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The significance of cordierite–hypersthene assemblages from the Beitbridge region of the Central Limpopo Belt; evidence for rapid decompression in the Archaean?

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Abstract

The following model equilibria have been observed in metapelites from the Diti Formation of the Central Limpopo belt.

$$\underset{\text{Orthopyroxene}}{(Mg,Fe)_2Si_2O_6} + \underset{\text{Sillimanite}}{2\ Al_2SiO_5} + \underset{\text{Quartz}}{SiO_2} = \underset{\text{Cordierite}}{(Mg,Fe)_2Al_4Si_5O_{18}}$$

$$\underset{\text{Garnet}}{2\ (Mg,Fe)_3Al_2Si_3O_{12}} + \underset{\text{Sillimanite}}{4\ Al_2SiO_5} + \underset{\text{Quartz}}{5\ SiO_2} = \underset{\text{Cordierite}}{3\ (Mg,Fe)_2Al_4Si_5O_{18}}$$

$$\underset{\text{Garnet}}{2\ (Mg,Fe)_3Al_2Si_3O_{12}} + \underset{\text{Quartz}}{3\ SiO_2} = \underset{\text{Cordierite}}{(Mg,Fe)_2Al_4Si_5O_{18}} + \underset{\text{Orthopyroxene}}{2\ (Mg,Fe)_2Si_2O_6}$$

$$\underset{\text{Orthopyroxene}}{5\ (Mg,Fe)_2Si_2O_6} + \underset{\text{Sillimanite}}{10\ Al_2SiO_5} = \underset{\text{Cordierite}}{4\ (Mg,Fe)_2Al_4Si_5O_{18}} + \underset{\text{Spinel}}{2\ (Mg,Fe)Al_2O_4}$$

An internally consistent thermodynamic data set in the MAS system for constituent phases has been derived which is also consistent with both calorimetric data and experimental equilibria. Microprobe analyses of the phases present in the Limpopo metapelites have been applied to equilibrium equations derived from this data set, and conditions of P = 4–5 kbar, $T > 670$°C are implied assuming $a_{H_2O} = 0$. Application of published cordierite-free thermobarometers both from the pelites and interlayered mafic granulites, provides similar P–T conditions (P = 3.5–5 kbar, $T = 750 \pm 50$°C) indicating that metamorphism was virtually dry. Conditions of $a_{H_2O} < 0.5$ are also implied by analysis of equilibria involving hydrous phases.

Decompression reactions are observed both in this and in earlier studies of supracrustal rocks from the Central Limpopo Belt, and a period of essentially isothermal uplift is suggested, which raised crustal blocks from depths of about 40 km to 15 km. One possible speculation is that such rapid uplift results from a phase of extensional tectonics which occurred between 2600 and 3150 Ma.

Introduction

The Limpopo Belt is an east-trending polymetamorphic terrain which separates the Rhodesian and Kaapvaal Archaean cratons. The belt has an extensive structural and metamorphic history which spans at least 1600 Ma from before 3600 Ma to 2000 Ma. It is bounded on its northern and southern margins by an orthopyroxene isograd so that metamorphism within the belt is characterized by the granulite facies. The belt is divided into three zones on the basis of fabric and lithology (Fig. 1). North and South Marginal zones consist of remnants of granite–greenstone terrains which are separated by major shear belts from the Central Zone. The Central Zone is characterized by a predominance of metasedimentary rocks which unconformably overlie a gneissic sialic basement (Mason, 1969; Barton and Key, 1981).

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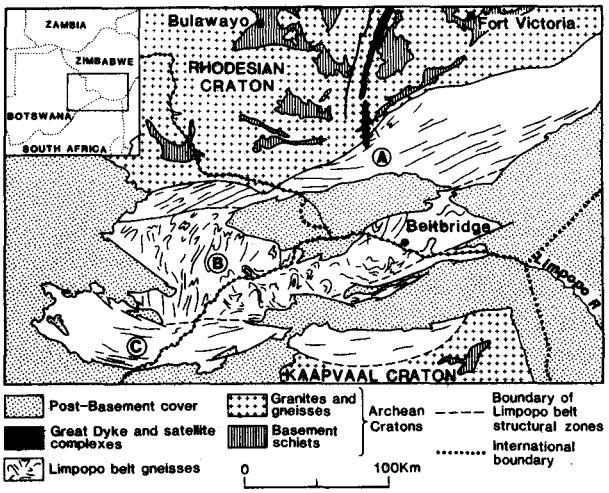

Fig. 1. Geological sketch map showing location of Beitbridge within the Limpopo Belt. A = North Marginal Zone, B = Central Limpopo Belt, C = South Marginal Zone.

Granulite-facies metamorphism in the belt has been defined as M_1, with retrogression ascribed to M_2 and subsequent shear-induced metamorphism in the marginal zones to M_3 (James, 1975). However, the age of granulite-facies metamorphism has not been uniquely defined. Within the Central Zone, gneisses and granulites from the Botswana region of the belt provide an Rb–Sr whole rock isochron age of 2600±40 Ma (Hickman and Wakefield, 1975) compared with an Rb–Sr age of 2900±130 Ma obtained from whole rock granulites from the North Marginal Zone (Hickman, 1978). Petrofabric analysis from the North Marginal Zone indicate two granulite events (Cliff, 1975): an earlier phase in which hornblende is stable and a subsequent event resulting in anhydrous two pyroxene granulites. Granulite metamorphism is probably more complex than this since Sm–Nd mineral ages from the North Marginal Zone indicate that some granulite facies assemblages equilibrated as recently as 1980 Ma (van Breeman and Hawkesworth, 1980). Because the diffusion of REE in granulites may be sufficiently rapid to prevent the preservation of crystallization ages (Humphries and Cliff, 1982) this young age may date the time of Sm–Nd closure at about 600°C. It appears that granulite-facies metamorphism within the belt was not restricted to its early history but was diachronous and possibly recurred over a period of several hundred million years.

Mineral assemblages from the Diti Formation

Assemblages from the Beitbridge region of the Central Zone are among the most complex examples of Limpopo Belt polymetamorphism. Watkeys (1979) and Broderick (1979) propose a deformation and metamorphic sequence which begins with a high-grade metamorphic event ($T >$ 860°C, $P >$ 11 kbar) and is terminated by a low-grade event ($T <$ 330°C) after a series of folding and decompressive metamorphic episodes. The lithological sequence which provides the most appropriate assemblages for petrogenetic study comprises a series of garnetiferous paragneisses (meta-arkoses and metapelites) interbanded with magnetite quartzites, calc-silicates and thin horizons of mafic granulites which has been mapped as the Diti Formation. These metamorphosed supracrustal rocks unconformably overlie basement gneisses (Macuville Group) and were deposited between 3570 and 3150 Ma (Barton and Ryan, 1977). The primary gneissic foliation of both supracrustal rocks and basement have been refolded by two periods of isoclinal folding (F_1 and F_2 of Watkeys, 1979). The granulite assemblages observed in the Diti Formation (from metamorphism M_2 of Watkeys, 1979) developed during F_2 which predates the synorogenic emplacement of the Bulai Gneiss at 2600 Ma (van Breemen and Dodson, 1972).

In the Beitbridge area, exposures of the Diti Formation provide four distinct cordierite-bearing assemblages (Table 1).

(i) Cordierite–Hypersthene–Garnet–Sillimanite–Quartz–Biotite±Perthite
(ii) Cordierite–Garnet–Sillimanite–Quartz–Biotite–Plagioclase±Perthite
(iii) Cordierite–Hypersthene–Sillimanite–Quartz–Biotite
(iv) Cordierite–Hypersthene–Sillimanite–Spinel–Plagioclase

Assemblage (i) occurs in mesocratic pelitic gneisses. Garnet is embayed and irregular in form and sometimes appears to be partially replaced by hypersthene or cordierite. Hypersthene occurs as skeletal porphyroblasts up to 1 cm across. Equant cordierite frequently shows a vermicular intergrowth with quartz. *Assemblage (ii)* includes coarse garnet porphyroblasts with abundant inclusions of sillimanite, biotite, plagioclase and quartz. The porphyroblasts are pre or syn-tectonic within a gneissic matrix of cordierite (often pinitised), sillimanite, biotite, oligoclase, microperthite and quartz. *Assemblage (iii)* is restricted to one occurrence of quartzose gneiss which contains porphyroblasts of pink hypersthene. Cordierite

Table 1. Pelitic assemblages from the Diti Formation

	Cord	Opx	Gnt	Sill	Qtz	Spinel	Biot	Plag	Kspar
BB22a*	✓	✓	✓	(✓)	✓	-	✓	-	✓
BB25c*	✓	✓	✓	(✓)	✓	-	✓	-	-
BB11d**	✓	-	✓	(✓)	✓	-	✓	✓	✓
BB13b**	✓	-	✓	✓	✓	-	✓	✓	-
BB20a*	✓	-	✓	✓	✓	-	✓	✓	✓
BB20b*	✓	-	✓	✓	✓	-	✓	✓	-
BB23b*	✓	✓	-	✓	✓	-	✓	-	-
BB17b (lens)***	✓	✓	-	✓	-	✓	✓	✓	-

