the crust as a function of time for different magma emplacement scenarios applicable to the Bushveld Complex (calculated using the programme THERMOS described in Cawthorn and Walraven, 1998). A and B:- thermal profiles of crust intruded by a 2 km thick felsic intrusion (the Lebowa Granite Suite - LGS) at a depth of 3-5 km 50 000 y and 500 000 y after emplacement. Liquidus temperature of the LGS assumed to be 800°C. C and D:- thermal profiles of crust that had been intruded by a 7 km thick mafic intrusion (representing the Rustenburg Layered Suite – RLS - with a liquidus temperature assumed to be 1300°C) 250 000 y before emplacement of the same felsic intrusion as represented in A and B immediately above the mafic sill. Restoration of the thermal profile to proportions similar to that in B in this case takes 4 my.*

behaviour in the LGS that promoted isotopic exchange for up to 1000 my after emplacement. U-Pb isotopic analyses of multiple zircon populations (Coertze *et al.*, 1978; Walraven *et al.*, 1981, 1982, 1983) typically provide highly discordant isotopic ratios reflecting metamictization and Pb loss, which in turn, provide minimum age estimates in the range 2000 Ma to 1500 Ma (Fig. 8). Whole rock Rb-Sr isotopic determinations for the LGS also typically yield either isochrons or errorchrons yielding young apparent ages which are considered to reflect selective removal of radiogenic Sr from feldspar by circulating late-stage fluids (Walraven *et al.*, 1990). Similarly, the Pb-Pb isotopic determinations on whole rocks and mineral separates from the host granites to the Zaaiplaats Mine yield apparent ages between 1187 Ma and 961 Ma (Fig. 8), some 1000 my younger than LGS emplacement. This has been interpreted to reflect continuous open

