Reviews in Garnet-Biotite Geothermometer: Its Versions, Inherent Problems of Accuracy and Precision, and Perspective for Further Research*

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Abstract: This paper briefly introduces twenty-one versions of garnet-biotite Fe-Mg exchange geothermometry, points out the sources of systematic errors inherent in geothermometry. The sources lie in that: (1) ideal Fe-Mg mixing in garnet and biotite is assumed; (2) non-ideal properties of both garnet and biotite are not considered completely; (3) minor elements—Mn, Ca in garnet, and Al^{VI}, Ti, Mn in biotite are almost neglected; (4) effects of pressure on equilibrium, although little, are almost not taken into consideration, and experiments were conducted at one fixed pressure; (5) the data used for regression analysis is too scarce in quantity; (6) internal consistency of the data used for empirical calibration is not fully guaranteed; (7) the authors take energy parameters W's (Margule's parameters) independent of pressure and temperature, though the former are actually the functions of the latter items; and (8) the most important composition-activity relationship is not consistent with the mineral compositions used for calibration. Furthermore, it is believed that empirical calibrations cannot avoid the following problems: (1) effect of retrograde metamorphism; (2) inconsistent data set; (3) effect of pressure on temperature estimation; and (4) effect of Fe^{3+} calculation. Exact experimental work should be done in order to accurately calibrate the geothermometer in which the Margule's parameters should be treated as the functions of pressure, temperature, and mineral compositions.

Key words: garnet-biotite Fe-Mg exchange geothermometer; systematic error; further research perspective

Introduction

Garnet-biotite pair is particularly useful in the study of metamorphism because this mineral pair commonly coexists in equilibrium over a wide range of metamorphic grades in rocks having a wide range of bulk-chemical compositions, and the partitioning of Fe and Mg between the coexisting biotite and garnet is known to be significantly dependent on the temperature of metamorphic equilibrium. The garnet-biotite Fe-Mg exchange geothermometer has found wide application and at present twenty-one different formulations (Perchuk, 1967, 1970, 1977, 1981; Perchuk et al., 1983, 1984; Saxena, 1969; Thompson, 1976; Goldman et al., 1977; Holdaway et al., 1977; Ferry et al., 1978; Lavrent'eva et al., 1981; Hodges et al., 1982; Ganguly et al., 1984; Indares et al., 1985; Aranovich et al., 1988; Dasgupta et al., 1991; Bhattacharya et al., 1992; Kullerud, 1995) are available [Indares and Martignole (1985) and

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Bhattacharya et al. (1992) each provided two versions, see Table 1].

In fact, some precursors (Kretz, 1959, 1964; Frost, 1962; Albee, 1965; Sen, S.K. et al., 1968; Hietanen, 1969; Dahl, 1969; Lyons et al., 1970) had found the relationship between Fe-Mg distribution coefficient K_d and metamorphic grade. Nowadays we have eighteen empirical calibrations (Perchuk, 1967, 1970, 1977, 1981; Perchuk et al., 1984; Saxena, 1969; Thompson, 1976; Goldman et al., 1977; Holdaway et al., 1977; Hodges et al., 1982; Ganguly et al., 1984; Indares et al., 1985; Aranovich et al., 1988; Dasgupta et al., 1991; Bhattacharya et al., 1992; Kullerud, 1995) and three experimental formulations (Ferry et al., 1978; Lavrent'eva et al., 1981; Perchuk et al., 1983).

