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PETROGENETIC SIGNIFICANCE OF THE GLIMMERITE OCCURRENCES IN THE OKIEP COPPER DISTRICT, NAMAQUALAND

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by

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ABSTRACT

Attention is focused on a rare rock type, rich in volatiles, associated with the Koperberg Suite, namely glimmerite (micaite). The objective of this study is to examine petrogenetic aspects of this minor but dispersed component and to elucidate its relationship with other members of the Koperberg Suite. From the study of xenoliths and internal contact phenomena in the Koperberg Suite bodies, it is known that the more mafic members (mica diorite, norite, orthopyroxenite, glimmerite) intruded after the more felsic members (anorthosite, mica leucodiorite, leuconorite).

Koperberg Suite glimmerite (KSG) is defined as a rock consisting of at least 70 volume percent mica, with apatite and Fe-oxides as the main accessory minerals. Micro-textures within KSG provide evidence of adaptations of the phlogopite crystal structure to minimize internal stress.

The chemical characteristics of biotite vary as a function of the composition of the host rock. Progressive crystallization of the magma appears to have controlled the elemental ratios. Although this implies that biotites from different rock types are unique, overlapping fields prohibit such an application for specific samples. Classification of biotites on the basis of locality and ore-bearing potential of the host rock, is restricted by overlapping chemographic domains. Geothermometric methods used, failed to establish an unequivocal temperature for the rocks under discussion. Instead a thermal range was encountered for Koperberg Suite rocks.

The behaviour of halogen elements within the micas (elevated Cl and F intercept values for all rock types) is supportive of a high-grade metamorphic event affecting the whole diversity of Koperberg Suite rocks. This is substantiated by high Ti values in micas from all the rocks. It is clear that a stable phlogopitic biotite crystallized from the original Koperberg magma and was subjected to mainly Mg-Fe and associated F-OH and Cl-OH exchange reactions. The Fe-Mg exchange relationship in phlogopitic Koperberg Suite micas with respect to variation in basicity is not a linear function. Phlogopite adopted a predominantly metamorphic affinity with high closing temperatures.

When the thermal history of the associated pelitic gneisses (Waters, 1986) is considered, the enrichment of Ti, Cl, and F, together with associated mineral textures and paragenesis, indicate that the magmatic Koperberg Suite was emplaced into a terrane that was experiencing upper amphibolite or granulite facies metamorphism. Although ore bodies show preference for mafic members of the Suite, such as glimmerite, the characteristics of biotite cannot be used to unequivocally determine ore potential.

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I. <u>INTRODUCTION</u>

The Okiep Copper District, South Africa's second largest copper
producing district, is situated in the arid Namaqualand region of the NW
Cape, demarcated by latitudes 29°25'S, 29°45'S and longitudes 17°30'E,
18°10'E and covering an area of approximately 3000km^2. A swarm of
transgressive composite intrusives, known as the Koperberg Suite, acts as
host to the copper deposits (Lombaard *et al* ., 1986).

Attention is focused on a rare rock type which is sporadically
present in the Koperberg Suite, namely glimmerite (micaite or biotitite).
This rock type consists almost entirely of biotite, *senso lato*, and apatite
and is closely associated with copper mineralization. It is useful for
reconstruction of the physicochemical conditions obtaining during genesis
of the Koperberg Suite, and subsequent events affecting it. Mica provides
a record of evolving magma composition and post magmatic events as
reflected by the variety of cations that readily substitute in its various
structural sites (Guidotti, 1984). The fundamental use of mica in this
context is that it enters into equilibrium relationships with other common
rock-forming minerals, thus indicating the degree to which halogen
fugacities were buffered and hence to which extent the oxidation state
within the entire system was affected. Contact zones consisting of narrow
mica laths, which developed between different rock types, are not regarded
as typical KSG as they are of dissimilar character and appearance.

II. <u>GEOLOGICAL SETTING</u>

The Okiep Copper District of South Africa is situated in the
Proterozoic Namaqua Mobile Belt. Currently the lithostratigraphic column
(Table 1) is subdivided (Holland and Marais, 1983) into a metavolcanic -
sedimentary succession (*viz.* the Okiep Group), which has been intruded by
three polyphase suites of granite gneisses (*viz.* the Gladkop Suite, the
Klein Namaqualand Suite and the Spektakel Suite). Emplacement of these
sheet-like bodies took place at different stages relative to the main
structural events (Joubert, 1971; Van Aswegen, 1988; Moore, 1989) and
occurs in an area of high-grade amphibolite to granulite facies
metamorphism (Joubert, 1986; Waters, 1986). The Koperberg Suite is the
youngest major group of intrusives in the district at 1042 ± 40 Ma
(Nicolaysen and Burger, 1965), 1072 ± 20 Ma (Clifford *et al*, 1975a) and
1100 Ma (Stumpfl *et al.*, 1976).

The Koperberg Suite occurs as a swarm of generally irregular,
easterly trending, steep north-dipping, dike-like bodies, usually 60 to
100m wide and rarely exceeding 1km in strike length. Associated with the
Koperberg Suite are enigmatic phenomena such as megabreccias and narrow,
linear, antiformal features along which continuity of the adjoining rocks
have been interrupted by piercement folding and shearing. The relative
volumetric frequency of occurrence of petrographic classes comprising the
Koperberg Suite is as follows: biotite leucodiorite 32%, leuconorite 26%,
anorthosite 26%, norite 5%, hypersthenite 4%, glimmerite 4% and mica
diorite 2%. The remainder consist of unusual rocks such as hornblendite,
nelsonite (Andreoli and Hart, 1987) and oxide diorite.

Table 1. Lithostratigraphy of the Okiep Copper District, after Holland and Marais (1983)

TABLE I
Lithostratigraphy of the Okiep Copper District

		Nama Group		Conglomerate, grit, quartzite, shale, limestone
				———————— Unconformity ————————
				Fault-associated dykes of pegmatite, syenite and diabase
		Koperberg Suite		Anorthosite, diorite, norite, hypersthenite
NAMAQUA-LAND COMPLEX	INTRUSIVE ROCKS	Spektakel Suite	Kweekfontein Granite	Quartz-microcline granite; some plagioclase; fine-grained
			Rietberg Granite	Quartz-microcline-biotite granite; typically porphyritic; phenocrysts of microcline-microperthite (Carlsbad twins) in even-grained groundmass of quartz, biotite and feldspar; accessory magnetite and zircon; grades to biotite-rich syenite
			Concordia Granite	Quartz-microcline granite; quartz, microcline-microperthite, subsidary oligoclase, variable biotite; garnet plentiful especially near contacts with schist (metapelite).
		Klein Namaqualand Suite	Modderfontein Granite-gneiss	Quartz-microcline granite-gneiss; quartz, microcline-microperthite, oligoclase, variable biotite, little garnet.
			Nababeep Granite-gneiss	Quartz-microcline-biotite granite-gneiss; some plagioclase; includes rocks formerly termed Nababeep gneiss, Brandberg gneiss, Leeupoort gneiss, Jakkalswater augen gneiss.
		Gladkop Suite	Noenoemaas-berg Granite-gneiss	Quartz-microcline granite-gneiss; quartz, microcline, minor plagioclase, biotite; fine-grained.
			Brandewyns-bank Granite-gneiss	Quartz-microcline granite-gneiss; quartz, microcline, minor plagioclase, variable biotite, hornblende; medium-grained.
	META-VOLCANO-SEDIMEN-TARY ROCKS	Okiep Group	Lammerhoek Subgroup	Quartz-feldspar-biotite granolite and gneiss; quartz, plagioclase and microcline, appreciable magnetite, minor hornblende, garnet; fine- to medium-grained; migmatitic; also hornblende gneiss, amphibolite, quartzite and calc-silicate.
			Khurisberg Subgroup	Diverse aluminous schists (metapelite), quartzite, leptite; includes the former Ratelpoort stage, Wolfram Stage and Springbok Stage.

