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GRANITIOD SERIES IN TERMS OF MAGNETIC SUSCEPTIBILTY: A CASE STUDY FROM THE ARCHAEAN BARBERTON REGION, SOUTH AFRICA

**SHUNSO ISHIHARA, LAURENCE J. ROBB,
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by

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ABSTRACT

Magnetic susceptibilities were measured on a representative collection of Archaean granitoids of the Barberton region using a portable KT5 magnetic susceptibility meter. The studied granitoids comprise: (1) syntectonic tonalite-trondhjemite-granodiorite (TTG) gneisses (132 samples); (2) late-tectonic calc-alkaline granitoids (402 samples); and (3) post-tectonic low- and high-Ca granitoids (12 samples). Most of the early-stage syntectonic granitoids (~3450 Ma) have low magnetic susceptibilities, less than 3 x 10^{-3} SI units, and correspond to ilmenite-series granitoids. The late-stage Kaap Valley tonalite pluton (~3230 Ma) contains sporadically distributed higher magnetic susceptibility values (greater than 3 x 10^{-3} SI units) corresponding to magnetite-series granitoids, although the magnetic susceptibilities are still less than one-third of typical magnetite-series TTGs of the Japanese Island Arc. The Barberton TTG suite is essentially derived from reduced magma that reflects the anoxic nature of the continental crust during the Archaean Eon. The more oxidized nature of parts of the Kaap Valley tonalite is enigmatic at this stage, but may be associated with later hydrothermal activity. Late-tectonic granodiorite-adamellite batholithic complexes (~3105 Ma) belong mostly to the magnetite series, and seem to suggest that relatively oxidized continental crust had evolved by this time. Post-tectonic granitic plutons, which formed largely between *c*. 2900 Ma and 2700 Ma, can be subdivided into low-Ca (akin to S-type) ilmenite series and high-Ca (akin to I-type) magnetite series rocks.

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GRANITOID SERIES IN TERMS OF MAGNETIC SUSCEPTIBILITY: A CASE STUDY FROM THE ARCHAEAN BARBERTON REGION, SOUTH AFRICA

INTRODUCTION

The direct measurement of magnetic susceptibility in rocks provides a powerful tool in field studies of granitic terranes since their magnetite contents have an important bearing on the redox state. The magnetic susceptibility technique has been successfully applied to the classification of many granite terranes, including the Phanerozoic granitoids of Japan (Kanaya and Ishihara, 1973; Ishihara, 1979) and South Korea (Ishihara et al., 1981; 2001a), North China (Ishihara et al., 2001b), the Malay Peninsula (Ishihara et al., 1979), the Lachlan Fold Belt of Australia (Tainosho et al., 1988), the Sierra Nevada (Bateman et al., 1991), the Peninsular Range batholith in California (Gastil et al., 1990; 1994) as well as the Andean granitoids of Northern Chile (Ishihara et al., 1981) and Peru (Ishihara et al., 1999). In all cases regional variations reveal the intrinsic redox state of the granitic magmas. However, to the author's knowledge similar studies have not been undertaken in Archaean granitic terranes.

In this study magnetic susceptibility measurements were made on some of the oldest Archaean granitoids in the world, namely, from the *c.* 3500 to 2700 Ma Barberton area of South Africa. A portable KT-5 magnetic susceptibility meter was used to obtain measurements on a systematic archival collection of hand specimens, representing the entire range of Archaean granitoids in the Barberton region, housed at the Economic Geology Research Institute of the School of Geosciences, University of the Witwatersrand, Johannesburg. Where the surfaces of the measured samples were found to be uneven, correction factors were used to normalize the measured values to those that would be obtained on smooth surfaces (Table 1). The measured values were also continuously cross-checked by referral to the GSJ house standard (Bison Model 3101).

Table 1: Correction factors used to normalize magnetic susceptibility measurements made on uneven rock surfaces

Surface unevenness (mm)	Correction factor (k)
1	1.07
2	1.15
3	1.24
4	1.35
5	1.44
6	1.55
7	1.66

[Correction factors supplied by manufacturers of the KT-5 magnetic susceptibility meter]

GEOLOGICAL OUTLINE

Archaean granitic magmatism surrounding the Barberton greenstone belt (Fig. 1) occurred episodically from *c.* 3550 to 2700 Ma (Kamo and Davis, 1994). The evolution of these granitoids can be broadly divided into three magmatic stages. These stages are: (1) syntectonic; (2) late-tectonic; and (3) post-tectonic, which broadly coincide with the lithological cycles originally identified by Anhaeusser and Robb (1980).

