



Valid garnet–biotite (GB) geothermometry and garnet–aluminum silicate–plagioclase–quartz (GASP) geobarometry in metapelitic rocks[☆]

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Abstract

At present there are many calibrations of both the garnet–biotite (GB) thermometer and the garnet–aluminum silicate–plagioclase–quartz (GASP) barometer that may confuse geologists in choosing a reliable thermometer and/or barometer. To test the accuracy of the GB thermometers we have applied the various GB thermometers to reproduce the experimental data and data from natural metapelitic rocks of various prograde sequences, inverted metamorphic zones and thermal contact aureoles. We have concluded that the four GB thermometers (Perchuk, L.L., Lavrent'eva, I.V., 1983. Experimental investigation of exchange equilibria in the system cordierite–garnet–biotite. In: Saxena, S.K. (ed.) *Kinetics and equilibrium in mineral reactions*. Springer-Verlag New York, Berlin, Heidelberg. pp. 199–239.; Kleemann, U., Reinhardt, J., 1994. Garnet–biotite thermometry revised: the effect of Al^{VI} and Ti in biotite. *European Journal of Mineralogy* 6, 925–941.; Holdaway, M.J., 2000. Application of new experimental and garnet Margules data to the garnet–biotite geothermometer. *American Mineralogist* 85, 881–892., Model 6AV; Kaneko, Y., Miyano, T., 2004. Recalibration of mutually consistent garnet–biotite and garnet–cordierite geothermometers. *Lithos* 73, 255–269. Model B) are the most valid and reliable of this kind of thermometer. More specifically, we prefer the Holdaway (Holdaway, M.J., 2000. Application of new experimental and garnet Margules data to the garnet–biotite geothermometer. *American Mineralogist* 85, 881–892.) and the Kleemann and Reinhardt (Kleemann, U., Reinhardt, J., 1994. Garnet–biotite thermometry revised: the effect of Al^{VI} and Ti in biotite. *European Journal of Mineralogy* 6, 925–941.) calibrations due to their small errors in reproducing the experimental temperatures and good accuracy in successfully discerning the systematic temperature changes of the different sequences. In addition, after applying the GASP barometer to 335 natural metapelitic samples containing one kind of aluminosilicate and 33 natural metapelitic samples containing two kinds of aluminosilicate, as well as to rocks in limited geographic areas and thermal contact aureoles, we propose that the calibrations of Holdaway (Holdaway, M.J., 2001. Recalibration of the GASP geobarometer in light of recent garnet and plagioclase activity models and versions of the garnet–biotite geothermometer. *American Mineralogist* 86, 1117–1129.) and Newton and Haselton (Newton, R.C., Haselton, H.T., 1981. Thermodynamics of the garnet–plagioclase–Al₂SiO₅–quartz geobarometer. In: Newton, R.C., Navrotsky, A., Wood, B.J. (eds.) *Thermodynamics of minerals and melts*. New York: Springer-Verlag. 131–147.); based on Kleemann and Reinhardt's (Kleemann, U., Reinhardt, J., 1994. Garnet–biotite thermometry revised: the effect of Al^{VI} and Ti in

[☆] In honour of Professor Xiuying Chen and in memory of Professor Youqin Yang. Except for Tables 1 and 2, all tables in this paper are deposited only in the Electronic appendix section. Additionally, in order to save space, only the relevant sub-figures are presented in the printed paper, and the full sub-figures can be found in the Electronic appendix section.

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biotite. *European Journal of Mineralogy* 6, 925–941.) thermometer) are the most valid GASP barometers. Other GB thermometers and GASP barometers are not recommended.

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1. Introduction

Valid geothermobarometry is a fundamentally important tool in deciphering the metamorphic conditions and understanding the evolution of the crust. Thanks to the great progress in petrological thermodynamics, now there are many geothermometers and geobarometers suitable to determine metamorphic conditions of rocks from greenschist to granulite facies and ultra-high temperatures, as well as from low- to medium- and ultra-high pressures. Either experimental or empirical or the combination of the two methods has been used in calibrating thermobarometry which results in the evolving versions of a given thermometer or a barometer.

