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An isotopic study on the volcanics of the Rooiberg Group: age implications and a potential exploration tool

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Abstract. Many geochronological studies on silicic magmatic rocks associated with the Bushveld Complex (rhyolitic lavas of the Rooiberg Group and granites of the Lebowa Granite Suite) have shown evidence of open-system behaviour of the Rb–Sr and Pb–Pb isotopic systems until 1600-1000 Ma, many hundreds of million years after crystallisation of these rocks. This pervasive open-system behaviour has been attributed to sustained hydrothermal circulation driven by the high heat productivity of the Bushveld granites. New Sr and Pb isotopic data are presented for basaltic to rhyolitic volcanics from the Rooiberg Group of the Transvaal Sequence in the Dullstroom-Loskop Dam area of the eastern Transvaal. These data show little evidence of open-system behaviour after about 1950 Ma and many sample suites retain ages which could reflect the formation of the Rooiberg Group i.e. older than 2070 Ma. It is argued that this preservation is due to the absence of fractionated, fluid/vapour-rich Bushveld granites in the immediate vicinity of the volcanic occurrences. Rooiberg Group volcanics with extensively perturbed Rb–Sr and particularly Pb–Pb isotopic systems reflect the action of granite-derived hydrothermal fluids. As a consequence, the isotope systematics in these volcanics could prove a useful exploration tool for sites of granite-derived metal deposits.

Deposition of the Transvaal Sequence terminated with the eruption of large volumes of volcanic and volcaniclastic rocks of basaltic andesite to rhyolitic composition which make up the Dullstroom Formation of the Pretoria Group and the Rooiberg Group stratigraphic units (terminology of SACS, 1980). These volcanics were intruded by components of the Bushveld Complex: mafic magmas which gave rise to the Rustenburg Layered Suite (RLS) and subsequently by granitic magmas which formed the sheeted granite of the Lebowa Granite Suite (LGS).

Several isotopic studies on the Rooiberg rhyolites and the LGS granites have yielded anomalously young dates. Low ages in the LGS rocks have been variously ascribed to post-crystallisation metasomatism by heated groundwaters producing selective loss of radiogenic ^{87}Sr (Walraven et al., 1985) or the maintenance of long-lived hydrothermal systems in the granites because of elevated K, U and Th concentrations (McNaughton et al., 1993). These post-solidification modifications are thought to have had an important influence in the mineralisation of the LGS granites (e.g. Walraven et al., 1990b; Robb et al., 1994).

This contribution presents new Sr and Pb isotope data on representative units of the Dullstroom-Rooiberg volcanic succession in the Dullstroom-Loskop Dam area of the eastern Transvaal (Fig. 1). This sample set provides several useful constraints on the post-solidification history of the volcanics which form both the roof and floor of the LGS in this region and has significance in the formulation of exploration models for ore concentrations deposited from granite-sourced hydrothermal fluids.

Stratigraphy of Rooiberg Group

Stratigraphic relationships between the Rooiberg Group and Dullstroom Formation

Until relatively recently, the Dullstroom Formation volcanics were regarded as the uppermost preserved unit of the Pretoria Group of the Transvaal Sequence (terminology and classification following SACS, 1980). Evidence collected during systematic studies of the volcanology and petrology of the Rooiberg felsites (Twist, 1985; Twist and Harmer, 1987) and Dullstroom eruptives (Schweitzer, 1985, 1986) in the Dullstroom-Loskop Dam area of the eastern Transvaal have indicated that the volcanics comprising these two stratigraphic units form part of a continuous eruptive sequence (Schweitzer, 1986; Harmer and Von Gruenewaldt, 1990; Eriksson et al., 1993).

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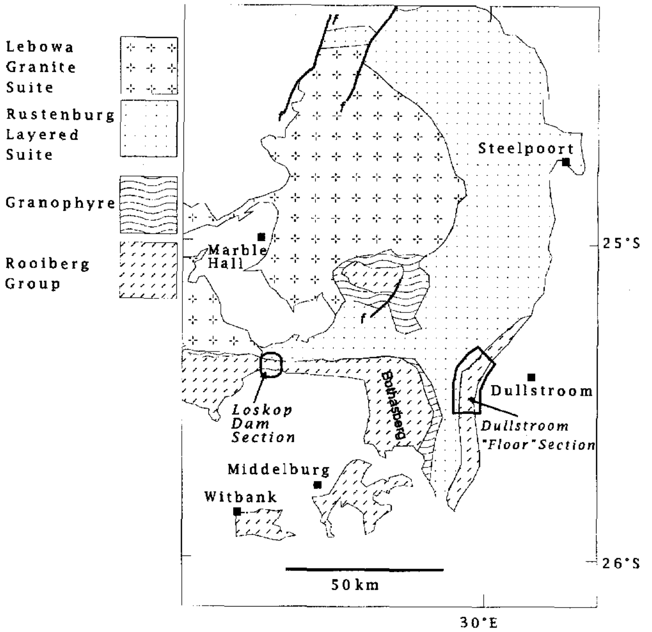