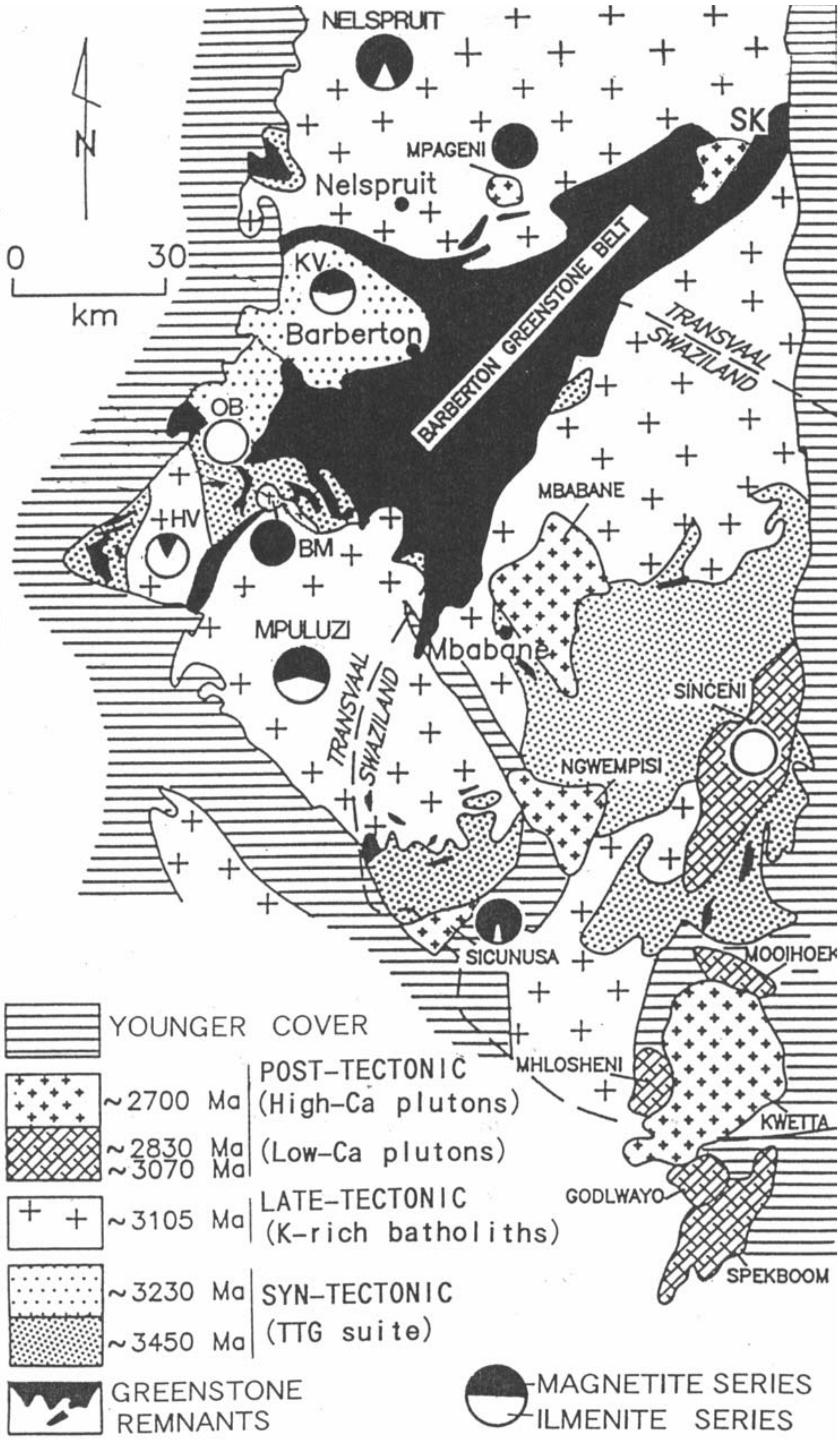

Figure 1: *Geological map of the Barberton region (modified after Anhaeusser and Robb, 1980) showing the ilmenite/magnetite-series ratios of major granitic bodies. KV = Kaap Valley pluton; OB = Other TTG bodies; HV = Heerenveen batholith; BM = Boesmanskop syenite pluton; SK = Salisbury Kop pluton.*