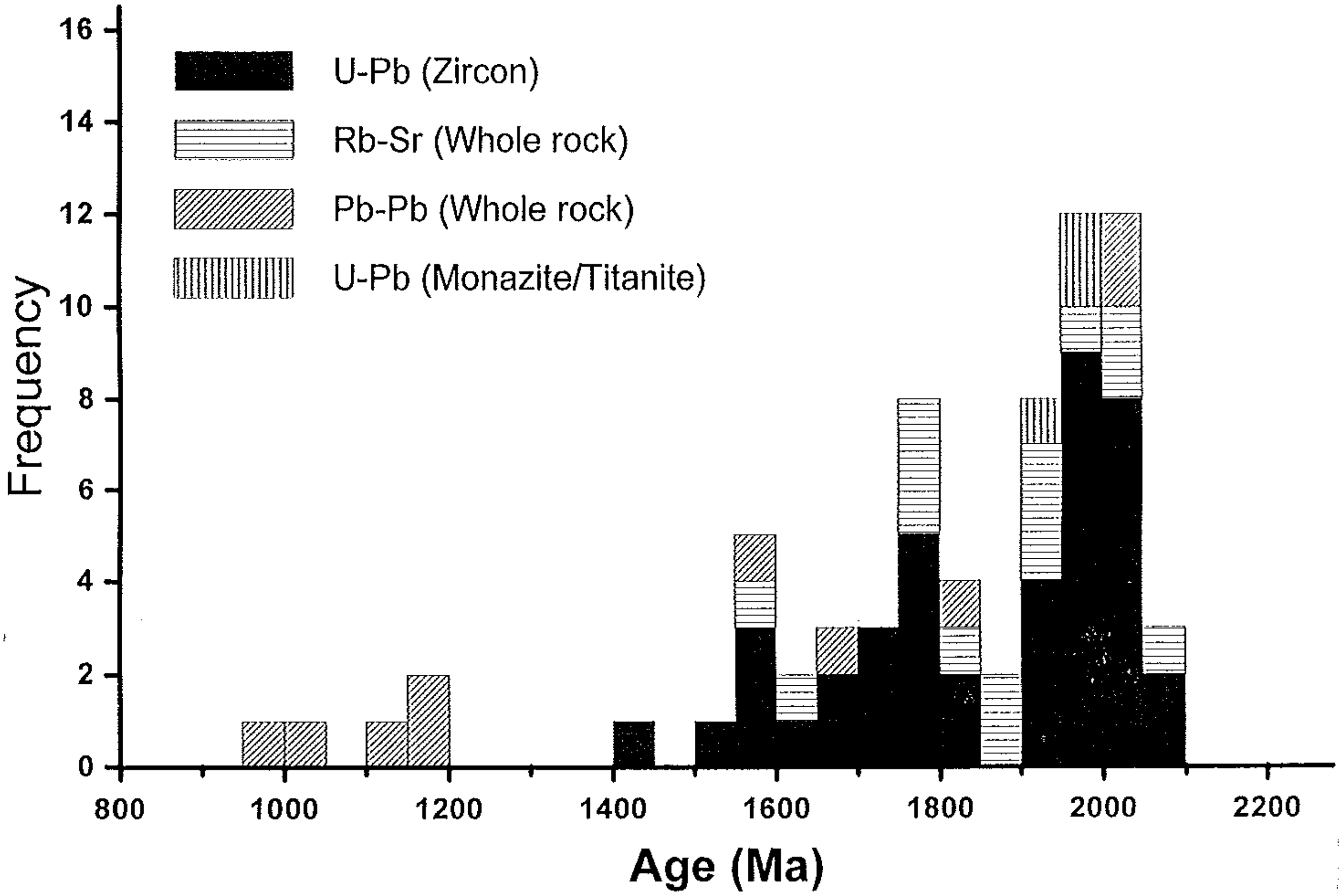

Figure 8: *Frequency histogram showing a compilation of all age determinations from the LGS and Rooiberg Group (U-Pb zircon data are from Coertze et al., 1978; Walraven et al., 1981,1982,1983; Walraven and Hattingh, 1993 and Walraven, 1997; Rb-Sr whole rock data are from Walraven et al., 1985; Walraven, 1987; Walraven et al., 1990; Hamilton, 1977 and Davies et al., 1970; Pb-Pb whole rock and mineral data are from Walraven, 1988 and McNaughton et al., 1993; U-Pb monazite data are from Nicolaysen et al., 1958 and Burger et al., 1967).*

system behaviour and Pb isotopic exchange, stimulated by the high heat productive capacity of the mineralized portions of the LGS, until the closure temperature for the Pb isotope system was reached during exhumation of the complex in the Mesoproterozoic (McNaughton *et al.*, 1993). These data do not provide anything more than broad constraints on the absolute timing of post-emplacement events, but they do collectively confirm the very long-lived and pervasive nature of fluid circulation and mineralization in the LGS.

AGE OF AUTHIGENIC ZIRCON IN MINERALIZED LGS

The study of the ore minerals in polymetallic, fracture-related, sulphide deposits of the LGS, using cathodoluminescence imagery, revealed the presence of very small (<100 micron) authigenic zircons which have grown together with late-stage hydrothermal quartz. A number of authigenic zircons, identified in samples from the Spoedwel Mine and surrounds, have been analysed for their U-Pb isotopic ratios on the SHRIMP at the Research School of Earth Sciences, Australian National University. The data is presented in Table 2 and plotted on a Tera-Wasserburg plot in Figure 9. The data show that the authigenic zircons all formed at

