A revision of the garnet-clinopyroxene Fe 2 + -Mg exchange geothermometer

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Abstract. A comprehensive experimental dataset was used to analyse the compositional dependence of the garnet-clinopyroxene Fe²⁺/Mg partition coefficient (K_d) . The Mg no. of garnet was found to have a significant effect on the K_d , in addition to calcium content of garnet. An empirical model was developed to relate these effects with equilibrium temperature and pressure in the form of a conventional geothermometer,

 $T(K) = \{-1629[X_{C_2}^{G_1}]^2 + 3648.55[X_{C_3}^{G_1}] - 6.59[Mg \text{ no. (Gt)}]$

 $+ 1987.98 + 17.66P(kbar)\}/(\ln K_d + 1.076)$.

Application of this thermometer produced reasonable temperature estimates for rocks from the lower crust (garnet amphibolites, granulites and eclogites) and the upper mantle (eclogite and lherzolite xenoliths in kimberlites, mineral inclusions in diamonds).

Introduction

Several experimental studies have demonstrated that $Fe²⁺/Mg$ partitioning between garnet and clinopyroxene is sensitive to external physical conditions and is strongly affected by internal compositional variations (e.g. Raheim and Green 1974a; Ellis and Green 1979; Pattison and Newton 1989). Råheim and Green (1974a) first calibrated the relationship between K_d [$=$ (Fe²⁺/Mg)^{Gt}/ $(Fe^{2+}/Mg)^{Cpx}$] and equilibrium temperature (T) and pressure (P) to form a geothermometer. Ellis and Green (1979) recognised a significant effect of Ca-content on K_d and quantified this effect to produce an improved version of the thermometer, which found a broad application in the field of petrology. Krogh (1988) further improved the thermometer by replacing the simple rectilinear relationship of $\ln K_d$ with $\bar{X}_{\text{Ca}}^{\text{Gt}}$ $\bar{E} = \frac{\text{Ca}}{\text{Ca} + \text{Mg}} + \text{Fe}^{2+} + \text{Mn}$ established by Ellis and Green (1979) with a curvilinear relationship. This latter improvement is confirmed by Brey and K6hler (1990), but the improvement is limited by the amount of experimental data used and by the exact mathematical model chosen to formulate the thermometer.

A recent experimental study by Pattison and Newton (1989) showed that the Mg no. $[= 100Mg/(Mg + Fe^{2+})]$ of garnet also had an important effect on K_d . They fitted their experimental data to a third order polynomial equation to account for this effect, and derived a new version of the thermometer that is applicable to rocks with Mg no. (Gt) ranging from 12.5 to 60. However, when applied to same mineral assemblages, this and other forms of the garnet-clinopyroxene Fe^{2+}/Mg exchange thermometer (e.g. Ellis and Green 1979; Ganguly 1979; Powell 1985) gave different temperature estimates.

Green and Adam (1991) specifically designed a new set of experiments to test various versions of this thermometer and found that none of the existing formulations could be safely applied to rocks of wide origin in terms of P, T and composition. In particular, Green and Adam (1991) found that the popular Ellis-Green thermometer reproduced their run temperatures for experiments at 30 kbar and $1150-1200$ °C but increasingly overestimated equilibrium temperatures at lower pressures and lower temperatures. They also found that the Pattison-Newton graphic thermometer consistently underestimated their experimental temperatures but the Pattison-Newton mathematical formulation overestimated their experimental temperatures with two exceptions.

It is clear that additional experimental data are needed for further evaluation and improvement of this useful geothermometer. In this paper, I present new experimental results and use these in conjunction with published data to develop a refined empirical model to describe the partitioning behaviour of $Fe²⁺/Mg$ between garnet and clinopyroxene.