Syntectonic granitoids

The earliest products of granitoid magmatism comprise pervasively foliated, tonalite/trondhjemites and granodiorites (the so-called TTG suite), which intruded pre- to syn-kinematically into the low-grade metavolcanic rocks of the Barberton greenstone belt and into sporadically preserved earlier high-grade gneisses (Robb and Anhaeusser, 1983). Their emplacement ages cluster at *c*. 3450 Ma and 3230 Ma (Kamo and Davis, 1994). Up to 13 TTG plutons have been recognized in the westernmost part of the Barberton greenstone belt, and the larger and better defined of these have been selected for study.

Most of these plutons comprise fine- to medium-grained, biotite-bearing (10-15%), tonalite and trondhjemite (Robb and Anhaeusser, 1983). However, the largest of these, the *c*. 3230 Ma old Kaap Valley pluton, is more melanocratic containing up to 25% hornblende, in addition to biotite (Robb et al., 1986).

Late-tectonic granitoids

The magmatic phase that superceded the widespread TTG plutonism in the region was characterised by the emplacement of large, multicomponent batholiths of granodiorite to adamellite/monzogranite (Anhaeusser and Robb, 1983; Robb et al., 1983). The batholiths were emplaced into pre-existing tonalite/trondjhemite crust at *c.* 3105 Ma, in a rather short time span of a few million years. Of these bodies the Nelspruit, Heerenveen, Mpuluzi and Boesmanskop bodies (Fig. 1) were selected for detailed measurement.

Post-tectonic granitoids

The final stages of granitic magmatism occurred over an extended period between *c.* 3074 Ma and 2690 Ma and are characterized by the episodic intrusion of discrete, post-tectonic granitoid plutons which cross-cut all the other Archaean rock types in this region (Fig. 1). These post-tectonic plutons have been subdivided into low- and high-Ca granitoids (Meyer et al., 1994), categories that are somewhat akin to the I- and S-type classification that has been widely applied in Phanerozoic granite terranes (Chappell and White, 1974, 1992). The low-Ca plutons are coarse-grained, porphyritic, biotite granites, which contain rare muscovite and are marginally peraluminous in character, while the high-Ca plutons are typically hornblende-biotite granites.

MAGNETIC SUSCEPTIBILITY MEASUREMENTS

Results of the magnetic susceptibility measurements (Table 2 and Fig. 2) are classified into two groups, viz., granitoids with values higher than 3.0×10^{-3} SI units and those with values lower than 3.0×10^{-3} SI units (Ishihara, 1990). This subdivision corresponds, respectively, to the magnetite-series and the ilmenite-series granitoids, as defined originally by Ishihara (1977).

Syntectonic granitoids

The oldest discrete TTG pluton in the Barberton region, the *c*. 3509 Ma old Steynsdorp pluton, is characterized throughout by low magnetic susceptibilities and is clearly an ilmenite-series granitoid. The *c*. 3450 Ma - old syntectonic TTG plutons in the Komati River valley, south of the Barberton greenstone belt (Fig. 1), are also generally low in magnetic