* Diti Dip, 20 km north-east of Beitbridge
** North bank of Limpopo River, 1 km south of Beitbridge
*** East of Beitbridge–Port Victoria Road, 3 km north of Beitbridge
(✓) Sillimanite rare and preserved as inclusions

Table 2. Thermochemical data for MAS system

	V_{298} cm^3	V_{1000} cm^3	S_{1000} JK^{-1}	$\Delta_f H^{*****}_{1000,ox}$ KJ	Calorimetry KJ
Corundum	25.58	26.02	180.2	0	0
β-Quartz	23.76	23.70	115.6	0	0
Sillimanite	49.90	50.41	295.1*	- 3.0	- 2.4 ±0.6
Spinel	39.71	40.43	267.9**	-23.45	-22.5 ±0.8
Forsterite	43.79	44.91	277.2	-62.0	-63.0 ±1.1
Enstatite	62.58	63.81	393.1	-69.16	-69.5 ±1.4***
Mg-Tschermak	58.95	59.75	393.2	+ 2.04	+ 2.0 ±8
Pyrope	113.27	115.26	777.5	-86.5	-84.6 ±2.0
Cordierite	233.2	235.6	1131.7****	-69.67	-68.1 ±2.5

* Ordered sillimanite
** Spinel + 3.4 JK^{-1}
*** Perkins et al. (1981), Wood and Holloway (1982)
**** Cordierite + 5.1 JK^{-1}, Newton and Wood (1979)
***** Enthalpies adjusted to experiments

and sillimanite are restricted to the matrix and are not contiguous with hypersthene.

The five phases cordierite–hypersthene–garnet–sillimanite–quartz from assemblage (i) suggest the following five four-phase equilibria in the system MgO–Al_2O_3–SiO_2 (MAS), each of which provides a potential barometer.

$$\underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + 2\ \underset{\text{Sillimanite}}{Al_2SiO_5} + \underset{\text{Quartz}}{SiO_2} = \underset{\text{Cordierite}}{Mg_2Al_4Si_5O_{18}} \quad (1)$$

$$2\ \underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} + 4\ \underset{\text{Sillimanite}}{Al_2SiO_5} + 5\ \underset{\text{Quartz}}{SiO_2} = 3\ \underset{\text{Cordierite}}{Mg_2Al_4Si_5O_{18}} \quad (2)$$

$$2\ \underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} + 3\ \underset{\text{Quartz}}{SiO_2} = \underset{\text{Cordierite}}{Mg_2Al_4Si_5O_{18}} + 2\ \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} \quad (3)$$

$$3\ \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + 2\ \underset{\text{Sillimanite}}{Al_2SiO_5} = 2\ \underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} + 2\ \underset{\text{Quartz}}{SiO_2} \quad (4)$$

$$5\ \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + 6\ \underset{\text{Sillimanite}}{Al_2SiO_5} = 2\ \underset{\text{Cordierite}}{Mg_2Al_4Si_5O_{18}} + 2\ \underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} \quad (5)$$

Equilibrium relation (2) pertains to assemblage (ii) above, and equilibrium (1) pertains to assemblage (iii).

Assemblage (iv) is a silica-deficient assemblage which occurs as lenses up to 5 mm across within a biotite–quartz–andesine leucogranite gneiss. Green spinel lies towards the center of these ocellae, with the rim containing fine-grained equant crystals of cordierite, pink hypersthene and sillimanite. Also present in this gneiss are garnet porphyroblasts with abundant inclusions of sillimanite. An equilibrium condition for the spinel-bearing ocellae may be written as follows:

$$5\ \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + 10\ \underset{\text{Sillimanite}}{Al_2SiO_5} = 4\ \underset{\text{Cordierite}}{Mg_2Al_4Si_5O_{18}} + 2\ \underset{\text{Spinel}}{MgAl_2O_4} \quad (6)$$

To evaluate the conditions of equilibrium defined by relations (1) to (6) it is necessary to secure experimental or thermodynamic information for these end member reactions in the MAS system.

Thermodynamic data for part of the MAS system

Calorimetric data now exist for all phases of interest to this study (Charlu et al., 1975; Kleppa and Newton, 1975) and an attempt to calculate the *P–T* projection of all relevant equilibria could be made. However, the quoted uncertainties in enthalpy data for phases makes this unsatisfactory, and so the available experimentally determined equilibria will be used to refine, perhaps somewhat arbitrarily, the data set so that internal consistency is obtained.

Following Wood and Holloway (1982) we assume that ΔS for solid–solid reactions remains constant in the temperature range of interest and choose 1000 K as a reference temperature for tabulation of entropies, volumes and enthalpies (Table 2). We also make the approximation that ΔV and ΔH remain constant so that for equilibrium we may write:

$$\Delta\mu = O \simeq \Delta H_{1000} - T\Delta S_{1000} + P\Delta V_{1000} + RT\ln K$$

Before refining the enthalpy data, some adjustments and assumptions were made as to the entropy and volume values used at 1000 K. Entropies were all taken from Robie et al. (1978) with the exception of orthoenstatite (Krupka et al., 1979) and pyrope (Haselton and Westrum, 1980). Our own work (Holland and Carpenter, 1983 and in prep.) indicates that sillimanite is highly ordered at all reasonable temperatures, but we have added 5.1 JK^{-1} mol^{-1} to the entropy of cordierite to allow for some configurational Al-Si disorder (Newton and Wood, 1979) and 3.4 JK^{-1} mol^{-1} to spinel for Mg-Al disorder. Volumes for all phases are tabulated and refer to end member compositions, with the exception of the two orthopyroxene end-members enstatite and magnesium tschermak's pyroxene for which relevant partial molar volumes are used (data of Danckwerth and Newton, 1978). All volumes at 298 K are from Robie et al. (1978) corrected to 1000 K using expansion data from Skinner (1966), Hazen (1976) and Winter and Ghose (1979).

Enthalpies of formation from the oxides at 1000 K are listed in Table 2 and were derived by fitting to four experimentally determined equilibria given by reactions (1) and (4) in addition to reactions (7) and (8) below:

$$\underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} = \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + \underset{\text{Orthopyroxene}}{MgAl_2SiO_6} \quad (7)$$

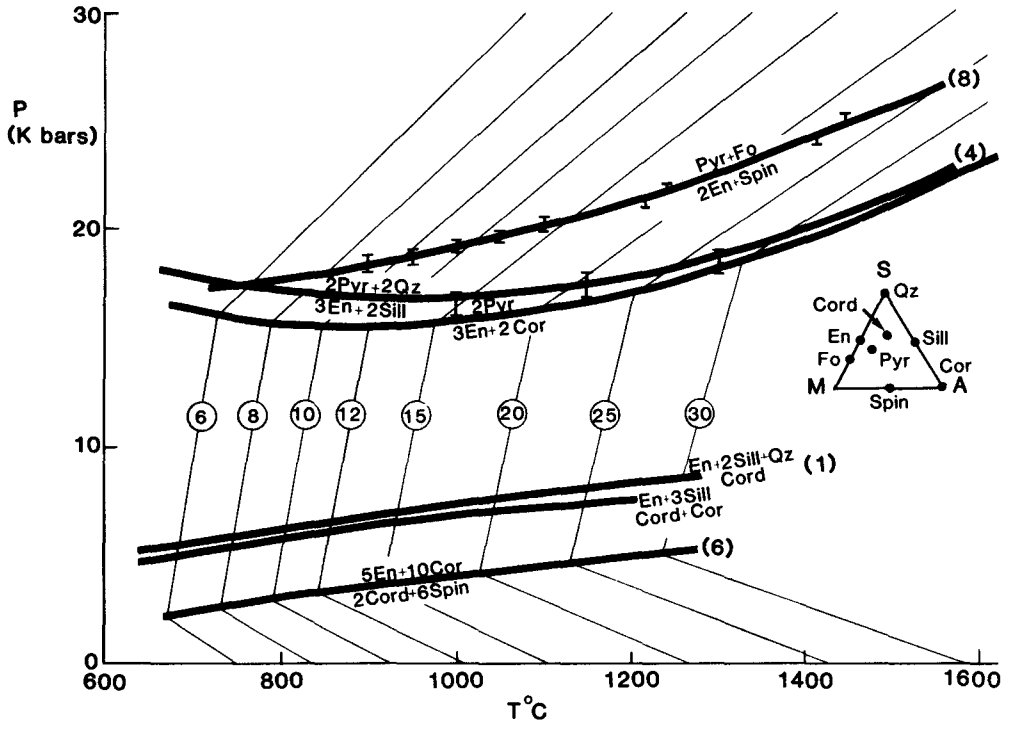

Fig. 2. Computed equilibria in the $MgO–Al_2O_3–SiO_2$ system using data of Table 2; experimental brackets are for reactions (4) and (8). Numbered lines represent saturation isopleths for X_{Al}^{m} in orthopyroxene with garnet at high pressures (through reaction 7)), with corundum at intermediate pressure (through the reaction Enstatite + 2 Corundum = 2 Mg – Tschermak), and with spinel and cordierite at low pressure (through the reaction Cordierite + 3 Spinel = 5 Mg – Tschermak).

$$\underset{\text{Garnet}}{Mg_3Al_2Si_3O_{12}} + \underset{\text{Olivine}}{Mg_2SiO_4} = 2\ \underset{\text{Orthopyroxene}}{Mg_2Si_2O_6} + \underset{\text{Spinel}}{MgAl_2O_4} \quad (8)$$

In the fitting procedure, entropies and volumes of reaction were taken from Table 2 and a value of ΔH assigned to each equilibrium such that the calculated curve passed through the pertinent experimental brackets. Sources of data are as follows: for (7), Perkins et al., (1981) from which $\Delta S_7 = 8.79$ JK^{-1} and $\Delta H_7 = 19.38$ KJ were derived; for (8), from Danckwerth and Newton, (1978) and Perkins et al. (1981) from which $\Delta H_8 = -13.27$ KJ was derived; for (4), from Perkins, (1983) from which $\Delta H_4 = -20.24$ KJ was derived (using higher temperature brackets only); and for (1), from the experimental data of Newton (1972) with additional experimental data reported in Perkins et al. (1981, p. 101) pertaining to two reactions involving cordierite and sapphirine which intersect at an invariant point through which (1) must pass, from which $\Delta H_1 = 5.49$ KJ was derived. In all cases variable Al_2O_3 content in orthopyroxene was taken into account by assuming ideal mixing of the $MgAl_2SiO_6$ component, following Wood and Banno (1973) and Wood and Holloway (1982).