Fig. 1. Geological sketch map showing the Rooiberg Group occurrences discussed in the text

Table 1. Comparison between SACS (1980) stratigraphic terminology and revised lithostratigraphy of the Dullstroom and Rooiberg volcanics (Schweitzer et al., in press)

SACS (1980)		Schweitzer et al. (in press)	
Rooiberg Group	Selons River Formation	Rooiberg Group	Klipnek Formation
	Damwal Formation		Schrikkloof Formation
Pretoria Group	Dullstroom Formation		Damwal Formation
			Dullstroom Formation

As a result, the stratigraphy of the Rooiberg Group has been re-evaluated by Schweitzer et al. (in press) who argue that the Dullstroom Formation should be considered part of the Rooiberg Group rather than the Pretoria Group. The recognition of an erosive base to the Dullstroom volcanics (Cheney and Twist, 1988) lends further support to this interpretation. Based on a regional correlation of the lithogeochemistry of the Rooiberg volcanics, a new stratigraphic subdivision of the Dullstroom-Rooiberg volcanics has been proposed (Schweitzer et al., in press) and this re-classification, presented in Table 1, will be adopted in this paper.

Volcanic stratigraphy of the Rooiberg Group

In the current (SACS, 1980) classification the Dullstroom Formation eruptives occur only below the level of intrusion of the RLS components and comprise about 80 flows of predominantly basaltic andesite composition with minor basalt, dacite and rhyolitic units. Two chemical sub-classes are distinguishable in the intermediate components: a high iron-titanium group ($Fe_2O_3^{Total}$ 11–15%; TiO_2 1.2–2.2%) and a more common low-titanium group having TiO_2 and $Fe_2O_3^{Total}$ below 1% and 11% respectively. The Rooiberg acid eruptives form the roof of the RLS in the Loskop Dam area where Twist (1985) subdivided the ca. 3.5 km thick volcanic succession into 9 units on the basis of colour, texture, phenocryst content and internal structure. Two contrasted compositional types of volcanic are recognised: a high magnesian variety (HMF = high magnesian felsite; MgO > 1.7%) and a low magnesian type (LMF = low magnesian felsite; MgO < 1.0%). LMF flows are found throughout the succession at Loskop Dam whereas HMF flows are found interbedded with LMF flows only in the lowest two volcanic units (i.e. Units #1 and #2).

Schweitzer (1985) demonstrated that the HMF flows in the Loskop Dam succession are the compositional equivalents of the low-TiO_2 of the Dullstroom Formation (terminology of SACS, 1980) whereas Harmer and Von Gruenewaldt (1991) highlighted the compositional similarity between the rhyolites erupted at the base of the Dullstroom and the HMF type. The persistence of the low iron-titanium magma type into the lower units of the Rooiberg Group has led to two important stratigraphic changes: firstly, the Dullstroom Formation is now considered a component of the Rooiberg Group and secondly, the Dullstroom Formation is extended to include the volcanics in Units #1 and #2 in the Loskop Dam area (Schweitzer et al., in press).

The age of the Rooiberg Group

Components of the RLS cut across the stratigraphic layering of the Pretoria Group and in the stratigraphically highest (southern) part of the eastern compartment of the Bushveld Complex, units of the RLS split the Dullstroom Formation of the Rooiberg Group. It is therefore apparent that the eruption of the lowest formations of the Rooiberg Group must have occurred prior to ca. 2050–2060 Ma, the currently accepted age of the RLS (Walraven et al., 1990a). Granites of the LGS were emplaced after the RLS mafic magmas and are thus younger than the Rooiberg volcanics. The Nebo Granite component of the LGS has been precisely dated by the zircon evaporation technique at 2054.4 ± 1.8 Ma by Walraven and Hattingh (1993). The Rooiberg volcanics must therefore be older than 2054 Ma. From field relationships, then, it is not possible that the Rooiberg Group can represent the volcanic equivalent of the Lebowa Granites. Significant compositional differences exist between the volcanics and granites (see discussion in Twist and Harmer, 1987) providing additional evidence against such a relationship.

In the Loskop Dam area, the uppermost Rooiberg Group and the overlying Loskop Formation sediments are intruded by a sheet of quartz porphyry (Clubley-Armstrong, 1977; Faurie, 1977) termed the Rooikop "Granophyre Porphyry" by SACS (1980). Zircons from this porphyry were analysed by Faurie (1977) and provide a date of $2072^{+23}/_{-22}$ Ma (errors are 95% confidence limits) when these data are recalculated following Eglington and Harmer (1993) by weighing the data for upper intercept. The data points show scatter in excess of the analytical uncertainty ("errorchron") and so the uncertainties are sensitive to the approach adopted to allow for this scatter. The age error limits quoted here were derived by augmenting the statistical uncertainties on the regression line by $(MSWD/F)^{1/2}$. This result constrains the age of the Rooiberg Group formations to be greater than 2050 Ma and possibly older than 2072 Ma.

Determining the absolute age of the Rooiberg Group is complicated by the fact that zircons are not found in the rhyolitic components: Zr increases progressively through the volcanic sequence at Loskop Dam (Twist, 1985; Twist and Harmer, 1987) indicating that, presumably because of elevated eruption temperatures, zircon crystallisation was suppressed in these lavas.

