A Pan-African zincian staurolite imprint on Namaqua quartz–gahnite–sillimanite assemblages

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Abstract

Quartz–gahnite–sillimanite asssemblages are described from a supracrustal enclave at Kraaifontein, 45 km south-west of Springbok in the Namaqualand Metamorphic Complex, South Africa. The assemblage formed by prograde reaction in the granulite facies zone during the 1100 Ma Namaqua event, possibly as the result of desulphidation of sphalerite. Subsequent lower-amphibolite-facies retrogression occurred in close proximity to shear zones during an early Pan-African metamorphic event at 700 Ma. Zincian staurolite formed as overgrowths on gahnite in a hydration reaction involving the consumption of gahnite, quartz, and sillimanite. Compositional zoning to more zinc-rich rims in gahnite at Kraaifontein is unrelated to the retrograde reaction, but is interpreted as a result of changing conditions during the prograde Namaqua event.

KEYWORDS: zinc, staurolite, gahnite, Kraaifontein, Namaqualand Metamorphic Complex.

Introduction

ZINC is known to show a preference for tetrahedrally-coordinated cation sites (Neumann, 1949; Albee, 1972; Griffen, 1981) that are commonly provided by staurolite in metapelitic assemblages at intermediate (lower amphibolite) metamorphic grades. In staurolite, Zn generally substitutes for tetrahedrally coordinated Fe^{2+} as a consequence of the similarity of their ionic radii (Miyake, 1985; Holdaway *et al.*, 1986*b*). Zinc partitions strongly into staurolite irrespective of the original concentrations in the host lithology (Stoddard, 1979; Tuisku *et al.*, 1987) due to the paucity of other suitable crystal sites in associated minerals.

Staurolite is a common constituent of metapelitic rocks in medium-grade amphibolite facies domains, with an upper stability limit that is generally defined by the reaction:

$$\text{staurolite} + \text{muscovite} + \text{quartz} \rightarrow \text{biotite} + Al_2SiO_5 + H_2O(\pm \text{garnet, Zn-rich spinel})$$

(Chinner, 1965; Guidotti, 1970; Schumacher, 1985). In muscovite-free assemblages, however, staurolite persists to somewhat higher upper-amphibolite grades (Ashworth, 1975; Yardley, 1981). At higher metamorphic grades (Ribbe, 1982), and at low modal proportions of staurolite (Guidotti, 1970), Zn is increasingly concentrated in staurolite.

Gahnite or zincian spinel $(Zn,Fe,Mg)Al_2O_4$ has been reported as a breakdown product of zincian staurolite in prograde metamorphic reactions at several localities (Atkin, 1978; Stoddard, 1979; Schumacher, 1985). Staurolite breakdown and desulphidation reactions involving sphalerite (Spry and Scott, 1986*b*; Moore and Reid, 1988) are common modes of formation of Zn-rich spinels. Zincian spinels, however, are stable over a much wider pressure–temperature range than zincian staurolite and are reported from phyllites in greenschist facies environments (Kramm, 1977) as well as from granulite facies rocks (Hicks *et al.*, 1985). In contrast to zincian staurolite, zincian spinels generally become progressively more Zn-rich as metamorphic grade decreases (Frost, 1973; Hicks *et al.*, 1985).

West of Springbok, Namaqualand, South Africa, a prograde Namaqua quartz–gahnite–sillimanite assemblage has been partially affected by a subsequent retrogressive Pan-African metamorphic overprint. The formation of zincian staurolite as a direct result of this event provides evidence of an interrelationship between zincian staurolite and zincian spinel that is the subject of this study.

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Geological setting

In the western part of the Namaqualand Metamorphic Complex (NMC), high-grade (upper amphibolite and granulite facies) assemblages formed during the Namaqua metamorphic event (Joubert, 1971; Waters, 1986) that attained peak conditions at approximately 1100 Ma (Clifford *et al.*, 1975). Quartz-gahnite ($\pm$sillimanite, garnet) assemblages represent a rare but widespread minor component (Hicks *et al.*, 1985; Spry and Scott, 1986*b*; Moore and Reid, 1988) of supracrustal sequences dominated by metaquartzitic, metapelitic and leucogneissic lithologies (Moore, 1986) that formed during this prograde event.