Table 2. Summary of SHRIMP U-Th-Pb zircon results

Grain spot	U (ppm)	Th (ppm)	Th/U	Pb* (ppm)	$^{204}Pb/^{206}Pb$	f_{206} %	Radiogenic Ratios $^{206}Pb/^{238}U$	±1σ	$^{207}Pb/^{235}U$	±1σ	$^{207}Pb/^{206}Pb$	±1σ	Ages (in Ma) $^{206}Pb/^{238}U$	±1σ	$^{207}Pb/^{235}U$	±1σ	$^{207}Pb/^{206}Pb$	±1σ	Conc. %
A-1.1	3018	77953	26	421	0.036334	54.9	0.0839	0.0035	0.930	0.119	0.0804	0.0093	519	21	668	64	1208	246	43
B-1.1	3647	670	0.2	861	0.003520	5.32	0.2072	0.0081	3.200	0.133	0.1120	0.0011	1214	44	1457	33	1833	19	66
B-2.1	3011	1783	0.6	1219	0.000727	1.10	0.3814	0.0152	6.362	0.286	0.1210	0.0020	2083	72	2027	40	1971	29	106
B-2.2	1353	365	0.3	755	0.000655	0.99	0.5308	0.0209	9.093	0.364	0.1243	0.0005	2745	89	2347	37	2018	6	136
B-2.3	1452	512	0.4	828	0.000539	0.82	0.5401	0.0217	9.418	0.437	0.1265	0.0024	2784	91	2380	44	2049	34	136
B-3.1	1391	537	0.4	273	0.000241	0.37	0.1859	0.0073	2.748	0.111	0.1072	0.0006	1099	40	1342	31	1753	10	63
C-1.1	1635	1922	1.18	297	0.015750	23.8	0.1276	0.0051	1.824	0.132	0.1036	0.0058	774	29	1054	49	1690	107	46

Notes: 1. Uncertainties given at the one σ level. 2. f_{206}% denotes the percentage of ^{206}Pb that is common Pb. 3. Correction for common Pb made using the measured $^{204}Pb/^{206}Pb$ ratio. 4. For % Conc., 100% denotes a concordant analysis.

approximately the same time, around 1957±15 Ma, some 100 my after emplacement of the LGS. These relatively precise data confirm the suggestion that hydrothermal activity was present in the granites at least 100 my after the cessation of orthomagmatic fluid flow and lend support to the suggestions that mineralization representing the intermediate, sulphide-dominated stages of the paragenetic sequence is related to the incursion of externally derived fluids. The exact relationship between the timing of authigenic zircon growth and its position in the paragenetic sequence is difficult to constrain. However, the quartz, which contains the authigenic zircons at the Spoedwel Mine, is associated with pyrite and chalcopyrite, suggesting that the 1960 Ma event may reflect the precipitation of base-metal sulphides in the metallotect and, therefore, predate the deposition of the late, oxidized Fe-F-U paragenesis.

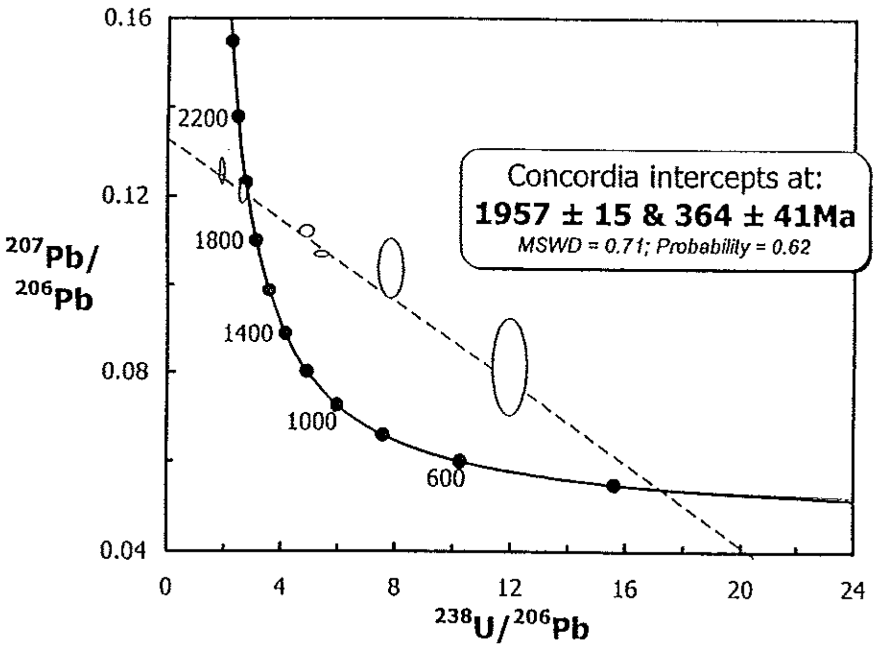

Figure 9: $^{207}Pb/^{206}Pb$ versus $^{238}U/^{206}Pb$ plot (Tera-Wasserburg concordia) for analyses of authigenic zircons associated with late hydrothermal quartz in the Spoedwel Mine (data from Table 3).