The dataset

The dataset used to analyse the Fe^{2+}/Mg partition between garnet and clinopyroxene consists of mineral equilibrium data in fifteen publications (Table 1) and new experimental data in ultramafic systems from Ai (1992, Table 2). The majority of these studies in the dataset are made on ultramafic compositions. Five series of experiments specifically address garnet-clinopyroxene equilibria. Two series of experiments are in mafic systems. They cover a wide range of "bulk rock" compositions. The experimental conditions range

Table 1. Reference sources for the experimental dataset

Reference no.	Source	Type of experiment	P (kbar)	$T^{\circ}C$	No. of gt-cpx pairs 39	
	Råheim and Green (1974a)	Gt-cpx equilibria	$20 - 40$	600-1500		
	Råheim and Green (1974b)	Gt-cpx equilibria	$12 - 30$	1100-1300	h.	
	Wood (1976)	Gt-cpx equilibria	$20 - 45$	1100-1400		
	Ellis and Green (1979)	Gt-cpx equilibria	$15 - 30$	750-1300	45	
	Pattison and Newton (1989)	Gt-cpx equilibria	$15 - 29$	$700 - 1200$	123	
b.	Johnston (1986)	Mafic system	$23 - 30$	1275-1455		
	Green and Adam (1991)	Mafic system	$10 - 30$	950-1200		
8	Hensen (1973)	Ultramafic system	$22.5 - 40.5$	1110-1410		
9	Akella and Boyd (1973)	Ultramafic system	$30 - 45$	1100		
10	Akella (1976)	Ultramafic system	$31 - 44$	1100-1300		
11	Mori and Green (1978)	Ultramafic system	$30 - 40$	950-1300	20	
12	Nickel (1983)	Ultramafic system	35	1000-1400	10	
13	Mengel and Green (1989)	Ultramafic system	$25 - 28$	975–1250	11	
14	Brey et al. (1990)	Ultramafic system	$28 - 60$	$900 - 1400$	21	
15	Wallace and Green (1991)	Ultramafic system	$20 - 30$	$950 - 1100$	15	
16	Table 2	Ultramafic system	$25 - 38$	1200-1500	47	

Gt, garnet; *cpx,* clinopyroxene

from 10 to 60 kbar in pressure and from 600 to 1500 $^{\circ}$ C in temperature, encompassing almost the entire lower crust-upper mantle region where garnet and elinopyroxene-bearing rocks originated. The dataset is therefore reasonably comprehensive and is meaningful for statistical analysis. However, the amount of data at low temperatures (below 900 °C) and high temperatures (above 1300 °C) is much less than in the middle temperature range (1000-1200 $^{\circ}$ C). The quality of these data is also not as good as that in the middle temperature range due to kinetic problems. At low temperatures, equilibrium is very difficult to achieve under laboratory conditions, especially in anhydrous systems, At high temperatures, the nucleation rate is so fast that metastable mineral compositions can often be preserved. Selection criteria such as cation totals to within ± 0.02 and weight percent of oxides to within ± 2.0 are employed to discriminate all experimental data available. The number of data is reduced to total 380 pairs.

Data analysis

Krogh (1988) proposed a general mathematical equation that governed the partitioning behaviour of $Fe²⁺/Mg$ between garnet and clinopyroxene:

$$
\ln K_d = \frac{\left\{a\left[X_{\text{Ca}}^{\text{Gt}}\right]^2 + b\left[X_{\text{Ca}}^{\text{Gt}}\right] + c + dP(\text{kbar})\right\}}{T(\text{K})} + e \tag{1}
$$

where a, b, c, d and e are constants. This agrees in principle with the forms of all previous derivations (Raheim and Green 1974a; Ellis and Green 1979, Ganguly 1979; Saxena 1979; Wells 1979; Dahl 1980; Powell 1985), except that of Pattison and Newton (1989) where the $\ln K_d$ -pressure relationship is a kind of reciprocal relationship and is in apparent contradiction with the experimental observation that K_d increases with increasing pressure (Raheim and Green 1974a; Green and Adam 1991). Nonetheless, the experimental results of Pattison and Newton (1989) are important because they showed a clear dependence of K_d on Mg no. (Gt), particularly in the low Mg no. (Gt) region.