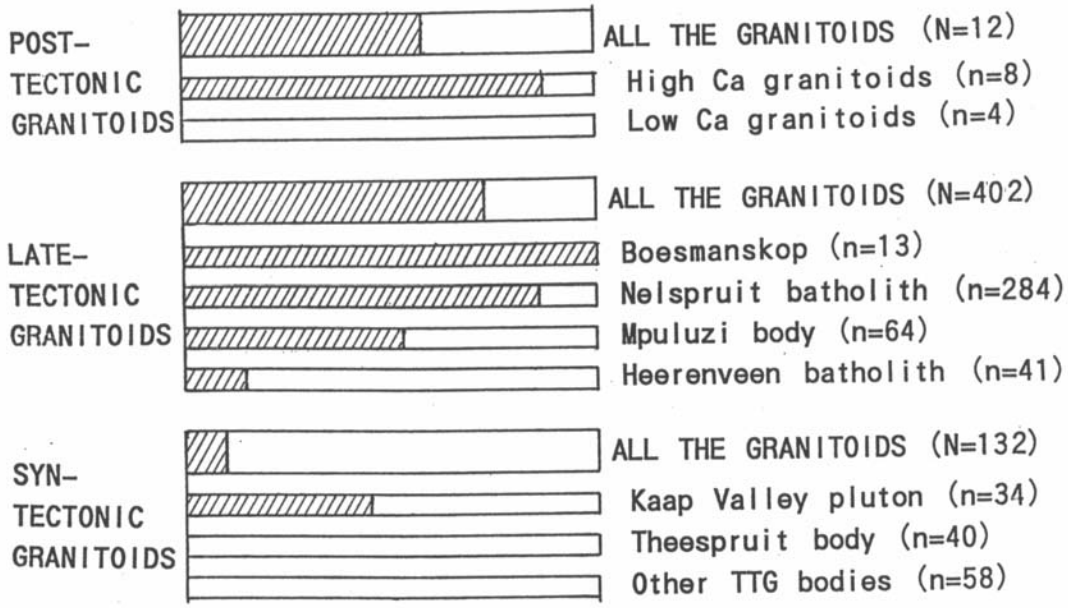

Figure 2: *Histograms of ilmenite-magnetite-series ratios of major granitic bodies. Magnetite series (shaded); Ilmenite series (open).*

susceptibility (i.e., less than 3.0 x 10^{-3} SI units) and only the slightly younger, more melanocratic, Kaap Valley tonalite pluton (*c*. 3230 Ma) demonstrated some magnetite-series values (45% of the measurements; Table 2). Hence, the majority of the TTG plutons exhibit ilmenite-series characteristics in terms of magnetic susceptibility and the ratio of magnetite- to ilmenite-series rocks among the syntectonic TTG suite is approximately 10:90 (Table 2).

Although some 45% of the samples measured from the Kaap Valley pluton are defined as magnetite-series rocks, the recorded intensities are, nevertheless, low and correlate more closely with the intermediate-series rocks defined by Ishihara et al. (1984). These vary from 4 x 10^{-3} to 21 x 10^{-3} SI units over a range of silica contents from 61 to 70 wt.%. These magnetic susceptibilities are about one-third of the values obtained for typical magnetite-series granitoids of the Miocene Tanzawa TTG suite in Japan, which vary between 20 x 10^{-3} and 50 x 10^{-3} SI units over a silica range of 59 to70 wt.%. Although the Kaap Valley pluton is the most oxidized of the Barberton TTG suite, its oxygen fugacity at the solidification stage is still much lower than Phanerozoic magnetite-series granitoids of equivalent composition.

Late-tectonic granitoids

The *c*. 3105 Ma old Nelspruit batholith is strongly magnetic and 86% of all measurements have values higher than 3.0 x 10^{-3} SI units, making this body distinctly magnetite-series in character. The granodiorite-adamellite rocks of the batholith have magnetic susceptibilities that typically range from 8 x 10^{-3} to 20 x 10^{-3} SI units, which is somewhat lower than 10 x 10^{-3} to 30 x 10^{-3} SI units of typical Phanerozoic magnetite-series granodiorite-monzogranite in Japan. Magnetic susceptibility values corresponding to ilmenite-series rocks occur only locally in the southeastern part of the Nelspruit batholith.

The Mpuluzi batholith, on the other hand, comprises an equal number of magnetite- and ilmenite-series values (53:47), whereas the smaller Heerenveen batholith is much less

Table 2: Barberton granitoids characterized in terms of magnetite- and ilmenite-series, as determined by magnetic susceptibility measurements

