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The Reaction



as a Geological Thermometer in the Opinicon Lake Region, Ontario

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Abstract. In equilibrated metamorphic rocks containing coexisting garnet, cordierite, quartz and sillimanite, the exchange of iron and magnesium between cordierite and garnet offers a highly favourable geological thermometer and barometer, because this exchange reaction is insensitive to pressure. Thermodynamic analysis shows that this thermometer may be calibrated from knowledge of the breakdown reactions for iron and magnesian cordierite end members to garnet. The thermometer was experimentally calibrated using cordierites of intermediate composition. When applied to rocks showing petrographic evidence of equilibrium, and chemical evidence of reaction between garnet and cordierite, the thermometer yielded temperatures of 600–750°C, and pressures of 5.7–6.7 kilobars. Similar conditions are indicated by other literature data on cordierite-garnet gneisses, and are believed to represent hornblende granulite grade of metamorphism.

One of the basic problems of metamorphic petrology is the determination of the peak pressure and temperature of metamorphism. If one assumes that the only relevant variables are temperature, total pressure and composition, this information can, in principle, be gained from the composition of appropriate coexisting mineral phases which are linked by a reaction relation dependent only on temperature and pressure. During a study of metamorphic rocks in southeastern Ontario, (Currie and Ermanovics, in press) petrographic evidence was discovered which makes it seem probable that garnet and cordierite are appropriate phases for this purpose.

The coexistence of cordierite and garnet in this region has been described in some detail by Wynne-Edwards and Hay (1963) and by Reinhardt (1968). In the Opinicon Lake region (Fig. 1) cordierite, together with quartz and potash feldspar, is present in the assemblages cordierite-biotite, cordierite-sillimanite, cordierite-garnet-biotite-sillimanite, and cordierite-garnet-sillimanite. One specimen contains cordierite-sillimanite-andalusite (69–117). Garnet free assemblages occur in the southeastern part of the area only, whereas to the northwest, garnet is found as porphyroblasts containing sillimanite and quartz inclusions. The amount of cordierite steadily declines to the north-west, and no cordierite is found northwest of Opinicon Lake. Plagioclase (An 38–55) is rarely present in cordierite bearing rocks and opaque minerals are present only as alteration products of mafic minerals.

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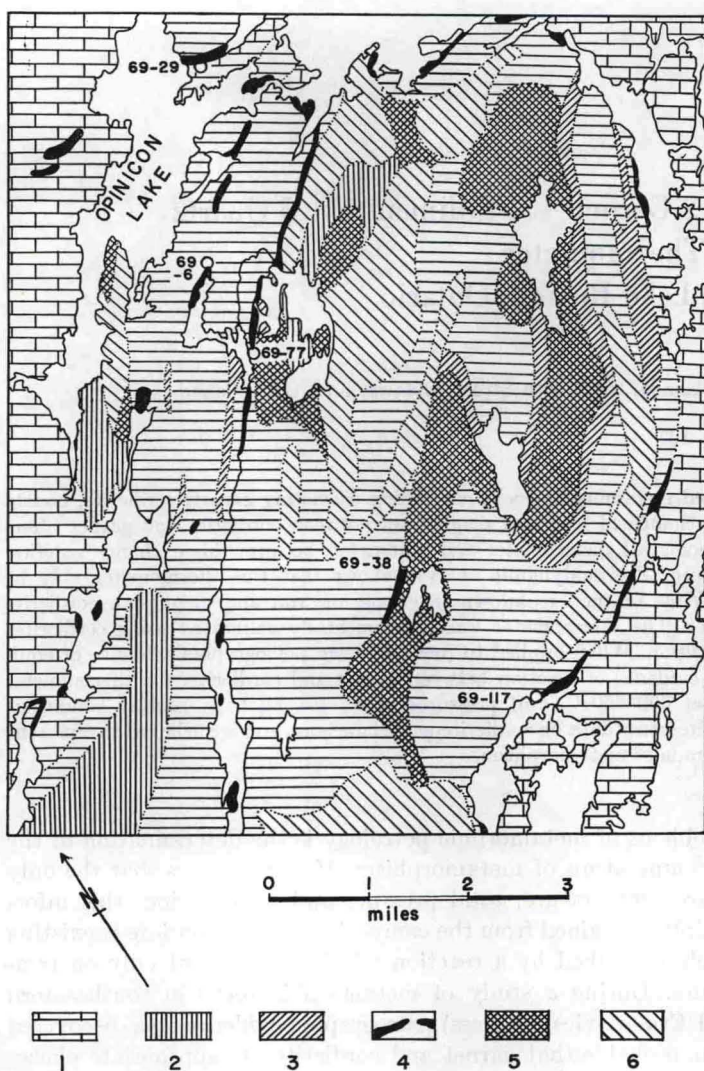