AGES OF MINERALIZING EVENTS IN THE LGS

In order to rationalize the plethora of radiometric isotope data applicable to the LGS and related rocks, an attempt is made in Figure 10 to compile age data that is based either on U-Pb or Pb-Pb isotope analyses of mineral separates (i.e. zircon, monazite, etc.), or on whole-rock Pb-Pb and Rb-Sr isotopic ratios which display reasonably well-constrained isochrons. Excluded from this selection are U-Pb isotopic determinations on metamict multiple zircon populations that are so discordant as to yield meaningless minimum ages. Although this process is subjective it does provide potential insights into the timing and episodicity of fluid flow and open system behaviour in the LGS.

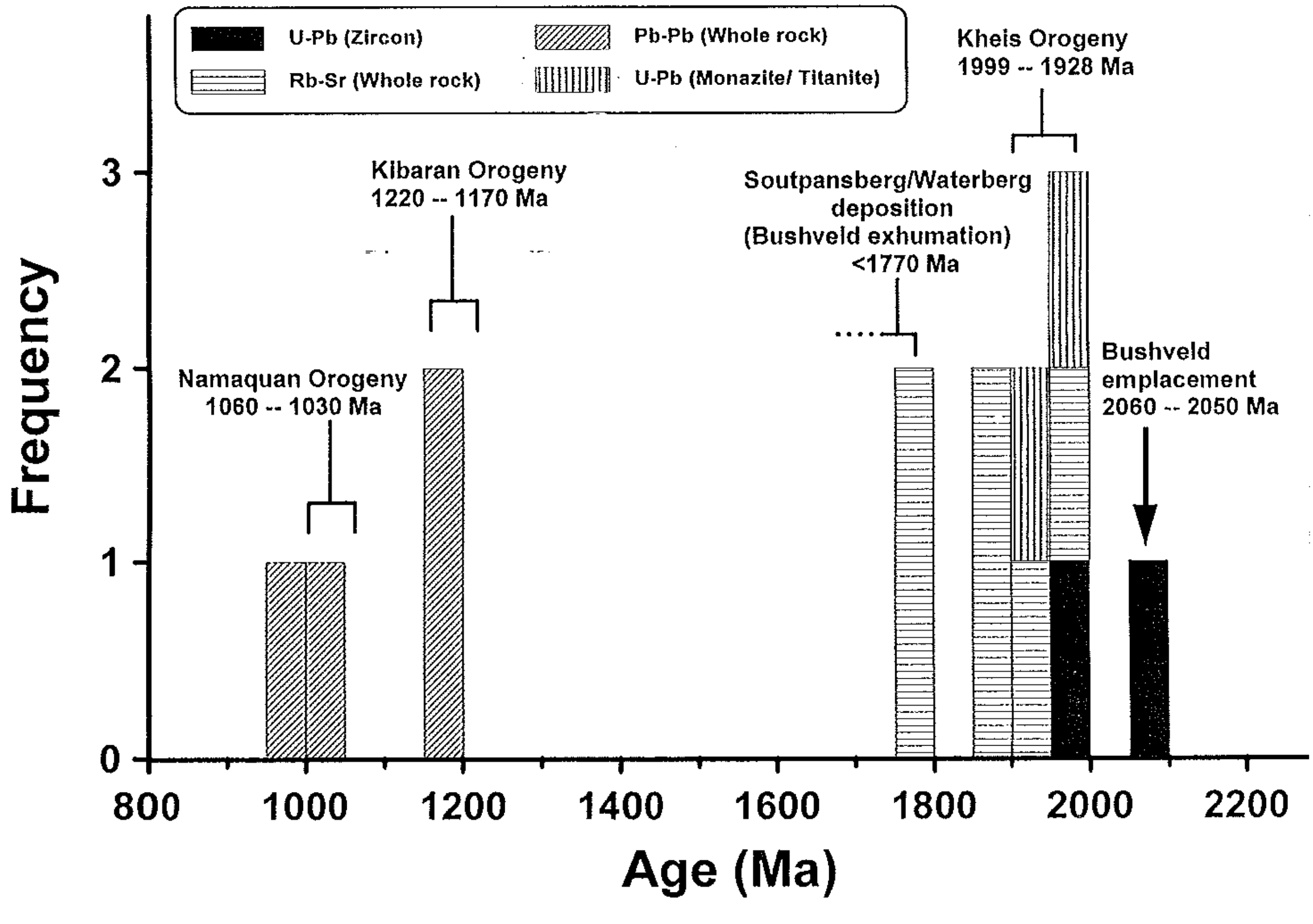

Figure 10: Frequency histogram showing ages for the LGS selected on the basis of accuracy and precision (i.e. single- or small-grain population U-Pb zircon or monazite ages or reasonably well constrained Rb-Sr and Pb-Pb isochrons). Also shown are the presently available age constraints for the Kheis orogeny (Kruger et al., 1999), Soutpansberg/Waterberg deposition (SACS, 1980), and the Kibaran and Namaquan orogenies (Robb et al., 1999).

In addition to the 1960 Ma age for authigenic zircons obtained in the present study, Nicolaysen *et al.* (1958) and Burger *et al.* (1967) produced U-Pb isotopic data for monazite separates from the LGS that yielded ages ranging from 1993+90/-78 Ma to 1955±37 Ma. Concentrations of REE are common in the polymetallic hydrothermal ores of the LGS and these data appear to support the suggestion made above that the principal period of mineralization occurred some

100 my after emplacement of the Complex. Studies that have yielded Rb-Sr isochrons from the LGS also point to apparent ages that are similar to the mineral ages mentioned above (i.e. 1982±63 Ma, Walraven *et al.*, 1985, and 1946±45, Davies *et al.*, 1970) and 70 -100 my younger than the emplacement age of the Bushveld Complex. It is pertinent to note that the period between 2000 Ma and 1900 Ma overlaps with the Kheis Orogeny (presently constrained between 1999 Ma and 1928 Ma; Kruger *et al.*, 1999), a major fold and thrust belt associated with continental accretion along the western margin of the Kaapvaal Craton.