Previous Calibrations

Perchuk (1967) initialized the garnet-biotite geothermometer. Saxena (1969) qualitatively calibrated the geothermometer by fitting only three sets of data to a "transformed K_d vs. $1/T$" curve (Perchuk, 1970). Thompson's empirical calibration (1976) was largely based on the comparison of natural assemblages with experimental phase equilibria by adopting an ideal Fe-Mg mixing model in both garnet and biotite. Perchuk (1977) empirically re-calibrated the geothermometer by hiring the amphibole-garnet geothermometer, and this is a refined version of an early geothermometer (Perchuk, 1970). Goldman and Albee (1977) based their calibration on temperatures derived from quartz-magnetite oxygen isotope exchange thermometry, and again, assumed ideal Fe-Mg mixing in both the minerals. Holdaway and Lee (1977) studied the stability relations of Fe-Mg cordierite with K-feldspar and empirically derived a garnet-biotite geothermometer from related equilibria. Ferry and Spear's calibration (1978) was based on reversed phase transition experiments in the purely binary Fe-Mg system of synthesized minerals, by holding $Fe/(Fe+Mg) = 0.9$, assuming Fe-Mg ideal mixing in both garnet and biotite. While the determination of Fe-Mg partitioning between mineral phases below 800℃ (Medaris, 1969; Gunter, 1974) had been unsuccessful in the past, Perchuk (1981) considered the effect of Mn in the mineral pair and revised Perchuk's early calibration (1977). Hodges and Spear (1982) presented their calibrations in the narrow ranges of pressures, temperatures and mineral compositions of the Al_2SiO_5 triple point (Holdaway, 1971) by adopting a consistent set of solution models. Perchuk and Lavrent'eva (1983) also calibrated the geothermometer on the basis of reversed experiments by using synthesized minerals, assuming ideal mixing of Fe-Mg between garnet and biotite and holding $Fe/(Fe+Mg) = 0.6$. Ganguly and Saxena (1984) empirically took into account the non-ideality of Fe-Mg-Mn-Ca garnet, assuming Fe-Mg ideal mixing in biotite. Later, Perchuk and Aranovich (1984) considered F content in biotite and presented a new version. Indares and Martignole (1985) empirically completed their calibrations by considering the non-ideal mixing properties of garnet and biotite, presenting two calibrations according to different composition-activity models. Dasgupta et al. (1991) derived the relevant energy parameters for garnet-biotite Fe-Mg exchange equilibrium through statistical regression of the experimental data (Ferry and Spear, 1987) (choosing eight sets) by taking into consideration the non-ideal terms in both garnet and biotite solid solutions. Bhattacharya et al. (1992) recalculated the non-ideal mixing properties of biotite, using Ferry-Spear's (1978) and Perchuk-Lavrent'eva's experimental data (1983), and derived two new versions of garnet-biotite geothermometer—the nineteenth version. Kullerud (1995) considered the effects of Ti, Cl and Ba on garnet-biotite Fe-Mg exchange and reformulated a new thermometer, which is of qualitative significance. Among the above formulations, Perchuk and Lavrent'eva (1983) and

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Bhattacharya et al. (1992) provided the relatively precise calibrations.