We stress that our data set (Table 2) is *not* intended to be a table of optimal quality information on enthalpies and entropies, but is designed to generate P–T equilibria consistent with experimental work. In defense of the approach taken, we point out that our data on the reaction

2 Pyrope = 3 Enstatite + 2 Corundum (Fig. 2)

are in very good agreement with a recent experimental determination by R. C. Newton (pers. comm.).

The data derived and tabulated in Table 2 were used to compute a P–T diagram for stable univariant equilibria involving quartz, cordierite, enstatite, sillimanite, pyrope, forsterite, spinel and corundum in the system $MgO–Al_2O_3–SiO_2$ in Figure 2. The Al_2O_3 isopleths for orthopyroxene are for co-existing garnet + orthopyroxene (high pressures), for coexisting corundum + orthopyroxene (intermediate pressures), and for coexisting spinel + cordierite + orthopyroxene (low pressures). Note that we have chosen to ignore the additional complexities involving sapphirine equilibria.

Additionally, we comment on the much debated position of reaction (2). This reaction can be written as a linear combination of reactions (1) and (4) both of which have an experimental basis and therefore we can derive $\Delta H_2 = -24.0 \pm 3.0$ KJ, $\Delta S_2 = 81.70$ JK^{-1}, and $\Delta V_2 = 15.614$ J bar^{-1} which lead to a positive dP/dT slope of 5.2

Table 3. Equilibrium equations for four phase assemblages from Diti Formation

(1) $P\ (\text{bars}) = \dfrac{(3.95 - \ln K_1)\ T\ (^{\circ}\text{K}) - 660}{0.569}$ where $K_1 = \dfrac{(a^{\text{cordierite}}_{Mg_2Al_4Si_5O_{18}})}{(a^{\text{hypersthene}}_{Mg_2Si_2O_6})}$

(2) $P\ (\text{bars}) = \dfrac{(9.83 - \ln K_2)\ T\ (^{\circ}\text{K}) + 2888}{1.878}$ where $K_2 = \dfrac{(a^{\text{cordierite}}_{Mg_2Al_4Si_3O_{18}})^3}{(a^{\text{garnet}}_{Mg_3Al_2Si_3O_{12}})^2}$

(3) $P\ (\text{bars}) = \dfrac{(1.94 - \ln K_5)\ T\ (^{\circ}\text{K}) + 4209}{0.741}$ where $K_3 = \dfrac{(a^{\text{hypersthene}}_{Mg_2Si_2O_6})^2\ (a^{\text{cordierite}}_{Mg_2Al_4Si_5O_{18}})}{(a^{\text{garnet}}_{Mg_3Al_2Si_3O_{12}})^2}$

(6) $P\ (\text{bars}) = \dfrac{(17.57 - \ln K_6)\ T\ (^{\circ}\text{K}) - 6040}{2.407}$ where $K_6 = \dfrac{(a^{\text{cordierite}}_{Mg_2Al_4Si_5O_{18}})^4 \times (a^{\text{spinel}}_{MgAl_2O_4})^2}{(a^{\text{hypersthene}}_{Mg_2Si_2O_6})^5}$

Activities of enstatite in orthopyroxene from Wood and Banno (1973), of the Mg-end member in spinel from Harris (1981) and of pyrope in garnet from Perkins and Newton (1981) and the Mg-end member in cordierite assuming ideal ionic solid solution.

bar K^{-1} passing through 800°C at 7.2±0.3 kbar. We place this metastable univariant curve at marginally higher pressures than Newton and Wood (1979), and in substantial agreement with the calculated curve of Martignole and Sisi (1981).

We feel that, in spite of somewhat arbitrary adjustments to individual enthalpies in Table 2 (pyrope somewhat more negative and forsterite somewhat more positive to gain reasonable values for enstatite, spinel and cordierite), the data set is in fair agreement with *both* the calorimetry *and* the experimental equilibria; we therefore feel confident in its use as a basis for calculating equilibria pertinent to granulite-facies rocks.

Cordierite–sillimanite–quartz assemblages from the Limpopo

Natural assemblages differ in two important respects from the MAS system considered so far. First, the introduction of Fe to the system will reduce the pressures at which garnet-bearing assemblages equilibrate due to the strong partitioning of Fe into garnet. Second, the presence of H_2O in the volatile phase will increase the equilibrium pressure of all reactions involving cordierite. Pressures are computed from the assemblages assuming $a_{H_2O} = 0$, and the effect of finite but low water activity is considered later. Equations defining the *P–T* relationships for four assemblages from the Diti Formation have been derived from the data in Table 2, and are given in Table 3.

Microprobe analysis of phases from the Diti Formation metapelites are given in Tables 4–9. Analytical conditions are described by Harris (1981). Cordierite and orthopyroxene are found to occur as unzoned crystals with little variation within a given section. Garnet is often slightly zoned (pyrope-rich core and almandine-rich rims) and in these cases, representative analyses of both core and rim

Table 4. Cordierite analyses

	+ Gnt, + Opx		+ Gnt				+ Opx	+ Spinel
	BB22a	BB25c	BB11d	BB13b	BB20a	BB20b	BB23b	BB17b
SiO_2	50.61	50.29	49.52	48.99	49.46	48.83	51.39	49.58
Al_2O_3	33.37	33.98	33.24	32.91	32.79	33.30	33.65	33.76
FeO*	3.22	3.42	4.59	4.30	3.98	4.67	3.68	4.81
MgO	11.30	11.42	10.39	10.34	10.94	10.50	11.08	10.06
Total	98.51	99.11	97.74	96.54	97.16	97.29	99.80	98.21
Atoms to 18(O)								
Si	5.07	5.01	5.03	5.03	5.04	4.99	5.09	5.02
Al	3.94	3.99	3.98	3.99	3.94	4.01	3.93	4.03
Fe	0.27	0.29	0.39	0.37	0.34	0.40	0.30	0.41
Mg	1.69	1.70	1.57	1.58	1.66	1.60	1.63	1.52

*All iron calculated as FeO

Table 5. Orthopyroxene analyses

	+ Gnt, + Cord BB22a	+ Cord BB25c	+ Cord BB23b	+ Spinel BB17b
SiO_2	50.80	52.32	51.85	48.21
TiO_2	-	-	-	0.17
Al_2O_3	6.57	5.24	6.65	8.26
Cr_2O_3	-	-	0.19	-
FeO*	19.84	19.89	16.69	26.25
MnO	-	-	0.17	0.16
MgO	22.73	23.47	25.09	17.45
CaO	-	-	-	0.09
ZnO	-	-	-	0.19
Total	99.94	100.91	100.64	100.78
Atoms to 6(O)				
Si	1.86	1.90	1.86	1.81
Al	0.28	0.22	0.28	0.37
Ti	-	-	-	0.01
Cr	-	-	0.01	-
Fe	0.61	0.60	0.50	0.83
Mg	1.24	1.27	1.34	0.98
Mn	-	-	0.01	0.01
Zn	-	-	-	0.01

* All iron calculated as FeO

are given (Table 7). For barometric purposes core analyses are used since these give higher pressures and therefore are a closer approximation to peak metamorphic conditions; maximum zoning observed in garnets from the Diti Formation pelites is $\Delta Mg/(Mg+Fe) < 5\%$ which only decreases computed pressures by about 600 bars if rim analyses are used.

Two samples from the Diti Formation contain the five-phase assemblage (in MAS) cordierite, orthopyroxene, garnet, sillimanite and quartz (Table 1). However, textural studies of these gneisses indicate that garnet is partially replaced by cordierite and orthopyroxene. Moreover, sillimanite is not found to be in contact with garnet. The textures are consistent with a decompression event in which garnet, sillimanite and quartz had initially formed cordierite by reaction (2), followed by garnet and quartz reacting to form cordierite and orthopyroxene by reaction (3). Similar decompression reactions have been described from garnet inclusions of Pyrenean gneisses (Vielzeuf, 1980). The positions of equilibria (1), (2) and (3) are plotted in Figure 3 for samples BB22a and BB25c using the equations formulated in Table 3. The data constrain the reactions to pass through a point in *P–T* space at which all five phases, of the observed compositions, are in equilibrium. This is an invariant assemblage in the model system only. The position of intersection MI lies between 4.1–4.5 kbar at T = 620–670°C. Since the five phases are *not* in equilibrium, but result from decompression reactions such as (2) and (3), the temperature of MI is a minimum estimate of equilibration temperatures. If the sillimanite-free four-phase assemblage represented by equation (3) is an equilibrium assemblage it places a maximum constraint of 4.5 kbar on the pressure at which these phases equilibrated.