In the coastal portions of the western NMC, the Namaqua lithologies were overprinted by an early Pan-African retrograde metamorphic event

FIG. 1. Regional map of the western Namaqualand Metamorphic Complex, showing distribution of supracrustal enclaves (dark shaded), intrusive rocks (unadorned) and overlying Pan-African metasediments (cross-hatching). Regional metamorphic zonation includes Namaqua upper amphibolite facies (1), cordierite-garnet subzone of granulite facies (2), spinel-quartz subzone of granulite facies (3), and Pan-African staurolite zone (4) with reported staurolite occurrences (stars). Localities mentioned in text are Steenbok (SB), Kraaifontein (KR), Zoutpan (ZP), Sout River (SR), Oranjefontein (OR) and Gamsberg (GAM). Towns are Steinkopf (SK), Aggeneys (AG), Springbok (SPR), Garies (GAR), and Bitterfontein (BI).

at about 700 Ma (Allsop *et al.*, 1979) that attained lower-amphibolite-facies grades at the coast, decreasing in intensity eastwards. At Sout River in southern Namaqualand (Fig. 1), garnet–cordierite–sillimanite gneisses of Namaqua age were altered to staurolite-bearing mica schists during this Pan-African tectono-metamorphic event in which large-scale sinistral rotation of basement structures occurred (Waters *et al.*, 1983). Along the Steenbok Shear, east of Port Nolloth, transformation of metapelitic rocks of the NMC supracrustal sequences to kyanite–staurolite schists is believed to have occurred during this episode (Joubert and Waters, 1980).

On the Kraaifontein farm, 45 km south-west of Springbok, quartz–gahnite ($\pm$sillimanite, galena) assemblages with minor rutile occur as a few thin lenses ($<$ 1 m thick and 20 m long) in a mixed sequence of granulite-facies supracrustal lithologies. The host rocks comprise K-feldspar-quartz-cordierite–garnet ($\pm$plagioclase, sillimanite) metapelitic rocks, orthopyroxene-bearing mafic granulites, minor quartzites and calc-silicate rocks, and varieties of biotite-bearing and biotite-free leucocratic quartzo-feldspathic gneisses (Cumming, 1975). The supracrustal remnant is exposed as a 2 km wide east–west striking, north-dipping belt within a terrain dominated by syntectonically intrusive augen gneisses (Fig. 1).

The sequence of supracrustal gneisses is cut by several north-east-striking shear zones at Kraaifontein with associated retrograde metamorphism (Cumming, 1975). The presence of kyanite, staurolite and garnet within similar shear zones has been reported from several localities along the west coast (Jack, 1980; Joubert and Waters, 1980), including an area to the immediate west of Kraaifontein (Theart, 1980) (Fig. 1). The formation of these shear zones and the associated retrogressive reworking of the Namaqua basement occurred during the initial phases of a major cycle of Pan-African deformation and sedimentation (Waters *et al.*, 1983; Jackson and Zelt, 1984).

Petrography and mineral chemistry

Samples were collected from three quartz–gahnite lenses in the supracrustal sequence at Kraaifontein. One of these (KR-2) contained staurolite as an overgrowth of gahnite, whilst in the other two (KR-1, KR-3) staurolite was absent. The prograde Namaqua mineral assemblages, common to all three lenses, comprise highly strained, coarse xenoblastic quartz grains and coarse-to-medium-grained, xenoblastic-to-interstitial, dark green gahnite. Sillimanite occurs as scattered clusters of coarse euhedral laths in association with gahnite concentrations or as fine needles enclosed in quartz. Rutile and zircon are present as minor and accessory constituents respectively. Coarse goethite/anglesite-filled vughs after oxidized galena occur in samples KR-1 and KR-2 and completely sericitized feldspar is present in sample KR-3.