Table 1. Summary of the twenty-one versions of garnet-biotite geothermometer

Author	Type of calibration	Mineral	Data set used	Garnet solid solution	Biotite solid solution	*K*d	Precision	Expression	Suitable for
Perchuk (1967)	Empirical	Natural		Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(a)		(1)	Qualitative estimation
Saxena (1969)	Empirical	Natural	3	Fe-Mg-Mn-Ca, ideal mixing	Fe-Mg, ideal mixing	(a)		(2)	Qualitative estimation
Perchuk (1970)	Empirical	Natural		Fe-Mg-Mn, ideal mixing	Fe-Mg-Mn, ideal mixing	(b)		(3)	Medium grade rocks
Thompson (1976)	Empirical	Natural	15	Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(c)		(4)	Medium- to high-grade rocks
Perchuk (1977)	Empirical	Natural	13	Fe-Mg-Mn, ideal mixing	Fe-Mg-Mn, ideal mixing	(a)	±50℃	(5)	Medium grade rocks
Goldman-Albee (1977)	Empirical	Natural	13	Fe-Mg-Mn-Ca, ideal mixing	Fe-Mg-Mn-Al-Ti, ideal mixing	(a)		(6)	Medium grade rocks
Holdaway-Lee (1977)	Empirical	Synthetic	65	Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(a)		(7)	Medium grade rocks with small amount of Mn, Ca, Ti, Al
Ferry-Spear (1978)	Experimental	Synthetic	12	Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(a)	±50℃	(8)	Medium- to high-grade rocks with small amount of Mn, Ca, Al, Ti
Lavrent'eva-Perchuk (1981)	Experimental	Synthetic	8	Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(d)	±40℃	(9)	Medium grade rocks with small amount of Mn, Ca, Ti, Al
Perchuk (1981)	Empirical	Natural	5	Fe-Mg-Mn, ideal mixing	Fe-Mg-Mn, ideal mixing	(d)	±7℃	(10)	Medium grade rocks
Hodges-Spear (1982)	Empirical	Natural	7	Fe-Mg-Mn-Ca, non-ideal mixing	Fe-Mg-Al-Ti, ideal mixing	(a)	±50℃	(11)	Medium- to low-grade rocks
Perchuk-Lavrent'eva (1983)	Experimental	Synthetic	62	Fe-Mg-Mn, ideal mixing	Fe-Mg-Mn, ideal mixing	(d)	±50℃	(12)	Medium- to high-grade rocks
Ganguly-Saxena (1984)	Empirical	Natural		Fe-Mg-Mn-Ca, non-ideal mixing	Fe-Mg, ideal mixing	(c)		(13)	Medium- to high-grade rocks
Perchuk-Lavrent'eva (1984)	Empirical	Natural	22	Fe-Mg, ideal mixing	Fe-Mg, ideal mixing	(d)		(14)	Granulite facies rocks
Indares-Martignole (1985)	Empirical	Natural	6	Fe-Mg-Mn-Ca, non-ideal mixing	Fe-Mg-Mn-Al-Ti, non-ideal mixing	(a)		(15-1)(15-2)	Granulite facies rocks
Aranovich et al. (1988)	Empirical	Natural		Fe-Mg-Ca, non-ideal mixing	Fe-Mg, ideal mixing	(d)		(16)	Medium grade rocks
Dasgupta et al. (1991)	Empirical	Synthetic	8	Fe-Mg-Mn-Ca, non-ideal mixing	Fe-Mg-Al-Ti, non-ideal mixing	(a)	±40℃	(17)	Medium grade rocks
Bhattacharya et al. (1992)	Empirical	Synthetic	78	Fe-Mg-Mn-Ca, non-ideal mixing	Fe-Mg, non-ideal mixing	(c)		(18-1)(18-2)	Medium- to high-grade rocks
Kullerud (1995)	Empirical	Natural	38	Fe-Mg, ideal mixing	Fe-Mg-Ti-Cl, non-ideal mixing	(a)		(19)	Qualitative estimation for Cl^-, Ti^-, and Ba-rich biotites

Note:

(a) $K\mathrm{d}=(\mathrm{Fe}/\mathrm{Mg})^{\mathrm{bio}}/(\mathrm{Fe}/\mathrm{Mg})^{\mathrm{grt}}$;

(b) $K\mathrm{d}=(\mathrm{Mg}/\mathrm{Mg}+\mathrm{Fe}+\mathrm{Mn})^{\mathrm{bio}}/(\mathrm{Mg}/\mathrm{Mg}+\mathrm{Fe}+\mathrm{Mn})^{\mathrm{grt}}$;

(c) $K\mathrm{d}=(\mathrm{Mg}/\mathrm{Fe})^{\mathrm{bio}}/(\mathrm{Mg}/\mathrm{Fe})^{\mathrm{grt}}$;

(d) $K\mathrm{d}=[X_{\mathrm{Mg}}/(1-X_{\mathrm{Mg}})]^{\mathrm{bio}}\times[(1-X_{\mathrm{Mg}})/X_{\mathrm{Mg}}]^{\mathrm{grt}}$;

(1) unavailable;

(2) graphic;

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(3) graphic;

(4) $T(1.56+3\ln K_d)=0.0234P+2739.62$;