Fig. 1. Geological sketch of the Opinicon Lake region (geology from Currie and Ermanovics, in press). 1 marble and related rocks; 2 pyroxene granulites; 3 quartzite, including migmatite; 4 cordierite-bearing gneisses, garnet bearing except in southeast corner of area; 5 biotite gabbro, diorite, and mafic gneiss; 6 granitoid plutons. Horizontal ruling — stratiform quartz-feldspar-biotite gneisses. The centre of the map area lies near $44^{\circ}30' \text{ N}$, $76^{\circ}20' \text{ W}$, about 28 miles north of the city of Kingston

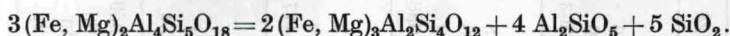
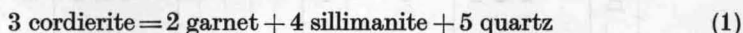
An electron probe study of the partition of iron and magnesium among the coexisting phases (Table 1) showed that cordierite, in addition to decreasing in amount, became steadily more magnesian from southwest to northeast, while garnet increased in amount and magnesium content. The $\text{Fe}/\text{Fe} + \text{Mg}$ ratio of biotite showed no very marked changes. These data strongly suggest that garnet

Table 1. *Compositions of coexisting garnet, cordierite, and biotite from the Opinicon Lake region*

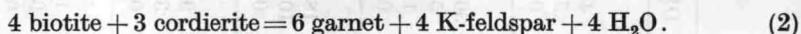
	H-70	H-29	H-66	69-117	69-38	69-77	69-6	69-29	D28	D56	D87	R114	R124	D102
Cordierite														
MgO	2.11	7.11	6.80	7.17	8.03	9.06	9.94	10.35	9.7	7.7	4.4	6.6	8.9	8.1
FeO	3.24	8.28	5.67	9.75	8.41	6.80	5.42	4.68	5.7	6.0	5.6	7.9	6.9	6.4
Fe/Fe + Mg	0.454	0.367	0.319	0.433	0.370	0.303	0.233	0.202	0.248	0.304	0.417	0.502	0.303	0.309
Garnet														
FeO	27.66	28.76	30.87	31.5	30.6	29.3	27.9	26.8	25.9	282	30.9	30.6	27.8	29.3
MgO	5.67	6.70	7.10	6.57	7.25	8.25	9.22	9.90	8.2	6.0	5.0	5.4	6.8	7.3
CaO	1.45	150	1.89	1.58	1.57	1.57	1.54	1.53	2.0	1.7	1.6	2.3	1.9	1.8
Fe/Fe + Mg	0.732	0.652	0.704	0.772	0.748	0.713	0.680	0.654	0.638	0.725	0.776	0.761	0.696	0.692
Biotite														
FeO	21.99	18.47	18.28	19.8	21.5	21.9	20.0	19.2	14.2	15.2	18.6	19.2	15.3	—
MgO	8.40	9.70	8.70	10.33	9.57	9.25	10.24	10.55	14.0	11.7	10.3	10.0	12.0	—
Fe/Fe + Mg	0.568	0.517	0.540	0.51	0.55	0.57	0.52	0.50	0.363	0.422	0.503	0.519	0.417	—
Bulk rock														
FeO	7.42	6.67	6.67	6.25	6.34	6.87	6.61	6.08	7.8	6.4	9.6	8.2	8.7	5.9
MgO	3.35	3.40	3.15	3.23	2.91	2.91	3.43	3.41	3.7	3.0	3.7	3.4	4.1	2.6
Fe/Fe + Mg	0.554	0.523	0.543	0.52	0.59	0.56	0.56	0.53	0.54	0.54	0.59	0.575	0.54	0.56
<i>K</i>	3.29	3.56	3.79	3.90	4.44	5.20	6.11	6.57	5.344	6.036	4.843	4.736	5.259	5.03
ln <i>K</i>	1.19	1.27	1.33	1.36	1.46	1.65	1.81	1.88	1.67	1.80	1.58	1.56	1.66	1.62
<i>T</i> (°A)	870	885	895	900	925	955	990	1005	960	989	942	940	959	950
<i>P</i> (Kbars)	5.76	6.04	6.04	5.68	6.16	6.19	6.46	6.69	6.37	6.37	6.16	5.85	6.22	6.22