A Rb-Sr study of 12 Rooiberg Group rhyolites from the Witbank and Bothasberg areas by Walraven et al. (1985) yielded a date of 1604 ± 28 Ma and extremely high initial $^{87}Sr/^{86}Sr$ of 0.732 ± 2 (re-calculation as quoted in Walraven et al., 1990a; MSWD = 3.75). This date is too young to represent the time of crystallisation of the rhyolites. The same samples yielded a highly imprecise Pb–Pb age estimate of $2003^{+289}/_{-360}$ (Walraven et al., 1990; MSWD = 9.43).

Isotope data

Sampling

Dullstroom Formation volcanics from the "floor" of the RLS. Rhyolitic samples were collected from the zone of basal rhyolites of the Dullstroom Formation which tend to form individual flows less than 8 km in strike length. The rhyolites are sparsely porphyritic with feldspar and augite phenocrysts variably replaced by sericite and chlorite, respectively. Lath-like augite phenocrysts are more abundant than the coarser-grained feldspar crystals. Swallow-tailed, polycrystalline quartz needles are common and presumably represent pseudomorphs after tridymite (Twist and French, 1983). The groundmass is wholly devitrified to a granular intergrowth of albite and quartz. Sericite and chlorite alteration is widespread throughout the groundmass and epidote is conspicuous. Basaltic andesite samples show ophitic textures with plagioclase and swallow-tailed amphibole crystals set in a fine, microlitic groundmass. Basalts tend to be porphyritic (phenocryst contents up to 20%) with plagioclase, clinopyroxene and amphibole grains frequently clustering in glommeroporphyritic textures. Magnetite phenocrysts are ubiquitous.
Rooiberg Group volcanics from the Loskop Dam area. Where possible, samples were taken from identifiable individual flows in the succession. Samples from the Loskop Dam section spanned most of the volcanic units delineated by Twist (1985) and include both high-MgO lava and low-MgO (LMF) types.

High-MgO samples were collected from flows in Unit #2 and are generally holocrystalline, consisting of a fine intergrowth of sheaf-like albite and augite grains. "Swallow-tailed" albite grains are present. Chlorite and hornblende pseudomorphs, presumably after augite, are evident in some sections. Titanomagnetite is ubiquitous and comprises up to 1% of the samples. Patchy chlorite and sericite alteration is common.

Low-MgO rhyolites from Unit #1 (RDW-1 to 10; Rb-11) have large lath-like albite phenocrysts in a devitrified groundmass of very fine quartz and feldspar. Unlike the previous sample set, no fresh pyroxene is found. Fine titanomagnetite is present throughout the groundmass constituting up to 1 modal percent. Some phenocrysts are sericitised and sericite and chlorite patches are evident in the groundmass. While still remarkably preserved for ancient fine-grained volcanics, these volcanics are more altered than those from the Unit #4 flow.

Samples from Unit #4 (DF-1 to 9 and 285/81, 313/81, 314/81) are dark grey low-MgO rhyolites containing 3–5% phenocrysts of augite, albite and titanomagnetite set in a groundmass of fine albite and augite laths with cryptocrystalline potash feldspar and quartz. Relict patches of volcanic glass are preserved in the groundmass of some samples. Excellent supercooling textures are noted including "swallow-tailed" albite crystals, sheaf-like aggregates of augite and elongate, skeletal titanomagnetite growths. Some albite phenocrysts exhibit minor sericitisation and epidote pseudomorphs are rarely seen. Preservation of augite and the barely devitrified glassy matrix testify to the essentially unaltered nature of the samples from this flow.

The remaining low-MgO volcanics are from Units 3,6,8 and 9 and most have a marked red colour in hand specimen. Phenocryst phases are extensively sericitised feldspar and, less commonly, quartz. The original glassy groundmass is wholly devitrified to a granophyric intergrowth of quartz and feldspar. Matrix quartz is coarser-grained than in the previously discussed sample groups and can be clearly distinguished microscopically. Sericite and chlorite alteration is widespread and epidote is also present; all mafic constituents are secondary. Red iron-oxide staining is pervasive throughout most samples and is presumably due to hematite exsolution during extensive devitrification.

Analytical methods

Samples were digested in full strength HF-HNO_3 mixtures in screw-top FEP Teflon vials and slowly taken to dryness with extra HNO_3. The dried sample was then dissolved in 6 M HCl and dried. Sr and Rb were separated on 10 mm internal diameter columns packed with 6 ml of AG50 W-$\times$12, 200–400# cation exchange resin using 2.5 M HCl as the eluant. For Pb analyses, the samples were converted to bromide with 1 M HBr and loaded onto quartz glass micro-columns packed with ca. 40 µl of purified AG1$\times$8 200–400# mesh resin in 0.5 M HBr. Other elements were removed by washing with 0.5 M HBr and the Pb stripped using H_2O. A second pass through the columns was used to further purify the Pb. All reagents were repeatedly distilled and, in the case of H_2O and HBr, additionally cleaned through ion exchange resins. Total method blanks never exceeded 1 ng for Sr and Rb, and 0.5 ng for Pb.