(5) $T(\ln K_d+2.57)=3650$;

(6) $\ln K_d=-0.177(\pm0.010)1000\ln\alpha_M^Q-1.22(\pm0.14)X_{Mn}^{grt}-2.14(\pm0.13)X_{Ca}^{grt}+1.40(\pm0.09)X_{Fe}^{bio}+0.942(\pm0.109)X_{Ti}^{bio}-1.59(\pm0.21)X_{Al^{VI}}^{bio}-0.492(\pm0.068)$, where $1000\ln\alpha_M^Q=5.57\times10^6/T^2$;

(7) $0=6150+0.0246P+(1.987\ln K_d-3.93)T$;

(8) $0=12454-4.662T+0.057P+5.961T\ln K_d$;

(9) $30176-0.318(P-6000)=T(8.313\ln K_d+21.04)$;

(10) $T=3650/(2.57+\ln K_d)+252.25(X_{Mn}^{grt}-0.035)$;

(11) $0=12454-4.662T+0.057P+5.961T\ln K_d+(9930-4.5T)\times(X_{gros}^2+X_{alm}X_{gros}+X_{gros}X_{sps}+X_{pyr}X_{gros})$;

(12) $T(1.987\ln K_d+5.699)=7843.7+\Delta V^0(P-6000)$;

(13) $T(1.987\ln K_d-\Delta S^0)=3306+0.0188P+6056(X_{Fe}^{grt}-X_{Mg}^{grt})+4912X_{Ca}^{grt}+3466X_{Mn}^{grt}$;

(14) $T(\ln K_d+2.868-3.27X_F^{bio}/n)=3772+P(0.029-0.02X_F^{bio}/n+2469X_F^{bio}/n$, where $X_F^{bio}=F/(F+OH+Cl)$, n= atom number of (Fe+ Mg) in biotite;

(15-1) $T(4.662-5.9616\ln K_d)=12454+0.057P-1392X_{Al^{VI}}^{bio}-20301X_{Ti}^{bio}+(9900+4.5T)X_{Ca}^{grt}$;

(15-2) $T(4.662-5.9616\ln K_d)=12454+0.057P-4770X_{Al^{VI}}^{bio}-22353X_{Ti}^{bio}+9000(X_{Ca}^{grt}+X_{Mn}^{grt})$;

(16) $T(\ln K_d+2.609+1.449X_{Ca}^{grt}+0.287N_{Al}^{bio}+0.503X_F^{bio})=3873.1+2871X_{Ca}^{grt}+0.0124P-957N_{Al}^{bio}$, where $X_{Ca}^{grt}=Ca/3$, $X_{Mg}^{grt}=Mg/3$, $X_{Fe}^{grt}=Fe/3$, $X_F^{bio}=F/(F+OH)$ and N_{Al}^{bio}= Al atom number in the octahedral coordination of biotite;

(17) $T(1.85-1.987\ln K_d)=4301+3000X_{Ca}^{grt}+1300X_{Mn}^{grt}-495(X_{Mg}^{grt}-X_{Fe}^{grt})-3595X_{Al^{VI}}^{bio}-4423X_{Ti}^{bio}+1073(X_{Mg}^{bio}-X_{Fe}^{bio})+0.0246P$;

(18-1) $T[(13.138+8.3143\ln K_d+6.276X_{Ca}^{grt}(1-X_{Mn}^{grt})]=20286+0.0193P-[2080X_{Mg}^2-6350X_{Fe}^2-13807X_{Ca}(1-X_{Mn})+8540X_{Fe}X_{Mg}(1-X_{Mn})+4215X_{Ca}(X_{Mg}-X_{Fe})]^{grt}+4441(2X_{Mg}^{bio}-1)$;

(18-2) $T[6.778+8.3143\ln K_d+6.276X_{Ca}^{grt}(1-X_{Mn}^{grt})]=13538+0.0193P-[837X_{Mg}^2-10460X_{Fe}^2-13807X_{Ca}(1-X_{Mn})+19246X_{Fe}X_{Mg}(1-X_{Mn})+5649X_{Ca}(X_{Mg}-X_{Fe})]^{grt}+7972(2X_{Mg}^{bio}-1)$;

(19) $\ln K_d=-1.82+2.60X_{Ti}^{bio}+5.67X_{Cl}^{bio}$

In the above formulations, T is in K and P is in bar.

