

IMPLICATIONS OF SPHALERITE INCLUSIONS IN GAHNITE FROM THE NAMAQUALAND METAMORPHIC COMPLEX, SOUTH AFRICA

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ABSTRACT

Inclusions of sphalerite in gahnite are reported from a quartzite at Kontorogab, 45 km northeast of Springbok in the Namaqualand Metamorphic Complex, South Africa. Gahnite occurs in a layer, 5 cm thick, in the basal portion of the quartzite. Gahnite concentrations are greatest along the upper (stratigraphic) contact and in cross-cutting veinlets within the layer. Sphalerite inclusions are restricted to larger gahnite grains in the central part of the layer. There is a progressive variation in gahnite composition from the upper contact to the central part of the layer where gahnite is more Fe-, Mg-rich and has a Zn/Fe value identical to that of the enclosed sphalerite. Gahnite is considered to have formed during a desulfidation reaction involving sphalerite and a metamorphic fluid. The reaction was controlled by the oxidized fluid along the major parting, where Zn-rich gahnite formed and sphalerite inclusions are absent. Penetration of the central part of the layer by the fluid was restricted, resulting in local preservation of sphalerite. Persistence of more reducing conditions due to desulfidation caused more Fe-, Mg-rich gahnite to crystallize in this region. Individual zoned grains reported from other localities may have formed by a similar process.

Keywords: sphalerite, gahnite, electron-microprobe analyses, metamorphic fluid, metamorphic reaction, Namaqualand, South Africa.

SOMMAIRE

Nous décrivons la présence d'inclusions de sphalérite dans la gahnite dans une quartzite échantillonnées à Kontorogab, situé à 45 km au nord-est de Springbok, dans le complexe métamorphique de Namaqualand (Afrique du Sud). On trouve la gahnite dans un horizon d'une épaisseur de 5 cm, dans la partie inférieure de la quartzite; ce minéral est concentré le long du contact stratigraphique supérieur et dans les veines qui recoupent ce niveau. Seuls les plus gros grains de gahnite possèdent des inclusions de sphalérite dans la partie centrale de cet horizon. La gahnite devient progressivement plus riche en fer et en magnésium du contact supérieur vers la partie centrale du niveau minéralisé; à cet endroit, gahnite et sphalérite partagent la même valeur Zn/Fe. La gahnite se serait formée au cours d'une réaction de désulfuration de la sphalérite en présence d'une phase fluide métamorphique. La réaction est régie par un fluide oxydant le long du plan de séparation important, où gahnite zincifère et sphalérite sont absentes. À cause de la

pénétration limitée de la phase fluide dans la zone interne du niveau, la sphalérite y est préservée. La persistance d'un milieu plus réducteur dû à la désulfuration donne une gahnite enrichie en Fe et en Mg dans cette région. La zonation chimique des cristaux que l'on a découvert ailleurs pourrait avoir la même origine.

(Traduit par la Rédaction)

Mots-clés: sphalérite, gahnite, analyses à la microsonde électronique, fluide métamorphique, réaction métamorphique, Namaqualand, Afrique du Sud.

INTRODUCTION

The common association of gahnite (Zn,Fe,Mg) Al₂O₄ with metamorphosed sulfide deposits (Plimer 1977, Sundblad 1982) has led to the proposal that zincian spinel forms by desulfidation of sphalerite in prograde metamorphic reactions with aluminosilicate minerals such as garnet, gedrite, kyanite or sillimanite (Wall & England 1979, Williams 1983, Spry & Scott 1986a,b). Although this hypothesis has a sound theoretical and experimental basis (Wall & England 1979, Spry 1984, Spry & Scott 1986b), field evidence for sphalerite breakdown is generally lacking (Sandhaus & Craig 1986, Spry & Scott 1986a) and gahnite is commonly found in natural occurrences as porphyroblasts showing textural equilibrium with sphalerite.

The formation of zincian spinel from Zn-bearing silicate phases (staurolite, biotite), generally as a minor constituent, during prograde or retrograde metamorphism in metapelitic rocks has also been demonstrated (Atkin 1978, Stoddard 1979, Dietvorst 1980, Schumacher 1985). In addition, Wall (1977) has proposed that fluids from a metamorphic or hydrothermal source can play a direct role in the precipitation of gahnite.

