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A Report on a Biotite-Calcic Hornblende Geothermometer

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Abstract This paper presents a biotite-calcic hornblende geothermometer which was empirically calibrated based on the garnet-biotite geothermometer and the garnet-plagioclase-hornblende-quartz geobarometer, in the ranges of 560–800°C (*T*) and 0.26–1.4 GPa (*P*) using the data of metadolerite, amphibolite, metagabbro, and metapelite collected from the literature. Biotite was treated as symmetric Fe-Mg-Al^{VI}-Ti quaternary solid solution, and calcic hornblende was simplified as symmetric Fe-Mg binary solid solution. The resulting thermometer may rebuild the input garnet-biotite temperatures well within an uncertainty of ±50°C. Errors of ±0.2 GPa for input pressure, along with analytical errors of ±5% for the relevant mineral compositions, may lead to a random error of ±16°C for this thermometer, so that the thermometer is almost independent of pressure estimates. The thermometer may clearly discriminate different rocks of lower amphibolite, upper amphibolite and granulite facies on a high confidence level. It is assumed that there is a ferric iron content of 11.6% in biotite, and that the iron content in calcic hornblende may be calculated according to the method of Dale et al. (2000). This thermometer can be used for medium- to high-grade metabasites and metapelites.

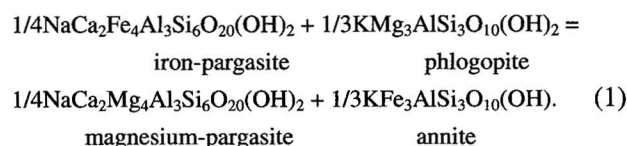
Key words: biotite-calcic hornblende geothermometer, calibration, random error, application

1 Introduction

There are a great variety of geothermometers based on the distribution of iron and magnesium atoms between coexisting ferromagnesian minerals in metamorphic rocks, e.g., the garnet-biotite geothermometer (e.g., Ferry and Spear, 1978; Hodges and Spear, 1982; Perchuk and Lavrent'eva, 1983; Holdaway, 2000), the garnet-clinopyroxene geothermometer (e.g., Ellis and Green, 1979; Berman et al., 1995), and the biotite-orthopyroxene geothermometer (e.g., Fomarev and Konilov, 1986). But, to our knowledge, the properties of iron and magnesium distribution between biotite and hornblende have not been well studied yet. In this paper, we try to calibrate an empirical biotite-calcic hornblende geothermometer, as empirically did by Dale et al. (2000) in empirically calibrating the geothermobarometers for the assemblage garnet + hornblende + plagioclase + quartz.

2 Thermodynamic Basis

The Fe-Mg distribution between biotite and hornblende can be described as



At equilibrium, the Gibbs free energy of reaction (1), ΔG , will be zero, which yields:

$$\Delta G = 0 = \Delta H^0 - T \cdot \Delta S^0 + (P-1) \Delta V^0 + RT \ln Kd(\gamma_{\text{Fe}}/\gamma_{\text{Mg}})^{\text{bio}} + RT \ln (\gamma_{\text{Mg}}/\gamma_{\text{Fe}})^{\text{hb}} \quad (2)$$

where ΔG is the Gibbs free energy of reaction (1); ΔH^0 , ΔS^0 and ΔV^0 are the standard enthalpy, entropic and volumetric variates of reaction (1) at 0.1 MPa and 298.15 K, respectively; Kd is the Fe-Mg distribution coefficient between biotite and hornblende ($Kd = X_{\text{Fe}}^{\text{bio}}/X_{\text{Mg}}^{\text{bio}} \cdot X_{\text{Mg}}^{\text{hb}}/X_{\text{Fe}}^{\text{hb}}$), and R the gas constant ($R = 8.3144 \text{ J K/mol}$). The last two items describe the non-idealities of Fe-Mg mixing in biotite and hornblende, respectively. Thermal expansion and compressibility coefficients of the mineral phases are neglected due to their negligible effects.