Table 6. Spinel analyses

	BB17b
SiO_2	0.58
Al_2O_3	60.33
FeO*	31.26
MgO	6.51
ZnO	2.51
Total	101.19
Atoms to 4(O)	
Si	0.02
Al	1.96
Fe	0.72
Mg	0.27
Zn	0.05

* All iron calculated as FeO

Cordierite–garnet–sillimanite–quartz (assemblage (iii)) represents the most common cordierite-bearing assemblage from the Limpopo metapelites. Calculation of the equilibria represented by equation (2) (Table 3) for four such assemblages from the Diti Formation provide pressures of 3.75–4.75 kbar in the temperature range 700–800°C (Fig. 3).

One sample containing cordierite, hypersthene, sillimanite and quartz (with biotite), but lacking garnet, has been found from Diti Dip (BB23b). Equilibrium pressures for this assemblage derived from equation (1), Table 3, are similar to those derived from the five-phage assemblages plotted in Figure 3. The barometric equations therefore indicate no significant difference in equilibrium pressures between the garnet and orthopyroxene-bearing assemblages. An A–F–M plot of bulk analyses (analysed by X-ray fluorescence) of cordierite-bearing gneisses from the Limpopo (Fig. 4) demonstrates that for Fe-rich bulk compositions ($Fe/Fe+Mg > 0.6$) cordierite and garnet coexist in the absence of orthopyroxene. For Mg-rich bulk compositions ($Mg/(Fe+Mg) > 0.75$) cordierite and orthopyroxene coexist in the absence of garnet. Only in intermediate compositions are all three femic phases (cordierite, garnet and orthopyroxene) present. A similar control has been noted in cordierite–orthopyroxene assemblages from the Indian Archaean (Harris and Jayaram, 1982). However, Figure 4 also indicates that although garnet–cordierite–hypersthene and garnet–cordierite–sillimanite assemblages are compatible, hypersthene–cordierite–sillimanite are incompatible as indicated by the crossed tie lines garnet–cordierite and hypersthene–sillimanite. This relationship is also expressed by reaction (5) where hypersthene and sillimanite represent the high pressure assemblage. This suggests

Table 7. Garnet analyses

	+ Cord, + Opx			+ Cord							+ Spinel
	BB22a	BB25c		BB11d		BB13b	BB20a		BB20b		BB17b
		core	rim	core	rim		core	rim	core	rim	
SiO_2	40.56	40.88	40.46	39.99	40.01	39.65	40.29	39.81	39.48	39.78	40.13
Al_2O_3	22.48	22.03	22.13	22.73	22.30	22.18	22.50	22.46	22.77	22.44	22.46
Cr_2O_3	-	-	-	-	0.11	0.11	-	-	-	0.13	-
FeO*	26.28	25.94	26.84	27.47	30.01	28.77	26.33	27.96	28.00	28.35	27.66
MnO	0.51	0.16	0.18	0.54	0.86	0.61	0.41	0.51	0.37	0.35	0.44
MgO	11.00	11.83	11.04	9.33	7.25	8.44	10.62	9.12	9.74	9.26	9.73
CaO	0.38	0.13	0.67	0.61	0.61	0.99	1.15	1.14	1.10	1.04	0.88
Total	101.21	100.97	101.31	100.67	101.2	100.75	101.29	101.00	101.46	101.35	101.30
Atom to 12(O)											
Si	3.04	3.06	3.04	3.03	3.06	3.03	3.03	3.02	2.99	3.02	3.03
Al	1.99	1.94	1.96	2.03	2.01	2.00	1.99	2.01	2.03	2.00	2.00
Cr	-	-	-	-	0.01	0.01	-	-	-	0.01	-
Fe	1.65	1.62	1.69	1.74	1.92	1.84	1.65	1.78	1.77	1.80	1.75
Mg	1.23	1.32	1.24	1.06	0.83	0.96	1.19	1.03	1.10	1.05	1.10
Mn	0.03	0.01	0.01	0.04	0.06	0.04	0.03	0.03	0.02	0.02	0.03
Ca	0.03	0.01	0.05	0.05	0.05	0.08	0.09	0.09	0.09	0.08	0.07

Core and rim analyses given where zoning exceeds 1%
* All iron calculated as FeO

Table 8. Biotite analyses

	+ Gnt, + Cord, + Opx		+ Gnt, + Cord				+ Cord, + Opx	+ Spinel
	BB22a	BB25c	BB11d	BB13b	BB20a	BB20b	BB23b	BB17b
SiO_2	38.15	38.49	37.36	36.72	37.25	36.84	38.65	36.51
TiO_2	4.97	5.39	5.14	5.76	4.94	6.04	4.88	5.28
Al_2O_3	15.87	16.12	16.10	15.47	16.08	15.93	16.28	15.39
Cr_2O_3	0.19	-	0.29	0.20	0.21	0.17	0.19	0.38
FeO*	11.99	11.47	15.73	14.04	12.73	14.42	13.30	15.37
MgO	14.69	15.87	11.74	13.30	14.79	11.68	14.19	11.90
CaO	-	0.09	-	-	-	-	-	0.11
Na_2O	-	0.44	-	-	-	-	0.40	-
K_2O	10.05	9.20	9.53	10.11	9.82	9.28	9.01	9.69
Total	95.90	97.09	95.89	95.60	95.81	94.36	96.90	94.63
Atoms to 22(O)								
Si	5.58	5.53	5.55	5.47	5.48	5.53	5.59	5.52
Al	2.74	2.73	2.82	2.72	2.79	2.82	2.78	2.74
Ti	0.55	0.58	0.58	0.65	0.55	0.68	0.53	0.60
Cr	0.02	-	0.03	0.02	0.02	0.02	0.02	0.05
Fe	1.47	1.38	1.96	1.75	1.57	1.81	1.61	1.94
Mg	3.20	3.40	2.60	2.95	3.24	2.61	3.06	2.68
Ca	-	0.01	-	-	-	-	-	0.02
Na	-	0.12	-	-	-	-	0.11	-
K	1.88	1.69	1.81	1.92	1.84	1.78	1.66	1.87

* All iron calculated as FeO

Table 9. Plagioclase analyses

	+ Gnt, + Cord				+ Spinel
	BB11d	BB13b	BB20a	BB20b	BB17b
SiO_2	65.30	60.69	60.21	60.96	61.95
TiO_2	-	-	-	-	-
Al_2O_3	22.19	23.79	24.28	25.06	24.31
FeO*	-	0.15	0.29	0.12	0.27
CaO	2.90	6.22	6.42	6.26	5.58
Na_2O	10.05	7.65	8.04	7.85	7.97
K_2O	0.19	0.43	0.08	0.23	0.32
Total	100.64	98.93	99.32	100.48	100.39
Atoms to 8(O)					
Si	2.86	2.73	2.70	2.70	2.74
Al	1.15	1.26	1.28	1.31	1.27
Ti	-	-	-	-	-
Fe	-	0.01	0.01	-	0.01
Ca	0.14	0.30	0.31	0.30	0.26
Na	0.83	0.67	0.70	0.67	0.68
K	0.01	0.03	-	0.01	0.02

* All iron calculated as FeO

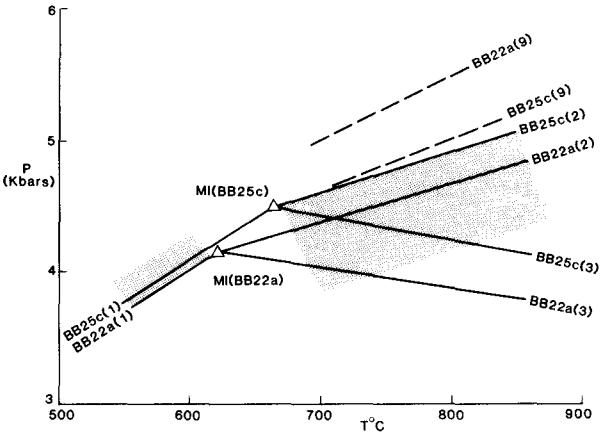

Fig. 3. Cordierite-bearing reactions for samples BB22a and BB25c. △ = model invariant point (MI) for five phase assemblages. Dashed lines represent equilibria derived from reaction (9) for these samples. Also shown are *P/T* bands for reactions (1) and (2) (shaded regions) from assemblages BB23b, BB11d, BB13b, BB20a, BB20b. Equilibria are identified by sample number, with reaction number given in parentheses.

that garnet–cordierite assemblages have equilibrated at lower pressures than sample BB23b in which hypersthene–sillimanite pairs are preserved. Since hypersthene is not seen to be in contact with either sillimanite or cordierite in this sample, reaction (5) has probably been frozen at slightly higher pressures by sluggish reaction rates.