In the western Namaqualand Metamorphic Complex, South Africa, gahnite occurs as a minor constituent of base-metal sulfide deposits at Aggeneys and Gamsberg (Ryan *et al.* 1986, Rozendaal & Stumpfl 1984, Spry & Scott 1986b) and also forms impersistent layers and lenses in quartzitic and garnet-rich rocks (Hicks *et al.* 1985, van der Westhuizen *et al.* 1986). Quartzites along the southern limb of the Geselskapbank synform, Namaqualand, contain narrow bands of gahnite of varied composition. Textural and compositional data obtai-

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ned from mineral assemblages at this locality have a direct bearing on proposals for the formation of gahnite by desulfidation of sphalerite and for local migration of Zn-, Al-bearing fluids.

GEOLOGICAL SETTING

At Naab se Berg Suid, one of a range of hills in the northern corner of the farm Kontorogab, 45 km northeast of Springbok, western Cape Province (Fig. 1), thin layers of blue-green gahnite are present within the basal portions of a white quartzite unit. The gahnite occurrences are located on the overturned limb of an isoclinal synform at, or close to, the contact with overlying, but stratigraphically lower, quartz-biotite-muscovite-sillimanite schist. The complex thrust-dominated structural-stratigraphic history of the area, situated on the southern limb of the major Geselskapbank synform, is described by Strydom & Visser (1986).

The host lithologies are mid-Proterozoic supracrustal metasediments of the Namaqualand Metamorphic Complex (NMC), and the regional stratigraphy at Kontorogab includes a basal gneiss

complex, an overlying biotite gneiss-schist-quartzite supracrustal succession, and younger intrusive leucogranitic rocks. The stratigraphic position of the gahnite lenses at the schist-quartzite contact is similar to that observed at Oranjefontein (Hicks *et al.* 1985), some 40 km due south. White quartzite units from this portion of the western NMC are interpreted as representing metamorphosed cherty sediments, and mica-sillimanite schists as potassic shales (Moore 1986) in a thin, but extensive sequence of supracrustal rocks that act as hosts to major deposits of base-metal sulfide at Aggeneys and Gamsberg (Rozendaal 1986, Ryan *et al.* 1986).

Mineral assemblages within the host metapelitic and amphibolitic lithologies indicate upper amphibolite-facies grades of metamorphism. *P-T* conditions are estimated at $650 \pm 50^\circ\text{C}$ and 4.5 kilobars (Grütter 1986) for garnet-cordierite-bearing metapelitic rocks at Geselskapbank, 5 km to the west. Similar temperatures and pressures are proposed for the Aggeneys-Gamsberg area, 60 km to the east (Moore 1977, Rozendaal 1978), whereas higher-temperature granulite-facies conditions are present at Oranjefontein to the south.