It is well known that the validity of a thermometer or barometer is a prerequisite in the study of metamorphism, and comparative studies of thermobarometry have accompanied the development of these thermobarometers. Such comparative thermobarometric studies include comparison of the various versions of the garnet–clinopyroxene thermometry (Jahnson et al., 1983; Fu et al., 1998), comparison of the phengite barometry (Gu et al., 2001), comparison of different conventional thermobarometers (Bohlen and Essene, 1980; Ferry, 1980; Applegate and Hodges, 1994; Essene, 1989; Essene and Peacor, 1995; Rathmell et al., 1999), comparison of various oxygen barometers (Harlov, 1992), and comparison of conventional and oxygen barometers (e.g., Dahl, 1979; Moecher and Sharp, 1999).

The study of the garnet–biotite (GB) geothermometry and garnet–aluminum silicate–plagioclase–quartz (GASP) barometry has a long history, and we now have several versions of these thermobarometers. However, these diverse calibrations may be confusing to petrologists in choosing a suitable version (e.g., Soto and Platt, 1999; Mishra et al., 2006). For example, Di Vincenzo et al. (2004) used several calibrations of the GB thermometer (Hodges and Spear, 1982; Ganguly and Saxena, 1984; Perchuk and Lavrent'eva, 1983; Kleemann and Reinhardt, 1994). Such dilemmas appear elsewhere in the literature.

With the development of geothermobarometric studies, it appears necessary to do a review for a given thermometer or barometer every decade or so. Years

ago, Chipera and Perkins (1988) applied the GB thermometers available then to the “thermal anticline” of the metapelites and metawackes of the English River subprovince, Ontario, Canada. Through trend surface analysis, they concluded that the Perchuk and Lavrent'eva (1983) calibration yielded the smoothest temperature contours and concluded that this calibration was the most precise and accurate version of GB thermometers available at that time.

Now, in this paper, we summarize and compare the available GB thermometers and GASP barometers in order to recommend the best calibrations to geologists. Throughout this paper, abbreviations are used to refer to the various GB thermometers and GASP barometers, as summarized in Table 1. Pre-1978 GB thermometers and pre-1981 GASP barometers are not considered.

2. Philosophy in deciding the validity of geothermobarometry

The study of thermobarometric validity is reviewed elsewhere (e.g., Chipera and Perkins, 1988; Applegate and Hodges, 1994). Realizing that a philosophy for testing the validity of a geothermometer or a geobarometer is necessary, we list hereafter the criteria for judging the validities of GB and GASP thermobarometry.

2.1. Criteria of valid GB geothermometry

A valid garnet–biotite thermometer should: (1) successfully reproduce the experimental garnet–biotite Fe–Mg exchange temperatures of Ferry and Spear (1978), Perchuk and Lavrent'eva (1983) and Gessmann et al. (1997); (2) successfully discern the systematic change of temperatures of metapelites from different zones of prograde metamorphic sequences; (3) clearly reflect the systematic change of temperatures of different zones of inverted metamorphic sequences; and (4) meaningfully discriminate the systematic change of temperatures of different zones of thermal contact aureoles.

2.2. Criteria of valid GASP geobarometry

The dP/dT slopes of the GASP barometer in P – T space are moderately steep, because they are largely

dependent on temperatures, which are usually determined by the garnet–biotite thermometer. A valid GASP barometry should meet the following criteria: (5) samples should plot in the correct Al_2SiO_5 polymorph stability field, and the transition boundaries of the Al_2SiO_5 polymorphs should be correctly discerned; (6) in general, rocks within a very limited geographic area without post-metamorphic structural discontinuity, should show no obvious pressure diversity, if GASP barometry is applied; and (7) rocks which formed at thermodynamic equilibrium within a limited contact aureole should have been metamorphosed at constant pressure, and the GASP barometry should confirm this phenomenon.

3. Experiments in calibrating the GB thermometry and GASP barometry

3.1. Fe–Mg exchange experiments between garnet and biotite

Ferry and Spear (1978) initiated Fe–Mg exchange experiments for calibrating the GB thermometer. They used synthetic garnet and biotite and equilibrated the phases at 600–800 °C at 2.07 kbar using the graphite–methane buffer. By using large garnet/biotite ratios (98:2), their garnets were kept at nearly the compositions of starting garnets ($\text{Alm}_{90}\text{Pyr}_{10}$) for the 12 tightly bracketed runs and $\text{Alm}_{80}\text{Pyr}_{20}$ for the 4 runs that were less tightly bracketed. Their GB thermometer was derived by assuming no ferric iron present in

the products and assuming ideal mixing for both phases.