Sr and Pb analyses were performed on a VG354 multi-collector mass spectrometer whereas Rb concentrations, and some Sr concentration and isotopic analyses were performed on a single-collector VG MM30 spectrometer. $^{87}Sr/^{86}Sr$ ratios were corrected for instrumental fractionation by normalising to $^{86}Sr/^{88}Sr$ of 0.1194. Fractionation of Pb isotopes was corrected by factors determined empirically from analyses of the NBS Pb standard SRM981 analysed in the same sample batch as the unknowns: at least two Pb standards were analysed per eight unknowns. Analytical uncertainties were assessed using duplicate analyses of rock standards and unknowns and are: 0.8% for $^{87}Rb/^{86}Sr$; 0.02% and 0.01% for $^{87}Sr/^{86}Sr$ determined on the MM30 and VG354 spectrometers respectively; 0.09% for $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ and 0.15% for $^{208}Pb/^{204}Pb$. The correlation between the errors in $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ was assessed as 0.95. Rb and Sr concentration measurements determined by X-ray fluorescence spectrometry have an uncertainty in $^{87}Rb/^{86}Sr$ of 1.5%. Repeated analyses of $^{87}Sr/^{86}Sr$ in the NBS Sr standard SRM 987 during the period of this study yielded 0.710271 ± 30 (1σ).

All regression calculations were performed following the recommendations described in Harmer and Eglington (1990). Uncertainties calculated for regression parameters where "geological scatter" is identified (i.e. "errorchrons" – tested using an F static based on errors determined on 60 replicates) are augmented by $(MSWD/F)^{1/2}$. All uncertainties are given as 95% confidence intervals.

Data

Results are presented in Table 2 and summaries of the various regression calculations discussed below are provided in Table 3.

1. Dullstroom Formation. Of the three sample suites analysed from the Dullstroom Formation volcanics cropping out below the RLS, only the basal rhyolites have sufficient range in Rb/Sr ratios to provide reasonably precise dates. All three suites yield Rb-Sr dates that are within error (admittedly extremely large) of 2080 Ma. Regressed on their own, the 10 basal rhyolite samples yield a date of 2037 ± 92 Ma and initial $^{87}Sr/^{86}Sr$ of 0.7071 ± 21. The Pb isotope data on 8 rhyolites yield an identical date of $2044^{+127}/_{-139}$. Considering the data sets together, it is clear that the high TiO_2 basalts have initial $^{87}Sr/^{86}Sr$ that are lower than the other groups (Fig. 2). Combining the low TiO_2 basaltic andesite with the rhyolite data yields a reasonably constrained (MSWD = 2.2) regression line date for 17 points of 2110 ± 31 Ma and initial $^{87}Sr/^{86}Sr$ of 0.7053 ± 5 (see Fig. 2).

The isotopic data for the sampled units from the Dullstroom Formation below the RLS, while providing only imprecise estimates of the possible primary age, show no evidence either of protracted cooling of the volcanics nor of pervasive isotopic/elemental disturbance at times

Table 2. Isotopic data for the Rooiberg Group Volcanics from the Dullstroom-Loskop Dam area