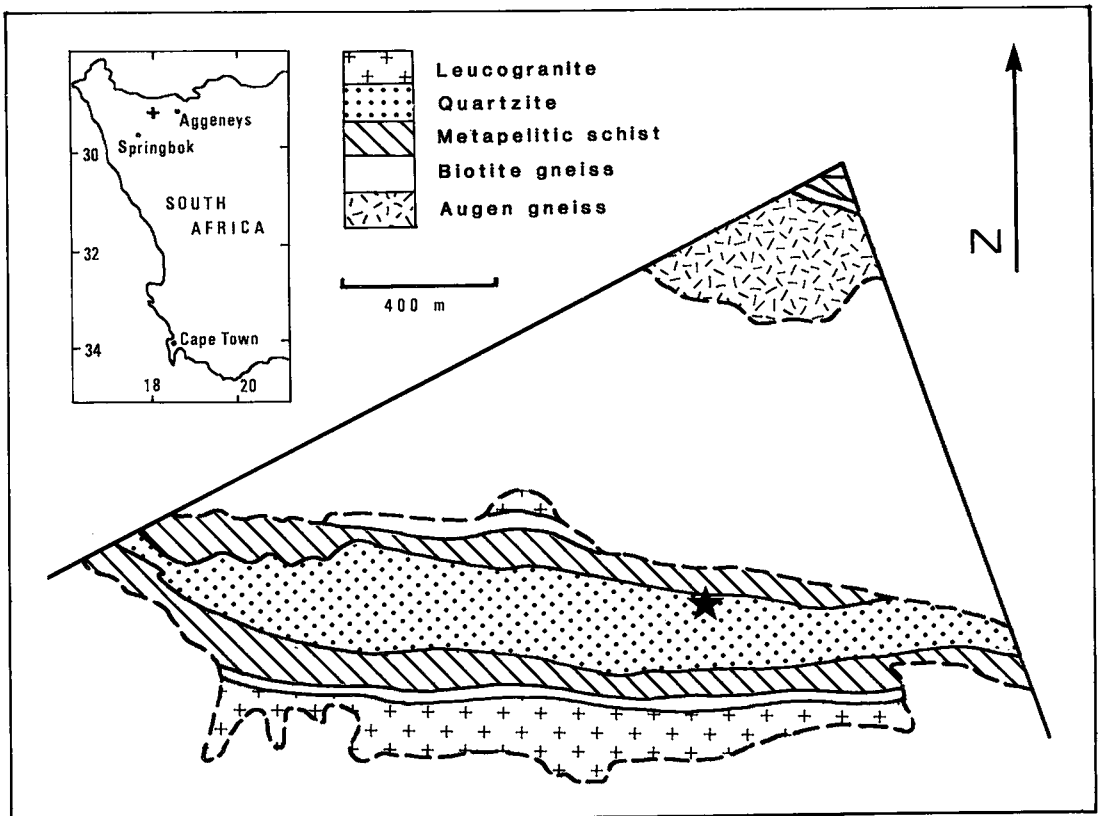


FIG. 1. Geological map of the Naab se Berg Suid locality on the farm Kontorogab (simplified from Strydom 1985). The sample site is marked by a star. Inset: map of western South Africa, showing the locality of Kontorogab farm (cross).

PETROGRAPHY

The white quartzite is a coarsely bedded (20 – 50 cm) unit with thin intercalated layers (1 – 15 cm) of mica-sillimanite schist and rarer fine layers or partings (1 – 10 cm) containing concentrations of gahnite, in places together with lesser sillimanite. The quartzite itself contains very minor amounts of disseminated muscovite, biotite, garnet, pyrite and magnetite.

Close examination of one specific gahnite-bearing layer revealed the following subdivisions: (i) a dense concentration of interlocking, anhedral grains of gahnite with minor interstitial quartz along a major parting, which is interpreted as a modified bedding plane; (ii) a layer, up to 2 cm thick, of disseminated anhedral to subhedral gahnite in quartz, together with minor concentrations of gahnite as thin, impermanent layers that are also interpreted as bedding foliations. Minor galena is present, and certain larger grains of gahnite contain small inclusions of sphalerite (Fig. 2); (iii) a 3-to-4-cm zone containing randomly oriented, disseminated phlogopite laths in a quartz matrix and isolated, commonly euhedral, octahedra of gahnite that become progressively rarer with increasing distance from zone (ii); and (iv) a few irregular, cross-cutting fracture planes within zone (iii) containing dense concentrations of coarse anhedral gahnite and small amounts of muscovite. The spatial relationship of the various zones is shown in Figure 3.

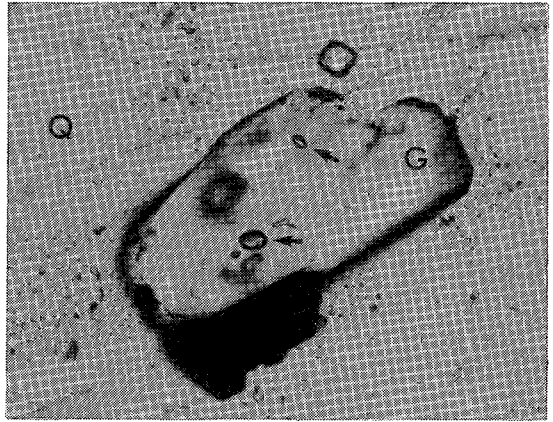


FIG. 2. Photomicrograph of sphalerite inclusions (arrows) in a gahnite grain (G) with associated galena (black) and surrounding quartz matrix (Q). Inclusions out of focus below surface of gahnite also appear to be sphalerite. Length of gahnite grain approximately 200 μm .

Quartz occurs as large recrystallized, relatively strain-free, anhedral grains in zones (ii) and (iii), totally enclosing the impermanent layers and disseminated grains of gahnite. Quartz is generally finer-grained and granular along the major parting. Rutile occurs as rare fine euhedral grains.