Glimmerite outcrops of the Koperberg Suite form elliptical bodies with easterly trending major axes orientated parallel to the prevalent direction of minimum stress. Individual occurrences rarely exceed 50m by 20m. The general sequence of intrusives of the composite Koperberg Suite bodies is that of increasing basicity (Boer *et al.*, 1988). Correspondingly, glimmerite occurs as a very late stage intrusive. Only rarely are glimmerite inclusions found in the more leucocratic Koperberg Suite rocks. The presence of these inclusions confirms the consanguineous relationship between glimmerite and other rock types of the Koperberg Suite.

III. GLIMMERITE PETROGRAPHY

Two types of glimmerite can be distinguished: (1) those with no enclaves; and (2) those with abundant inclusions. The most common type of glimmerite is a well-foliated, homogeneous, monomineralic rock composed essentially of fine-grained biotite. Kink folding is sometimes conspicuous in this type of glimmerite which is devoid of inclusions. The second kind is typified by abundant inclusions of recrystallized grano- blastic plagioclase and quartz. A variation of this spotted glimmerite reveals numerous small greenish quartzose nodules in which Ni proved to be the chromofore.

Textures present in glimmerite vary from decussate to parallel (Fig. 1a,b). Both these textures indicate adaptations of the biotite crystal lattice to minimize internal stress, and are regarded as typical products of thermal metamorphism. However, these textures can also be interpreted as indicative of external strain at the time of crystallization. The effect of differential stresses are preserved in biotite as is evident in the bending and fracturing of flakes as well as undulatory extinction in both mica and quartz.

Most striking aspect of Koperberg Suite glimmerite (KSG) modal analysis (Table 2) is the abundance of apatite (as much as 15 vol%). Apatite chadacrysts are anhedral, while the crystals developed on the mineral contacts are the only euhedral crystals present in glimmerite. Enstatite (En_{58-65}) and plagioclase (An_{40-52}) occur as subhedral grains with a granoblastic texture. Abundant rutile in biotite and conspicuous ilmenite exsolution lamellae in magnetite, orientated parallel to the octahedron faces of the latter, testify to the titanium enrichment of the glimmerites. As a rule the interstitial Cu-Fe sulphides predominate over oxides (magnetite, hercynite, ilmenite and hematite) except when there is an increase in enstatite content. Accessory minerals present in glimmerite are quartz, zircon, sphene, hornblende, and goethite in decreasing order of abundance. Pleochroism of the biotite in glimmerite varies in intensity, decreasing markedly with increasing alteration. No reverse pleochroism was encountered.

Table 2 *Average modal composition of 9 glimmerite samples (± standard error of the mean, 2s/√n) compared with previous results for some of the associated rock types. The number of samples for the mica diorite, hypersthene diorite and anorthosite are 8, 25, and 5, respectively.*

	Glimmerite		Mica Diorite*	Norite*	Anorthosite*
Biotite	75.70	(0.57)	28.8	2.3	3.6
Plagioclase	13.96	(0.67)	44.2	52.9	94.7
Enstatite ⌀	1.41	(0.32)	3.1	35.6	0.5
Apatite	4.03	(0.31)	7.7	0.7	0.4
Opaques	3.93	(0.25)	15.8	8.5	0.7
Rest	1.19	(0.17)	0.4	–	0.1

* Values from Conradie and Schoch (1986).
⌀ Nomenclature after Morimoto *et al.* (1988).

Although hydrothermal action is of secondary importance, some indications of circulating fluids (metamorphic, hydrothermal or deuteric) are present. Biotite flakes in contact with quartz are less altered than the flakes associated with feldspar crystals. Chloritization (penninitization) of biotite occurs to a limited extent. A brown chloritic phase, which resembles chlorite in optical properties, was detected by systematic electron microprobe (EM) scanning. This secondary phase suggests that post-emplacement thermobarometric conditions did not proceed sufficiently far as to cause development of propylitic associations. Serrated intergrowths between biotite and secondary talc were observed by
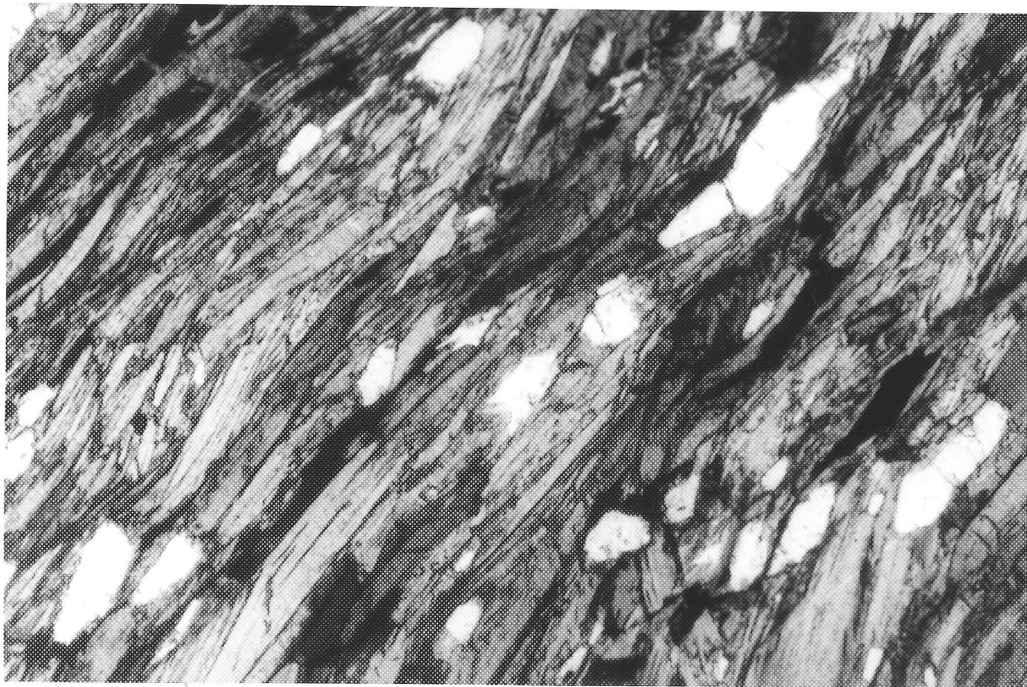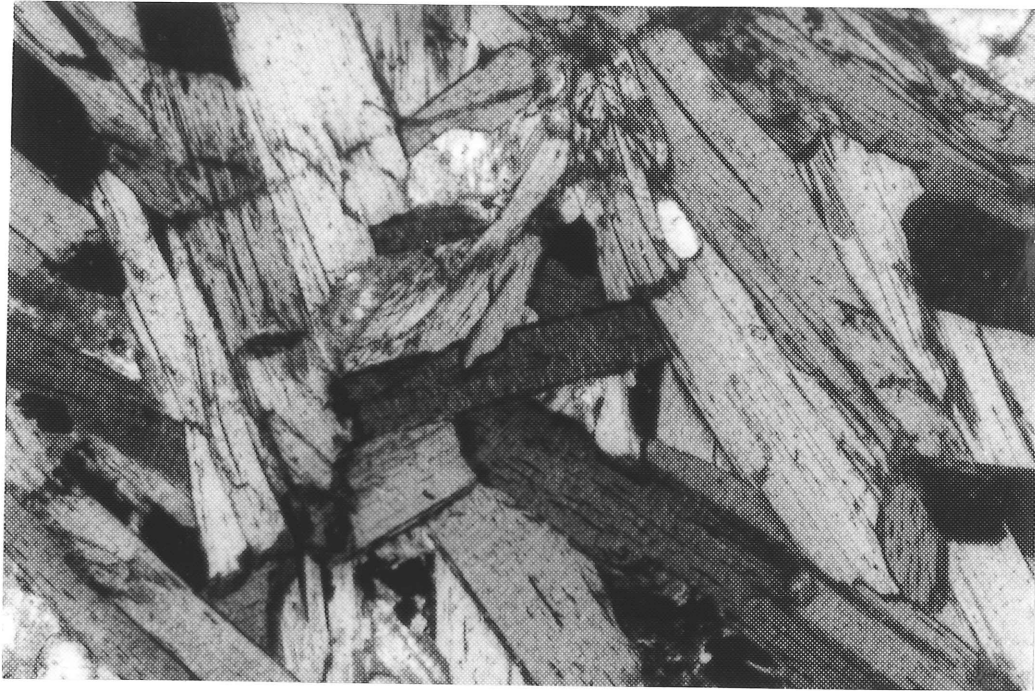
Figure 1: (a) Typical decussate texture and (b) parallel texture in
          glimmerite. Area covered by each photomicrograph:
          2.55 x 1.8mm.