Analyses H-70, H-29, H-66 from Wynne-Edwards and Hay (1963); analyses D28, D56, D87, R114, R124, R102 from Reinhardt (1968).

and cordierite are in a reaction relation experimentally realized by Richardson (1968) and by Hirschberg and Winkler (1968), namely



If the rocks have equilibrated, and all phases in Eq. (1) are present and stoichiometric, the presence of other iron and/or magnesium bearing minerals, such as biotite or hypersthene, does not change the equilibrium constant defined by (1), but alters the amounts of garnet and cordierite to permit appearance of minerals with Fe/Fe + Mg ratios controlled by equations analogous to (1), for example



Since Al_2O_3 and SiO_2 are present in excess of the amounts needed to form the iron and iron magnesium silicates in Eq. (1), it follows that variation in Al/Fe + Mg and Si/Fe + Mg ratios do not affect Eq. (1) unless they result in the disappearance of one of the phases. These considerations suggest Eq. (1) as a possible geothermometer, assuming that (a) oxygen fugacity is low enough to prevent oxidation of significant amounts of iron in the garnet and cordierite, (b) cordierite is anhydrous (Schreyer and Yoder 1954), making the reaction independent of water fugacity, (c) cordierite and garnet contain no contaminants replacing significant amounts of iron and magnesium. Whole rock compositions (Wynne-Edwards and Hay, 1963; Reinhardt, 1968; Currie and Ermanovics, in press) show that the contents of the most probably contaminants, Ca and Mn, are notably low in garnet-cordierite gneisses.

Thermodynamic Considerations

The operation of the garnet-cordierite geothermometer may be displayed as follows. Consider the exchange reaction



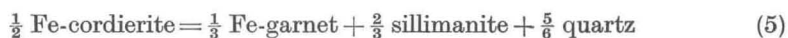
The theory of such reactions has been treated by Kretz (1961) and others, assuming that each mineral forms an ideal solution of iron and magnesium end members. The cell volumes of both garnet and cordierite are linearly dependent on the mol fraction of the end members (Iiyama, 1956; Rickwood, 1968) which is a necessary but not sufficient condition for ideality. We shall assume ideality, and show below that the reaction experimentally shows almost ideal behaviour.

For given (P , T) conditions, reaction (3) will reach an equilibrium described by the equilibrium constant K . The change in Gibbs free energy associated with the reaction may be written

$$\begin{aligned} dF &= -RT \ln K = -RT \ln (X_m^c / 1 - X_m^c) (X_f^g / 1 - X_f^g) \\ &= \frac{1}{2} F_m^c + \frac{1}{3} F_f^g - \frac{1}{2} F_f^c - \frac{1}{3} F_m^g \end{aligned} \quad (4)$$

where F is the Gibbs free energy, T the absolute temperature, R the gas constant, X_j^i the mol fraction of component j in phase i , and m — Mg, f — Fe, g — garnet, c — cordierite. The volume change in this reaction is negligible (see p. 4), hence K is not explicitly dependent on pressure.