Sphalerite inclusions in gahnite from zone (ii) commonly have a rounded to spheroidal shape (5 – 24

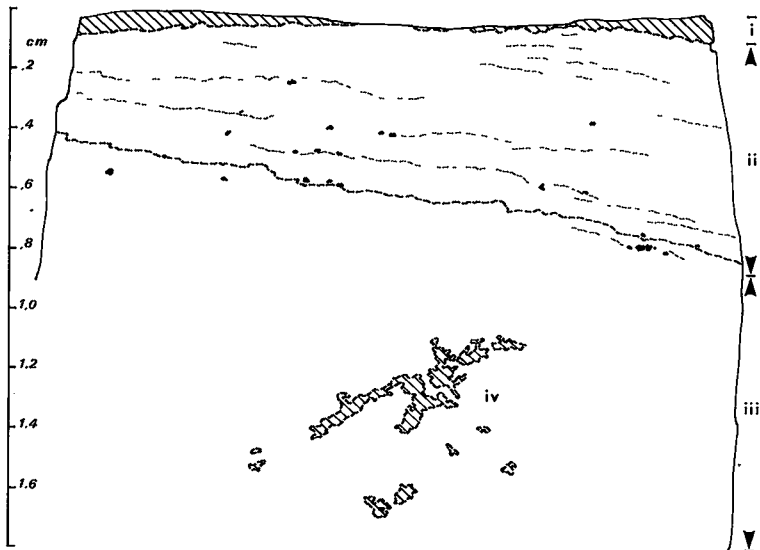


FIG. 3. Sketch of thin-section AMP2E2, showing the four zones (i,ii,iii,iv) containing gahnite. In zone (ii), gahnite grains with sphalerite inclusions are emphasized (black), together with foliation planes (broken lines). The thin section is cut at right angles to foliation. Scale shows distance from top of major parting.

μm diameter) and are totally enclosed in the gahnite host. They occur either as single inclusions or clusters of several grains and are unrelated to fractures or other defects in the gahnite. In addition to sphalerite, quartz is a common inclusion in gahnite throughout all four zones.

MINERAL CHEMISTRY

Gahnite compositions from Kontorogab are presented in Figure 4 in terms of molar proportions of end-member gahnite, hercynite and spinel. The compositional range is $\text{Ghn}_{83-92}\text{Hc}_{3-8}\text{Spl}_{5-9}$, and there is a systematic variation from Zn-rich gahnite along the major parting [zone (i)] to more Fe-, Mg-rich compositions in zones (ii) and (iii) (Fig. 5, Table 1). Gahnite from Kontorogab is more Zn-rich than that reported from other localities in western Namaqualand (Hicks *et al.* 1985, Spry & Scott 1986b) with the exception of gahnite from the retrograde greenschist-facies zone at Oranjefontein (Hicks *et al.* 1985). The gahnite from Kontorogab is relatively Mg-rich compared with that from other localities, excluding Oranjefontein. The Fe/Mg value of gahnite from Kontorogab remains constant despite

variation in Zn content. Mineral zoning was not observed in individual grains.

Sphalerite inclusions in gahnite from zone (ii) have a minor Fe content (range from 5.3 to 6.8 wt.% Fe, 6 samples) and have Zn/Fe values (8 – 12) identical to that of enclosing gahnite. Along the major parting [zone (i)], where sphalerite inclusions are absent, the Zn/Fe values of gahnite are considerably higher (approximately 20). Sphalerite shows an extremely wide range in Fe content at Gamsberg (0.2 to 11.6% Fe, Rozendaal & Stumpfl 1984) and at Aggeneys (1.0 to 13.7% Fe, estimated from De Waal & Johnson 1981). The most common values from these two localities are in the 8 to 10% Fe range, significantly higher than at Kontorogab.

Phlogopite laths from zone (iii) have Mg/(Mg + Fe) values near 0.77 and contain minor amounts of Zn (mean 0.48, range 0.29 to 0.71 wt.% ZnO, 12 samples), similar to that reported from Oranjefontein (Hicks *et al.* 1985). The micas have low K contents and relatively high Al contents (Table 1), indicating that a certain degree of alteration (chloritization) has probably taken place. Phlogopite from Kontorogab has a distinct minor F content (mean 1.92, range 1.75 to 2.26 wt.% F).