means of EM. During alteration the release of excess free iron yielded goethite which occurs as a reddish, fine-grained overgrowth.

1V. GEOCHEMICAL CHARACTERISTICS OF KOPERBERG SUITE GLIMMERITE (KSG)

Whole rock analyses were performed using a Philips PW 1404 X-ray spectrometer. All the mineral analyses considered in this paper were obtained by wavelength dispersion electron microprobe analysis (Cameca Camebax Microbeam). For the phyllosilicates a slightly defocussed electron beam was used under 15kV and 20nA operating conditions. Duplicated runs compared well in all cases. The machine settings, and standards used for analyzing minerals were routine.

Glimmerite whole rock analyses are extremely rare in international literature. An average analysis compiled from 11 KSG samples is presented in Table 3. When compared with whole rock analyses from elsewhere, a remarkable titanium enrichment of KSG is apparent. This, however, is not a chemical characteristic of glimmerite as such. The glimmerites described in the literature are predominantly associated with kimberlitic and carbonatitic rocks. The chemically related but mica-poor lamprophyric magmas and other alkali ultramafic magmas are generally distinguished by a high volatile content (Wilshire, 1985; Bailey, 1982). The latter magmas are furthermore characterized by low SiO_2 contents and enrichment in incompatible and other complex-forming elements of high ionic potential. The differences apparent from Table 3 could quite feasibly reflect different geological settings and/or modes of occurrence, rather than a fundamental disparity.

Chemographic methods used to distinguish biotite from various rock types do not give unambiguous distinction. However, KSG biotite conforms to the general rule that biotites with a plutonic affinity have $Ti/(Ti+Mg+Al^{VI}) < 0.20$ and $Ti/(Ti+Mg+Fe^{VI}) < 0.10$ (Brigatti and Gregnanin, 1987), contrary to the tendency of biotite from volcanic rocks (Fig. 2a, b).

38 Samples

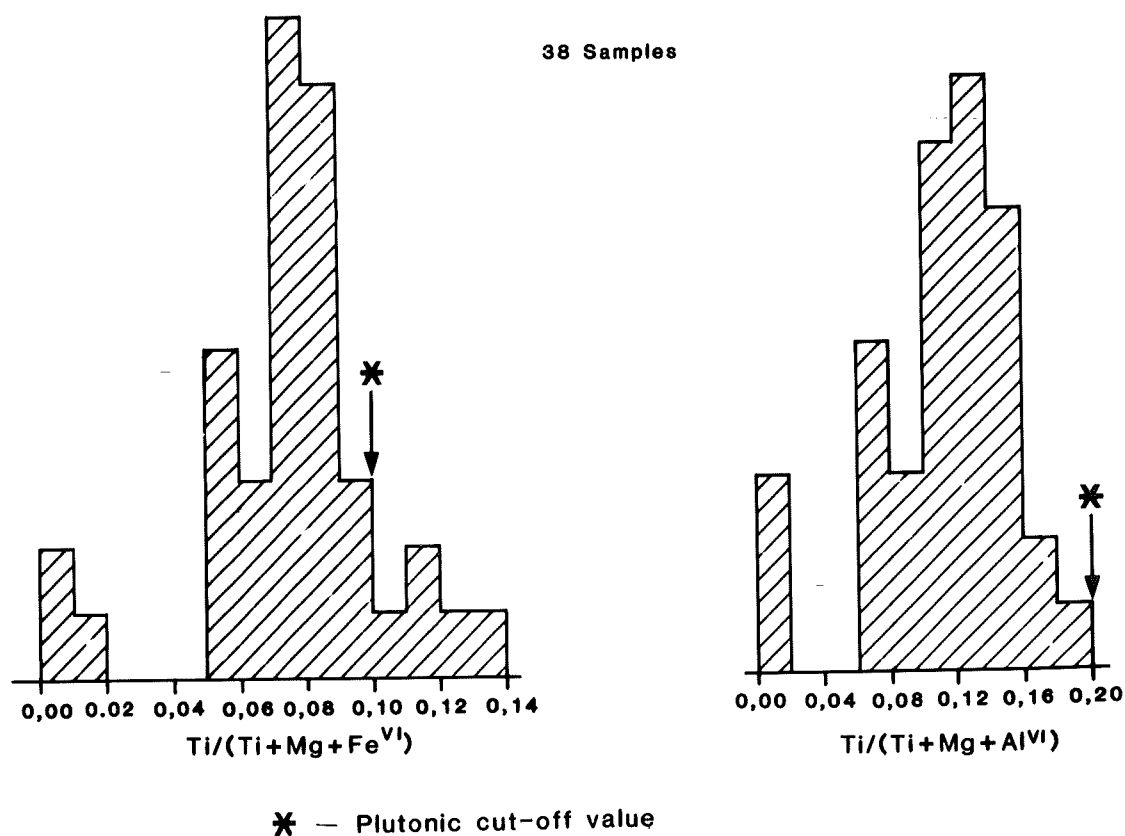**✳ — Plutonic cut-off value**

Figure 2. Histograms of (a) $Ti/(Ti+Mg+Fe^{VI})$ and (b) $Ti/(Ti+Mg+Al^{VI})$ ratios for KSG biotites.