Information on the value of K may be obtained by study of the reaction



which has been realized by Richardson (1968). This reaction is at equilibrium along a given curve in the P - T plane, at any point of which

$$\left(\frac{1}{2} F_f^c - \frac{1}{3} F_f^g\right) = \left(\frac{2}{3} F_{\text{sill}} + \frac{5}{6} F_{\text{quartz}}\right) \quad (6)$$

Now for a given temperature T , if the equilibrium pressure for Eq. (5) is p_e^a , then at any other pressure, P

$$\left(\frac{1}{2} F_f^c - \frac{1}{3} F_f^g\right) = \frac{2}{3} F_{\text{sill}} + \frac{5}{6} F_{\text{quartz}} + (P - P_e^a) dV \quad (7a)$$

where the possible difference in compressibility between phases is ignored. dV is the volume change in reaction (5).

Now an exactly analogous equation to (7) holds for the conversion of magnesian cordierite. Using the data of Robie and Waldbaum (1968), Iiyama (1956), Schreyer (1965), Deer *et al.* (1963, vol. 1) and Rickwood (1968), the change in volume of this reaction (26.35 cc per mol) is virtually identical to that of the iron analogue, (26.49 cc per mol). The equation analogous to (7a) is therefore

$$\left(\frac{1}{2} F_m^c - \frac{1}{3} F_m^g\right) = \frac{2}{3} F_{\text{sill}} + \frac{5}{6} F_{\text{quartz}} + (P - P_e^m) dV \quad (7b)$$

subtracting (7a) from (7b) and substituting in (4) we obtain

$$\begin{aligned} \left(\frac{1}{2} F_m^c - \frac{1}{3} F_m^g\right) - \left(\frac{1}{2} F_f^c - \frac{1}{3} F_f^g\right) &= -(P_e^p - P_e^a) dV \\ &= -RT \ln (X_m^c/1 - X_m^c) (X_f^g/1 - X_f^g). \end{aligned} \quad (8)$$

From Eq. (8) it is clear that if the composition of coexisting garnet and cordierite are known, and the pressures at which pure cordierite end members are converted to garnet are known, the temperature can be found. (8) may be rearranged to give

$$T = (dP \cdot dV/R) (1/\ln K) \quad (9)$$

where dP is $(P_e^p - P_e^a)$.

It may also be shown, given ideal solution in garnet and cordierite, that the pressure is fixed for a particular coexisting pair. Consider pure Fe-cordierite in equilibrium with pure almandine. Now suppose that magnesium is added to the system. The magnesian component of the cordierite and garnet would be in equilibrium with each other at P_e^p . Thus for garnet to remain in equilibrium with cordierite in the magnesium bearing system, the pressure must rise in such a way that the change in free energy due to the magnesium components is balanced by the change in free energy of the iron components. Since the volume change for conversion of cordierite to garnet is invariant with composition, this condition may be expressed by

$$(P_e^p - P) (X_m^g + X_m^c) dV = (P - P_e^a) (X_f^g + X_f^c) dV$$

By algebraic rearrangement, and substitution of $(1 - X_m^g)$ for X_f^g and $(1 - X_m^c)$ for X_f^c , this may be reduced to

$$P = P_e^a + (dP/2) (X_m^g + X_m^c) \quad (10b)$$

Given appropriate experimental data, it is thus possible to determine both temperature and pressure from the compositions of coexisting garnet and cordierite.

Experimental Considerations

The variation with temperature of the equilibrium pressure between Fe-cordierite and almandine is known from the work of Richardson (1968), but similar data is not available for pyrope. In fact the work of Schreyer (1970) and Henson and Green (1970) strongly suggests that the direct breakdown of magnesian cordierite to pyrope can not be experimentally realized because of the appearance of hypersthene and/or sapphirine as a breakdown product of cordierite. This difficulty does not invalidate Eq. (9), but it requires experimental work to establish the value of dP . From Eq. (9), it may be seen that for a given T , one value for K would establish dP . Other values of K at the same temperature would serve to demonstrate the internal consistency of the model, that is the validity of the ideal solution assumption for garnet and cordierite, and the neglect of compressibilities in calculating dP .