An analog of equation (3), but in the FeO–MgO–Al_2O_3–SiO_2 system, provides an independent check on the barometric reliability of equation (3).

$$\underset{\text{Garnet}}{4\ Fe_3Al_2Si_3O_{12}} + \underset{\text{Garnet}}{2\ Mg_3Al_2Si_3O_{12}} + \underset{\text{Quartz}}{9\ SiO_2} = \underset{\text{Orthopyroxene}}{6\ Fe_2Si_2O_6} + \underset{\text{Cordierite}}{3\ Mg_2Al_4Si_5O_{18}} \quad (9)$$

The large volume change of this reaction makes it particularly appropriate for barometry. Combining the standard state data of Table 2 with the experimental results on the reactions

$$\underset{\text{Almandine}}{2\ Fe_3Al_2Si_3O_{12}} + \underset{\text{Forsterite}}{3\ Mg_2SiO_4} = \underset{\text{Pyrope}}{2\ Mg_3Al_2Si_3O_{12}} + \underset{\text{Fayalite}}{3\ Fe_2SiO_4} \quad (10)$$

$$\underset{\text{Fayalite}}{Fe_2SiO_4} + \underset{\text{Quartz}}{SiO_2} = \underset{\text{Ferrosilite}}{Fe_2Si_2O_6} \quad (11)$$

using data given by Wood and Holloway (1982) provides the following equation for the assemblage cordierite–garnet–orthopyroxene–quartz:

$$P\ (\text{bars}) = \frac{(7.28 - \ln K_9)\ T\ (°\text{K}) + 535}{2.33}$$

$$\text{where } K_9 = \frac{(a^{\text{cordierite}}_{Mg_2Al_4Si_5O_{18}})^3\ (a^{\text{hypersthene}}_{Fe_2Si_2O_6})^6}{(a^{\text{garnet}}_{Mg_3Al_2Si_3O_{12}})^2\ (a^{\text{garnet}}_{Fe_3Al_2Si_3O_{12}})^4}$$

The positions of this equilibrium for assemblages from BB22a and BB25c are plotted in Figure 3 and indicate slightly higher pressures than those derived from the MAS system. Assuming $T < 800°C$, pressures are consistent to within ±0.8 kbars.

We can therefore conclude from this study of the cordierite–sillimanite–quartz assemblages from the Central Limpopo that assemblages record pressures of 4.5±0.8 kbar assuming anhydrous conditions, with $T > 670°C$. Lack of zoning within the garnets is to be expected at these high temperatures of equilibration. The assemblage cordierite–garnet–orthopyroxene–quartz (reaction (3)) has also been reported from pelites of the northern Transvaal region of the South Marginal Zone (Van Reenan and du Toit, 1978). Sillimanite is either absent or present as fibrolite inclusions in biotite. Application of equation (3) to published analyses of the femic phases cordierite, hypersthene and garnet implies pressures of 4.0±0.8 kbar similar to conditions computed from the Beitbridge samples of the Central Limpopo Belt.

Independent *P–T* determinations from the Diti Formation

Fe–Mg exchange reactions are commonly invoked to constrain metamorphic temperatures of pelitic gneisses. In the Limpopo samples, biotite–garnet pairs provide very scattered temperatures since the composition of biotite is highly variable within a given section (see Ghent et al., 1982 for discussion of biotite–garnet thermometry in high grade rocks). Biotite inclusions in garnet are Mg-rich and provide temperatures of 600–650°C based on the experimental calibration of Ferry and Spear (1978). Contiguous garnets and biotites provide temperatures of 650–

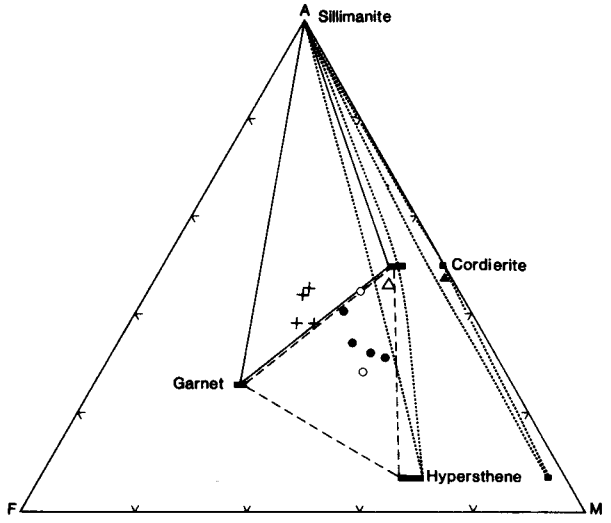

Fig. 4. Phase relations and bulk analyses of pelitic assemblages from the Limpopo plotted on a simplified AFM ternary diagram. A = $Al_2O_3/(Al_2O_3 + MgO + FeO)$. M = MgO/(FeO + MgO). All Fe calculated as FeO. Solid line indicates cordierite–garnet–sillimanite (+ = Beitbridge samples). Dashed line indicates cordierite–hypersthene–garnet (○ = Beitbridge samples, ● = Transvaal samples after Van Reenan and Du Toit, 1978). Dotted line indicates cordierite–hypersthene–sillimanite (△ = Beitbridge sample, this study, ▲ = Beitbridge sample, from Chinner and Sweatman, 1968). An AFM projection from potassium feldspar was not applicable since perthite is only present in three samples.

750°C and for non-contiguous grains, temperatures of up to 950°C (outside the calibration range of the thermometer) are indicated. These temperatures may well all be overestimated because all Fe in biotite is treated at Fe^{2+}. If as much as 15% of Fe in biotite from high grade gneisses is actually Fe^{3+} (Grew, 1981), temperatures would be reduced by about 100°C. It is clear that the mineral pairs have re-equilibrated during cooling, the rate of equilibration being controlled by, among other things, the shared surface area. Similar re-equilibration has been inferred by Ghent et al. (1982) in granulite-facies metamorphism. It could be argued that garnet cores and non-contiguous biotites should be selected, but this procedure may provide an *over*-estimate of temperature unless garnets are small and their retrogressed rims are narrow (Tracy et al, 1976). For cordierite–garnet pairs, re-equilibration appears even more complete as temperatures above 650°C are not recorded regardless of the calibration used (Wells, 1979; Martignole and Sisi, 1981; Lonker, 1981). These observations imply Fe–Mg exchange thermometers involving biotite, garnet and cordierite do not realistically constrain peak metamorphic conditions in high-grade pelitic gneisses.

The aluminum distribution between garnet and orthopyroxene has recently been calibrated as a barometer (Harley and Green, 1982). However, the steep slope of this reaction with the temperature axis makes it more suitable as a thermometer provided the approximate pressure is known. Mineral pairs from samples BB22a and BB25c provide temperatures in the range 740–820°C, assuming P = 4.0–5.0 kbar (Fig. 5).

A barometer is provided by the assemblage garnet–plagioclase–sillimanite–quartz which has been calibrated by Newton and Haselton (1981). A large uncertainty stems from the activity of grossular in garnet of very low CaO contents. Another uncertainty in plagioclase-bearing reactions comes from the activity coefficient of anorthite in plagioclase. Recent experimental studies on ordering and mixing in plagioclase (Carpenter, pers. comm.) suggests that the aluminum-avoidance model applied by Newton and Haselton may underestimate the magnitude of γ_{An}. For the present we take a nominal value for γ_{An} of 2 ± 1 for temperatures in the range 700–800°C. (The Al-avoidance model leads to values <1.3). This uncertainty propagates an error of ±2 kbar in the barometer. Pressures derived from samples BB13b, BB20a and BB20b lie in the range 3.5–5.0 kbar for temperatures of 780±40°C (Fig. 5). Sample BB11d indicates somewhat higher pressures, but is excluded since sillimanite is only present as inclusions in garnet, and therefore was not in equilibrium with other phases.

Interlayered with pelitic gneisses in the Diti Formation, horizons of mafic granulites contain the assemblage gar-

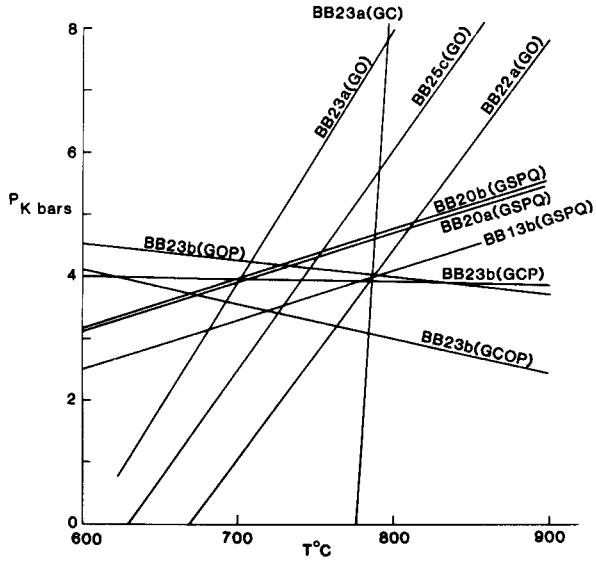

Fig. 5. Cordierite-absent reactions from the Diti Formation. Mineral phases given in parentheses. GO = garnet–orthopyroxene (Harley and Green, 1982); GSPQ = garnet–sillimanite–plagioclase–quartz (Newton and Haselton, 1981); GCOP = garnet–clinopyroxene–orthopyroxene–plagioclase (Hansen, 1981); GCP = garnet–clinopyroxene–plagioclase, and COP = garnet–orthopyroxene–plagioclase (Perkins and Newton, 1981); GC = garnet–clinopyroxene (Ellis and Green, 1979).