Aside from the sericitization of feldspar in sample KR-3 (which may be due to weathering processes), there is no evidence of retrograde metamorphism in samples KR-1 and KR-3. In sample KR-2, however, gahnite grains are clouded by opaque inclusions and rimmed by staurolite. All gradations from dusty gahnite 'ghosts' in the cores of staurolite clusters to fine staurolite rims surrounding coarse gahnite grains are observed. The staurolite grains show distinct crystal faces in contact with quartz (Fig. 2) but a poorly defined, amorphous contact with gahnite, indicating growth of staurolite from gahnite nuclei towards quartz. Discrete lath-like grains of staurolite are also present. Coarse intergrowths between staurolite and ilmenite occur in places.

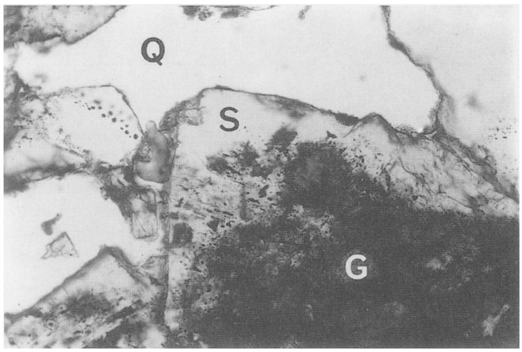

FIG. 2. Photomicrograph showing zincian staurolite (S) overgrowing gahnite (G) in the presence of quartz (Q) in sample KR-2. Note the well-formed crystal faces that staurolite presents to quartz, and the irregular outline of the relict gahnite with the contact obscured by fine Fe oxides. Photo length 1.5 mm.

Compositions of gahnite grains from Kraaifontein are plotted in Fig. 3 as end-member components (molecular %) of gahnite, hercynite, and spinel after correction for a magnetite component assuming ideal stoichiometry. The galaxite component ($<$ 1 mol. %) was ignored due to insignificant levels of Mn. The compositional range from samples with or without associated zincian staurolite is essentially identical ($Ghn_{56-67}Hc_{24-31}Sp_{9-13}$ in sample KR-2 and $Ghn_{58-66}Hc_{21-30}Sp_{12-14}$ in sample KR-1 respectively), although the mean Zn content of spinel in the absence of zincian staurolite is slightly lower (60.2 mol. % Zn, 26 analyses) than that associated with zincian staurolite (62.9 mol. % Zn, 25 analyses). The reality and/or significance of this difference is obscured by the fact that gahnites show compositional zoning in both rocks, although core and rim analyses were included in the above estimations of mean compositions.

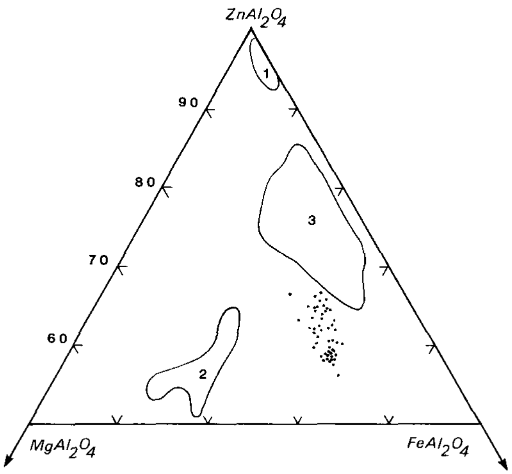

FIG. 3. Triangular plot showing the composition (molecular proportions) of gahnite from Kraaifontein (dots) in comparison with fields for gahnite from the greenschist facies at Oranjefontein (1), the granulite facies at Oranjefontein (2) (Hicks *et al.*, 1985) and the upper amphibolite facies at Aggeneys (Spry, 1987*b*).

Zoned gahnites have Fe-, Mg-rich cores and Zn-rich rims (Table 1), typical of most reported occurrences of compositional zoning in gahnite (Spry, 1987*a*). In sample KR-2 (zincian staurolite-bearing), Al is also slightly higher in rim analyses, whereas in sample KR-1, Al and Mg remain constant. Maximum measured compositional variation from core to rim in an individual grain is 8.1 mol. % Zn. The magnitude of compositional zoning does not appear to be affected by the presence or absence of zincian staurolite (Fig. 4).