Figure 1 further demonstrates the negative correlation between $\ln K_d$ and Mg no. (Gt) (only experimental data at 30 kbar are shown for clarity). Although the restricted Mg no. range and scatter of data points obscured any correla-

Fig. 1. The effect of Mg no. garnet, on $\ln K_d$. The correlation trends for most data at constant pressure and temperature are negative. For reasons of clarity, only data at 30 kbar are shown in this diagram (and Figs. 2, 3)

tion trend for data at 900 °C and 1500 °C, the trend is very obvious for data $1000-1400$ °C. The effect of aluminium cations in clinopyroxene on K_d is insignificant, and is perhaps more dependent on bulk rock composition than temperature and pressure. A correlation between $\ln K_d$ and X_{Ca}^{Cpx} [= Ca/(1 – Na)] (Ca and Na are cations in clinopyroxene for a 6-oxygen formulation) seems to be evident from Fig. 2, but this simply reflects close correlations of temperature with both $\ln K_d$ and $X_{\text{Ca}}^{\text{Cpx}}$ (Bertrand and Mercier 1985; Brey and Köhler 1990). Although Ellis and Green (1979) implied that correction of the $X_{\text{Ca}}^{\text{Cpx}}$ effect on K_d was accomplished by an $X_{\text{Ca}}^{\text{Gt}}$ term, there does not seem to be a simple relationship between $X_{\text{Ca}}^{\text{Gt}}$ and $X_{\text{Ca}}^{\text{Cpx}}$ (Fig. 3). Therefore the dependence of K_d on $X_{\text{Ca}}^{\text{Cpx}}$ is effectively taken into account by the temperature term and it follows that the composition terms which should be included in modelling the $\ln K_d - T$ relation are X_{Ca}^{ct} and Mg no. (Gt). Presence of $Fe³⁺$ in both garnet and clinopyroxene poses an additional problem for interpreting natural mineral assemblages (Luth et al. 1990), but at present this could not be evaluated with experimental data obtained from microprobe analysis.

Fig. 2. The ln K_d – $X_{\text{Ca}}^{\text{Cpx}}$ relationship. The parameter $X_{\text{Ca}}^{\text{Cpx}}$ essentially reflects temperature. The three parameters, $\ln K_d$, X_{Ca}^{Cpx} and the reciprocal of temperature, are positively correlated. Only 30 kbar data are shown

Fig. 3. The $X_{\text{Ca}}^{\text{Gt}} - X_{\text{Ca}}^{\text{Cpx}}$ correlation diagram. There is no clear, straight-forward relationship between $X_{\text{Ca}}^{\text{Gr}}$ and $X_{\text{Ca}}^{\text{Cpx}}$. Only 30 kbar data are shown

Following Krogh (1988) and adding a Mg no. (Gt) term in addition to X_{Ca}^{Gt} , I derived a modified model to describe the partitioning behaviour of $Fe²⁺/Mg$ between garnet and clinopyroxene:

 $\ln K_d =$

$$
\frac{\left\{a\left[X_{\text{Ca}}^{\text{Gt}}\right]^2 + b\left[X_{\text{Ca}}^{\text{Gt}}\right] + c\left[\text{Mg no. (Gt)}\right] + d + eP(\text{kbar})\right\}}{T(\text{K})} + f
$$
\n(2)

where a, b, c, d, e and f are constants. A statistical analysis of the 380 pairs of data in Tables I and 2 suggests that in a normal probability plot, data points which are inconsistent with the general trend may be in error. A total of 109 pairs of such erroneous data points are detected and excluded. The final dataset is left with 271 pairs of data points. Multiple linear regression analysis of the 271 pairs of coexisting garnet and clinopyroxene to fit the above
model gave: $a = -1629$ (± 636), $b = 3648.55$ model gave: $a = -1629$ (± 636) , $b = 3648.55$ $(\pm 391.72), c = -6.59 (\pm 0.35), d = 1987.98 (\pm 79.81),$ $e = 17.66 \ (\pm 0.68), f = -1.076 \ (\pm 0.036) \ (R^2 = 0.981).$ Regression with a $\left[\text{Mg} \text{ no. } (\text{Gt})\right]^2$ term included did not produce a better fit, so that this term was excluded. Consequently the revised garnet-clinopyroxene geothermometer becomes:

$$
T(K) = \{ -1629[X_{Ca}^{Gt}]^{2} + 3648.55[X_{Ca}^{Gt}] - 6.59[Mg\text{ no. (Gt)}] + 1987.98 + 17.66P(kbar)\} / (\ln K_d + 1.076). \tag{3}
$$