Granitic bodies	Measurements	Magnetite-series	Ilmenite-series
Syntectonic stage (TTG gneisses)			
Kaap Valley (3227 Ma)	34	15 (44%)	19 (56%)
Nelshoogte (3212 Ma)	6	0 (0%)	6 (100%)
Stolzburg (3481 Ma)	4	0 (0%)	4 (100%)
Theespruit (3437 Ma)	40	0 (0%)	40 (100%)
Steynsdorp (3509 Ma)	19	0 (0%)	19 (100%)
Rooihoogte	9	0 (0%)	9 (100%)
Others	20	0(0%)	20 (100%)
Total	132	15 (11%)	117 (89%)
Late-tectonic stage (3105 ± 5 Ma)			
Heerenveen	41	6 (15%)	35 (85%)
Mpuluzi	64	34 (53%)	30 (47%)
Nelspruit	284	243(86%)	41 (14%)
Boesmanskop	13	13(100%)	0 (0 %)
Total	402	296 (74%)	106 (26%)
Post-tectonic stage			
Sicunusa (high-Ca – 2723 Ma)	4	3 (75%)	1 (25%)
Mpageni (high-Ca – 2740 Ma)	3	3 (100%)	0 (0 %)
Salisbury Kop (high-Ca – 3079 Ma)	1	1 (100%)	0 (0 %)
Total (high Ca)	8	7 (88 %)	1 (12 %)
Sinceni (low Ca – 3074 Ma)	4	0 (0 %)	4 (100%)

magnetic with only 15% of the rocks measured having magnetite-series characteristics. The small, *c.* 3105 Ma old, Boesmanskop syeno-granite body adjacent to the Mpuluzi batholith (Fig. 1) is composed only of magnetite-series rocks. In summary, the late-tectonic granitoids are strongly biased towards magnetite-series characteristics, with an overall ratio of magnetite- to ilmenite-series rocks of 73:27.

Post-tectonic granitoids

Meyer et al. (1994) reported an assemblage comprising titanite, allanite and magnetite in the high-Ca granitoids, a feature which is characteristic of magnetite-series granitoids. Samples from three high-Ca granites were measured and 88% of these do, indeed, preserve high-magnetic susceptibilities and are consistent with magnetite-series granitoids. Measurements of the high-Ca Sicunusa granite in Swaziland (Fig.1) suggested a component (25%) of ilmenite-series characteristics, although it is clear that more measurements are needed to substantiate this trend.

The low-Ca Sinceni granite, also in Swaziland (Fig.1), is characterised entirely by low-magnetic susceptibilities indicative of an ilmenite-series granite. Meyer et al. (1994) reported an accessory mineral assemblage of monazite and ilmenite in the low-Ca granitoids, which typifies ilmenite-series granitoids (Ishihara, 1977).

MINERAL CHEMISTRY OF BIOTITE AND APATITE

The principal mafic silicate mineral in the majority of granitoids studied from the Barberton region is biotite, whose chemical composition is dependant upon the presence or absence of magnetite (Czamanske et al., 1981; Wones, 1981). For this reason, biotites from a small subset of the syn-, late-, and post-tectonic Barberton granitoids were analysed mainly to confirm the observations regarding the contrasting redox state of the granites in the Barberton area. The study of the mineral chemistry of redox-sensitive minerals in granites of the Barberton area remains in a very preliminary stage and work is still in progress to shed additional light on the nature and significance of the contrasting redox state of these rocks.

The following comments reflect the preliminary nature of this aspect of the study:

The XMg value (i.e., Mg/[Mg+Fe] atomic ratio) of mafic silicate minerals is a sensitive indicator of the redox conditions under which they crystallized (Czamanske and Wones, 1973). For example, the XMg value of biotite, which equilibrated with magnetite and K-feldspar, a common assemblage in magnetite-series granite, is controlled by the following reaction for the annite component in the biotite solid-solution series (Wones and Eugster, 1965):

$$\underset{annite}{KFe_3AlSi_3O_{11}.H_2O} \leftrightarrow \underset{magnetite}{Fe_3O_4} + \underset{K\text{-}feldspar}{KAlSi_3O_8} + H_2$$

Thus, biotites that crystallized under oxidizing conditions tend to show higher XMg values than those that formed in a reducing environment.

Chemical compositions of biotite and apatite were determined by a JEOL 733mkII electron probe microanalyzer at the Geological Institute, University of Tokyo. Analyses were done at 1.2 x10-8 Å and an accelerating voltage of 15kV with acquisition time of 20 seconds. The results were calibrated using conventional ZAF correction with factors and programs supplied by JEOL, and listed in Tables 3 and 4.