Sample	Rb(ppm)	Sr(ppm)	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$ Precision	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
Dullstroom Formation (Basalt: High-Ti Group)							
D-139a[b]	44.5	383	0.3363	0.71322 ± 5			
D-139c[b]	47.9	374	0.3711	0.71468 ± 7			
D-139d[b]	47.6	397	0.3477	0.71396 ± 7			
D-139f[b]	53.9	410	0.3802	0.71474 ± 6			
D-139g[b]	47.0	397	0.3432	0.71426 ± 5			
D-139h[b]	55.2	413	0.3872	0.71513 ± 6			
D-139i[b]	38.6	439	0.2541	0.71154 ± 5			
D-139k[b]	46.9	412	0.3297	0.71340 ± 6			
Dullstroom Formation (Basaltic andesite: Low-Ti Group)							
D-143a[b]	56.3	264	0.6173	0.72404 ± 5			
D-143b[b]	61.6	261	0.6850	0.72548 ± 7			
D-143d[b]	49.7	253	0.5701	0.72231 ± 6			
D-143e[b]	52.4	278	0.5470	0.72197 ± 5			
D-143g[b]	80.4	263	0.8872	0.73218 ± 5			
D-143h[b]	66.2	273	0.7038	0.72704 ± 8			
D-143i[b]	40.1	267	0.4361	0.73018 ± 8			
D-143k[b]	79.0	264	0.8667	0.72109 ± 6			
Dullstroom Formation (Basal rhyolites)							
D-14[b]	80.8	205	1.146	0.74235 ± 5			
D-53	104.4	227.4	1.333	0.745351 ± 13[a]	18.358	15.592	38.340
D-92a	112.5	202.9	1.612	0.754231 ± 14[a]	19.305	15.742	39.531
D-92d[a]	145	186	2.265	0.773961 ± 13[a]	19.690	15.785	39.933
D-92g	132.7	200.9	1.921	0.762721 ± 17[a]	17.639	15.505	37.615
D-92h	116.9	195.7	1.737	0.758131 ± 14[a]	17.497	15.503	37.554
D-92i[b]	133	194	1.993	0.768421 ± 13[a]			
D-92k	135.5	207.9	1.896	0.76295 ± 2	16.706	15.420	36.675
D-161	96.6	230.0	1.219	0.74297 ± 7	18.180	15.573	38.795
D-163[b]	144	208	2.008	0.76474 ± 4	17.517	15.503	37.544
D-4[b]	79.4	271	0.8496	0.73152 ± 9			
D-23[b]	86.7	237	1.060	0.73875 ± 5			
Dullstroom Formation (Loskop Dam: Unit #1): Low-MgO type							
RDW-1	166.6	120.1	4.058	0.82407 ± 6	19.213	15.765	39.817
RDW-2A	176.1	118.4	4.355	0.83114 ± 4	18.458	15.696	38.860
RDW-3[b]	182	91.0	5.889	0.87002 ± 9	17.980	15.612	38.148
RDW-4	189.9	115.5	4.822	0.84289 ± 5	18.920	15.733	39.332
RDW-5	183.2	84.13	6.409	0.88266 ± 16	19.753	15.808	40.387
RDW-6	188.6	110.5	5.008	0.85044 ± 3	19.983	15.851	40.669
RDW-7A	189.0	112.4	4.932	0.85096 ± 10	17.786	15.654	37.811
RDW-8	192.6	137.5	4.096	0.81856 ± 3	17.374	15.576	37.366
RDW-9[b]	164	105	4.554	0.83072 ± 4	18.462	15.683	38.738
RDW-10	152.5	189.6	2.344	0.77668 ± 3			
RB-11	188.9	129.2	4.280	0.83049 ± 1	17.738	15.586	38.031
Dullstroom Formation (Loskop Dam: Unit #2): High-MgO type							
RB-46					16.554	15.439	36.401
RB-52					15.740	15.361	35.493
RB-56					15.647	15.348	35.416
RB-68					16.232	15.386	35.434
RB-69[b]	137	267	1.4930	0.752460 ± 10[a]	15.696	15.363	35.505
Damwal Formation (Loskop Dam: Unit #3)							
RB-95	156.4	213.9	2.1287	0.77277 ± 2	17.089	15.576	37.037
301/81	169.5	139.9	3.5394	0.80685 ± 1			
318/81	49.72	154.6	0.9329	0.73525 ± 1			
Damwal Formation (Loskop Dam: Unit #4)							
DF-1	97.0	159	1.774	0.758273 ± 12[a]	20.390	16.044	40.981
DF-2	85.0	166	1.488	0.751528 ± 12[a]	20.990	16.047	41.590
DF-3	111	113	2.865	0.790917 ± 15[a]	18.879	15.793	39.091
DF-4	168	176	2.783	0.787900 ± 12[a]	20.068	15.918	40.688
DF-5	129	149	2.523	0.780283 ± 16[a]	19.960	15.906	40.431
DF-6	134	142	2.752	0.787470 ± 9[a]	18.962	15.793	39.346

Table 2. Continued

Sample	Rb(ppm)	Sr(ppm)	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$ Precision	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
285/81	148	141	3.063	0.794043 ± 11[a]	18.454	15.727	38.533
313/81	41.0	172	0.6911	0.728994 ± 11[a]	24.758	16.503	46.686
314/81	–	–	–	–	23.264	16.342	44.378
Damwal Formation (Loskop Dam: Unit #6)							
RB-152					19.396	15.883	39.724
L-140					16.766	15.487	36.658
L-144					16.282	15.408	35.992
L-153					18.963	15.753	38.939
Kwaggasnek Formation (Loskop Dam: Unit #8)							
RB-181					17.078	15.542	36.823
RB-178					16.539	15.426	36.290
Schrikkloof Formation (Loskop Dam: Unit #9)							
RB-193					20.827	15.879	40.251
RB-196					17.571	15.614	37.235
RB-201					20.826	15.914	41.543

[a] $^{87}Sr/^{86}Sr$ determined on multi-collector VG354 spectrometer; unflagged values measured on single-collector MM30 spectrometer (see text)
[b] Rb, Sr determined by XRF