In comparison with gahnite from other localities in the Namaqualand Metamorphic Complex, Kraaifontein gahnites are relatively Fe-, Mg-rich and Zn-poor, similar to those at other prograde granulite facies localities such as Oranjefontein (Hicks *et al.*, 1985). They differ from gahnites at amphibolite-facies localities, including the Aggeneys and Gamsberg ore deposits (Spry and Scott, 1986*b*; Spry, 1987*b*; Moore and Reid, 1988) in their higher Mg and relatively low Zn contents (Fig. 3).

Zn-rich staurolites at Kraaifontein have consistent compositions that vary only within a narrow range (Table 1). Their SiO_2 content is similar to that for staurolite from other localities, whereas their Al_2O_3 and TiO_2 contents are slightly lower (Holdaway *et al.*, 1986*b*). The total cation content per 46 oxygens, however, is relatively high for staurolite (mean 29.52 per formula unit compared to the range 29.33–29.58 obtained by Griffen and Ribbe, 1973). High R^{2+} components ($Fe+Mg+Mn+Zn$) and low Al(VI)—defined as ($Al+Si-8$)—per formula unit, may be indicative of the presence of Fe^{3+} in staurolite at Kraaifontein.

In the absence of garnet or biotite, staurolite can accommodate relatively high H_2O contents (2.25 wt. % H_2O, Holdaway *et al.*, 1986*a*). Zn-rich staurolite at Kraaifontein may contain, by difference, up to 2.50 wt. % H_2O (Table 1). One explanation that has been offered for high H_2O contents in staurolite is the homogeneous substitution reaction reported by Lonker (1983): $Al^{3+}+3(O^{2-}) \rightleftharpoons \text{vacancy}+3(OH^-)$. Low Al_2O_3 contents in Zn-rich staurolite at Kraaifontein may, therefore, be explained by the presence of Fe^{3+} and/or high levels of H_2O. Zn-rich staurolites reported by Spry and Scott (1986*a*) also have low Al_2O_3 contents. No relationship, however, was detected between Zn content and OH content by Lonker (1983).

Staurolite from Kraaifontein contains from 4.35 to 4.90 wt. % ZnO. This is anomalously high for staurolite although values in excess of 7.0 wt. % ZnO have been reported (Griffen and Ribbe, 1973; Miyake, 1985; Spry and Scott, 1986*a*). Zn^{2+} substitution for Fe^{2+} in the tetrahedral site of staurolite predominates over all other substitutions (Griffen, 1981; Miyake, 1985). End-member zincian staurolites have been synthesized (Griffen, 1981) but have not been reported from natural occurrences. Many naturally occurring zincian staurolites with ZnO contents greater than 6 wt. % are thought to have formed by desulphidation of sphalerite (Spry and Scott, 1986*a*). Staurolites with ZnO contents lower than 6 wt. % generally formed in reactions that did not involve sphalerite.

In contrast to its common occurrence in regions affected by the Pan-African event, staurolite is a rare mineral in earlier prograde Namaqua assemblages in the western NMC. Occurrences are generally restricted to the lower amphibolite facies in the Vioolsdrif–Steinkopf area where staurolite occurs in metastable assemblages in cordierite-bearing metapelitic rocks (Ward, 1977; P. Booth,