Other Rb-Sr data, however, suggest a significantly younger resetting event in the range 1870 Ma to 1780 Ma (Hamilton, 1977; Walraven, 1987). It is suggested that these ages might be related to the progressive exhumation of the Bushveld Complex in the Palaeoproterozoic. This is supported by the fact that the LGS is unconformably overlain in several places by conglomerates of the Waterberg Group, the deposition of which, although not well constrained, was initiated at or around 1770 Ma (SACS, 1980). Uplift and exhumation of the granites would have exposed them to circulation of meteoric fluids, which might have been responsible for the formation of the late Fe-U-F paragenesis in high-level, mineralized portions of the LGS. The Rb-Sr age data may, therefore, be reflecting the closure of this isotope system as the Complex passed through the relevant blocking temperatures during exhumation.

A final event in the LGS appears to be reflected in the Pb-Pb isochrons obtained from a variety of the host granites at the Zaaiplaats tin mine. Although the cassiterite mineralization represents an early stage in the paragenetic sequence and is associated with magmatic fluids, the Pb-Pb isotope systematics clearly identify a pervasive, but very late, set of events at between 1187±51 Ma and 961±129 Ma (McNaughton *et al.*, 1993). The fluid inclusion studies of Ollila (1981) and Pollard *et al.* (1991) identified a low-temperature secondary fluid population that could conceivably have been associated with re-setting of the Mesoproterozoic Pb-Pb ages observed at Zaaiplaats. It is also pertinent to note that the Zaaiplaats Pb-Pb ages are very similar to the ages of the two major Mesoproterozoic orogenic events that occurred to the south and west of the Kaapvaal Craton, termed the Kibaran and Namaquan orogenies. These have recently been constrained chronologically to 1220-1170 Ma and 1060-1030 Ma, respectively (Robb *et al.*, 1999).