Table 3. Summary of regressions and age calculations

Sample	Age (Ma) ± 95%	$^{87}Sr/^{86}Sr_i$ or μ	MSWD/n (F)	Comment
Dullstroom Fm				
Low-Ti (bas. andesites & rhyolites)	2110 ± 31	Sr 0.7053 ± 5	2.2/17 (1.84)	Exclude D-14, D-143i, D-143K
Rhyolites	2037 ± 92	Sr 0.7071 ± 21	4.0/10 (2.10)	
	$2044^{+127}/_{-139}$	Pb 9.88 ± 0.17	1.4/8 (2.25)	
	$2137^{+143}/_{-158}$	Pb 9.88 ± 0.24	0.83/7 (2.39)	Exclude D92K
Loskop Dam: Unit #1	1933 ± 32	Sr 0.7095 ± 12	8.5/13 (1.95)	
	$1677^{+170}/_{-192}$	Pb 10.17 ± 0.14	2.4/10 (2.1)	
Damwal Fm				
Unit #4	1946 ± 23	Sr 0.7096 ± 5	2.5/8 (2.25)	
	$2007^{+90}/_{-96}$	Pb 10.5 ± 0.2	5.3/9 (2.17)	
	$2018^{+58}/_{-60}$	Pb 10.4 ± 0.1	0.77/8 (2.25)	Exclude DF-1
All Rhyolites from Loskop Dam	$2075^{+49}/_{-51}$	Pb 10.4 ± 0.2	4.7/29 (1.67)	Exclude DF-1, L193, L201, RB-152, RB-95

Notes: All uncertainties are given as 95% confidence intervals; where MSWD of regression exceeds the relevant F statistic, uncertainties have been augmented by $(MSWD/F)^{1/2}$
μ values are calculated for reservoirs of the second stage of the Stacey and Kramers (1975) model; i.e. reservoirs generated at 3700 Ma with ratios of $^{206}Pb/^{204}Pb = 11.152$ and $^{207}Pb/^{204}Pb = 12.998$

substantially younger than the time of emplacement of the RLS magmas.

2. Rooiberg Group rhyolites from Loskop Dam. Both Rb-Sr and Pb isotopic data are available for these samples.

A date of 1946 ± 23 Ma is reflected by the Rb-Sr data (8 points) for the petrographically freshest low-MgO rhyolites from Unit #4 whereas the Pb analyses for the same samples (9 data points) yield a statistically equivalent date of $2007^{+90}/_{-96}$ Ma. Excluding sample DF-1 from the calculation improves the goodness of fit (see Table 3) and provides an identical, but more precise, isochron date of $2018^{+58}/_{-60}$ Ma. The rather imprecise Pb–Pb estimate is within-error of 2080 Ma whereas the more precise Rb-Sr date is not. Data for the less fresh rhyolites from Unit #1 give a Rb-Sr date of 1933 ± 32 Ma which is comparable to the Unit #4 rhyolites while the Pb isotopic data for the same samples indicate a significantly younger date of $1677^{+170}/_{-192}$.

If the data sets are combined, the Rb-Sr data yield a date of 1946 ± 22 (on 21 samples) whereas a date of $2075^{+49}/_{-51}$ Ma may be recovered from 29 of the 34 Pb isotope analyses (see Fig. 3a and 3b). Interestingly, the samples excluded from this regression calculation are not from the data set giving the younger date: i.e. the "younger" data set are accomodated within the average spread of the data. Samples DF-1, Rb-95 and Rb-152 fall significantly above the best fit line whereas the two samples from Unit #9 (L-193, L201) fall below.

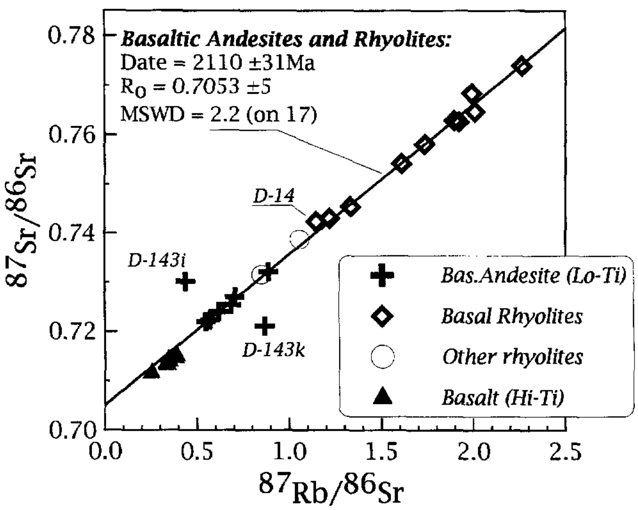

Fig. 2. Rb-Sr isochron plot of data for the Dullstroom Formation of the Rooiberg Group. Labelled samples and the basalt data were excluded from the regression

On average, then, the rhyolite succession at Loskop Dam remained closed systems to Rb and Sr at temperatures below the blocking temperature for the Rb-Sr isotopic system at least since 1950 Ma. The U-Pb isotopic system was possibly perturbed in those rhyolites from the lowermost volcanic unit at Loskop Dam. Considered as a group, however, the Pb isotopic data for the Rooiberg rhyolites from Loskop Dam largely preserve evidence of the assumed crystallisation age of the Rooiberg Group.

Discussion

Ages calculated from isotopic decay systems (e.g. Rb-Sr, U-Pb) reflect the times at which diffusion of parent and daughter nuclides ceased in the mineral or rock system under study i.e. the time at which the system last passed below the blocking temperature and "closed" (e.g. York, 1978). Isotope decay schemes offer the possibility, then, of detecting, and evaluating the scale of, post-crystallisation hydrothermal circulation and/or re-heating of igneous bodies.