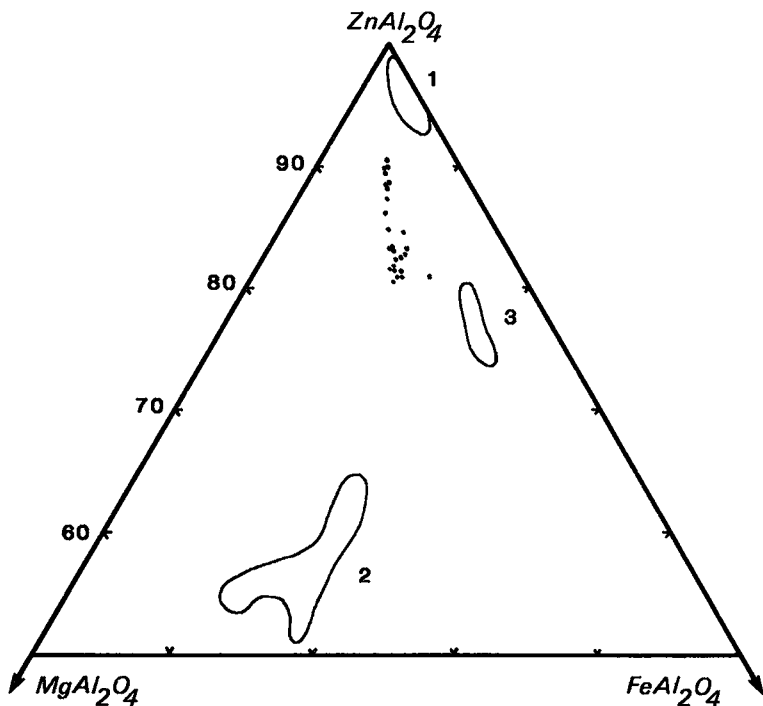


FIG. 4. Triangular plot showing the composition of gahnite from Kontorogab, in comparison with fields obtained for blue gahnite (1) and green gahnite (2) at Oranjefontein, and gahnite from the Broken Hill orebody, Aggeneys (3) (data from Hicks *et al.* 1985). Compositions have been normalized to end-member gahnite, spinel and hercynite after removal of calculated magnetite.

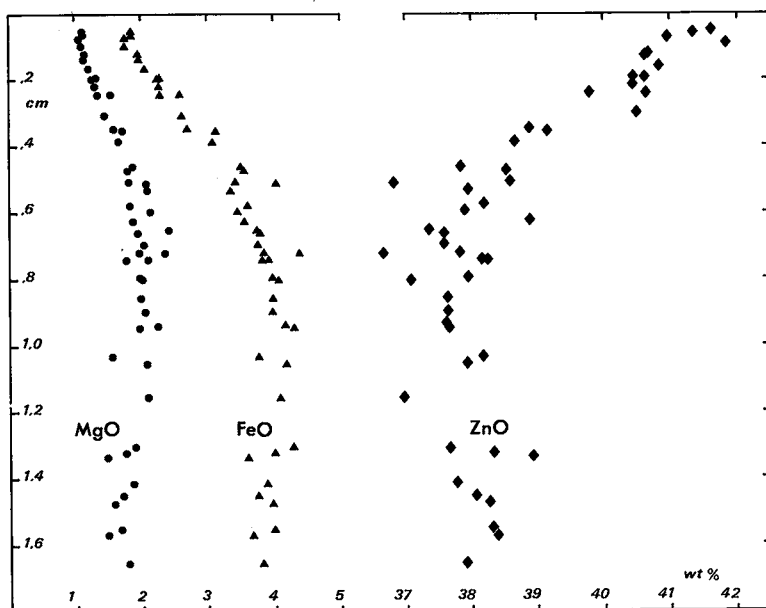


FIG. 5. Compositional profile of gahnite across thin-section AMP2E2. Plotted data (wt.% MgO, FeO, ZnO) are taken from complete electron-microprobe analyses from a series of traverses. Scale shows distance from top contact of major parting for direct comparison with Figure 3.

DISCUSSION

The presence in zone (ii) of sphalerite inclusions in the core of the larger grains of gahnite, together with the similar Zn/Fe values of sphalerite and enclosing gahnite, indicates that the gahnite formed during reactions involving the desulfidation and breakdown of sphalerite. These reactions require the participation of Al- and Mg-bearing phases that were not observed as mineral constituents in zone (ii). Besides gahnite, zone (ii) comprises quartz, galena, sphalerite and rare rutile. It is conceivable that pre-existing aluminosilicate may have been consumed in the gahnite-forming reaction, leaving excess Zn as sphalerite inclusions.