net–hornblende–clinopyroxene–orthopyroxene–plagioclase (BB23a). Such assemblages are useful because they may constrain both temperatures and pressures of equilibration. Garnet–two pyroxenes–plagioclase–quartz (Hansen, 1981), garnet–orthopyroxene–plagioclase–quartz and garnet–clinopyroxene–plagioclase–quartz (Perkins and Newton, 1981) barometers are all sensitive to the activity coefficient of anorthite in plagioclase. If a value of 2 ± 1 is chosen, this generates an uncertainty of ±2 kbar which, if combined with the quoted error of ±1.5 kbar, results in an overall uncertainty of ±2.5 kbar. The three barometers, when aplied to sample BB23a, indicate maximum pressures of 3.5–4.0 kbar, (the maximum constraint results from the absence of quartz). Temperatures derived from experimentally based calibrations of Fe–Mg distribution between garnet and clinopyroxene (Ellis and Green, 1979) indicate conditions of 790 ± 50°C. The aluminum distribution between garnet and orthopyroxene (Harley and Green, 1982) indicates temperatures of about 700°C. Two pyroxene thermometry is erratic, providing temperatures between 800°C and 975°C using the calibration of Wood and Banno (1973), Wells (1977) and Powell (1978). A recent study of two pyroxene thermometry (Fornarev and Graphchikov, 1982) concludes that published calibrations overestimate temperatures by 50 to 150°C. Lindsley (1983) and Bohlen and Essene (1979) discuss the problems of two pyroxene thermometry and conclude that present calibrations are inappropriate for granulites which have equilibrated at $T < 900$°C. The mafic assemblage of sample BB23a is therefore believed to have equilibrated at pressures below about 5 kbar and at temperatures of 750 ± 50°C. Such conditions are consistent with those determined from the cordierite-bearing reactions derived in this paper.

The presence of water during metamorphism

The presence of H_2O in the volatile phase will increase the equilibrium pressure of all reactions involving cordierite since water may enter the cordierite structure and stabilise it to higher pressures than under anhydrous conditions. Three dehydration reactions are observed from the Diti formation assemblages which will allow some estimate of water activity to be made.

$$\underset{\text{Biotite}}{2\,KMg_3AlSi_3O_{10}(OH)_2} + \underset{\text{Quartz}}{6\,SiO_2} = \underset{\text{Orthopyroxene}}{3\,Mg_2Si_2O_6} + \underset{\text{Potassium Feldspar}}{2\,KAlSi_3O_8} + 2\,H_2O \quad (12)$$

$$\underset{\text{Biotite}}{2\,KMg_3AlSi_3O_{10}(OH)_2} + \underset{\text{Quartz}}{9\,SiO_2} + \underset{\text{Sillimanite}}{6\,Al_2SiO_5} = \underset{\text{Cordierite}}{3\,Mg_2Al_4Si_5O_{18}} + \underset{\text{Potassium Feldspar}}{2\,KAlSi_3O_8} + 2\,H_2O \quad (13)$$

$$\underset{\text{Amphibole}}{2\,Ca_2Mg_5Si_8O_{22}(OH_2)} + \underset{\text{Garnet}}{2\,Mg_3Al_2Si_3O_{12}} = \underset{\text{Orthopyroxene}}{7\,Mg_2Si_2O_6} + \underset{\text{Clinopyroxene}}{2\,CaMgSi_2O_6} + \underset{\text{Plagioclase}}{2\,CaAl_2Si_2O_8} + 2\,H_2O \quad (14)$$

Estimates of a_{H_2O} in sample BB22a from reaction (12) indicate values of 0.5 if calculations are based on the experimental studies of Hoffer and Grant (1980), who used femic phases of intermediate composition, or much lower values of <0.1 if based on Wones and Dodge (1977) who used Mg-end member phases. Reaction (13), observed in sample BB20a, is not strongly sensitive to changes in a_{H_2O} since cordierite is one of the products and is itself a hydrate. Reaction (14), sample BB23a, may be computed from the experimental work on tremolite dehydration by Boyd (1959) with the data of Perkins and Newton (1981) and implies $a_{H_2O} = 0.2$ for BB23a, although this is somewhat unsatisfactory since the observed hornblende has a very low tremolite component. We conclude that $a_{H_2O} < 0.5$, but that more precise estimates are as yet not possible. Studies of natural assemblages in granulite terrains almost invariably place $a_{H_2O} < 0.3$ (see Touret, 1974, Martignole and Nantel, 1982 for discussion of a_{H_2O} during granulite metamorphism), and low a_{H_2O} is implied by the apparent absence of melting in these Limpopo samples assuming that metamorphic conditions reached 800°C. The assemblage alkali feldspar and quartz, for example, will be constrained from melting at these temperatures only if $a_{H_2O} < 0.5$ (Bohlen et al., 1983).

An alternative semi-quantitative estimate of a_{H_2O} can be derived from the oxide totals of cordierite analyses. These lie between 0.8–2.6% below 100% (averaging grains from each section) which, if it is assumed to be due entirely to the presence of water (and ignoring the imprecision of the analyses), suggests $a_{H_2O} = 0.1$–0.6 obtained from the hydration isotherms of Martignole and Sisi (1981). In fact the optically positive nature of cordierite indicates that some, if not most, of these channel volatiles will be CO_2 (Armbruster and Bloss, 1982) so that this semiquantitative estimate of a_{H_2O} must be an over-estimate.

The effect of $a_{H_2O} = 0.5$ on reaction (2) can also be estimated. Pressures are increased by about 700 bars relative to the anhydrous reaction if the Newton and Wood (1979) hydration theory is used. Alternatively, treating cordierite hydration as in Martignole and Sisi (1981) raises the pressure by about 1200 bars. However, the pressures computed from cordierite-bearing reactions (Fig. 3) assume that $a_{H_2O} = 0$, and these lie in the same field as those computed from equilibria which do not involve cordierite (Fig. 5). It can be concluded that the effect of water during metamorphism of these samples

was negligible in comparison with other uncertainties in the geobarometry.

Cordierite–sillimanite–spinel assemblages from the Limpopo

The assemblage cordierite–hypersthene–sillimanite–spinel (reaction (6)) occurs as lenses within leucogneiss from the Limpopo Belt (BB17b, Table 1). This assemblage is mantled by plagioclase and biotite but is not in contact with quartz which is present in the matrix of the rock. Also present are porphyroblasts of garnet with sillimanite inclusions, but these are not found in contact with the spinel-bearing ocellae. The significance of these assemblages is important since they occur in the same locality from which sillimanite–orthopyroxene assemblages have been described from an unusually Mg-rich sample (Chinner and Sweatman, 1968) and within which sillimanite has apparently pseudomorphed kyanite. If this is the case, then the textures represent significant decompression from pressures in excess of 10 kbar.

The phases cordierite–hypersthene–sillimanite–spinel may have formed an equilibrium assemblage represented by equation (6) (Table 3). This yields conditions of P = 3.75 – 4.5 kbar, for T = 750±50°C (Fig. 6). The garnet porphyroblasts provide pressures in the range 3.5–4.25 kbar derived from Newton and Haselton's garnet–sillimanite–plagioclase–quartz barometer (assuming a value of two for the activity coefficient of anorthite). The two assemblages therefore imply similar pressures well within error of the two barometers. The garnet porphyroblasts have a more ferruginous composition than the cordierite-bearing lenses which might represent decompression pseudomorphs of an Mg-rich aluminous phase such as sapphirine.

Suitable mineral thermometers do not occur in this gneiss. Garnet–biotite pairs give erratic results for reasons already discussed. Fe-Mg distribution between cordierite and spinel has been calibrated as a thermometer (Vielzeuf, 1983) but application to the Limpopo lenses indicates a temperature of ~650°C which lies below the range for which Vielzeuf's K_D vs. T plots provide a reasonable correlation. However, garnet–pyroxene pairs are present in granulites from the same locality which contains the assemblage garnet–orthopyroxene–biotite–plagioclase–quartz (BB16d). These indicate temperatures of 700–740°C after Harley and Green (1982). This granulite also provides the assemblage garnet–orthopyroxene–plagioclase–quartz which indicates pressures of about 4 kbar at temperatures of about 720°C. It can be seen therefore that both pelitic and semi-pelitic granulites from this locality indicate pressures of about 4 kbar, and temperatures of about 720°C. No indication has been found of the much higher pressures reported by Chinner and Sweatman (1968). However, if the equation (1) (Table 3) is applied to the phases analyzed in sample 93543 (Chinner and Sweatman, 1968), pressures are obtained of

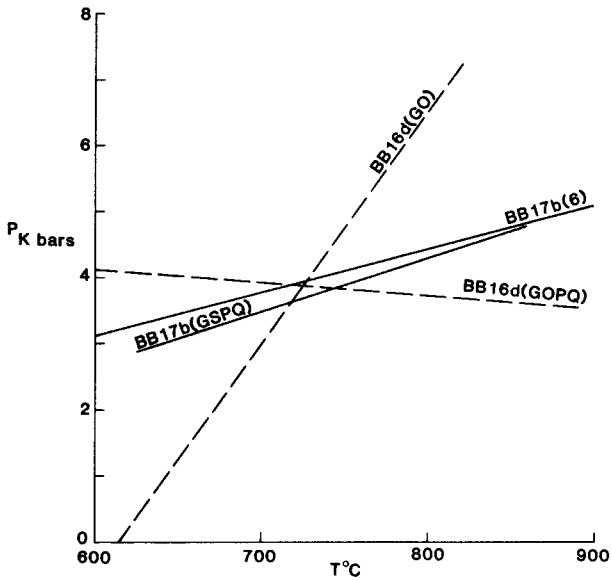

Fig. 6. P–T field of equilibration for lenses from sample BB17b (solid lines). Reaction (6) computed from data in Table 3. GSPQ = garnet–sillimanite–plagioclase–quartz (Newton and Haselton, 1981): reactions from sample BB16d are GO = garnet–orthopyroxene (Harley and Green, 1982); GOPQ = garnet–orthopyroxene–plagioclase–quartz (Perkins and Newton, 1981).