Magnetite in the high-magnetic susceptibility part of the Kaap Valley TTG suite occurs in polygonal to granular shapes, often associated with biotite and/or hornblende. The magnetite may also contain inclusions of ilmenite, chalcopyrite and even hematite, which develop along 111 cleavage planes. Secondary epidote and titanite are common in the magnetite-series TTG suite, but ilmenite is rare. XMg values in the magnetite-series Kaap Valley tonalite are relatively high and range between 0.555 and 0.575. The XMg values of biotites from the late- and post-tectonic magnetite-series granitoids (i.e., the Boesmanskop and Salisbury Kop plutons, respectively; Table 3) are likewise high and range between 0.582 to 0.588. By contrast, the XMg values of biotite in a sample of the Theespruit pluton, with a distinctly ilmenite-series character, are lower, with values of 0.305 and 0.525 (Table 3).

Table 3: Chemical composition of biotite

stage	syn-tectonic granitoids								post-tectonic granitoids			
series	ilmenite-series				magnetite-series				magnetite-series			
pluton	Theespruit				Kaap Valley				Boesmankop		Salisbury Kop	
magnetic susceptibi	0.12×10^{-3}		0.05×10^{-3}		53.0		19.9		24.0		6.5×10^{-3}	
Sample Number	23-4		23-5		24-6		LKV-19		22-3		24-7B	
	TTG-5		TTG-6		TTG-1		TTG-3					
number of analysis	85		22		123		16		26		22	
	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)
SiO_2 (wt%)	36.99	(0.36)	35.73	(0.31)	37.16	(0.23)	37.08	(0.54)	39.53	(0.35)	37.66	(0.42)
Al_2O_3	14.70	(0.28)	17.26	(0.26)	15.04	(0.23)	14.91	(0.19)	11.25	(0.17)	14.31	(0.41)
TiO_2	1.65	(0.29)	1.70	(0.13)	1.74	(0.22)	2.26	(0.60)	0.87	(0.06)	1.41	(0.17)
FeO*	19.01	(0.47)	24.07	(0.42)	17.34	(0.44)	17.11	(0.34)	17.79	(0.82)	16.59	(0.37)
MnO	0.24	(0.04)	0.44	(0.04)	0.79	(0.06)	0.28	(0.04)	0.55	(0.04)	0.68	(0.04)
MgO	11.79	(0.32)	5.92	(0.16)	12.15	(0.37)	12.99	(0.50)	13.91	(0.64)	13.26	(0.41)
CaO	0.08	(0.05)	0.04	(0.03)	0.04	(0.03)	0.14	(0.32)	0.01	(0.01)	0.02	(0.01)
Na_2O	0.06	(0.03)	0.05	(0.02)	0.08	(0.23)	0.06	(0.03)	0.03	(0.02)	0.06	(0.02)
K_2O	9.16	(0.42)	9.28	(0.21)	9.53	(0.27)	9.37	(0.50)	9.76	(0.09)	9.82	(0.11)
Cl	0.02	(0.01)	0.01	(0.01)	0.08	(0.01)	0.05	(0.01)	0.22	(0.02)	0.02	(0.01)
=O	0.00		0.00		-0.02		-0.01		-0.05		0.00	
F	0.25	(0.10)	0.25	(0.13)	0.45	(0.13)	0.13	(0.09)	1.78	(0.21)	1.06	(0.16)
=O	-0.06		-0.06		-0.10		-0.03		-0.40		-0.24	
total	93.88		94.70		94.29		94.33		95.27		94.63	
Si (atom. O=22)	5.736	(0.035)	5.630	(0.023)	5.740	(0.029)	5.667	(0.050)	6.222	(0.027)	5.844	(0.062)
Al	2.688	(0.045)	3.206	(0.039)	2.739	(0.036)	2.686	(0.030)	2.087	(0.032)	2.617	(0.075)
Ti	0.192	(0.034)	0.201	(0.015)	0.203	(0.026)	0.260	(0.070)	0.104	(0.007)	0.164	(0.020)
Fe(2+)*	2.465	(0.060)	3.172	(0.068)	2.240	(0.058)	2.187	(0.038)	2.343	(0.118)	2.152	(0.046)
Mn	0.032	(0.006)	0.059	(0.006)	0.104	(0.008)	0.036	(0.006)	0.074	(0.006)	0.089	(0.005)
Mg	2.725	(0.077)	1.390	(0.036)	2.797	(0.082)	2.960	(0.115)	3.264	(0.139)	3.066	(0.095)
Ca	0.013	(0.009)	0.007	(0.004)	0.007	(0.005)	0.023	(0.053)	0.002	(0.002)	0.003	(0.002)
Na	0.017	(0.008)	0.016	(0.007)	0.023	(0.007)	0.018	(0.008)	0.010	(0.005)	0.017	(0.007)
K	1.812	(0.080)	1.866	(0.036)	1.878	(0.048)	1.826	(0.089)	1.961	(0.020)	1.943	(0.018)
Cl	0.006	(0.003)	0.002	(0.002)	0.021	(0.003)	0.012	(0.002)	0.060	(0.006)	0.004	(0.002)
F	0.123	(0.051)	0.126	(0.064)	0.222	(0.062)	0.063	(0.045)	0.885	(0.107)	0.520	(0.077)
Al(IV)	0.264		0.371		0.260		0.333		-0.222		0.156	
XMg	0.525	(0.010)	0.305	(0.008)	0.555	(0.013)	0.575	(0.007)	0.582	(0.022)	0.588	(0.012)