2.1 Hornblende activity model

Amphibole is undoubtedly one of the most complex minerals (Leake et al. 1997), so that a thorough description of the non-ideal mixing properties of amphibole requires a great amount of data of coexisting

biotite and hornblende pairs, which are not available for us at present. In the present study, hornblende is treated as a simple symmetric Fe-Mg binary solution, thus we have

$$RT \ln(\gamma_{\text{Mg}} / \gamma_{\text{Fe}})^{\text{hb}} = W_{\text{FeMg}}^{\text{hb}} (X_{\text{Fe}}^{\text{hb}} - X_{\text{Mg}}^{\text{hb}}) \quad (3)$$

where $X_{\text{Fe}}^{\text{hb}}$ and $X_{\text{Mg}}^{\text{hb}}$ are mole fractions of Fe and Mg atoms in hornblende, defined as $X_{\text{Fe}}^{\text{hb}} = \text{Fe}^{2+} / (\text{Fe}^{2+} + \text{Mg})$ and $X_{\text{Mg}}^{\text{hb}} = \text{Mg} / (\text{Fe}^{2+} + \text{Mg})$, respectively, and $W_{\text{FeMg}}^{\text{hb}}$ is the Margules parameter describing non-ideality of hornblende and is treated as a constant.

2.2 Biotite activity model

Natural biotite may be treated as a symmetric Fe-Mg-Al^{VI}-Ti quaternary solution (Indares and Martignole, 1985), with Fe, Mg, Al^{VI} and Ti mixing in the octahedral sites. The activity-composition relationship of biotite can thus be described as

$$RT \ln(\gamma_{\text{Fe}} / \gamma_{\text{Mg}})^{\text{bio}} = W_{\text{FeMg}}^{\text{bio}} (X_{\text{Mg}}^{\text{bio}} - X_{\text{Fe}}^{\text{bio}}) + \Delta W_{\text{AlVI}}^{\text{bio}} \cdot X_{\text{AlVI}}^{\text{bio}} + \Delta W_{\text{Ti}}^{\text{bio}} \cdot X_{\text{Ti}}^{\text{bio}} \quad (4)$$

where $W_{\text{FeMg}}^{\text{bio}}$, $\Delta W_{\text{AlVI}}^{\text{bio}}$, and $\Delta W_{\text{Ti}}^{\text{bio}}$ are Margules parameters, and $X_{\text{Mg}}^{\text{bio}}$, $X_{\text{Fe}}^{\text{bio}}$, $X_{\text{AlVI}}^{\text{bio}}$, and $X_{\text{Ti}}^{\text{bio}}$ are ion fractions of biotite, defined as $X_i^{\text{bio}} = i / (\text{Fe}^{2+} + \text{Mg} + \text{Al}^{\text{VI}} + \text{Ti})$, in which i stands for Fe²⁺, Mg, Al^{VI}, and Ti ions, respectively.

2.3 Regression model

Inserting Equations (3) and (4) into (2) yields

$$T = (\Delta H^0 / \Delta S^0) + (P-1) \cdot (\Delta V^0 / \Delta S^0) + (W_{\text{FeMg}}^{\text{bio}} / \Delta S^0) \cdot (X_{\text{Mg}}^{\text{bio}} - X_{\text{Fe}}^{\text{bio}}) + (\Delta W_{\text{AlVI}}^{\text{bio}} / \Delta S^0) \cdot X_{\text{AlVI}}^{\text{bio}} + (\Delta W_{\text{Ti}}^{\text{bio}} / \Delta S^0) \cdot X_{\text{Ti}}^{\text{bio}} + (W_{\text{FeMg}}^{\text{hb}} / \Delta S^0) \cdot (X_{\text{Fe}}^{\text{hb}} - X_{\text{Mg}}^{\text{hb}}) + (1 / \Delta S^0) \cdot RT \ln Kd \quad (5)$$

which is a temperature-dependent regression model of the thermometer.