Later, Perchuk and Lavrent'eva (1983) calibrated the GB thermometer in the temperature range of 575–950 °C at 6.0 kbar using natural Ca- and Mn-poor garnets and biotites under mostly the NNO and rarely the QFM buffer. They devised a GB thermometer by assuming no ferric iron in the product phases and also assuming ideal mixing for both phases. Unfortunately, Ca and Mn contents were not reported in their 60 run products but they reported that Mn and Ca were dissolved into the fluid, and thus these components were in lower concentrations in the products. Kleemann and Reinhardt (1994) reported Ca and Mn contents of the product garnets for the 35 runs and Aranovich et al. (1988) reported Al^{VI} contents of the product biotites for these runs.

The Gessmann et al. (1997) Fe–Mg exchange experiments between garnet and biotite have been done at 2.07 kbar and 600–800 °C using large garnet/biotite ratios (95:5) under QFM and CoCoO buffers. They assumed 2% Fe^{3+} contents in the product garnets. Garnet was synthetic $\text{Alm}_{80}\text{Pyr}_{20}$ and $\text{Alm}_{70}\text{Pyr}_{30}$. Biotite was also synthetic over a wide composition range. Small amounts of Al^{VI} in their product biotites were considered to have resulted from the starting garnets. Assuming all Fe to be Fe^{2+} for the product biotite, they devised two formulations using Berman's (1990) garnet activity model and two different activity models of biotite, based on their data and data of Ferry and Spear (1978). Four runs (70/5, 70/12, 70/9, 70/12) of

Table 1
Abbreviations of the various GB thermometers and GASP barometers

Abbreviation	Reference	Abbreviation	Reference
GB thermometers			
T(78FS)	Ferry and Spear (1978)	T(90B)	Berman (1990)
T(81P)	Perchuk (1981)	T(90WG)	Williams and Grambling (1990)
T(81LP)	Lavrent'eva and Perchuk (1981)	T(Mc91)	McMullin et al. (1991)
T(82PG)	Pigage and Greenwood (1982)	T(91D)	Dasgupta et al. (1991)
T(82HS)	Hodges and Spear (1982)	T(92BA)	Bhattacharya et al. (1992) Model A
T(83PL)	Perchuk and Lavrent'eva (1983)	T(92BB)	Bhattacharya et al. (1992) Model B
T(84GS)	Ganguly and Saxena (1984)	T(94KR)	Kleemann and Reinhardt (1994)
T(85P)	Perchuk et al. (1985)	T(95K)	Kullerud (1995)
T(85IMA)	Indares and Martignole (1985) Model A	T(97GA)	Gessmann et al. (1997) Model A
T(85IMB)	Indares and Martignole (1985) Model B	T(97GB)	Gessmann et al. (1997) Model B
T(86H)	Hoinkes (1986)	T(00H)	Holdaway (2000)
T(86PA)	Perchuk and Aranovich (1986)	T(04A)	Kaneko and Miyano (2004) Model A
T(88A)	Aranovich et al. (1988)	T(04B)	Kaneko and Miyano (2004) Model B
GASP barometers			
P(81NH)	Newton and Haselton (1981)	P(88KN)	Koziol and Newton (1988)
P(82HS)	Hodges and Spear (1982)	P(90B)	Berman (1990)
P(84GS)	Ganguly and Saxena (1984)	P(01H)	Holdaway (2001)
P(85P)	Perchuk et al. (1985)		

their experiments were discarded from the regression due to possible disequilibrium.

In the following discussion, the above three experimental data sets are abbreviated as FS78, PL83 and G97, respectively.

3.2. GASP experiments

Numerous experimental determinations of the anorthite breakdown univariant reaction have been made in various P – T ranges by different authors (Hays, 1966; Hariya and Kennedy, 1968; Schmid et al., 1978; Goldsmith, 1980; Gasparik, 1984; Koziol and Newton, 1988; Wood, 1988). After inspecting the convergence of P – T lines in PT space, Koziol and Newton (1988) regarded that the most precise experiments before 1988 are the determinations of Goldsmith (1980) and Gasparik (1984). The five experimental data sets except for that of Schmid et al. (1978) have been shown to be a self-consistent set by McKenna and Hodges (1988).