* indicates that the total Fe contents are expressed as FeO or Fe(2+).

In oxidized and reduced magmas sulphur is typically accommodated as SO_4^- and HS^- species, respectively (Carroll and Rutherford, 1988). Magmatic apatite, that crystallizes from an oxidized magma, generally contains appreciable sulphur as SO_3, while the SO_3 content in apatites that crystallized from a reduced magma is negligible. The SO_3 contents in apatites from the magnetite-series granitoids of the syntectonic Kaap Valley tonalite pluton, as well as from the post-tectonic Salisbury Kop pluton average 0.12 wt.% (range 0.04 - 0.40 wt.%) and 0.25 wt.% (range 0.19 - 0.36 wt.%), respectively. On the other hand, the SO_3 contents in apatite from the ilmenite-series Theespruit trondhjemite pluton are <0.02 wt.%. These values confirm the contrasting redox state of the magnetite- and ilmenite-series granites identified in the Barberton region.

The SO_3 contents of apatites in the post-tectonic magnetite-series Boesmanskop syeno-granite complex (sample 22-3, Table 4) are variably low and range between 0.04 and 0.11 wt.%. These uncharacteristically low values can, perhaps, be explained by subsolidus recrystallization, and a subsequent change from the primary redox condition of the magma. The biotite composition of this sample shows negative Al (IV) contents, and excess Si contents, with respect to the ideal stoichiometry of phlogopite-annite solid solution. In addition, these biotites also exhibit the highest Cl (0.22 wt.%; c = 0.02 wt.%) and F (1.78 wt.%; c = 0.21 wt.%) contents. The biotites may, therefore, be subsolidus in origin as suggested by their low TiO_2 contents, relative to those in the other samples (Table 3).

DISCUSSION AND CONCLUSIONS

The existence of both magnetite- and ilmenite-series granitoids is an intrinsic feature of Phanerozoic granitic terranes. Magnetite-series granitoids reflect an original high Fe_2O_3/FeO

Table 4: Cl, F and SO_3 contents of apatite

stage				syn-tectonic granitoids							post-tectonic granitoids			
series		ilmenite-series				magnetite-ser		magnetite-series				magnetite-series		
pluton		Theespruit					Kaap Valley				Boesmankop		Salisburgkop	
magnetic susceptibi	0.12×10-3		0.05×10-3		53.0		21.2		19.9		24.0		6.5×10-3	
Sample Number	23-4		23-5		24-6		LKV-7		LKV-19		22-3		24-7B	
	TTG-5		TTG-6		TTG-1		TTG-2		TTG-3					
number of analysis	85		27		104		57		23		130		42	
	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)	ave	(±1σ)
Cl (wt.%)	0.02	(0.01)	0.01	(0.01)	0.03	(0.01)	0.04	(0.01)	0.11	(0.05)	0.01	(0.01)	0.01	(0.01)
F	4.21	(0.31)	4.72	(0.40)	4.47	(0.35)	4.24	(0.32)	3.63	(0.33)	4.69	(0.37)	4.86	(0.34)
SO3	0.02	(0.02)	0.01	(0.01)	0.40	(0.19)	0.12	(0.05)	0.13	(0.04)	0.04	(0.11)	0.36	(0.25)