Sources of Uncertainties Inherent in the Former Formulations

But, when applying the above formulations to the same mineral pair at equilibrium, one may usually find large scatters in temperature estimation. This is because of the inherent problems of accuracy and precision of geothermometry. In fact, the usefulness of a given geothermometer or geobarometer in determining "peak" metamorphic conditions for natural assemblages depends largely on the inherent accuracy of the calibration used.

The above versions have the following problems because: (1) ideal Fe-Mg mixing in garnet and biotite is assumed (Perchuk, 1967, 1970, 1977, 1981; Perchuk et al., 1983, 1984; Saxena, 1969; Thompson, 1976; Goldman et al., 1977; Holdaway et al., 1977; Ferry et al., 1978; Lavrent'eva et al., 1981); (2) non-ideal properties of both garnet and biotite are not considered completely (Perchuk, 1967, 1970, 1977, 1981; Perchuk et al., 1983, 1984; Saxena, 1969; Thompson, 1976; Goldman et al., 1977; Holdaway et al., 1977; Ferry et al., 1978; Lavrent'eva et al., 1981); (3) minor elements—Mn, Ca in garnet, and Al^{VI}, Ti, Mn in biotite are almost neglected (Perchuk, 1967; Saxena, 1969; Thompson, 1976; Holdaway et al., 1977; Ferry et al., 1978; Lavrent'eva et al., 1981; Perchuk et al., 1984); (4) effects of pressure on equilibrium, although little, are almost not considered (Perchuk, 1967, 1970, 1977, 1981; Saxena, 1969; Goldman et al., 1977), and specifically, experiments are conducted at one fixed pressure (Ferry et al., 1978; Lavrent'eva et al., 1981; Perchuk et al., 1983); (5) the amount of the data used for regression analysis is too scarce (Perchuk, 1967, 1970, 1977, 1981; Perchuk et al., 1984; Saxena, 1969; Thompson, 1976; Goldman et al., 1977; Ferry et al., 1978; Lavrent'eva et al., 1981; Hodges et al., 1982; Ganguly et al., 1984; Indares et al., 1985; Aranovich et al., 1988; Dasgupta et al., 1991); (6) inter-

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nal consistency of the data used for empirical calibration is not fully guaranteed (Perchuk, 1977, 1981; Perchuk et al., 1984; Goldman et al., 1977; Holdaway et al., 1977; Hodges et al., 1982; Ganguly et al., 1984; Indares et al., 1985; Aranovich et al., 1988; Dasgupta et al., 1991; Bhattacharya et al., 1992; Kullerud, 1995). It should be pointed out that elemental and isotopic partitioning does not have the same temperature dependency since $\ln K_d$ is a function of $1/T$ whereas $\ln\alpha_M^Q$ is expressed as a function of $1/T^2$, Fe-Mg and $^{18}O-^{16}O$ exchange may have occurred at different rates during cooling, but such a phenomenon was neglected in one calibration (Goldman and Albee, 1977). Dasgupta et al. (1991) studied the relatively complex garnet-biotite Fe-Mg exchange equilibrium through statistical regression of the experimental data (Ferry and Spear, 1978) of pure chemical composition; (7) the authors take the energy parameters W's (Margule's parameters) independent of pressure and temperature (Hodges et al., 1982; Ganguly et al., 1984; Aranovich et al., 1988; Dasgupta et al., 1991; Bhattacharya et al., 1992), while the former are actually the functions of the latter items; and (8) the most important composition-activity relationship is not consistent with mineral compositions (Hodges et al., 1982; Ganguly et al., 1984; Indares et al., 1985; Aranovich et al., 1988; Dasgupta et al., 1991; Bhattacharya et al., 1992; Kullerud et al., 1995) used for calibration.