3 Data Set

Data of natural rock samples, including metapelite, metagabbro, amphibolite and metadolerite, were collected from different literature (Table 1), which should meet the following conditions: (1) coexisting garnet, biotite and hornblende at equilibrium are reported; (2) electron microprobe analyses of the

minerals are listed, with at least nine major kinds of oxides; (3) temperatures computed by different thermometers are generally in agreement, which in turn reflects possible equilibrium. When chemical zoning in garnet is reported, only the rim compositions were used. All the hornblendes collected from literature are classified as calcic amphiboles (Leake et al., 1997).

An important step in utilizing the chemical compositions for geothermobarometric calculations is to correctly determine the ferric iron contents of minerals. From electron microprobe analyses of relatively simple minerals it is possible to calculate the Fe₂O₃ content with certain reliability, on conditions of charge balance and ideal formula. But, for more complex minerals with vacant cation sites and variable H₂O contents (e.g., micas), such a calculation is probably meaningless (Krogh and R  heim, 1978). As for the case of biotite, an average of 13.35% ferric iron is measured by M  ssbauer spectroscopy in 51 natural reduced biotite samples from Maine (Guidotti and Dyar, 1991), and this amount is supported by wet chemical analyses (Guidotti, 1984). Similarly, Will (1998) suggested that natural biotite contains as much as 15% total iron as ferric. In our study we assumed that all biotite contains 11.6% ferric iron, as Holdaway (2000) did. For amphibole, the situation is much more complicated due to its complex crystallography (Leake et al., 1997), and now a new procedure has been developed by Dale et al. (2000), and we adopted their method in calculating the ferric iron contents in calcic

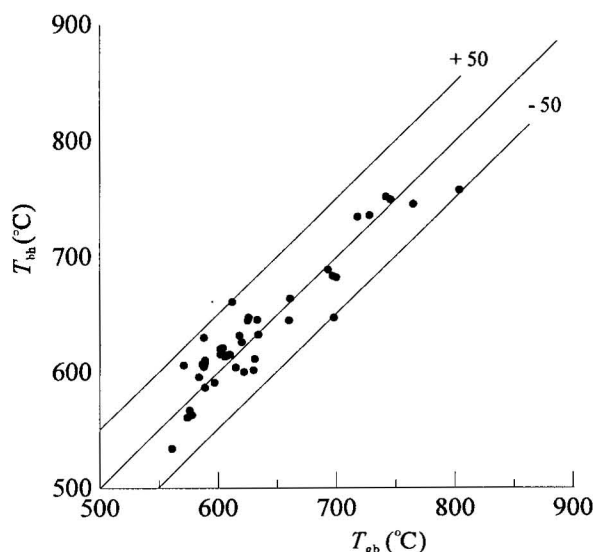


Fig. 1. Comparison of the garnet-biotite and biotite-calcic hornblende thermometers.

(The data are taken from Tables 1 and 3.)

hornblende.

Input temperatures and pressures were calculated by simultaneously applying the garnet-biotite geothermometer (Holdaway, 2000) and the garnet-plagioclase-hornblende-quartz geobarometer (Kohn and Spear, 1990), and the resulting data are listed in Table 1. The rocks listed in Table 1 include metadolerite, amphibolite, metagabbro and metapelite.