ratio of the source rock, whereas ilmenite-series granitoids are considered to have been derived either from low Fe_2O_3/FeO source rocks or were reduced by carbon contained in pelitic source rocks (Ishihara, 1984). Bateman et al. (1991) argued that ilmenite-series tonalites in the western foothills of the Sierra Nevada batholith originated from reduced mafic protoliths in the lower crust, rather than from locally derived carbonaceous sedimentary wall rocks (Ishihara and Sasaki, 1989). Many S - type or ilmenite-series granitoids, which do not contain pelitic enclaves, may also have been reduced by C- and S-bearing fluids derived from pelitic source rocks (Takagi and Tsukimura, 1998).

Syntectonic granitoids

In the Barberton region, syntectonic TTG-suite granitoids are believed to have formed by fractional melting of amphibolites and/or quartz eclogites of basaltic composition (Arth and Hanson, 1975; Robb, 1983). Such source rocks may have been derived from the base of the greenstone belt itself, and were probably represented by komatiitic/komatiitic basaltic volcanics together with a minor component of chemical sediment. Komatiitic rocks usually record Fe_2O_3/FeO ratios lower than 0.3 (Hunter, 1974; Hawkesworth and Onions, 1977). Felsic partial melts derived from komatiitic precursors are, therefore, also likely to reflect low Fe_2O_3/FeO ratios and be classified as ilmenite-series granitoids. In addition, komatiitic rocks also occur with intercalated cherty sediments, which are not infrequently also enriched in ferric iron and organic carbon. These ingredients perhaps also contributed to a reducing environment in the source and it is, therefore, not surprising that most of the older TTG rocks in the Barberton region are ilmenite-series in nature.

The *c*. 3230 Ma old Kaap Valley pluton is more than 200 million years younger than the remainder of the TTG suite studied in the Barberton region. Whereas the latter are predominantly ilmenite-series in character, the Kaap Valley tonalite has a magnetite- to ilmenite-series ratio of 45:55 and is markedly more oxidized than the other members of the syntectonic suite. Faure and Harris (1991) estimated that the original $\delta^{18}O$ values for the Kaap Valley tonalite were 7.6 per mil, which is about 1 per mil higher than typical mantle values, and also suggests a more $\delta^{18}O$-enriched source. Other reasons which might explain the higher proportion of magnetite-series values in the Kaap Valley pluton include the fact that it is characterized by a higher mafic mineral content, which points to a higher intrinsic water content of the parental tonalite magma. The Kaap Valley pluton is also believed to have been emplaced at a relatively shallow crustal level and was also subjected to localized hydrothermal fluid flow and sporadic mineralization. Together, these factors may have contributed to the more oxidized character of this intrusion. The younger *c*. 3230 Ma age of the Kaap Valley pluton does not seem to be an adequate explanation for its partial magnetite-series character since the adjacent Nelshoogte pluton is of a similar age, but is entirely ilmenite-series in nature (Table 2).

Late- to post-tectonic granitoids

The majority of the late and post-tectonic granite magmas, including the *c.* 3105 Ma Nelspruit, Mpuluzi and Heerenveen batholiths and the *c.* 2700 Ma Mpageni and Sicunusa plutons, but excluding the low-Ca granites, are represented by oxidized magnetite-series granitoids. These are dominantly of crustal derivation and the source rocks are believed to be represented by pre-existing crustal granitoids such as the *c.* 3450 to 3230 Ma TTG suites (Anhaeusser and Robb, 1983; Robb, 1983; Robb et al., 1983). It is perhaps surprising to note that oxidized granitic magmas were forming in the continental crust as early as *c.* 3105 Ma ago, since this implies that parts of the Archaean continental crust were substantially oxidized by this time. This suggestion does, however, raise pertinent questions regarding the debate around atmospheric evolution and, more specifically, the timing of oxygen augmentation in the Archaean Earth (Ohmoto, 1996, 1997).

The marginally peraluminous and muscovite-bearing characteristics of the low-Ca granitoids suggest that they may represent S-type, ilmenite-series magmas, generated within the continental crust from sedimentary protoliths. In this regard it is pertinent to note that the *c.* 2850 Ma Sinceni pluton is stanniferous, which is also consistent with its reduced S-type character.

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