Some Problems that Empirical Calibration Cannot Resolve

Generally speaking, in the process of empirical calibration, the partitioning of Fe and Mg between biotite and garnet has been investigated by measuring the compositions of biotite and garnet from natural rock specimens for which the temperatures of crystallization were estimated by independent means (Perchuk, 1977; Goldman et al., 1977; Indares et al., 1985). However, empirical calibration has its own problems in the following that can be neither avoided nor resolved:

Effect of retrograde metamorphism

In empirical calibration natural garnet-biotite pairs are always used. But, garnet rims and the biotite at their contact, or garnet cores near biotite inclusions and the included biotite, have undergone Fe-Mg re-equilibration during the retrograde metamorphic process. From the new compositions, the only temperature that can be obtained corresponds to the same stage of the retrograde event. From this point of view, P and T estimates from the mineral pairs by applying other geothermobarometry to those sites of garnet and biotite do not necessarily stand for the real "peak" conditions, and thus we may misuse the P, T data as those under peak conditions.

Inconsistent data set

When empirically calibrating garnet-biotite geothermometry, researchers usually extract mineral data not only directly from natural mineral pairs, but also from literature reported by other researchers; P and T values used for calibration come not only from other geothermobarometry, but also from the available experimental results. All these would bring about an inconsistent data set, that is, the accuracy and precision of the data are not of the same magnitude.

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Effect of pressure on temperature estimation

In the empirical calibrating process, it is not convenient to choose miscellaneous P–T–X data, and pressures used often cluster to a narrow range.

Effect of Fe^{3+} calculation

One of the principal problems with any thermometer involving biotite is its Fe^{3+} content. If Fe^{3+} is removed from the total Fe, the Kd will be changed, but there will be an additional parameter to account for the non-ideal mixing of Fe^{3+}. The net effect is difficult to predict. Because any natural garnet-biotite pair contains Fe^{3+} cations in the crystals, how to properly calculate its content becomes a tiresome piece of work.

Perspective for Further Research

Experimental calibration can avoid all the shortcomings of the empirical calibrations stated above. Thanks to the development of high-P and T techniques, today we may do exact experiments over wide ranges of P and T, and mineral compositions in which the "minor elements" Mn, Ca, Al, Ti, and F are also included to refine garnet-biotite geothermometry.

Garnet and biotite have complex mixing properties due to their multicomponent isomorphs, $(Mg, Fe, Mn, Ca)_3^{VIII}(Al, Fe^{3+})_2^{VI}(Si^{IV}O_4)_3$, and $(K, Na)^{VIII}(Mg, Fe, Mn)_{3y}^{VI}(Al, Fe^{3+}, Ti)_y^{VI}[(Al, Si)_4^{IV}O]_{10}(OH, O, F, Cl)_2$, respectively, where the Roman numerals represent cation coordination numbers. Exact mineral composition—activity relations should be established for both garnet and biotite prior to experiments for the sake of accuracy and precision. We should consider the interactive properties of elements including "minor elements" in building a-X expressions in which the Margules parameters are treated as the functions of P and T, and mineral compositions. In this respect, the geothermometer will be multicomponent rather than only an Fe-Mg exchange geothermometer. This requests a large amount of data sets which can only be provided by experiments. Furthermore, Fe^{3+} contents will be very low when synthetic substances are used, and can be controlled by experiments and monitored by Mossbauer spectrometer. In the experiments, P, T, and mineral compositions must be measured accurately.

If the multicomponent a-X relations are too complex to model over wider ranges of P, T and X, we may divide the ranges to narrow ones. But this will result in many "subgeothermometers" which do not assemble the traditional ones, and simultaneously break the continuity of the P–T–X–G space.

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