Sr and Pb isotopic systems have been used to demonstrate that the LGS granites in the Zaaiplaats area were subjected to protracted periods of fluid circulation at temperatures in excess of about 300 °C (Walraven et al., 1990; McNaughton et al., 1993). McNaughton et al., 1993) demonstrated that the U-Pb isotopic system in the Zaaiplaats granites was last disturbed as recently as ca. 1000 Ma, i.e. over 1 Ga after crystallisation of the LGS granites. These authors argue that the protracted circulation of hydrothermal fluids, driven by heat generated by radioactivity of the high U, Th and K contents in the granites, was responsible for maintaining the granites at temperatures above the blocking temperature for U-Pb over this time period. Walraven et al. (1990b) also demonstrated substantial post-crystallisation open-system behaviour in the Rb-Sr system in the Zaaiplaats granites in close proximity to zones of Sn mineralisation.

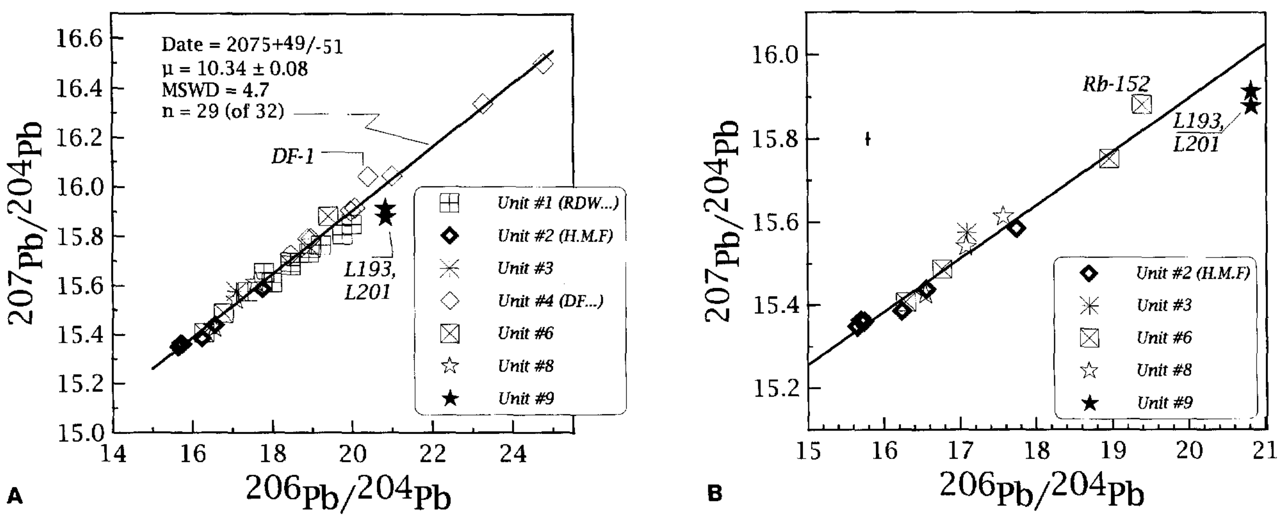

Fig. 3A,B. Plot of $^{206}Pb/^{204}Pb$ *versus* $^{207}Pb/^{204}Pb$ for data from silicic lavas from the Rooiberg Group in the Loskop Dam area. Labelled samples were excluded from the regression. Enlarged portion of the plot shown in **A**. The regressed isochron line from **A** is shown

Circulation of hydrothermal fluids, both within the crystallized RLS granites and into their enclosing rocks, is of obvious importance in mineralisation. Robb et al. (1994) regard both endo-granitic and exo-granitic polymetallic mineralisation related to the Bushveld Granites as the result of evolving hydrothermal systems which persisted for "several hundred million years" after the granites solidified.

The isotopic data for the Rooiberg Group presented in this communication can be usefully applied to investigate whether the Rooiberg volcanics, particularly those of rhyolitic composition, were also subjected to protracted post-solidification open system behaviour and, if present, whether the isotopic disturbance can be attributed to sustained hydrothermal circulation (either within the volcanics or from the granites) or to the thermal effects (possibly with associated fluid circulation) induced by the intrusion of the Rustenburg Layered Suite mafic magmas.

As detailed in previous sections, the isotopic data for volcanics from the Rooiberg Group from several areas suggest some post-crystallisation disturbance. However, the pervasive "re-setting" of the Rb-Sr systematics apparent in Rooiberg rhyolites sampled from Witbank and Bothasberg to give *isochron* ages of ca 1600 Ma (Walraven et al., 1985) is not noted in the sample sets from the Loskop Dam-Dullstroom area.

Results from the rhyolites from Unit #1 suggest that the Rb-Sr and U-Pb decay systems responded in a different way to the post-crystallisation "disturbance": the younger Pb–Pb age indicating that the U-Pb system remained open (i.e. above the relevant blocking temperature) longer (i.e. to lower temperatures?) than the Rb-Sr. It is of interest to note that comparable differences in the response of Rb-Sr and U-Pb systems are also reflected in the data from the Zaaiplaats granites. Model dates calculated from the Rb-Sr data by Walraven et al. (1990b) indicate dates of final closure predominantly in the range 1.3–1.7 Ga (summarised in Fig. 4), several hundred million years prior to the 1.0 Ga closure times for the U-Pb system deduced by McNaughton et al. (1993).