An experimental program was undertaken to establish the value of dP for various temperatures in the metamorphic range. Gels with the stoichiometric composition of cordierite with Fe/Fe + Mg ratios 0.8, 0.6, 0.4 and 0.2 were prepared by dissolving appropriate amounts of standardized ferrous oxalate, magnesium oxalate, and aluminum wire in nitric acid, and gelling with tetraethyl orthosilicate. After dessication, the iron in the products was reduced to the ferrous state by passing CH_4 over it at 800°C for 8 hours. Analysis of the products after this treatment showed that they were 100% cordierite and that the ferrous/ferric ratio exceeded 20 in all materials. Pure cordierite with Fe/Fe + Mg > 0.8 cannot be prepared by this method due to appearance of a spinel phase.

The experiments were carried out in an internally heated pressure vessel similar to that described by Goldsmith and Heard (1961), using methane as a pressure medium. Temperatures were measured with 3 Pt - Pt/10% Rh thermocouples, placed at 0.5 cm intervals along the sample capsules, and pressure was measured by a resistance piezometer standardized against a dead-weight tester. The specimens were welded into 3 mm diameter by 0.1 mm wall Pt tube with about 5 weight percent H_2O , and four samples of different Fe/Fe + Mg ratio were run at one time, permitting the simultaneous determination of the garnet cordierite equilibrium over a range of compositions. The empty space in the furnace was filled by graphite rods, assuring maintenance of the oxygen fugacity at the graphite-methane buffer. Quoted temperature are believed to be accurate within $\pm 4^\circ$, and pressure within 0.1 kilobar.

All runs in this apparatus were 48 hours in length. Sixty day runs in standard Tuttle bombs, using the graphite-methane buffer, gave results identical within measurement errors to the 48 hour runs at 600° , 6 kilobars, 40 mol per cent magnesium molecule, and 800° , 5 kilobars, 20 mol percent magnesium molecule.

The experimental products were X-rayed in 114.6 mm. Debye-Scherrer cameras using silica as an internal standard, and examined in immersion oils. The first appearance of garnet was more easily detected optically. The composition of the garnet was determined by assuming that the cell edge and refractive index were

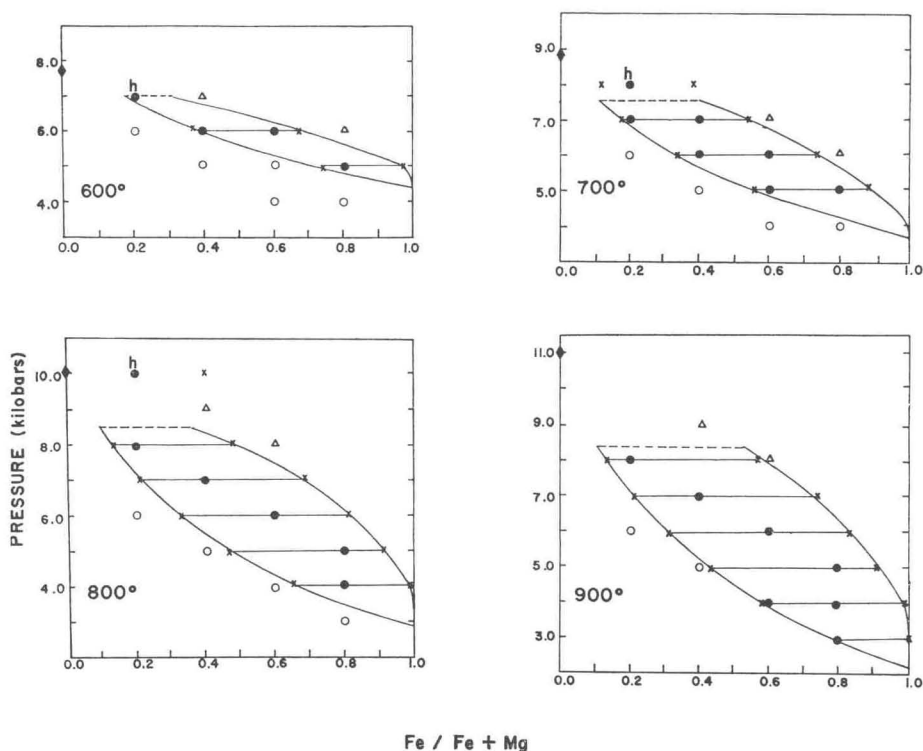


Fig. 2. Experimental results in the System Garnet-cordierite. ○ only cordierite in products; △ only garnet in products; ● coexisting garnet and cordierite in products, with composition given by the x . The runs shown by solid circles marked "h" contained hypersthene in the products, plus garnet and/or cordierite with the composition shown by the x