It should be noted that the GASP barometer was experimentally calibrated using nearly pure anorthite and nearly pure grossular at high pressures (>12.3 kbar) and high temperatures (>900 °C), except for run 98 conducted at 600 °C (Hariya and Kennedy, 1968).

4. Natural medium- to high-grade metapelitic samples

4.1. General description

To test the validity and applicability of GB and GASP thermobarometry, we have collated additionally nearly 500 natural metapelitic samples from the literature all over the world. They cover a temperature range of 450–880 °C and a pressure range of 0.8–11.9 kbar (Tables 4–10 in Appendix A).

Selection of samples fit the following criteria: (1) there is a clear description of textural equilibrium among garnet, biotite, plagioclase, quartz and/or aluminosilicate in the literature; (2) the samples did not undergo retrogressive metamorphism; (3) there are detailed and high quality electron microprobe analyses of the minerals involved, at least SiO_2 , TiO_2 , Al_2O_3 , FeO , MnO , MgO , CaO , Na_2O and K_2O , and stoichiometry of the analyzed minerals was confirmed; and (4) if there is growth zoning in garnet, only the rim composition was used, and accordingly, only the (rim) compositions of matrix biotite and plagioclase not in contact with garnet were used. The nearly 500 natural metapelitic samples

listed in the Electronic appendix section fall in the mineral composition ranges: $X_{\text{Fe}}=0.32$ – 0.90 (mostly between 0.60 and 0.80), $X_{\text{Mg}}=0.02$ – 0.44 (mostly between 0.10 and 0.20), $X_{\text{Ca}}=0.01$ – 0.29 (mostly between 0.10 and 0.20) and $X_{\text{Mn}}=0.001$ – 0.60 (mostly between 0.10 and 0.20) in garnet; $X_{\text{Ca}}=0.01$ – 0.93 (mostly between 0.20 and 0.40), $X_{\text{Na}}=0.07$ – 0.99 (mostly between 0.60 and 0.80) and $X_{\text{K}}=0.001$ – 0.07 (mostly between 0.03 and 0.10) in plagioclase; $X_{\text{Fe}}=0.12$ – 0.58 (mostly between 0.30 and 0.45), $X_{\text{Mg}}=0.17$ – 0.73 (mostly between 0.30 and 0.50), $X_{\text{AlVI}}=0.02$ – 0.32 (mostly between 0.10 and 0.20) and $X_{\text{Ti}}=0.01$ – 0.12 (mostly between 0.03 and 0.05) in biotite.

When applying the GASP barometers, only those samples with $X_{\text{Ca}} > 0.03$ in garnet and $X_{\text{Ca}} > 0.17$ in plagioclase are considered, as suggested in Todd (1998) and Holdaway (2001).

4.2. Ferric iron contents in natural garnet and biotite

It is assumed in this study that there is 3 mol% ferric iron in metapelitic garnet, and 11.6 mol% ferric iron in biotite for ilmenite-bearing natural metapelites (Tables 4–10 in Appendix A) collected from the literature all over the world. For the magnetite-bearing natural biotite samples, 20 mol% ferric iron content in biotite is assumed. This is the same treatment as in Holdaway (2000) and Wu et al. (2004).

4.3. Calcium contents in natural garnet and plagioclase

After theoretical modelling and analyses of natural metapelites, Todd (1998) suggested that the GASP barometer should be used with great caution when the product $X_{\text{an}} \cdot X_{\text{gros}}$ is < 0.05 . Holdaway (2001) found that GASP can be used for $X_{\text{an}} > 0.17$ and $X_{\text{gros}} > 0.03$. In this paper, for the nearly 400 natural aluminosilicate-bearing metapelites (Tables 4–10, in Appendix A), the composition criteria of $X_{\text{an}} > 0.17$ and $X_{\text{gros}} > 0.03$ have been applied.