Table 3. Major and trace element composition of analysed glimmerite (respectively weight per cent and ppm), compared to average minette and lamproite analyses. A - Glimmerite, B - Minette, C - Lamproite and D - Vaugnerite

| | A | | | | B | C | D |
	1	2	3	4	5	6	7
SiO_2	33.69	27.60	38.85	40.93	52.60	53.30	57.48
TiO_2	4.57	0.54	0.90	1.88	1.90	3.00	1.06
Al_2O_3	13.18	16.30	17.03	12.53	12.60	9.10	15.14
$Fe_2O_3(\Sigma Fe)$				6.05			
Fe_2O_3	5.64	5.30	2.10				2.85
FeO	14.50	1.10	0.83		7.60	6.30	4.43
MnO	0.12	0.05	0.05	0.05	0.13	0.10	0.09
MgO	11.12	16.80	24.43	22.88	8.00	12.10	5.93
CaO	3.73	11.90	1.27	1.29	7.90	5.80	4.57
Na_2O	0.48	0.10	tr	0.53	2.00	1.40	2.78
K_2O	5.79	6.70	9.48	8.93	6.00	7.20	3.75
P_2O_5	2.16	9.00	0.51	0.37	1.20	1.30	0.52
CuO	1.42						
S	0.76	0.02					
CO_2	0.49	0.07			1.80	2.80	
H_2O+	2.04	2.90					1.14
Total	99.69	98.38	95.45	95.44	101.7	102.4	99.74
LOI	1.99				2.20	2.70	0.16
K/Al	0.49	0.41	0.56	0.71			
K/Na	28.88	67.00	>900	16.84			
Zn	276			56	110	84	
Ni	458			803	155	420	
Rb	389			622	115	272	
Sr	184			82	1010	1530	
Y	74			8	36	27	
Zr	220			239	350	920	
Ba	5291			3755	1345	5120	

1. "Average" Koperberg Suite glimmerite, South Africa, (Boer, 1989). 2. Glimmerite from Västervik, Sweden (Kersten, 1976). 3. Glimmerite, Kiya Alkalic Intrusion, Yenisei Ridge, USSR (Belov, 1970). 4. Glimmerite, Kapirikamodzi, Malawi (Morel, 1988) 5. "Average minette" (Bergman, 1987). 6. "Average lamproite" (Rock, 1984). 7. Vaugnerite from Massif Central Francais (Sabatier, 1980; 1984).

V. EXCHANGE REACTIONS IN BIOTITE

A. Fe-Mg Exchange in Biotite

Biotite compositional variation can, to a large extent, be described in terms of end members. Chemical controls on the mineral compositions are, however, imperfectly known. The relative importance of individual substitutions, producing the chemical composition of particular phases and the way in which biotite composition should be expressed in terms of end members, is still a matter of dispute (Hewitt and Abrecht, 1986; Labotka, 1983). An end member projection plane, in which the only

allowed compositional variations are $Fe^{2+} = Mg^{2+}$ and the Tschermak exchange $(Mg,Fe^{2+}) + Si^{IV} = Al^{IV} + Al^{VI}$, is used for Koperberg Suite biotite (Fig. 3) following Guidotti (1984). The term biotite pertains to any general position on the projection plane, but end member terms are used when the attention is focused on a specific sample. It should be pointed out that the terms Al-annite and Al-phlogopite used here, were formerly termed siderophyllite and eastonite, respectively (see Deer *et al.*, 1966). Although the term siderophyllite is still used, it is now applied to a more Al-rich composition than previously. The term eastonite is obsolete.

Two main characteristics of Koperberg Suite biotites are apparent from Figure 3 (average analysis for various rock types presented in Table 4):

(i) a significant aluminium enrichment in certain samples, due to Tschermak exchange; and

(ii) the restricted linear substitution between Mg^{2+} and Fe^{2+} for various rock types.
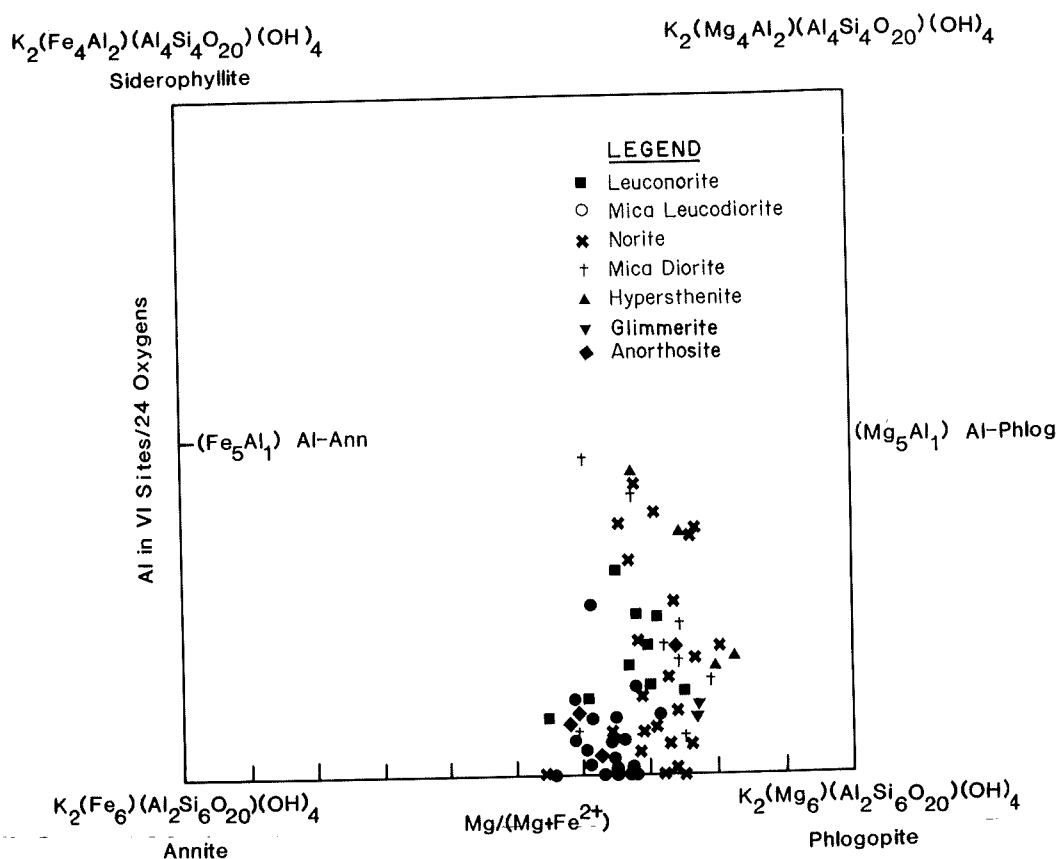

Figure 3. *Koperberg Suite biotite data from various rock types plotted on a biotite end member diagram (after Guidotti, 1984). The end member with composition $K_2(Mg_4Al_2)(Al_4Si_4O_{20})(OH)_4$ was known as eastonite in the old nomenclature system. Each plotted point represents the average of at least ten point analyses obtained with the use of Cameca Camebax Microbeam apparatus.*

The restricted variation Mg/(Mg+Fe$_T$) biotite composition amongst Koperberg Suite rocks can be interpreted in a number of ways. Either the biotites crystallized at constant oxygen fugacity with limited external buffering during cooling or all biotites re-equilibrated to the same extent during a uniform post-crystallization event. The high Ti content of the Mg-rich biotites points towards a high-grade metamorphic imprint. The latter is in agreement with observations made by various other authors (Engel and Engel, 1960, Guidotti *et al*, 1977, etc.) if the assumption is

made that Ti in biotite is quidrivalent and substitutes into octahedral sites. The Fe and Mg compositional micromaps of a biotite crystal (Fig. 4) confirms the important role of $Fe^{2+} = Mg^{2+}$ exchange. Figure 4 directs attention to the heterogeneous distribution of the two elements under discussion.