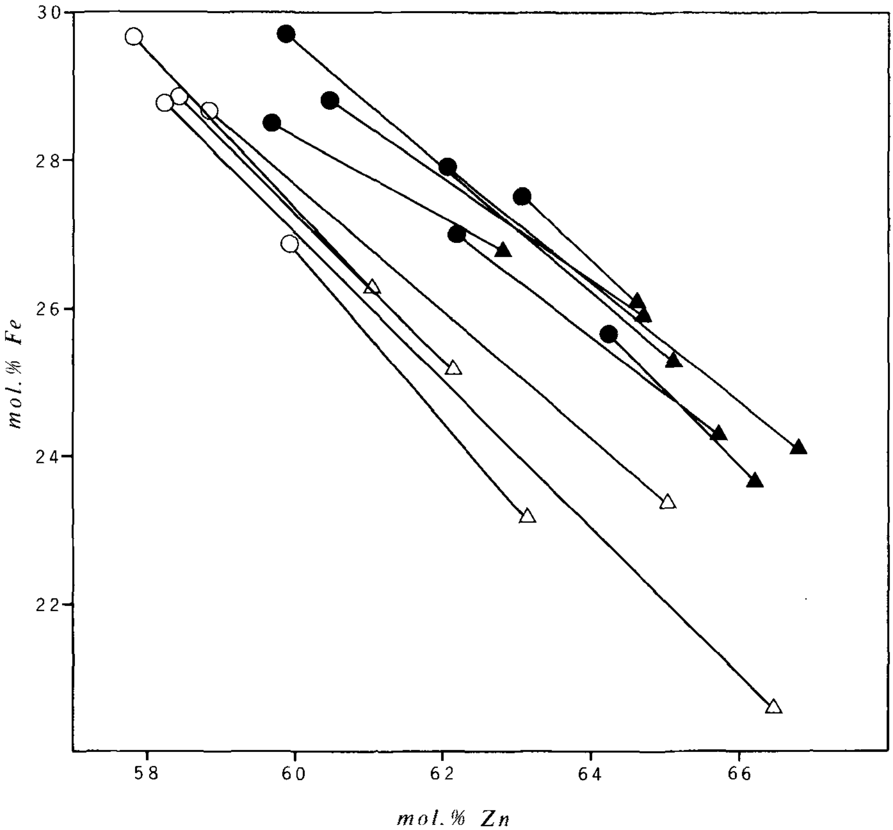

FIG. 4. Plot showing the variation in Fe and Zn content between cores (circles) and rims (triangles) of zoned gahnites from Kraaifontein. Gahnites from sample KR-2 (solid) and sample KR-1 (open).

pers. comm.). Rare examples of zincian staurolite are reported from Gamsberg (Spry and Scott, 1986*b*), containing 2.8 wt. % ZnO, and Swartkoppies, north of Gamsberg (A. Willner, pers. comm.). In the latter two instances, remnant zincian staurolite appears to have survived breakdown reactions involving the formation of zincian spinel in an area where upper amphibolite facies P–T conditions (700 °C and 6 kbar) prevailed (Moore, 1977).

Discussion

Zn-rich staurolite at Kraaifontein is observed to have formed as a result of the superimposition of a localized retrograde amphibolite facies Pan-African metamorphic imprint on a high-grade Namaqua quartz–gahnite–sillimanite assemblage. This reaction involves hydration, and may be written for the Zn end-member components as follows:

$$\underset{\text{gahnite}}{4ZnAl_2O_4} + \underset{\text{quartz}}{3SiO_2} + \underset{\text{sillimanite}}{5Al_2SiO_5} + \underset{\text{fluid}}{H_2O} \rightarrow \underset{\text{staurolite}}{Zn_4Al_{18}Si_8O_{46}(OH)_2}.$$

A close relationship between zincian spinel and zincian staurolite has been previously observed (Atkin, 1978; Stoddard, 1979; Schumacher, 1985), but in prograde reactions where staurolite breaks down to produce gahnite.

The Zn/Fe ratios of gahnite (± 2.15) and of staurolite (± 0.35) are markedly different (Table 1), requiring either the addition of Fe to, or the removal of Zn from the above equation, or partitioning of the two elements between newly formed staurolite and relict gahnite during the reaction. It could be argued that evidence for partitioning is provided by the compositional zoning to Zn-rich rims in the relict gahnite. However, comparable zoning is also present in sample KR-1 which lacks staurolite (Fig. 4). Compositional zoning at Kraaifontein, therefore, appears

TABLE 1. Analyses of zincian staurolite and gahnite from Kraaifontein, Namaqualand Metamorphic Complex, South Africa.