An alternative possibility is that Al and Mg were introduced as components of the metamorphic fluid that infiltrated along grain boundaries or within pore spaces. This proposal is supported by the occurrence of gahnite along cross-cutting fractures in zone (iii). These dense concentrations in a zone otherwise containing very minor disseminated gahnite provide evidence of the local migration of Al, Zn, Fe and Mg. The observation that gahnite surrounds sphalerite, and not an aluminosilicate mineral, implies that the Al-bearing constituent was more mobile than the Zn-bearing one.

In the absence of complexing agents, the solubility of Al_2O_3 in metamorphic fluids is extremely low. Significant amounts of Al are generally found only

in solution as alkali- Al_2O_3 complexes (Anderson & Burnham 1983) under the metamorphic conditions existing in the western NMC at the time of gahnite formation. Reactions such as the following may have been responsible for the formation of gahnite:

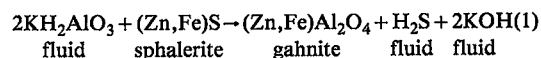


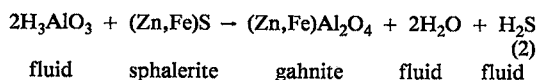
TABLE 1. CHEMICAL COMPOSITION OF GAHNITE AND PHLOGOPITE FROM KONTOROGAS

	1. Gahnite	2. Gahnite	3. Phlogopite
SiO_2	0.06	0.00	37.16
TiO_2	0.00	0.05	1.89
Al_2O_3	56.25	55.61	16.87
Cr_2O_3	0.00	0.19	
FeO^*	3.91	1.73	9.29
MnO	0.00	0.08	0.02
MgO	2.10	1.12	17.85
Na_2O			0.16
K_2O			0.61
ZnO			0.29
F			2.26
TOTAL:	100.49	99.41	94.39
O	4	4	22
Si	0.002	0.000	5.552
Ti	0.000	0.001	0.212
Al	1.970	1.988	2.970
Cr	0.000	0.002	
Fe	0.098	0.044	1.161
Mn	0.000	0.002	0.002
Mg	0.093	0.051	3.975
Na			0.046
K			1.641
Zn	0.838	0.912	0.032
F			1.066
	3.001	3.000	

* Total iron as FeO. 1. Gahnite containing sphalerite inclusion from zone (ii), sample AMP2E2. 2. Gahnite from the major parting (zone (i)), sample AMP2E2. 3. Phlogopite from zone (iii), sample AMP2E2. Analyses by electron microprobe (Cameca, accelerating potential 15 kV, sample current 40 nA, against natural and synthetic mineral standards).

K-, Na-bearing gneisses and metapelitic schists predominate in the lithological suite of the western NMC (Moore 1986). Breakdown of K-feldspar to sillimanite-quartz intergrowths along grain boundaries (Grütter 1986) and as discrete nodules (Moore 1977) is a relatively common phenomenon in leucocratic quartzo-feldspathic gneisses and certain metapelitic rocks. In these de-alkalization reactions, K and Na are removed by metamorphic fluids as ionic species (Eugster 1970) that could well include alkali-Al₂O₃ complexes.

In low-pH metamorphic fluids, particularly in the presence of fluoride in solution (Hem 1970), H₃AlO₃ may be present as a dissolved Al species, although probably at lower concentrations than the alkali complexes of reaction (1). Fluorite is a widespread trace constituent of the leucogneisses of the western NMC (Moore 1986), and the presence of substantial amounts of fluorine in phlogopite at Kontorogab suggests that F-bearing fluids may have been present during the major metamorphic reactions. A low value of the ratio X_K^+/X_{H^+} in the metamorphic fluid could result in the following reaction:



Low-pH metamorphic fluids are considered to have been active elsewhere in the western NMC (Waters & Moore 1985).

The concentration levels of Mg in the metamorphic fluid necessary to form gahnite of the composition recorded at Kontorogab are insignificant compared to those of Al (Table 1). Mg (and Fe) are commonly transported in pore solutions as chloride complexes (Roedder 1972, Vidale 1983) with Mg partitioning preferentially into coexisting solid phases in certain instances (Schuilen 1980).