4 Calibration

The unknown constants in Equation (5) can be determined by taking an iterative regression procedure. With such a procedure, temperatures calculated using the regressed variables are the same as those used to compute the values of $RT \ln Kd$. This continues until the differences between temperatures calculated from regressed variables in successive iterations are less than 0.001°C for all samples. The results of the final regression, on a 95% confidence level, are $\Delta H^0/\Delta S^0 = 884.9 (\pm 37.5) \text{ K}$, $\Delta V^0/\Delta S^0 = -2.0 (\pm 2.0) \text{ K mol/GPa}$, $W_{\text{FeMg}}^{\text{bio}}/\Delta S^0 = -782.4 (\pm 486.2) \text{ K}$, $\Delta W_{\text{AlVI}}^{\text{bio}}/\Delta S^0 = -201.8 (\pm 204.3) \text{ K}$, $\Delta W_{\text{Ti}}^{\text{bio}}/\Delta S^0 = 1177.3 (\pm 267.6) \text{ K}$, $W_{\text{FeMg}}^{\text{hb}}/\Delta S^0 = -809.9 (\pm 428.1) \text{ K}$, and $1/\Delta S^0 = -0.043 (\pm 0.026) \text{ K mol/J}$, and a multiple correlation coefficient of $R = 0.95$, respectively.

Inserting the regressed constants into Equation (5), we have the biotite-calcic hornblende geothermometer:

$$T(\text{K}) = \frac{8849 - 2.0P(\text{GPa}) + 782.4(X_{\text{Fe}}^{\text{bio}} - X_{\text{Mg}}^{\text{bio}})}{1 + 0.043R \ln Kd} + \frac{-201.8X_{\text{AlVI}}^{\text{bio}} + 1177.3X_{\text{Ti}}^{\text{bio}} - 809.9(X_{\text{Fe}}^{\text{hb}} - X_{\text{Mg}}^{\text{hb}})}{1 + 0.043R \ln Kd} \quad (6)$$

The thermometer rebuilds the input garnet-biotite temperatures well within $\pm 50^\circ\text{C}$ (Fig. 1).

5 Error Analysis

The applicability of a geothermometer depends largely on its systematic and random errors. Due to lack of experimental data, the systematic error of this thermometer cannot be deciphered. However, through numerical modelling, we may discuss its random error. The sources of this kind of uncertainty are errors of input pressure, and errors of uncertainties of relevant mineral compositions.

The total random error of the thermometer may be

expressed as

$$\Delta T = \left(\frac{\partial T}{\partial P}\right) \cdot \Delta P + \left(\frac{\partial T}{\partial X_{\text{Fe}}^{\text{hb}}}\right) \cdot \Delta X_{\text{Fe}}^{\text{hb}} + \left(\frac{\partial T}{\partial X_{\text{Mg}}^{\text{hb}}}\right) \cdot \Delta X_{\text{Mg}}^{\text{hb}} + \left(\frac{\partial T}{\partial X_{\text{Fe}}^{\text{bio}}}\right) \cdot \Delta X_{\text{Fe}}^{\text{bio}} + \left(\frac{\partial T}{\partial X_{\text{Mg}}^{\text{bio}}}\right) \cdot \Delta X_{\text{Mg}}^{\text{bio}} + \left(\frac{\partial T}{\partial X_{\text{Al}}^{\text{bio}}}\right) \cdot \Delta X_{\text{Al}}^{\text{bio}} + \left(\frac{\partial T}{\partial X_{\text{Ti}}^{\text{bio}}}\right) \cdot \Delta X_{\text{Ti}}^{\text{bio}} \quad (7)$$

We used data from Tables 1 and 3 to calculate the random error of the thermometer, and each item is listed in Table 2. From the table we may conclude that: (1) pressure estimates have negligible effects on temperature estimates, and (2) the total random error of the thermometer is merely $\pm 16^\circ\text{C}$, thus making it a high-quality thermometer.

6 Applications of the Thermometer

Rocks listed in Table 3 were not included in calibrating the thermometer, and thus can be used to test the accuracy and precision of the present thermometer. Biotite-calcic hornblende and garnet-biotite temperatures are well within the range $\pm 35^\circ\text{C}$ (Table 3, Fig. 1).