FeO(wt%)

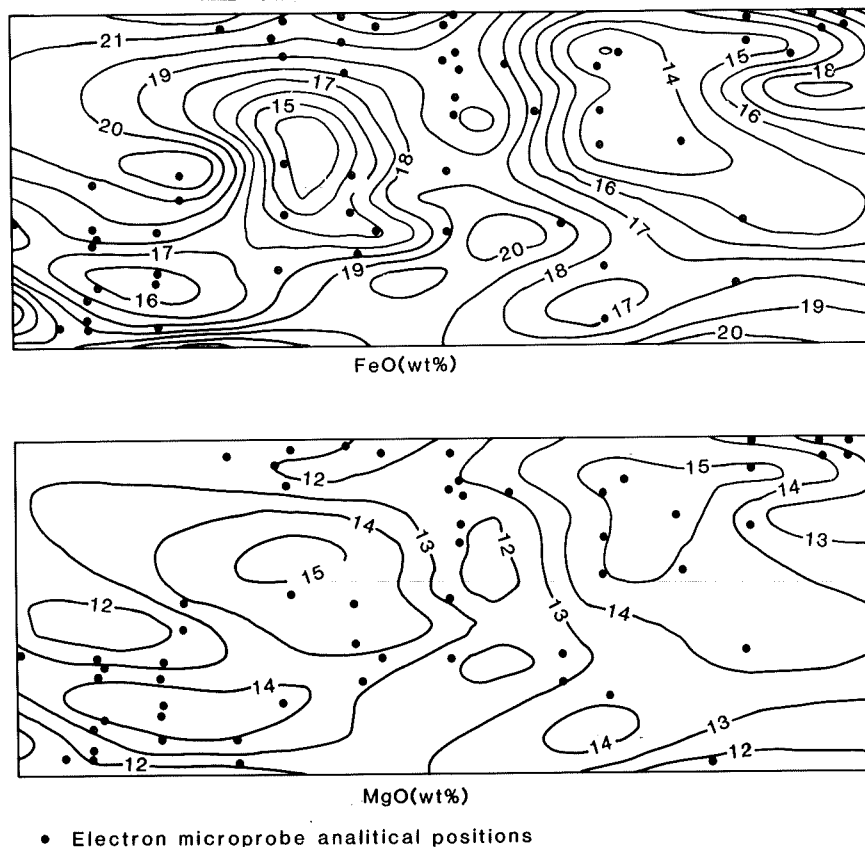

MgO(wt%)

• Electron microprobe analitical positions

Figure 4. Representative Fe and Mg micromaps, generated by electron microprobe, of a biotite crystal. The crystal outline roughly coincides with borders of the microgrid. Area covered by micromap: 400μm x 150μm.

B. Halogen – OH Exchange in Biotite

Experiments indicate that the F=OH and Cl=OH exchange in biotite is governed by several independent factors including temperature, the activities of halogen acids, and the compositions of biotite (Munoz and Swenson, 1981; Volfinger *et al.*, 1985). It is a known fact that biotites with a high Mg/Fe ratio tend to have higher maximum F-contents than ferroan biotites (Boer, 1989). Rosenberg and Foit (1977) used crystal field theory to account for "Fe-F avoidance". The crystal field splitting parameter of Fe^{2+} octahedrally co-ordinated to F- is significantly smaller than its value when (OH)- is the co-ordinating anion. Thus, the presence of Fe^{2+} is not favoured at sites where F- substitutes for (OH)- due to smaller crystal field stabilization energy. Munoz and Ludington (1974) showed experimentally that annite is much less effective in removing F from the fluid phase than phlogopite.

The above-mentioned principle is illustrated for Koperberg Suite rocks in Figure 5 in which a positive correlation is apparent between wt% F and X_{Mg}. The mole fraction of the phlogopite component (X_{Mg}) versus weight percent fluorine for biotites from the Koperberg Suite corroborates the

Table 4. Electron microprobe analysis of average major and trace elements for biotites from different Koperberg Suite rock types. A - Anorthosite, B - leuconorite, C - mica leucodiorite, D - norite, E - mica diorite, F - oxide diorite, G - hypersthenite, H - glimmerite.

	A	B	C	D	E	F	G	H
SiO_2	37.21	37.45	38.07	36.59	35.83	36.05	38.36	35.72
TiO_2	4.49	4.53	4.17	4.68	4.34	4.25	4.10	4.58
Al_2O_3	12.44	13.26	12.93	13.55	12.40	12.44	13.92	12.48
Cr_2O_3	0.17	0.14	0.24	0.34	0.12	0.24	0.31	0.30
FeO	12.88	11.65	11.30	11.06	14.63	15.27	8.56	15.01
MnO	0.08	0.06	0.06	0.06	0.08	0.04	0.02	0.08
MgO	13.89	14.72	14.77	15.92	13.33	13.05	16.48	13.27
CaO	0.01	0.06	0.28	0.02	0.02	0.01	0.00	0.03
Na_2O	0.06	0.09	0.09	0.14	0.09	0.07	0.14	0.12
K_2O	10.13	9.91	9.43	9.79	10.17	10.23	9.88	9.19
H_2O	1.93	2.04	1.97	1.99	1.81	1.89	2.05	1.75
F	2.54	3.20	2.90	3.64	2.16	2.83	3.49	2.33
Cl	2.53	1.25	2.09	0.57	3.37	1.99	0.99	3.40
$O=F$	1.07	1.35	1.22	1.53	0.91	1.19	1.47	0.98
$O=Cl$	0.57	0.28	0.47	0.13	0.76	0.45	0.22	0.77
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Si IV	5.79	5.74	5.84	5.59	5.67	5.68	5.78	5.64
Al IV	2.21	2.26	2.16	2.40	2.30	2.30	2.22	2.30
T site	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00
Al VI	0.07	0.14	0.18	0.04	0.01	0.01	0.25	0.03
Ti VI	0.52	0.52	0.48	0.53	0.49	0.49	0.46	0.49
Cr	0.02	0.02	0.03	0.04	0.01	0.03	0.04	0.04
Fe^{2+}	1.68	1.49	1.45	1.41	1.94	2.01	1.08	1.99
Mn^{2+}	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01
Mg	3.22	3.36	3.37	3.62	3.14	3.07	3.70	3.13
O site	5.52	5.54	5.52	5.65	5.61	5.61	5.53	5.68
Ca	0.00	0.01	0.05	0.00	0.00	0.00	0.00	0.01
Na	0.02	0.03	0.03	0.04	0.03	0.02	0.04	0.04
K	2.01	1.94	1.84	1.91	2.05	2.06	1.90	1.85
A site	2.03	1.97	1.92	1.95	2.08	2.08	1.94	1.90
O	20.00	20.00	20.00	20.00	20.00	20.00	20.00	20.00
OH	2.08	2.12	2.05	2.09	2.01	2.06	2.08	1.93
F	1.25	1.55	1.41	1.76	1.08	1.41	1.66	1.16
Cl	0.67	0.32	0.54	0.15	0.90	0.53	0.25	0.91
X_{phlog}	0.52	0.56	0.57	0.59	0.48	0.46	0.66	0.47
X_{sid}	-	0.05	0.01	0.07	0.03	0.03	0.05	0.04
X_{ann}	0.48	0.39	0.42	0.34	0.49	0.51	0.29	0.49