In Fig. 4, the new thermometer $[Eq. (3)]$ is compared with previous formulations by Ellis and Green (1979) [Fig. 4a] and Krogh (1988) [Fig. 4b] by plotting the difference between computed temperatures by each method for synthetic garnet-clinopyroxene pairs and the experimental temperatures against the relevant experimental run temperatures. Equation (3) reproduces all experimental run temperatures within $\pm 100^{\circ}$ C, and gives more accurate temperature estimates, with less scatter for temperatures below 1200 °C. Both the Ellis-Green and Krogh expressions tend to overestimate temperatures below 1200 °C. Whereas estimates above 1200 °C for all three methods are similar, the Krogh expression is most discrepant and the Ellis-Green expression shows a trend of underestimation with increasing experimental temperature (Fig. 4a, b).

Applications

Equation (3) is constructed on the basis of a large amount of experimental data over a wide range of P, T, and composition, therefore it should be applicable to rocks of wide origins and may be successfully extrapolated to natural environments below the experimental temperature range. This is tested in Table 3 by calculation of equilibrium temperatures for rocks from the lower crust (amphibolites, granulites, crustal eclogites) and the upper mantle (eclogite and lherzolite xenoliths in kimberlites, mineral inclusions in diamonds). The new thermometer generally gives more realistic temperature estimates than previously published garnet-clinopyroxene thermometers. For instance, when applied to the metamorphic suites listed by Pattison and Newton (1989), the new thermometer not only produces reasonable temperatures, but also gives narrower ranges of the estimated temperatures for each suite than the Pattison-Newton formulation. Other thermometers (e.g. Krogh 1988; Powell 1985; Ellis and Green 1979) tend to yield temperatures outside the range expected for the corresponding metamorphic grade.

Krogh (1988) calculated equilibrium temperatures in the range of 690–790 °C for a suite of eclogitic rocks from the Tromso nappe complex within the northern Scandinavian Caledonides. Equation (3) suggests a relatively lower equilibrium temperature range of $636-740$ °C, which may be more appropriate for these rocks because eclogites generally have lower equilibrium temperatures than granulites and temperatures higher than 700° C are normally considered to belong to the granulite facies.

Application of Eq. (3) to four diamondiferous eclogites from southern Africa (Smith et al. 1989), one diamondiferous lherzolite from Sloan, USA (McCallum and Eggler 1976), and two diamondiferous lherzolites from Finsch, South Africa (Shee et al. 1982) generates equilibrium temperatures consistent with diamond stability (Table 3).

Unreliable estimate of $Fe³⁺$ content in garnet and clinopyroxene, and non-equilibrium between garnet and clinopyroxene are two major sources of errors in estimating equilibrium temperatures of natural rocks. For example, Koons (1984) applied the Ellis-Green thermometer to a suite of eclogites from the Sesia zone of the western Alps. Italy and calculated their temperatures

^a Data from Ai (1992)

ranging from 400 to 1040 $^{\circ}$ C. He suggested that the spread was caused by the high-jadeite pyroxene solid solutions. Equation (3) gives an even larger temperature range (316-1065 °C) for these rocks. However, if all Fe is allocated to $Fe²⁺$ in both garnets and clinopyroxenes, the calculated temperatures for half of the rocks reported by Koons (1984) fall in the range of $604-649$ °C. Within the other half, two have relatively low calculated temperatures (475 and 384 °C) and two have relatively high calculated temperatures (1083 and 817 \degree C, Table 3). There is no clear relation between the calculated temperatures and the $Na₂O$ content of the clinopyroxenes. Koons (1984) mentioned that these garnets are usually zoned so the anomalously wide range of temperature estimates may

also be a result of non-equilibrium between garnet and clinopyroxene. A suite of eclogite from Cima di Gagnone, Switzerland (Evans et al. 1979) provides another example. The garnets in this suite are zoned, but the clinopyroxenes are not, except in one metarodingite (163-M). If the unzoned clinopyroxene compositions and the garnet rim compositions in samples other than 163-M are used to calculate the equilibrium temperature, a range of 657-716 °C is obtained from Eq. (3) (Table 3). If the garnet core compositions are used, the calculated temperatures range from 582 to 864 °C. In the metarodingite (163-M) the garnet is not zoned but the clinopyroxene is. The clinopyroxene rim composition corresponds to a calculated temperature of $1\overline{3}43^{\circ}$ C; and the core composition,