11.456 and 1.714 for pyrope, and 11.526 and 1.830 for almandine and that both varied linearly with composition. The cell edge was determined to $\pm 0.003 \text{ \AA}$. Corresponding to about 5% error in the almandine content, while the refractive index was determined within 0.003, corresponding to a 3% error in almandine content. The two determinations agreed within 5% in all cases, and this is believed to be the approximate limit of error. Cordierite composition was determined by comparison of the X-ray powder patterns and refractive indices of the run products with the series of artificial cordierites of known composition used as starting materials. Both "d" spacings and maximum refractive index appear to vary linearly with composition, and, with the accuracy of determination noted above, give a determination of composition within 7%. The two determinations agreed within 9% in all cases.

The only phases other than garnet and cordierite detected were sillimanite, quartz, and hypersthene. The latter appeared only in three runs in composition $\text{Fe/Fe} + \text{Mg} = 0.20$. The results of the 46 runs are presented graphically in Fig. 2¹, which incorporates the data of Richardson (1968).

¹ A table of the experimental results is available on request from the author.

Table 2. Calculation of dP from experimental runs

T (Å)	P (kbars)	X_m^c	X_m^g	K	$\ln K$	dP	avg. dP	avg. K
873	6.0	0.64	0.32	3.18	1.156	3.24	3.24	3.18
973	5.0	0.41	0.11	5.62	1.726	5.27		
	6.0	0.66	0.26	5.52	1.708	5.21	5.27	5.62
	7.0	0.83	0.46	5.73	1.746	5.33		
1073	5.0	0.52	0.11	8.76	2.170	7.31		
	6.0	0.67	0.19	8.65	2.157	7.26	7.28	8.68
	7.0	0.80	0.32	8.50	2.140	7.21		
	8.0	0.86	0.41	8.83	2.178	7.33		
1173	5.0	0.56	0.09	12.87	2.555	9.40		
	6.0	0.69	0.16	11.68	2.457	9.04	9.28	1246
	7.0	0.80	0.24	12.67	2.359	9.35		
	8.0	0.85	0.31	12.61	2.534	9.33		

$$dP = \frac{RT \ln K}{dv} = 0.003138 T \ln K.$$

The results suggest that garnet-cordierite equilibrium, except for a relatively small region at the magnesium-rich end, can be represented by simple loops in P - X projection (constant T). This is the prediction made by the thermodynamic analysis and Fig. 2 shows the computed loops, from Eq. (9) and (10) using experimental P_e^p . The experimental work therefore strongly supports the hypothesis of ideality for Eq. (3). In P - X projection the area occupied by coexisting garnet and cordierite becomes larger with increasing temperature. Presumably the garnet-cordierite volume is terminated at high temperature by other reactions, probably involving hypersthene, (Henson and Green 1970).

Application of the Garnet-Cordierite Geothermometer

The experimental data may be applied to natural assemblages where garnet and cordierite are the only Fe and Mg bearing phases, or to groups of specimens of fixed mineral composition where the compositions of garnet and cordierite change composition in various specimens, but the other phases do not. In such assemblages, it is obvious from Fig. 2 that at a given temperature and pressure the compositions of coexisting garnet- and cordierite are fixed. However it may also be possible to find garnet-free or cordierite-free assemblages at the given P , T . The value of K from the coexisting garnet and cordierite may be converted directly to T , by noting that the relation between dP and T in Table 2 is linear. It may be expressed by

$$dP = 0.020 T - 14.18 \quad (11)$$

where dP is in kilobars, and T in absolute temperature. Substituting this in Eq. (9) and rearranging yields

$$T = \frac{14.18}{0.020 - \frac{R \ln K}{dv}} \quad (12)$$

substituting the appropriate values for R and dV into this equation gives

$$T = 4515 / (6.37 - \ln K). \quad (12a)$$

It can readily be seen that this geothermometer is relatively insensitive for Fe/Fe + Mg ratios greater than about 0.7, because the garnet produced is very close to pure almandine (Fig. 2). For ratios lower than this it can be computed that an error in $\ln K$ of 0.2, is equivalent to a temperature error of 50°. For a typical K of 6, such an error would correspond to an error of roughly 1.1 in determination of K . If this error in turn is assigned entirely to analytical error, it would correspond roughly to an error of 10% in the Fe/Mg + Fe ratio of both garnet and cordierite. Determination of this accuracy does not appear to present any problems in the present state of the art, either by electron probe methods, or by concentration of minerals. It follows that for favourable compositions the method should give results accurate within 50°, and probably considerably better.