relatively high F-content of Mg-enriched phases. Despite the high degree of overlapping amongst biotites from the different Koperberg Suite rock types, it is evident that glimmerite is more ferroan than biotite from norite. There seems to be no significant characterization of Fe-F avoidance in terms of locality (Fig. 5). It is furthermore of interest to note that no linear Mg-Fe exchange relationship exists with respect to variation in basicity for Koperberg Suite biotites, as was considered by Conradie (1983) and Schoch and Conradie (1990).
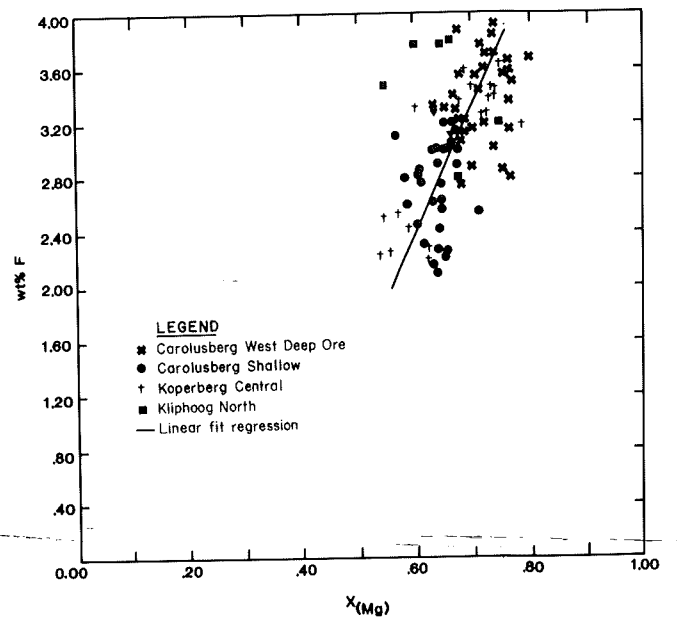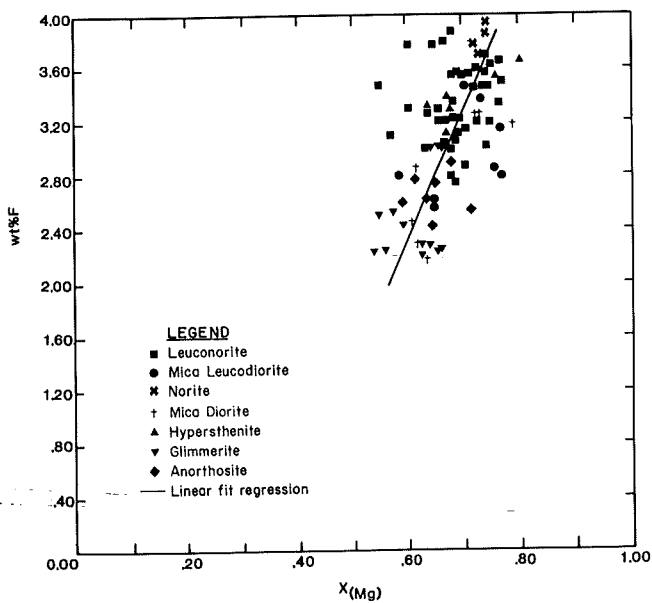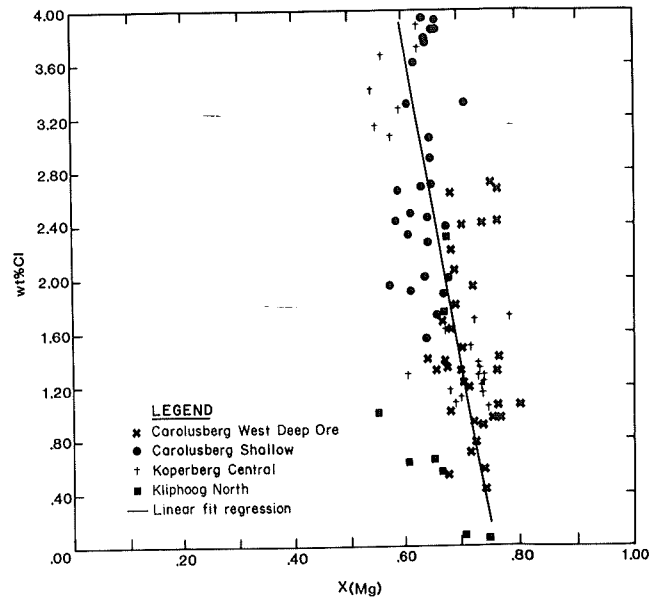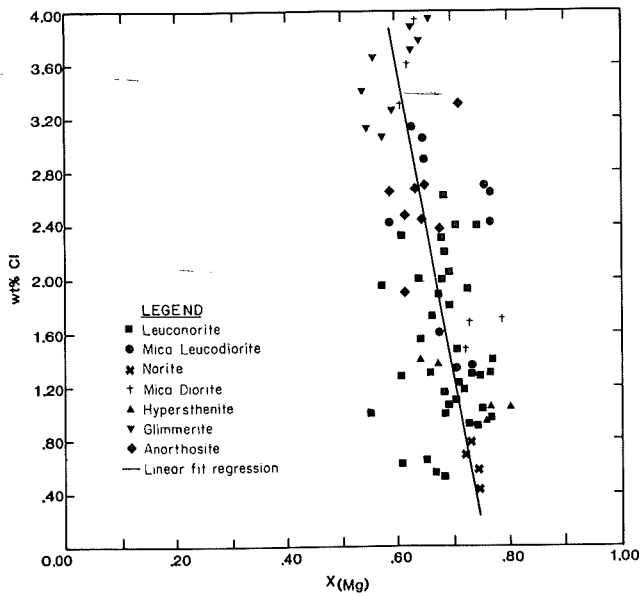
Figure 5: The relationship between chlorine and fluorine content of Koperberg Suite biotites and the iron-magnesium ratio $(X_{(Mg} = MgO/MgO + FeO)$. Overlapping in terms of field site is so extensive that no locality or rock type characterization is possible.

Substitution of F for OH markedly increases the thermal stability of phlogopite. Fluorphlogopite melts at 1390°C at 1 bar (Munoz, 1984), whereas hydroxyl phlogopite dehydrates at 950°C for $P(H_2O) = 100$ bars (Wones, 1967). Munoz (1984) showed that the substitution of F for OH has a destabilizing influence on the annite structure, which is in striking contrast to the increase in stability for phlogopitic compositions.