Modena and Potenza (1989) studied chemical compositions of coexisting biotites and amphiboles in metabasites from Ivrea-Verbano Zone, Italian Western Alps, and concluded that from southeast to northwest in the 12 km-long section, the metamorphic grade gradually changed from lower amphibolite, upper amphibolite, to granulite facies, much more like a prograde sequence. But, due to lack of sufficient mineral data, they were unable to give a quantitative specification. Applying the biotite-hornblende thermometer to these rocks, we may find that the metamorphic temperatures range from 592 to 684°C for the lower amphibolite-facies rocks, 694°C for the upper amphibolite-facies rocks, and 750 – 798°C for the granulite-facies rocks (Table 3). The new thermometer thus clearly discriminated rocks of different metamorphic grades.

Stephenson (1977) studied the compositions of coexisting hornblende and biotites from amphibolite and granulite-facies gneisses from the south coast of Western Australia, and found that they are controlled by host rocks' composition, paragenesis, metamorphic grade, pressure and oxygen fugacity. Again, due to lack of sufficient mineral data, they could not give quantitative evidence. By applying the biotite-

hornblende thermometer to these rocks, we have found that the metamorphic temperatures of amphibolite-facies rocks range from 651 to 706°C (averaging 677°C), whereas for the granulite-facies rocks they range from 630 to 733°C (averaging 688°C). The difference is also distinct.

7 Discussion

It should be stated that the present thermometer is very primitive due to our simplification of the mixing properties of hornblende. We hope that warm-hearted readers send us more data concerning this topic. Anyway, such a thermometer can be used when some minerals such as garnet and/or muscovite are absent in the rocks studied.

As long as sufficient data of coexisting biotite and hornblende are available, we may thoroughly re-consider the non-idealities of calcic hornblende, including the within-site and cross-site interactions, as was done by Holland and Blundy (1994) and Dale et al. (2000).

This thermometer can be used for medium- to high-grade metabasites and metapelites.

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Table 1 Natural rock samples for calibrating the biotite-calcic hornblende geothermometer

Reference	Sample	Rock	P_{90} (MPa)	T_{gb} (°C)	T_{bh} (°C)
Glassley (1980)	91156	Metadolerite	759.6	718	734
	91154	Metadolerite	738.2	728	735
Zhao et al. (1999)	910	Amphibolite	1227.0	589	586
	1004	Amphibolite	1279.4	576	566
	1010	Amphibolite	1294.8	602	620
	1011	Amphibolite	1349.3	574	560
	1102	Amphibolite	1269.6	588	629
	1103	Amphibolite	1391.2	607	614
	1308	Amphibolite	1151.3	606	613
	1312	Amphibolite	1055.0	578	562
Pattison (1991)	Hu1 (dark)	Metagabbro	810.6	746	749
	Hu1 (margin)	Metagabbro	862.7	765	745
	P262 (margin)	Metagabbro	955.8	804	757
	HU15a (dark)	Metagabbro	858.0	742	751
Boyle & Westhead (1992)	70954	Metapelite	838.3	589	610
	69291	Metapelite	738.3	610	615
	69305	Metapelite	815.4	588	604
	70901	Metapelite	823.9	589	608
	70899	Metapelite	861.6	631	611
	70895	Metapelite	943.7	604	620
	70894	Metapelite	1048.2	622	600
	75176	Metapelite	1027.8	602	615
	75772	Metapelite	967.2	615	603
	70893	Metapelite	968.3	634	632
	70891	Metapelite	964.4	630	601
	70888	Metapelite	983.8	620	625
Kohn & Spear (1993)	K87-83N	Amphibolite	626.9	561	533
Kohn et al. (1993)	SP10-A1	Metapelite	421.1	661	663
	SP-10A2	Metapelite	262.3	618	631
Spear (1982)	73-29D	Amphibolite	616.9	597	590
	73-30N	Amphibolite	660.3	698	646
	73-30S	Amphibolite	615.3	612	660

Note: T_{gb} and P_{90} are input temperatures and pressures calculated by simultaneously applying the garnet-biotite geothermometer (Holdaway, 2000) and the garnet-plagioclase-hornblende-quartz geobarometer (Kohn and Spear, 1990), and T_{bh} denotes temperatures computed using the biotite-calcic hornblende thermometer developed in this work.