~6 kbar at 750°C, 1–2 kbar higher than for samples discussed in this paper. Chinner and Sweatman report that the distribution of quartz is very irregular and we suggest that progression of the continuous reaction (1) has been frozen by lack of quartz.

Implications of granulite metamorphism

The assemblages observed from the Diti Formation indicate metamorphism under conditions of P = 4.0±0.8 kbar, T = 750±50°C and low a_{H_2O}. Textures observed in both this study and previous work (Robertson, 1977; Horrocks, 1980, 1983) suggest that the assemblages result from decompression reactions, and one study (Chinner and Sweatman, 1968) suggests that an early assemblage was formed at pressures of 10–12 kbar. This implies that rapid and virtually isothermal crustal uplift has occurred, bringing up a crustal block from a depth of about 40 km to 15 km. Rapid uplift would also account for the extremely high geothermal gradient implied by the cordierite–orthopyroxene gneisses which is in excess of any steady state geotherm, but could be derived as a transient geotherm by rapid decompression without loss of temperature.

Tectonic studies of the Limpopo Belt generally concur that the observed structures result from continental collision between the Kaapvaal and Rhodesian cratons (Coward and Fairhead, 1980; Barton and Key, 1981; Light, 1982). The Diti Formation is ascribed to the lower crust within the overthrust block of the Kaapvaal plate (Light, 1982). Thermal modelling of overthrust orogenic zones

(England and Richardson, 1977), does suggest that a period of essentially isothermal uplift follows as a consequence of erosion of the tectonically thickened crust regardless of whether the rocks in question come from the upper or lower thrust slice. However, another possible explanation which merits discussion is that the crust was subject to extension and rapid crustal thinning during a period which post-dates the formation of the high pressure assemblages. A phase of extensional tectonics would allow regions of the lower crust between the cratons to rise rapidly without perceptible cooling.

The time of the cordierite–orthopyroxene granulite metamorphism in the Beitbridge region has been placed between 2600 and 3050 M (Watkeys, 1979). The timing of the high pressure kyanite–orthopyroxene assemblages is not known since the assemblage has not been observed but is inferred from pseudomorphs. It must however postdate the deposition of the Beitbridge Group between 3570 and 3150 Ma and subsequent tectonic overthrusting which resulted in metamorphism at depths of about 40 km. At about 3150 Ma the Central Limpopo Belt was intruded by ultramafic complexes, anorthosites and mafic dikes. These intrusives have been interpreted as indicators of a suture zone within a collision environment (Light, 1982) but could equally result from a period of mantle-derived magmatism during extensional tectonics (Barton and Key, 1981). The association of the anorthosites with riebeckite gneisses in the Beitbridge area (Light, 1982) favors the latter interpretation since peralkaline magmatism is almost entirely restricted to regions of extensional tectonics. Certainly, if the Limpopo Belt was subject to an extensional regime prior to the cordierite–orthopyroxene granulite metamorphism, it would provide a mechanism for the elevated transient geotherms implied by their assemblages and for the rapid decompression implied by their textures. The distinction between these two possible models of decompression, (a) tectonic thickening and erosion, and (b) extensional uplift, may in principle be made by detailed investigation of the mechanisms and kinetics of the decompression reactions. We would expect the former model to involve decompression over a longer time-scale than the latter.

Conclusions

Metapelitic granulites from the Diti Formation of the Central Limpopo Belt provide several mineral barometers which include the following four phase assemblages: cordierite–garnet–sillimanite–quartz, cordierite–orthopyroxene–sillimanite–quartz, cordierite–garnet–orthopyroxene–quartz and cordierite–orthopyroxene–sillimanite–spinel. Appropriate equilibrium conditions have been calculated in the MAS system based on internally consistent standard state data. Application of the equilibria to the Beitbridge assemblages indicates that metamorphic pressures lay in the range of 4–5 kbar (assuming $a_{H_2O} \approx 0$ during metamorphism), similar to pressures derived from cordierite-bearing assemblages in the South Marginal Zone. Application of published thermobarometers to more basic assemblages from the Diti Formation indicates peak metamorphic conditions given by $P = 4.0 \pm 0.8$ kbar, $T = 750 \pm 50$°C. The close agreement between cordierite-bearing and cordierite-absent barometers provides some indication that water activity was very low during metamorphism. No evidence was found for high-pressure granulite metamorphism, although textural evidence of decompression reactions does allow the interpretation that earlier, high pressure metamorphism occurred, but whose products have been imperfectly preserved. The high temperatures maintained at relatively low pressures suggest that decompression was essentially isothermal and therefore unusually rapid. One possible hypothesis, which requires further testing, is that the assemblages record a period of extensional tectonics with associated crustal thinning and rapid uplift of the lower crust to shallow crustal levels.

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References

Armbruster, T. and Bloss, F. D. (1982) Orientation and effects of channel H_2O and CO_2 in cordierite. American Mineralogist, 67, 185–291.

Barton, J. M. and Key, R. M. (1981) The tectonic development of the Limpopo Mobile belt and the evolution of the Archaean Cratons of Southern Africa. In A. Kroner, Ed., Precambrian Plate Tectonics, p. 185–212. Elsevier, Amsterdam.

Barton, M. J. and Ryan, B. (1977) A review of the geologic framework of the Limpopo Mobile Belt. Botswana Geological Survey Bulletin, 12, 183–200.

Bohlen, S. R. and Essene, E. J. (1979) A critical evaluation of two-pyroxene thermometry in Adirondack granulites. Lithos, 12, 251–350.

Bohlen, S. R., Boettcher, A. L., Wall, V. J. and Clemens, J. D. (1983) Stability phlogopite and quartz and sanidine–quartz: A model for melting in the lower crust. Contributions to Mineralogy and Petrology, 83, 270–277.

Boyd, F. R. (1959) Hydrothermal investigations of amphiboles. In P. H. Abelson, Ed., Researches in Geochemistry, 1. Wiley, New York.

Broderick, T. J. (1979) Explanation of the geological map of the country South of Nuanetsi, Nuanetsi and Beitbridge Districts. Rhodesian Geological Survey Short Report 46.

Charlu, R. V., Newton, R. C. and Keppa, O. J. (1975) Enthalpies of formation at 970 K of compounds in the system MgO–

Al_2O_3–SiO_2 from high temperature solution calorimetry. Geochimica et Cosmochima Acta, 39, 1487–1497.

Chinner, G. A. and Sweatman, T. R. (1968) A former association of enstatite and kyanite. Mineralogical Magazine, 36, 1052–1068.

Cliff, R. A. (1975) Two granulite facies events near the northern margin of the Limpopo Belt. 19th Annual Report res. Inst. African Geology, University of Leeds, 68–69.

Coward, M. P. and Fairhead, J. D. (1980) Gravity and structural evidence for the deep structure of the Limpopo Belt, southern Africa. Tectonophysics, 68, 31–43.

Danckwerth, P. and Newton, R. C. (1978) Experimental determination of the spinel peridotite to garnet peridotite reaction in the system MgO–Al_2O_3–SiO_2 in the range 900°C–1100°C and Al_2O_3 isopleths of enstatite in the spinel field. Contributions to Mineralogy and Petrology, 66, 189–201.

Ellis, D. J. and Green, D. H. (1979) An experimental study of the effect of Ca upon garnet-clinopyroxenes Fe–Mg exchange equilibria. Contributions of Mineralogy and Petrology, 71, 13–22.

England, P. C. and Richardson, S. W. (1977) The influence of erosion upon the mineral facies of rocks from different metamorphic environments. Journal of the Geological Society of London, 134, 201–213.

Fonarev, V. I. and Graphchikov, A. A. (1982) Experimental study of Fe–Mg and Ca distribution between coexisting ortho and clinopyroxenes P = 294 MPa and T = 750 and 800°C. Contributions to Mineralogy and Petrology, 79, 311–318.

Ferry, J. M. and Spear, F. S. (1978) Experimental calibration of the partitioning of Fe and Mg between biotite and garnet. Contributions to Mineralogy and Petrology, 66, 113–117.