5. Valid GB geothermometry in medium- to high-grade metapelites

5.1. Brief history of GB thermometry

The development of GB thermometry has a 40-year history. Kretz (1959) first suggested that it is possible to establish distribution coefficient–temperature relationships by experimental methods. Frost (1962) had used the Fe–Mg distribution between garnet and biotite to

qualitatively determine the metamorphic grade of metamorphic rocks. Kretz (1964) pointed out that the Fe–Mg distribution between garnet and biotite may be a function of pressure and temperature, but did not give a formula. Such a Kd–T relationship was subsequently confirmed by several authors (Albee, 1965; Sen and Chakraborty, 1968; Dahl, 1969; Hietanen, 1969; Lyons and Morse, 1970; Dallmeyer, 1974). However, at this early stage the Russian geologist Perchuk's (1967) work was still neglected by scientists outside the former USSR.

Since Perchuk's (1967) empirical calibration this thermometer has repeatedly been calibrated either theoretically or experimentally or theoretically and experimentally. Saxena (1969) gave an empirical graphic presentation, at the same time Perchuk (1970) proposed another empirical calibration. Later, empirical calibrations were proposed by several scientists (e.g., Thompson, 1976; Goldman and Albee, 1977; Holdaway and Lee, 1977; Perchuk, 1977, 1981) until Ferry and Spear (1978) and Perchuk and Lavrent'eva (1983) presented their experimental thermometers. Lavrent'eva and Perchuk (1981) presented their 575–900 °C experimental data and gave a new formalism.

From the year of 1978 this thermometer has been continuously revised by petrologists such that at present this thermometer has been calibrated in at least 32 papers. Most of the authors incorporated only the FS78 data set in their calibrations (e.g., Hodges and Spear, 1982; Pigage and Greenwood, 1982; Ganguly and Saxena, 1984; Thompson, 1984; Indares and Martignole, 1985; Hoinkes, 1986; Bhattacharya and Raith, 1987; Bhattacharya et al., 1992; Berman, 1990; Williams and Grambling, 1990; Dasgupta et al., 1991; McMullin et al., 1991), while some authors reformulated the thermometer based preferentially on the PL83 data set (e.g., Perchuk and Aranovich, 1986; Perchuk et al., 1985; Aranovich et al., 1988; Kullerud, 1995). Other authors included both the FS78 and PL83 data set (e.g., Kleemann and Reinhardt, 1994; Holdaway, 2000; Holdaway et al., 1997; Kaneko and Miyano, 2004). When Gessmann et al. (1997) published their experimental data, this thermometer was recalibrated incorporating this G97 data set (e.g., Gessmann et al., 1997; Holdaway, 2000).

Most early GB thermometers consider only the ideal Fe–Mg binary biotite and/or ideal Fe–Mg binary garnet. Saxena (1969) considered Fe–Mg–Ca–Mn quaternary garnets and Fe–Mg–Al^{VI}–Ti quaternary biotites. However, the nonideal mixing properties of quaternary biotite have not been addressed until Indares and Martignole (1985) first considered the

effects of Ti and Al^{VI} in biotite as well as Ca and Mn in garnet. Almost all the later calibrations consider the nonideal, symmetric quaternary biotite and non-ideal, symmetric or even asymmetric quaternary garnet, and these successive calibrations take into account the successive improvements in Margules parameters for the various interactions in garnet and biotite, which in turn lead to generally better and better thermometers.

In the revision of the GB thermometer, most authors argued that ferric iron contents in garnet and biotite may be neglected. Williams and Grambling (1990) first considered Fe³⁺ in biotite, later Fe³⁺ was taken into account in several calibrations (Holdaway, 2000; Holdaway et al., 1997; Kaneko and Miyano, 2004).

It should be stated that McMullin et al. (1991) incorporated the FS78 data and the melting experimental data of LeBreton and Thompson (1988). Patiño Douce et al. (1993) proposed two formulations of GB thermometers based on their earlier partial melting experiments on a natural metapelitic rock. However, it is generally believed that equilibrium was not achieved in these melting experiments.

5.2. GB thermometry versus experimental GB Fe–Mg exchange temperatures

To test the validity of the various garnet–biotite thermometers, we first apply these calibrations to reproduce the experimental temperatures (FS78, PL83, G97) to see which calibrations show the smallest deviation.