DISCUSSION AND CONCLUSIONS

The timing of fluid flow events in the LGS is difficult to constrain, although radiogenic isotope data clearly point to long-lived, open-system chemical re-equilibration that selectively homogenizes isotopic ratios for up to 1000 my after granite emplacement. Constraining the isotopic data by selection of ages on the basis of accuracy/precision and analytical technique provides a framework which suggests that the extended paragenetic sequence in the LGS is related to episodic fluid flow, the latter related to tangible geological events that occurred on or off the Kaapvaal Craton during the Palaeo- and Mesoproterozoic. Magmatic fluid dominated hydrothermal circulation was active for less than 5 my after granite emplacement (i.e. between 2054 and 2050 Ma) as suggested by thermal modelling of the Bushveld intrusion and the age data for cassiterite from the Rooiberg Mine (2049 Ma). The early Sn-W-Mo-F magmato-hydrothermal mineralization is, therefore, attributed to this time period and duration.

However, age data for authigenic zircon associated with polymetallic sulphide mineralization representing the intermediate stage of the paragenetic sequence, indicates that these ores formed some 100 my after granite emplacement, at around 1960 Ma. The precipitation of these ores is clearly unrelated to magmatic fluids (*sensu stricto*) and processes are possibly related to mingling between the evolved, residual magmatic fluid repository and externally derived fluids of connate origin. The timing of this fluid flow event is broadly coincident with the Kheis Orogeny, which occurred along the western margin of the Kaapvaal Craton between about 1999 Ma and 1928 Ma (Kruger *et al.*, 1999).

The timing of uplift and exhumation of the Bushveld Complex is also difficult to constrain although it is well known that conglomerates of the Waterberg Group, laid down at or around 1770 Ma, unconformably overlie the LGS. Rb-Sr age data, as well as other re-set isotopic systems, indicate that closure of these isotope systems occurred at around 1800 Ma. Exposure of the upper portions of the LGS to the surface at this time could conceivably have been responsible for the introduction of meteoric fluids into existing conduits created during earlier stages of fluid flow and mineralization. It is tentatively suggested that this fluid regime might have been responsible for the formation of the late, oxidized, Fe-F-U ore paragenesis in the LGS. The timing of meteoric fluid circulation is uncertain and may well have persisted, albeit episodically, right through the Mesoproterozoic when these fluids were responsible for the re-setting of Pb isotopic systems in the host granites of the Zaaiplaats Mine between 1200 Ma and 1000 Ma (McNaughton *et al.*, 1993). It is also possible that the very late fluid fluxes were unrelated to mineralization *per se*, but were nevertheless responsible for pervasive open system chemical behaviour long after the mineralizing systems had closed down.

Causes of Late-stage, Externally Derived Fluid Flow in the LGS

The scenario being advocated in the present study differs from the steady-state model proposed by McNaughton *et al.* (1993) in which an internally derived fluid circulated through the high heat-productive portions of the LGS for up to 1000 my after emplacement. The present study suggests that, although fluid flow events did indeed persist in the LGS for considerably longer periods of time than is conventionally accepted for magmato-hydrothermal systems, the fluids were externally derived and flowed episodically during specific extrinsic geological events that not only affected the Bushveld Complex, but also affected other portions of the craton. Large-scale fluid circulation in the mid- to upper-portions of the crust are known to occur in response to several factors, the most important of which are regional thermal gradients, crustal advection, regional dehydration reactions related to metamorphism, topographic head and deformation-induced hydraulic head (Oliver, 1996). The concept of deformation-induced, or "orogeny-driven", fluid flow has gained widespread acceptance since the idea was first proposed by Oliver (1986). Koons and Craw (1991), for example, have shown that zones of major compression-related orogeny, such as the Alps and Himalayas, are characterized by complex fluid flow regimes, the driving forces of which combine topographic head-driven meteoric flow, compaction-related connate flow and disequilibrium dehydration related to the accompanying metamorphism. Particularly relevant is the observation that the outboard side of a collisional orogen is characterized by meteoric and connate fluids, the circulation of which can extend for hundreds of kilometres from the thrust front. Such a scenario has also been advocated for the development of large mineralized districts such as the Pb-Zn and petroliferous deposits of the southwestern USA (Oliver, 1986; Ge and Garven, 1992).