Gunow *et al.* (1980) defined a single numerical value to express the relative degree of fluorine enrichment in a mica. Because of Fe-F avoidance the wt% F is in itself inadequate to measure relative enrichment. The fluorine intercept value (IV(F)) for biotites is:

$$IV(F)_{biotite} = 1.52X_{Phlog} + 0.42X_{Ann} + 0.20X_{Sid} - \log(X_F/X_{OH}) \quad (1)$$

where $X_{Phlog} = X_{Mg} = Mg/(Mg + Fe_T)$

$X_{Sid} = [(3 - Si/Al)/1.75][1 - X_{Plog}]$ and
$X_{Ann} = 1 - (X_{Phlog} + X_{Sid})$.

The fluorine intercept value is related through the equilibrium constant to relative HF activity by the equation (Gunow *et al.*, 1980):

$$\log[f(H_2O)/f(HF)] = 2100/T + IV(F) \quad (2)$$

When compared with some other base metal occurrences the Koperberg Suite exhibits relatively high degrees of fluorine enrichment. This can probably be accounted for using the above-mentioned avoidance rules. Other factors that are known to influence the F content of biotite are temperature, the H_2O/F ratio in the fluid phase, and the Al-content of the biotite (Valley *et al*, 1982). The close clustering of values for the Koperberg Suite suggests that halogen activity was relatively uniform. Furthermore, the narrow $IV(F)_{biotite}$ range accentuates the similitude of Mg/Fe ratios amongst biotites studied for a broad spectrum of Koperberg Suite rock types. It is not possible to classify any Koperberg Suite sample according to locality or rock type on grounds of either IV(F) values or Mg/Fe ratios.

The fluorine intercept value alone cannot distinguish between micas in mineralized copper systems and those from barren systems, a fact which was also noted by Kesler *et al.* (1975).

The behaviour of fluorine in Koperberg Suite biotites reflects small differences in halogen acid activity in very stable phlogopitic micas. It is unusual for a diverse suite of rocks to have such a homogeneous halogen distribution. The compositional control of Cl in biotite is not well understood. By analogy to the fluorine intercept value Munoz (1984) developed a chlorine intercept value (IV(Cl)):

$$IV(Cl) = -5.01 - 1.93X_{Phlog} - \log(X_{Cl}/X_{OH}) \quad (3)$$

The chlorine intercept value is related through the equilibrium constant to HCl activity;

$$\log[f(H_2O)/f(HCl)] = 5151/T + IV(Cl) \quad (4)$$

The available Koperberg Suite IV(Cl) data does not conform with values for known "mineralized" localities, but are instead very much enriched in Cl content.

In order to eliminate OH from intercept equations, an F/Cl inter-
cept value [IV(F/Cl)] was developed (Munoz, 1984). The numerical value is
used to measure F/Cl ratios in biotite which are corrected for the opposing
effects of Fe-F and Mg-Cl avoidance, or as a measure of log[f(HCl)/f(HF)].
By subtracting equation (4) from (2);

$$\log[f(HCl)/f(HF)] = -3051/T + IV(F) - IV(Cl) = -3051/T + IV(F/Cl) \quad (5)$$

where

$$IV(F/Cl) = 3.45X_{Phlog} + 0.41X_{Ann} + 0.20X_{Sid} - \log(X_F/X_{Cl}) + 5.01 \quad (6)$$

It is obvious that equation (6) does not contain a term for XOH
which makes the IV(F/Cl) value insensitive to the analytical uncertainties
for OH anion occupancy. Thus IV(F/Cl) values are likely to be more
accurate than either IV(F) or IV(Cl) on their own (Fig. 6). Wide-ranging
overlap of fields for different rock types could probably be ascribed to a
pervasive event which influenced these biotites. The high F/Cl intercept
values for the Koperberg Suite biotites correspond to lower F/Cl ratios, as
IV(F) is directly linked to the fugacity ratio f(HCl)/f(HF) at constant
temperature. The relatively low F/Cl ratios for Koperberg Suite biotites
is remarkable and points towards a significant Cl-enrichment rather than
decreasing F content during crystallization and/or closing equilibration.
The chlorine enrichment can be ascribed to post-crystallization metamorphic
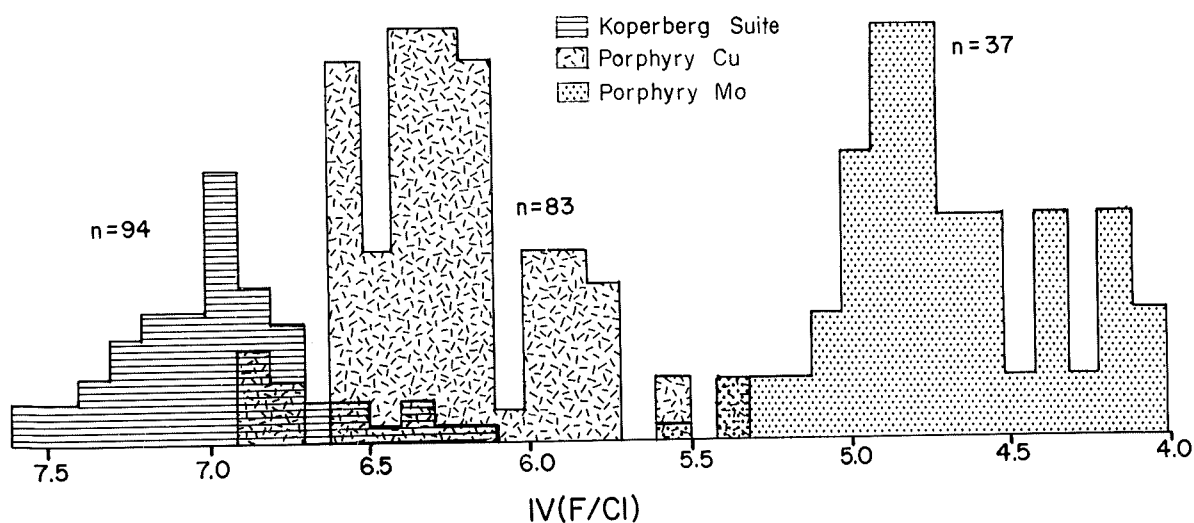fluid circulation.

Figure. 6. *Fluorine/chlorine intercept values for different groups
compared to Koperberg Suite values. The data for the porphyry
Mo (Henderson) deposit are from Gunow et al. (1980) and the
data for porphry copper deposit (Santa Rita) are from Jacobs
and Parry (1979).*

C. The Effect of Contamination

Owing to the re-equilibration of magnetite during cooling
(Czamanske et al., 1981) and the consequent interpretive problems of Fe-Ti
oxide geothermometry and oxygen barometry (Spencer and Lindsley, 1981),
little is known about the oxygen fugacities during Koperberg Suite
crystallization (Conradie and Schoch, 1986).