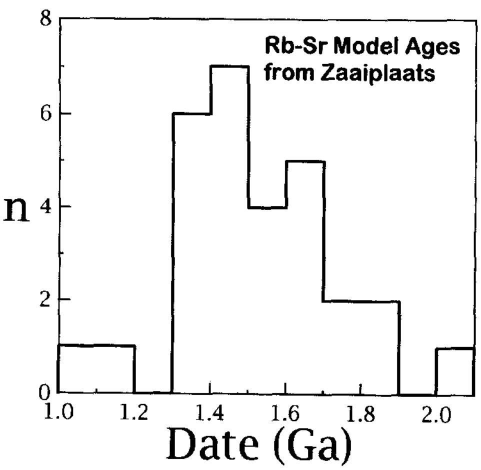

Fig. 4. Histogram summary of Rb-Sr model ages for the mineralised Zaaiplaats granite data of Walraven et al. (1990b)

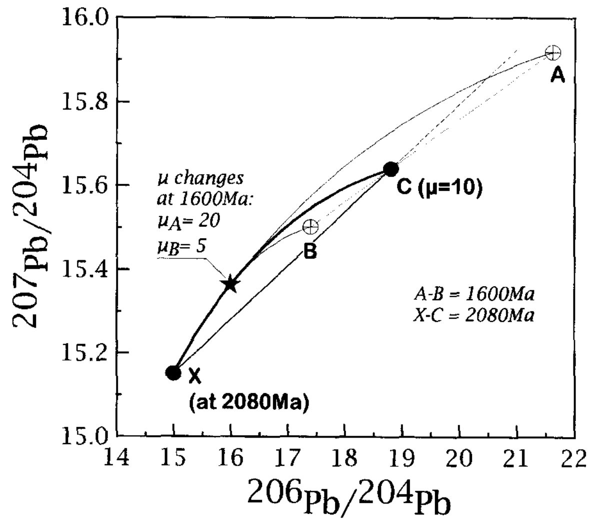

Fig. 5. $^{206}Pb/^{204}Pb$ *versus* $^{207}Pb/^{204}Pb$ plot to show the effect of changing μ at 1600 Ma on a sample formed at 2080 Ma (see text for discussion)

"Re-setting" of the Rb-Sr dates in certain Rooiberg volcanics and the Zaaiplaats granites has been attributed to selective loss of radiogenic ^{87}Sr. Anomalously young Pb–Pb ages are produced through changes in sample $^{238}U/^{204}Pb$ ($= \mu$). This is demonstrated in Fig. 5 which depicts a sample formed at 2080 Ma which is subsequently altered (at 1600 Ma). Sub-sample B has the μ ratio halved whereas A has μ increased (doubled). The present day Pb ratios (A–C–B) fall on an array of age 1600 Ma: the sample with U/Pb increased by alteration plotting *below* the 2080 Ma isochron whereas the sample which suffered reduced U/Pb plots *above* the primary age isochron. Twist (1985) argued that alteration of the rhyolites at Loskop Dam tended to increase with stratigraphic height (i.e. to higher unit numbers) and that the uppermost units had suffered silicification and substantial U loss. It is apparent from Figs. 3a and 3b, however, that samples L193 and L201 from Unit #9 plot below the regressed isochron which suggests that, if these samples were erupted with the same initial Pb ratios as the other rhyolites, U was *enriched* relative to Pb subsequent to formation! The apparently systematic chemical variations in the LMF volcanics with height through the Loskop Dam succession, and limited initial Nd isotopic data (Twist and Harmer, 1987; unpublished data), provides no evidence of a change in source for the Unit #9 eruptives.

The lack of pervasive re-setting of the isotopic systems in the Loskop Dam section is in marked contrast to the near-isochron 1600 Ma systematics noted for the Witbank and Bothasberg sample sets (Walraven et al., 1985). It is noteworthy that Bushveld granites do not occur in the Dullstroom-Loskop Dam region sampled for this study whereas granites are closely associated with the Rooiberg volcanics in the Witbank area. These observations possibly suggest that hydrothermal fluid circulation sustained by the high heat productivity of the LGS granites, particularly in the highly fractionated fractions of the sheet, may be responsible for any observed disturbance of isotopic systematics in the Rooiberg lavas. Radioactive elements were apparently not sufficiently concentrated in the rhyolites to sustain hydrothermal activity long after the cooling of the rhyolites and/or the mafic magmas of the RLS.

Anomalously young isotopic ages encountered in Rooiberg Group volcanics may therefore be utilised to identify the influence of granite-driven hydrothermal activity. In view of the importance of such hydrothermal activity in the generation of polymetallic ore concentrations (e.g. Robb et al., 1994), Pb isotope studies should prove useful as an exploration tool in locating regions of higher mineralisation potential in exposures of Rooiberg Group volcanics underlain by LGS granites.

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