Duane and Kruger (1991) have previously related the Kheis Orogeny to the generation of tectonically driven fluids responsible for pervasive isotope re-setting and Pb-Zn mineralization over large areas of the western Kaapvaal Craton. Although there are no accurate age constraints for the Kheis Orogeny, all available isotopic data straddle the 1960 Ma age suggested in this study to represent the timing of the intermediate, base-metal sulphide stages of the mineralizing system in the LGS. Support for the existence of pervasive, craton-wide, orogeny-driven, fluid flow at this time is provided by the widespread re-setting of Rb-Sr isotope systematics in a variety of Palaeoproterozoic rock sequences on the craton (Duane and Kruger, 1991). The fact that the fluid itself was meso- to epithermal and connate in character is supported by the association of Mississippi Valley type Pb-Zn deposits, in dolomites of the Transvaal Supergroup along the western edges of the craton, with this event (Duane and Kruger, 1991). The circulation of this fluid is considered to be related to the creation of a deformation-induced hydraulic head during the stage of thin-skin fold-and-thrust tectonics that deformed Kheis-related sequences along the western edge of the craton in the Palaeoproterozoic (Hartnady *et al.*, 1985). The extent of the circulation, and the nature of the fluid conduits, for this orogeny-driven, fluid-flow event are, however, not well understood at this stage.

Incursion of oxidizing meteoric fluids during exhumation of the Bushveld Complex is considered here to have been temporally related to deposition of the intra-continental red-bed sediments of the Waterberg Group at or around 1770 Ma. This link remains tenuous at this stage as the actual timing of the late-stage paragenetic sequence in the LGS is not known. Very late fluid flow and chemical homogenization, at circa 1100 Ma, as indicated in the Pb-Pb isotopic studies at Zaaiplaats, is probably not related to uplift of the Bushveld Complex at this stage, as suggested by McNaughton *et al.* (1993). Rather, it may again be related to the circulation of deformation-driven fluids during the major Mesoproterozoic orogenic events to the S and SW of the craton. The tectonic model of Jacobs *et al.* (1993) identifies the Kaapvaal Craton as a southerly directed indentor into existing Palaeoproterozoic crust at this time, a scenario that would, in all likelihood, have resulted in circulation of deformation-driven fluids on the craton itself (Thomas *et al.*, 1994).

In summary, magmato-hydrothermal input to mineralization in the LGS is now considered to have been restricted to the dominantly Sn ores formed within about 5 my of emplacement of the granite. Later, polymetallic, base-metal sulphide-dominated stages of the paragenetic sequence are suggested to have been related to an event in which the residual magmatic fluid repository mingled with externally derived fluids of possible connate character circulating on a craton-wide scale in response to the Kheis Orogeny some 100 my after granite emplacement. Subsequent to this, oxidizing meteoric fluids are considered to have prevailed, giving rise to the late Fe-F-U dominated paragenesis. These fluids circulated during exhumation of the Bushveld Complex at about 1770 Ma, but may also have been stimulated by an orogeny-driven hydraulic head as late as the Mesoproterozoic, some 1000 my after granite emplacement. If correct, this scenario supports models which recognize a link between the formation of major mineral provinces and episodic, regional tectonism/deformation/uplift through the medium of craton-wide fluid flow in the mid- and upper crust.

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