Table 2 Random error analyses of the biotite-calcic hornblende thermometer

	ΔP	ΔX_{Fe}^{hb}	ΔX_{Mg}^{hb}	ΔX_{Fe}^{bio}	ΔX_{Mg}^{bio}	ΔX_{Al}^{bio}	ΔX_{Ti}^{bio}
+ 0.2 GPa	-4°C						
- 0.2 GPa	+4°C						
+ 5%		-1°C	-1°C	-1°C	+3°C	-3°C	+2°C
- 5%		+2°C	+1°C	+1°C	-3°C	+3°C	-3°C
Total error	$\pm 16^\circ\text{C}$						

Table 3 Application of the biotite-calcic hornblende geothermometer

Reference	Sample	Rock	Grade/Facies	P_{90} (MPa)	T_{gb} (°C)	T_{bh} (°C)
Todd & engi (1997)	Ma9331	Amphibolite		708.4	626	647
	Ma9332	Amphibolite		1000.0	571	606
	Ma9341	Amphibolite		984.3	625	644
	Ma9342	Amphibolite		839.6	697	683
	Ma9345	Amphibolite		823.8	700	682
	Mag539	Amphibolite		593.2	693	688
	Mag539	Amphibolite		826.7	660	644
Epard et al. (1995)	As93126	Metapelite		600.0*	584	595
	As93221	Metapelite		600.0*	633	645
	As93254	Metapelite		600.0*	587	606
Modena & potenza (1989)	2	Metabasite	Lower amphibolite	500.0*		643
	25b	Metabasite	Lower amphibolite	500.0*		663
	26	Metabasite	Lower amphibolite	500.0*		592
	193	Metabasite	Lower amphibolite	500.0*		662
	213	Metabasite	Lower amphibolite	500.0*		679
	143	Metabasite	Lower amphibolite	500.0*		684
	165	Metabasite	Upper amphibolite	500.0*		694
	14	Metabasite	Granulite	500.0*		750
	21	Metabasite	Granulite	500.0*		798
Stephenson (1977)	1	Basic gneiss	Amphibolite	500.0*		691
	2	Basic gneiss	Amphibolite	500.0*		706
	3	Basic gneiss	Amphibolite	500.0*		701
	4	Basic gneiss	Amphibolite	500.0*		651
	5	Basic gneiss	Amphibolite	500.0*		666
	6	Basic gneiss	Amphibolite	500.0*		659
	7	Basic gneiss	Amphibolite	500.0*		667
	8	Basic gneiss	Amphibolite	500.0*		688
	9	Basic gneiss	Amphibolite	500.0*		669
	10	Basic gneiss	Amphibolite	500.0*		674
	11	Basic gneiss	Amphibolite	500.0*		694
	12	Basic gneiss	Amphibolite	500.0*		657
	13	Basic gneiss	Granulitic	500.0*		707
	14	Basic gneiss	Granulitic	500.0*		638
	15	Basic gneiss	Granulite	500.0*		661
	16	Basic gneiss	Granulite	500.0*		694
	17	Basic gneiss	Granulite	500.0*		718
	18	Basic gneiss	Granulite	500.0*		721
	19	Basic gneiss	Granulite	500.0*		719
	20	Basic gneiss	Granulite	500.0*		717
	21	Basic gneiss	Granulite	500.0*		733
	22	Basic gneiss	Granulite	500.0*		671
	23	Basic gneiss	Granulite	500.0*		649
	24	Basic gneiss	Granulite	500.0*		684
	25	Basic gneiss	Granulitic	500.0*		630

Note: Pressures with asterisks are assumed values according to the respective description in the literature. Others are similar to those in Table 1.

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