Eq. 12(a) has been applied to the data in Table 1. These data indicate that the temperature reached about 600° in the southeastern part of the area, and almost 700° in the northwestern part of the area, at pressures of 5.1 and 5.8 kilobars respectively.

We have also applied the geothermometer to the data of Reinhardt (1968), and Wynne-Edwards and Hay (1963) (Table 1). For the simplest mineral assemblage garnet-cordierite-sillimanite quartz (Reinhardt's specimen D-102) the computed conditions are 675°C and 6.2 kilobars. For his other specimens containing biotite, excepting specimen D-56, the garnet-cordierite pair, indicates a temperature of $675 \pm 10^\circ$, and pressure of 6.1 ± 0.3 kbars, in agreement with the conditions indicated by specimen D-102. This agreement strongly suggests that the geothermometer is applicable to nearly all cordierite-garnet-sillimanite-quartz gneiss. It is possible that D-56 illustrates the partial breakdown of one of the assumptions, but it is also possible that an analytical error is involved. The other specimens which show significant variations in Fe/Mg ratios in biotite, demonstrate the independence of the thermometer from other Fe and Mg bearing phases.

Throughout this analysis the pressure of H_2O has been ignored. In Fig. 3 we have combined data for the garnet-cordierite equilibrium with data for the aluminosilicate triple-point (Gilbert *et al.*, 1968), the biotite breakdown curve (Eugster and Wones, 1964), and the muscovite breakdown curve (Evans, 1965). The following conclusions, although based on the Gilbert *et al* triple point, are also applicable to most of the other suggested triple points discussed by Zen (1969). The occurrence of andalusite in equilibrium with sillimanite, as observed in the southeastern part of the region, and the total absence of muscovite cannot be explained if $P_{H_2O} = P_{total}$. However if P_{H_2O} is much less than total pressure, the field data may be reconciled with experimental data. The data of Reinhardt (1968) require very low water pressures, approaching 0.2 total pressure. According to Gilbert *et al.* (1968), for the deduced P - T conditions these rocks would contain sillimanite as the stable polymorph. Under these conditions, garnet and cordierite can coexist with biotite, in the absence of muscovite, and in the presence of either andalusite and sillimanite. These latter polymorphs can both be present in equilibrium with compositions having Fe/Fe + Mg greater than 0.8. In order for the observed coexistence of cordierite with Fe/Fe + Mg less than 0.3 with garnet (Fe/Fe + Mg < 0.7) to take place, as it does north of Loughborough Lake, both temperature and total pressure must increase to roughly 700° and 6.5 kilobars

on the basis of one group of samples labeled "high grade", which show K values similar to those in the Opinicon Lake region, and another group labeled "low grade" which show substantially higher K values of 10–12. These latter rocks are suggested to have been metamorphosed at temperatures of roughly 400°C. No mineral assemblages are quoted by Saxena and Hollander (1969), but examination of the original sources shows that the supposedly "low grade" rocks all contain sillimanite, which according to the data of Gilbert *et al.* (1968) implies a temperature greater than 622°C. Several contain kyanite in addition, implying high pressures. Clearly such rocks cannot be correctly termed low grade metamorphic rocks. Similar K values of 10–12 are found by Gable and Sim (1969) for part of the Front Range in Colorado, where migmatite is abundant, and it is expressly stated that the rocks must have been above the melting temperature of granitic compositions. The temperatures implied by the data of Gable and Sim (1969) are approximately 850° at pressures of 6.3 kb. Significantly, the non-migmatitic areas have lower K values of 6–8, corresponding to temperatures of roughly 760° at 6.0 kb.

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