	1		2	3	4	5
SiO_2	27.40	(27.41, 26.97 - 27.89)	-	-	-	-
TiO_2	0.29	(0.29, 0.14 - 0.45)	-	-	-	-
Al_2O_3	52.58	(52.08, 50.92 - 52.84)	54.11	55.93	56.01	55.94
FeO*	10.70	(10.96, 10.28 - 11.47)	14.18	11.21	13.79	12.58
MgO	1.75	(1.77, 1.40 - 1.99)	2.29	2.08	2.88	2.93
MnO	0.25	(0.24, 0.16 - 0.29)	0.16	0.07	0.41	0.40
ZnO	4.63	(4.66, 4.35 - 4.90)	28.52	30.68	26.75	28.21
Total	97.60		99.26	99.97	99.84	100.06
	Cations per 46 Oxygens		Cations per 4 Oxygens			
Si	7.714		-	-	-	-
Ti	0.061		-	-	-	-
Al	17.448		1.928	1.965	1.957	1.954
Fe	2.519		0.358	0.279	0.342	0.312
Mg	0.962		0.103	0.092	0.127	0.129
Mn	0.059		0.004	0.002	0.010	0.010
Zn	0.735		0.637	0.675	0.585	0.617
Total	29.498		3.030	3.013	3.021	3.022

* Total iron as FeO. 1. Zincian staurolite, sample KR-2. Numbers in brackets are mean compositions and range for 19 analyses. 2. Gahnite core, sample KR-2. 3. Gahnite rim, same grain, sample KR-2. 4. Gahnite core, sample KR-1. 5. Gahnite rim, same grain, sample KR-1.

Analyses by electron microprobe (Cameca, accelerating potential 15 kV, sample current 40 nA, against natural and synthetic mineral standards).

to be unrelated to staurolite formation. The slightly higher overall Zn content of gahnite associated with staurolite (± 2.7 mol. % Zn, see also Fig. 4) may be a consequence of the difference in Zn/Fe ratios between staurolite and gahnite. However, these small differences could equally represent primary variations between and within the quartz–gahnite–sillimanite lenses. The presence of minor rutile in the prograde assemblage and ilmenite in the retrograde assemblage is consistent with a change in the Fe–Zn budget with the introduction of Fe.

Retrograde metamorphic effects have also been observed in quartz–gahnite rocks at Oranjefontein, 90 km east of Kraaifontein (Fig. 1). Here prograde granulite-facies quartz–gahnite ($\pm$garnet, phlogopite) assemblages were replaced by retrograde greenschist facies assemblages containing end-member gahnite, chlorite, hematite and rutile (Hicks *et al.*, 1985). Hydration occurred at Oranjefontein under varying degrees of open-system conditions, associated with east–west-striking shear zones.

Pressure–temperature conditions prevailing during the retrograde event at Kraaifontein were apparently considerably higher than those at Oranjefontein. Temperatures of 585 °C and pressures of 5 kbar are reported from garnet–biotite and garnet–staurolite pairs at Zoutpan, 30 km south of Kraaifontein (Joubert and Waters, 1980) and similar, or slightly lower *P–T* conditions probably prevailed at Kraaifontein given the general east–west zonation of Pan-African metamorphism along the west coast (Fig. 1). Temperatures in excess of 540 °C are generally required for the formation of staurolite (Hoschek, 1969). In the NMC, zincian spinels have formed under low-grade (greenschist) and high-grade (upper amphibolite/granulite) metamorphic conditions, whereas zincian staurolite was preferred at intermediate (lower amphibolite) *P–T* conditions.

Compositional zoning in zincian spinel has been described as resulting from partitioning of Zn, Fe and Mg between the spinel and adjacent sulphides or ferromagnesian silicate minerals during retrograde metamorphism (Spry, 1987*a*). At Kraaifontein, compositional zoning is present in gahnite in sample KR-1 which lacks other ferromagnesian minerals and also lacks visible evidence of the retrograde Pan-African imprint. Identical zoning is present in relict gahnite rimmed by staurolite in sample KR-2. The zoning is thus thought to be a prograde rather than a retrograde phenomenon at Kraaifontein. Processes whereby compositional zoning formed as a result of progressive changes in oxygen and sulphur fugacity during the course of prograde desulphidation reactions, as proposed by Moore and Reid (1988), are therefore favoured. The presence of oxidized coarse blebs after galena in the quartz–gahnite–sillimanite rocks at Kraaifontein, indicate that desulphidation of sphalerite was a likely mode for formation of gahnite during the prograde Namaqua metamorphic event.

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