Table 3. Results of calculated equilibrium temperatures for a range of natural rocks

Sample	$\ln K_d$	$X_{\rm Ca}^{\rm Gr}$	Mg. no. (gt)	No. of samples	P (kbar) $T3(^{\circ}C)$		$TK(^{\circ}C)$ $TP(^{\circ}C)$		$T(EG)$ ^o C	$T(PN)$ ^o C $T(BM)$ ^o C
Amphibolite and granulite (Pattison and Newton 1989)										
Mica Creek Eastern Dalradian Kapuskasing Central Gneiss Belt 1.50-1.73 Adirondack	$2.13 - 2.30$ $1.63 - 2.26$ $1.25 - 2.13$ $1.12 - 2.00$	$0.29 - 0.37$ $0.24 - 0.32$ $0.18 - 0.30$ $0.17 - 0.22$ $0.19 - 0.21$	12.07-18.97 15.38-18.84 13.64 - 42.86 16.22-32.05 13.51-36.99	5 6 11 8 11	6 8 6.5 10 8	596-693 631-756 554-808 707-971 595-894	645-744 680-799 590-894 727-986 638-951	663-765 697-806 635-904 752-1027 670-957	682-779 714-821 657-917 771-1031 691-967	521-654 583-752 517–758 625-777 568-916
Eclogite (Tromsø, North Norway; Krogh 1988)										
	$1.50 - 2.26$	$0.15 - 0.44$	17–55	17	15	631–740	688–790	705–832	725-844	428-783
Eclogite (Sesia Zone of the Western Alps, Italy; Koons 1984)										
(With Fe^{3+}) (Without $Fe3+$)	$0.87 - 3.51$ $0.85 - 3.04$	$0.13 - 0.30$ $0.13 - 0.30$	12.12-28.15 12.06-28.13	8 8	15 15		316-1065 324-987	$381 - 1024$ 384-1083 387-1002 445-1037	406-1035 471-1047	$220 - 1330$ 287-1362
Eclogite (Cima di Gagnone, Ticino, Switzerland; Evans et al. 1979)										
(Gt. core) (St. rim) 163-M. Cpx rim 163-M. Cpx core	1.88-2.90 $1.82 - 2.49$ 1.34 2.02	$0.25 - 0.43$ $0.17 - 0.42$ 0.77 0.77	25.25-46.68 37.56-53.61 56.02 56.02	4 4 1 $\mathbf{1}$	25 25 25 25	582-864 657-716 1343 987	608-899 693-766 1066 798	636-929 734-779 1501 1178	655-941 757-796 1481 1176	
All Fe in garnet allocated to $Fe2+$										
163-M. Cpx rim 163-M. Cpx core	2.62 1.95	0.71 0.71	40.98 40.98	$\mathbf{1}$ $\mathbf{1}$	25 25	789 1028	670 862	937 1161	944 1159	
Diamondiferous eclogite (southern Africa; Smith et al. 1989)										
EX ₂ EX10 JJG144 XM23	1.09 1.14 1.25 1.19	0.24 0.27 0.19 0.23	59.08 55.45 51.67 52.74	1 $\mathbf{1}$ 1 $\mathbf{1}$	50 50 50 50	1237 1247 1088 1180	1196 1204 1027 1129	1161 1165 1030 1105	1172 1175 1048 1119	
Diamondiferous lherzolite (Sloan, Wyoming, USA; McCallum and Eggler 1976)										
S1L102	0.96	0.13	83.67	1	51 ^a	1091	1063	1097	1113	1056
Diamondiferous lherzolite (Finsch, South Africa; Shee et al. 1982)										
XM46 XM48	1.02 1.06	0.15 0.16	83.94 82.83	1 $\mathbf{1}$	63 ^a 57 ^a	1185 1126	1125 1103	1138 1110	1153 1126	1173 1089
Diamond inclusions (Sloan, Wyoming, USA; Otter and Gurney 1989)										
A37 A73 $1 - 10$ $1 - 15(1)$ $1 - 15(2)$	1.28 1.16 1.03 0.61 1.15	0.26 0.19 0.16 0.09 0.09	44.33 65.02 55.84 66.23 78.34	1 $\mathbf{1}$ 1 $\mathbf{1}$ 1	50 50 50 50 50	1176 1106 1171 1357 929	1113 1077 1085 1192 884	1087 1072 1097 1250 967	1101 1088 1113 1259 990	
Diamond inclusions (Koffiefontein, South Africa; Rickard et al. 1989)										
K _{2a} K8a K16a K41a K42a K43a K 56a	0.79 1.39 1.72 0.67 0.74 0.65 0.97	0.10 0.22 0.21 0.09 0.12 0.09 0.17	87.41 62.56 36.41 49.09 45.87 63.21 68.14	$\mathbf{1}$ $\mathbf{1}$ 1 1 1 1 $\mathbf{1}$	50 50 50 50 50 50 50	1141 1013 922 1361 1370 1325 1199	1093 1000 864 1142 1177 1153 1152	1152 997 875 1209 1208 1220 1147	1167 1015 897 1221 1219 1231 1160	
Diamond inclusions (Western Australia; Jaques et al. 1989)										
Argyle E4/17	$0.69 - 1.30$ 1.17	$0.28 - 0.36$ 0.22	34.86-66.11 52.40	6 $\mathbf{1}$	50 50	1174	1116	1281-16971213-16211184-1520 1192-1508 1097	1112	