The above fluid-solid reactions differ significantly from the solid-solid reactions proposed for the desulfidation process by Spry & Scott (1986b). These involve the breakdown of aluminosilicate (Al₂SiO₅) and sphalerite to form gahnite and quartz, or garnet and sphalerite to form gahnite, pyrrhotite + pyrite and quartz. Whereas it is conceivable that aluminosilicate (sillimanite) originally coexisted with sphalerite along the major parting and subsequently reacted to form gahnite, it seems unlikely that an Al-silicate phase coexisted with sphalerite in zone (ii). Aluminosilicate-sphalerite breakdown reactions, moreover, do not account for the presence of Mg in the gahnite of either zone.

Owing to the disseminated nature of gahnite in zone (ii), it is improbable that an Al-silicate phase was invariably in contact with scattered sphalerite and was consumed by a solid-solid reaction. The

gahnite-forming reaction in zone (ii) required the migration of Al (and Mg) by diffusion or fluid infiltration, an apparently inefficient process that allowed for the preservation of sphalerite remnants in the cores of some of the larger grains of gahnite and resulted in a distinct Al deficiency in the octahedral site of gahnite from zone (ii) compared to gahnite from zone (i) (Table 1).

The hercynite and spinel components of zincian spinel coexisting with quartz are limited by the prevailing *P-T* conditions (Bohlen *et al.* 1986, Seifert & Schumacher 1986). Zincian spinel, as a result, generally becomes progressively more Zn-rich at lower metamorphic grades (Frost 1973, Kramm 1977, Hicks *et al.* 1985). In comparison with gahnite coexisting with quartz at other localities in the western NMC (Fig. 4), the gahnite from Kontorogab is relatively Zn-rich. This may imply that slightly lower *P-T* conditions existed at Naab se Berg Suid compared to those at Aggeneys, Gamsberg and Geselskapbank.

The systematic, mm-scale compositional variation observed at Kontorogab (Fig. 5), however, cannot be attributed to *P-T* differences but is more likely the result of variation in oxygen fugacity at constant *P-T* between the various zones. Zone (i) represents the major parting, where advection or flow of fluid and relatively high externally controlled $f(\text{O}_2)$ conditions might be expected. In contrast, zone (ii) is a region where fluid flow was replaced by diffusive transport, reaction with sphalerite was incomplete, and internally controlled $f(\text{O}_2)$ was reduced as a direct result of the more dominant role assumed by desulfidation. Under conditions of higher $f(\text{O}_2)$ at constant *P* and *T*, Spry & Scott (1986b) have calculated that gahnite formed by breakdown of sphalerite in solid-solid reactions is richer in Zn. Similar trends may be anticipated in the above fluid-solid reactions (1) and (2).

The variation in composition of the zincian spinel at Kontorogab, from more Fe-, Mg-rich compositions in association with sphalerite in zone (ii) to more Zn-rich compositions at the major parting, is similar to the zoning observed from core to rim in individual grains of gahnite at other localities (Spry 1987). This zonation is ascribed to re-equilibration of gahnite in the presence of sphalerite, Fe sulfides and silicates as the assemblage cooled, or during a retrograde metamorphic event (Dietvorst 1980, Spry 1987). At Oranjefontein, however, retrograde metamorphism has resulted in a discrete overgrowth of one zincian spinel phase on another, and continuous mineral zoning is absent (Hicks *et al.* 1985).

Some of the more spectacular examples of mineral zoning are to be found in systems such as veins and cavity fillings where mineral growth occurs in the presence of a fluid phase. An alternative suggestion, therefore, based on the above model for the origin of gahnite from Kontorogab, is that the more Fe-

rich core of zoned grains formed in the early stages of a prograde metamorphic reaction (desulfidation) under internally controlled diffusive conditions with low $f(\text{O}_2)$ due to the desulfidation process. The Zn-rich rim, however, equilibrated at a slightly later stage in the metamorphic event, when externally controlled metamorphic fluids of markedly different $f(\text{O}_2)$ had infiltrated along grain boundaries.

This type of mineral zonation should be most obvious in zincian spinels formed during desulfidation reactions due to potentially large variations in $f(\text{O}_2)$. Significantly, all the occurrences of zincian spinel showing compositional zoning reported by Spry (1987) are from metamorphosed massive-sulfide deposits. The lack of zonation at many localities (Spry 1987) could be attributed to the absence of significant volumes of an advective fluid phase at the grain boundaries. Rare occurrences of reversed zoning would be due to low $f(\text{O}_2)$ conditions associated with an externally controlled fluid of unusual composition.

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