Ghent, E. D., Knitter, C. C., Raeside, R. P. and Stout, M. Z. (1982) Geothermometry and geobarometry of pelitic rocks, upper kyanite and sillimanite zones, Mica Creek area, British Columbia. Canadian Mineralogist, 20, 295–305.

Grew, E. S. (1981) Granulite facies metamorphism at Mobdezhanaza Station, East Antarctica. Journal of Petrology, 22, 297–336.

Hansen, B. (1981) The transition from pyroxene granulite facies to garnet clinopyroxene granulite facies. Experiments in the system CaO–MgO–Al_2O_3–SiO_2. Contributions to Mineralogy and Petrology, 76, 234–242.

Harley, S. L. and Green, D. H. (1982) Garnet–orthopyroxene barometry for granulites and peridotites. Nature, 300, 697–701.

Harris, N. B. W. (1981) The application of spinel-bearing metapelites to P/T determinations: an example from South India. Contributions to Mineralogy and Petrology, 76, 229–233.

Harris, N. B. W. and Jayaram, S. (1982) Metamorphism of cordierite gneisses from the Bangalore region of the Indian Archean. Lithos, 15, 89–98.

Haselton, H. T. and Westrum, E. F. (1980) Low temperature heat capacities of synthetic pyrope, grossular and $pyrope_{60}$ $grossular_{40}$. Geochimica et Cosmochimica Acta, 44, 701–709.

Hazen, R. M. (1976) Effects of temperature and pressure on the crystal structure of forsterite. American Mineralogist, 61, 1280–1293.

Hickman, M. H. (1978) Isotopic evidence for crustal reworking in the Rhodesian Archean craton, southern Africa. Geology, 6, 214–216.

Hickman, M. H. and Wakefield, J. (1975) Tectonic implications of new geochronological data from the Limpopo belt at Pikwe, Botswana, southern Africa. Geological Society of America Bulletin, 86, 1468–1472.

Hoffer, E. and Grant, J. A. (1980) Experimental investigation of the formation of cordierite–orthopyroxene paragenesis in pelitic rocks. Contributions to Mineralogy and Petrology, 73, 15–22.

Holland, T. J. B. and Carpenter, M. A. (1983) Order–disorder and melting in sillimanite. Terra Cognita, 3, 162.

Horrocks, P. C. (1980) Ancient Archaean supracrustal rocks from the Limpopo Mobile Belt. Nature, 286, 596–599.

Horrocks, P. C. (1983) A corundum and sapphirine paragenesis from the Limpopo mobile belt, S. Africa. Journal of Metamorphic Geology, 1, 13–23.

Humphries, F. J. and Cliff, R. A. (1982) Sm–Nd dating and cooling history of Scourian granulite, Sutherland. Nature, 295, 515–516.

James, P. R. (1975) A deformation study across the northern margin of the Limpopo Belt, Rhodesia. Ph.D. Thesis, University of Leeds, England.

Kleppa, O. J. and Newton, R. C. (1975) The role of solution calorimetry in the study of mineral equilibria. Fortschritte der Mineralogie, 52, 3–20.

Krupka, K. M, Kerrick, D. M. and Robie, R. A. (1979) Heat capacities of synthetic orthoenstatite and natural anthophyllite from 5 to 1000 K. EOS, Transactions of the American Geophysical Union, 60, 405.

Light, M. P. R. (1982) The Limpopo mobile belt: a result of continental collision. Tectonics, 1, 325–342.

Light, M. P. R. and Watkeys, M. K. (1977) An outline of the Archaean and early Proterozoic geological history of the region around Beitbridge. Annual Rhodesian Geological Survey Report, 3, 35–40.

Lindsley, D. A. (1983) Pyroxene thermometry. American Mineralogist, 68, 477–493.

Lonker, S. W. (1981) The P–T–X relations of the cordierite–garnet–sillimanite–quartz equilibrium. American Journal of Science, 281, 1056–1090.

Martignole, J. and Nantel, S. (1982) Geothermometry of cordierite-bearing metapelites near the Morin anorthosite complex, Grenville province, Quebec. Canadian Mineralogist, 20, 307–318.

Martignole, J. and Sisi, J. C. (1981) Cordierite–Garnet–H_2O equilibrium: a geological thermometer, barometer and H_2O fugacity indicator. Contributions to Mineralogy and Petrology, 77, 38–46.

Mason, R. (1969) The Limpopo mobile belt—Southern Africa. Philosophical Transactions of the Royal Society of London, Series A, 273, 463–485.

Newton, R. C. (1972) An experimental determination of the high pressure stability limits of magnesian cordierite under wet and dry conditions. Journal of Geology, 80, 398–420.

Newton, R. C. and Haselton, H. J. (1981) Thermodynamics of the Garnet–Plagioclase–Al_2SiO_5–Quartz geobarometer. In R. C. Newton and B. J. Wood Eds., Thermodynamics of Minerals and Melts, p. 129–145. Springer-Verlag, New York.

Newton, R. C. and Wood, B. J. (1979) Thermodynamics of water in cordierite and some petrologic consequences of cordierite as a hydrous phase. Contributions to Mineralogy and Petrology, 68, 391–406.

Perkins, D. III (1983) The stability of Mg-rich garnet in the system CaO–MgO–Al_2O_3–SiO_2 at 1000–1300°C and high pressure. American Mineralogist, 68, 355–364.

Perkins, D. III and Newton, R. C. (1981) Charnockite geobarometers based on coexisting garnet–pyroxene–plagioclase–quartz. Nature 292, 144–146.

Perkins, D. III, Holland, T. J. B. and Newton, R. C. (1981) The Al_2O_3 contents of enstatite in equilibrium with garnet in the system MgO–Al_2O_3–SiO_2 at 15–40 kbar and 900–1600°C. Contributions to Mineralogy and Petrology, 78, 99–109.

Powell, R. (1978) The thermodynamics of pyroxene geotherms. Philosophical Transactions of the Royal Society of London, Series A, 288, 457–469.

Robertson, I. D. M. (1977) Some granulite facies metasediments of the Rhodesian part of the North Marginal zone of the Limpopo mobile belt. Geological Survey of Botswana Bulletin 12, 157–177.

Robie, R. A., Hemingway, B. S. and Fisher, J. R. (1978) Thermodynamic properties of minerals and related substances at 298.15 K and 1 bar (10^5 pascals) pressure and at higher temperatures. Bulletin of the U.S. Geological Survey 1452.

Skinner, B. J. (1966) Thermal expansion. In S. P. Clark Ed., Handbook of Physical Constants, p. 75–96. Geological Society of America Memoir 97.

Touret, J. (1974) Faciès Granulite et Fluides Carboniques. Centenaire de la Société Géologique Belgique. Géologic des domaines Cristallins, Liege, 267–287.

Tracy, R. J., Robinson, P. and Thompson, A. B. (1976) Garnet composition and zoning in the determination of temperature and pressure of metamorphism, central Massachusetts. American Mineralogist, 61, 762–775.

Van Breemen, O. and Dodson, M. H. (1972) Metamorphic chronology of the Limpopo Belt, southern Africa. Geological Society of American Bulletin, 83, 2005–2018.

Van Breemen, O. and Hawkesworth, C. J. (1980) Sm–Nd isotopic study of garnets and their metamorphic host rocks. Transactions of the Royal Society of Edinburgh: Earth Sciences, 71, 97–102.

Van Reenen, D. D. and du Toit, M. C. (1978) The reaction Garnet + Quartz = Cordierite + Hypersthene in granulites of the Limpopo Metamorphic Complex in Northern Transvaal. Geological Society of South Africa Special Publication, 4, 149–177.

Vielzeuf, D. (1980) Orthopyroxene and cordierite secondary assemblages in the granulitic paragneisses from Lherz and Saleix (French Pyrenees). Bulletin de Minéralogie, 103, 66–78.

Vielzeuf, D. (1983) The spinel and quartz associations in high grade xenoliths from Tallante (SE Spain) and their potential use in geothermometry and barometry. Contributions to Mineralogy and Petrology, 82, 301–311.

Watkeys, M. K. (1979) Explanation of the Geological Map of the Country west of Beitbridge. Rhodesian Geological Survey Short Report, 45.

Wells, P. R. A. (1977) Pyroxenes thermometry in simple and complex systems. Contributions to Mineralogy and Petrology, 62, 129–139.

Wells, P. R. A. (1979) Chemical and thermal evolution of Archaen sicilic crust, Southern West Greenland. Journal of Petrology, 20, 187–226.

Winter, J. K. and Ghose, S. (1979) Thermal expansion and high temperature crystal chemistry of the Al_2SiO_5 polymorphs. American Mineralogist, 64, 573–586.

Wones, D. R. and Dodge, F. C. W. (1977) The Stability of phlogopite in the Presence of quartz and diopside. In D. G. Fraser, Ed., Thermodynamics in Geology, p. 229–248. Reidel Publishing Company, Dordrecht, The Netherlands.

Wood, B. J. and Banno, S. (1973) Garnet–orthopyroxene and orthopyroxene–clinopyroxene relationships in simple and complex systems. Contributions to Mineralogy and Petrology, 42, 109–124.

Wood, B. J. and Holloway, J. R. (1982) Theoretical prediction of phase relationships in planetary mantles. Proceedings of the 13th Lunar and Planetary Scientific Conference Part 1, Journal of Geophysical Research, 87, A10–A30.

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