T3, equation (3) of this study; TK, Krogh (1988); TP, Powell (1985); T(EG), Ellis and Green (1979); T(PN), Pattison and Newton (1989); T(BM), the two-pyroxene thermometer of Bertrand and Mercier (1985)

a Pressures are calculated from the barometer of Nickel and Green (1985). Others are assumed

Fig. 4. Comparison of temperatures calculated from Eq. (3) of this study, *solid squares,* the Ellis-Green thermometer, *empty triangles* in (a), and the Krogh formulation, *empty diamonds,* in (b) with the experimental run temperatures. This is shown by the difference between calculated temperatures from each thermometer and the experimental run temperatures. Equation (3) reproduces experimental run temperatures within 100 °C. Both Ellis-Green and Krogh formulations tend to overestimate the experimental run temperatures (in the temperature range of 900 and 1300 $^{\circ}$ C)

987 \degree C. Both are significantly higher than the estimates for other samples, which is a direct consequence of the high $Fe³⁺$ given to the garnet by Evans et al. (1979). Assuming no $Fe³⁺$ in the garnet, the calculated temperatures for the corresponding clinopyroxene rim and core compositions are 789 and $1028 \degree C$ (Table 3). It is noted that the garnet in 163-M contains exceptionally high $Fe³⁺$ compared with other samples from the same locality and that the given cation total per 12 oxygens for this garnet in 8.053.

Mineral inclusions in single diamonds may not necessarily be in mutual chemical equilibrium. If not, the calculated temperatures may be abnormal. This is best illustrated by Otter and Gurney (1989) and Rickard et al. (1989) and further elaborated by Gurney (1989). Equation (3) is employed to estimate equilibrium temperatures for eclogitic inclusions in diamonds from three localities (Sloan, USA, Otter and Gurney 1989; Koffiefontein, South Africa, Rickard et al. 1989; and Western Australia, Jaques et al. 1989). Most these calculated temperatures are within the range of 1013–1370 \degree C, with two exceptionally low values (one in the Sloan diamonds, one in the Kof**fiefontein diamonds) and one extremely high value (in the Western Australia diamonds). These exceptions are probably a result of disequilibrium between garnet and clinopyroxene.**

Most calculated temperatures from Eq. (3) are in accordance with estimates from other constraints [e.g. (1) phase relations such as the metamorphic grade and the presence of kyanite; (2) other thermometers based on different reaction schemes such as the two-pyroxene thermometer]. The improvement of the new thermometer over previously published garnet-clinopyroxene thermometers lies in the low temperature range (below 1200 ~ and for rocks with low Mg no. of garnet (mostly high grade metamorphics). For high pressure mantle rocks with temperature above 1200 $\mathrm{°C}$, the new thermometer produces similar temperature estimates to the Ellis and Green thermometer.

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