Log (f_{HF}/f_{H2O}) and log (f_{HF}/f_{HCl}) ratios exhibit little variation amongst Koperberg Suite biotites. The subordinate role of Fe-F and Mg-Cl avoidance is noticeable when the distribution of biotite end members is taken into consideration (Fig. 3). From Figure 7 it is apparent that biotite values plot in the field defined for strongly contaminated primary igneous melts (I-SC) (Ague and Brimhall, 1988). The above mentioned argument concerning Fe-F and Mg-Cl avoidance is confirmed, indicating the controlling contribution of f_{HF}/f_{H2O} during crystallization and metamorphism. Only a slight increase in F/OH with increase in Mg is evident, as indicated by the low angle positive slope of the Koperberg Suite field in Figure 7. This trend is similar to the mineralization trend of Brimhall and Ague (1988) for various porphyry Mo, porphyry Cu and hydrothermal base metal deposits. The position of the Koperberg Suite data on Figure 7 could thus not solely be attributed to contamination, but also demonstrate out the influence of mineralization and metamorphic effects.
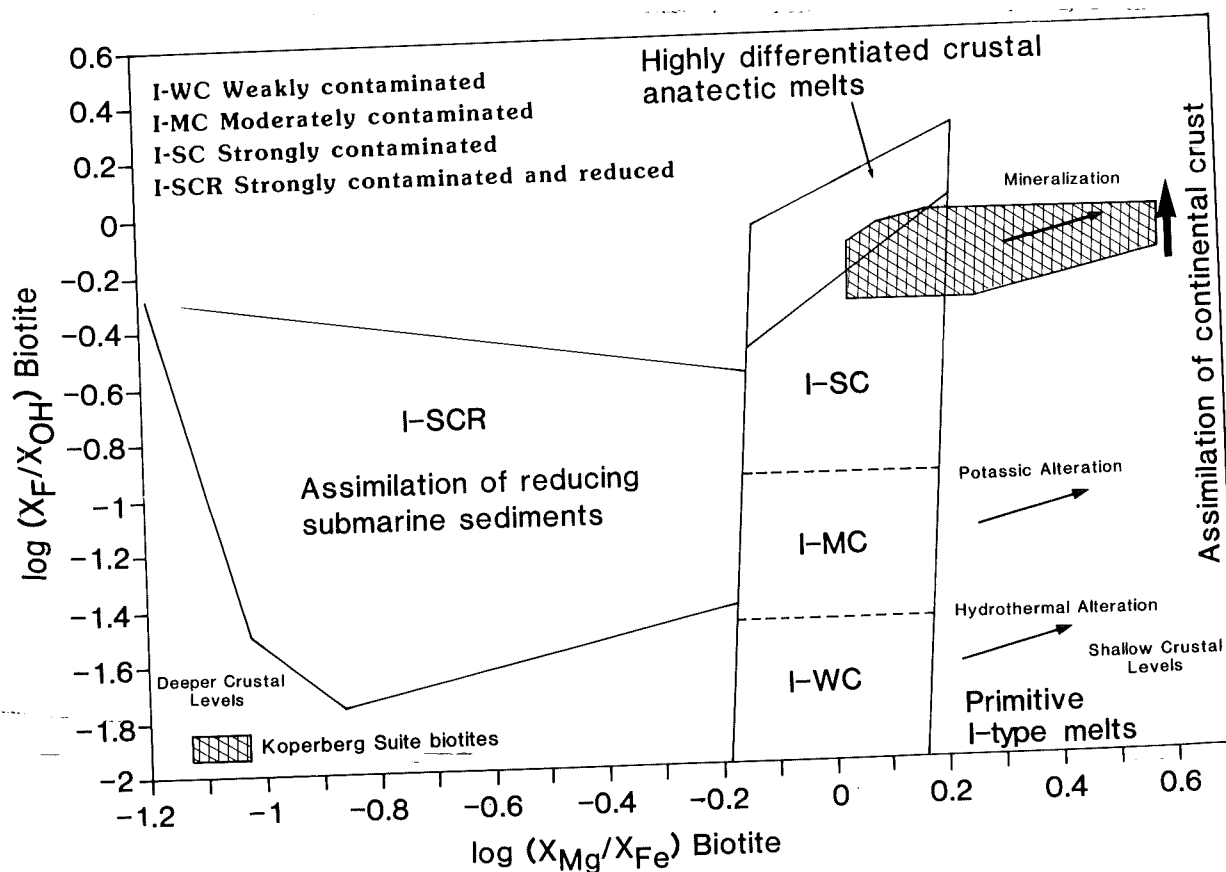

Figure 7. Binary diagram summarizing the effects of contamination and the relationship between igneous and hydrothermal biotites. Field boundaries are after Ague and Brimhall (1988).

VI. DISCUSSION AND INTERPRETATION

The petrogenesis of the glimmerite (and by implication the Koperberg Suite) is complex in spite of the relatively small number of rock-forming minerals involved. The applicable range in the values of several critically intensive and extensive variables is still imperfectly known and difficult to determine. A number of contrasting petrogenetic hypotheses have been presented in the past for the Koperberg Suite (Benedict *et al.*, 1964, Van Zyl, 1967, Clifford *et al.*, 1975a, b, 1981, McIver *et al.*, 1983, Conradie and Schoch, 1986), reflecting considerable divergence of opinion.

In terms of biotite chemistry it is critical to decide whether the halogen content in the Koperberg Suite micas represents a pristine crystallization condition or whether the original halogen contents have been overprinted by secondary (metamorphic or deuteric) fluids. From the data given in this paper it is obvious that the existing halogen contents of the KSG biotites do not represent pristine compositions. The calculation of a meaningful exchange temperature from analytical data is fraught with imponderables. Amongst others, Van Aswegen (1988) indicated T_{max} for the Okiep Copper District to be below 800°C and the pressure to vary between 5.1 and 5.5 kb.

It is evident that Cl fractionates into biotite with decreasing Mg/Fe ratio while the opposite is true for F. As is the case at Santa Rita and Henderson (Munoz, 1984), the halogen content in Koperberg Suite biotites is dependent on the relative halogen acid activity during OH=halogen exchange. It is, however, difficult to define an activity-T_{HF} or activity-T_{HCl} path for fluids cooling from high temperatures. It is clear (Guidotti, 1984, Mora and Valley, 1989) that increasing metamorphic grade results in the concentration of Cl and F into hydrous phases, because dehydration of the rock results in fewer hydrous minerals. How- ever, partial melting events also tend to increase the halogen content of the hydrous phase in the residue (Holloway, 1987). The concurrent Mg and F enrichment apparent in Figure 7 probably indicates that the biotites were stabilized at high temperatures by low-energy Mg-F bonds. The conclusion reached above is supported by X-ray powder diffraction patterns identifying the highly refractory fluorphlogopite as one of the polytypes present in the Koperberg Suite (Boer, 1989). From the facts noted it becomes clear that an initially stable phlogopitic biotite crystallized from the original Koperberg magma and was subsequently subjected to mainly Mg-Fe and associated F-OH and Cl-OH exchange reactions. Relative hydrofluoric acid fugacity, $\log[f(H_2O)/f(HF)]$, calculated for the rim portions of KSG biotites (3.93, standard deviation = 0.0072) assuming 800°C and 5kb (Van Aswegen, 1988, McIver *et al.*, 1983), compares favourably with background values of the two main host rocks, namely the Concordia and Nababeep granite gneisses at about 3.85. These values were corroborated by Willner (pers. comm. 1988) in their work on the peraluminous metapelites (cordierite-biotite gneiss background value at 4.6) of the Bushmanland Sequence of central Western Namaqualand with a rather uniform ratio around 3. When the thermal history of the associated pelitic gneisses (Waters, 1986) is considered, this enrichment of F and Cl, together with associated mineral textures and paragenesis, indicate that the magmatic Koperberg Suite has been affected by a upper amphibolite or granulite facies metamorphic event.

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