We have selected the suitable experimental data from the three experimental data sets. The four Alm₈₀Pyr₂₀ runs (153, 149, 152, 150) of FS78 are not tightly reversed, as stated by the authors. However, incorporation of these four wider reversals will not significantly hurt the regression calculations. As for the experiments of PL83, only 35 of the total 60 runs were used, because only in these runs were Al^{VI} contents in product biotite reported by Aranovich et al. (1988). Gessmann et al. (1997, corrected 1998) discarded four runs (70/5, 70/12, 70/9, 70/12) from their experiments, but there are two more runs (80/1, 80/11) which show large deviations in reproducing the experimental temperatures using the G97 thermometer, so these six runs were also discarded from our consideration. Thus there remain 59 runs over the wide range of 575–950 °C and 2.07–6.00 kbar and which cover a wide spectrum of mineral compositions.

Different treatments regarding the experimental data have been made for various thermometers and these assumptions should be briefly summarized here:

- (1) Fe^{3+} content of product biotite: most GB thermometers assume all iron in the product biotite is ferrous. Williams and Grambling (1990) assumed 10% ferric iron in FS78 biotite. Gessmann et al. (1997) did not analyze the Fe^{3+} contents of their product biotite but assumed that ferric iron was negligible. Holdaway et al. (1997), Holdaway (2000) and Kaneko and Miyano (2004; Case A) assumed that the FS78 product biotite contains 7% Fe^{3+} , whereas the PL83 product biotite contains 15% Fe^{3+} . Additionally, Holdaway (2000) assumed that the G97 biotite contains 7% ferric iron.
- (2) Al^{VI} content of product biotite: Holdaway (2000) and Kaneko and Miyano (2004) assumed that the FS78 product biotite contains 0.1 Al^{VI} apfu (atoms per formula unit). Aranovich et al. (1988) measured Al^{VI} contents in the PL83 experiments and estimated Al^{VI} as half of the total ($\text{Al} - 1$), i.e., $\text{Al}^{\text{VI}} = (\text{total Al} - 1)/2$ apfu, but Holdaway et al. (1997) believed that this term should be corrected by a factor of 1.25. Later this factor was assumed to be 1.2 in Holdaway (2000) and this factor was also adopted by Kaneko and Miyano (2004). Other GB thermometers do not consider the Al^{VI} content in product biotite.
- (3) Ti content of product biotite: no Ti is present in the FS78 and G97 product biotite. The average Ti content of 0.056 apfu in biotite was assumed for PL83 experiments by Holdaway et al. (1997) and Holdaway (2000) and 0.053 by Kaneko and Miyano (2004).
- (4) Fe^{3+} content of product garnet: Holdaway et al. (1997), Holdaway (2000) and Kaneko and Miyano (2004; Case A) assumed that all product garnet of the three Fe–Mg exchange experimental data sets contains 3 mol% Fe^{3+} of the total Fe. The other GB thermometers assumed that all Fe is ferrous in the product garnet.
- (5) Ca and Mn contents of product garnet: no Ca and Mn exist in the FS78 and G97 product garnet. Kleemann and Reinhardt (1994) assumed that the final Ca and Mn contents were identical to those of the starting garnets of PL83 experiments, but Holdaway et al. (1997), Holdaway (2000) and Kaneko and Miyano (2004) assumed that these values should be corrected by a factor of 0.6 to that assumed by Kleemann and Reinhardt (1994).

Other GB thermometers do not include the Ca and Mn contents in the product garnets.

We strictly adopted the various treatments of the experimental data for each thermometer when applying it to reproduce the experimental temperatures, although different thermometers adopt different assumptions concerning the GB experiments. Such an approach most effectively reflects the real precision of each GB thermometer, because the uncertainties may be compensated for by the thermodynamic parameters derived in the calibration. The reproduced temperatures by different GB thermometers are listed in Table 3 in Appendix A, and the goodness and standard error of each of the GB thermometers in reproducing the experimental temperatures are listed in Table 2 and are depicted in Fig. 1.

From Table 2 and Fig. 1 we may easily see that, most of the GB thermometers show large deviations in reproducing the three sets of experimental temperatures. It is obvious that most of the GB thermometers show good reproducibility to the FS78 experimental temperatures, this is not surprising because these thermometers were calibrated on the basis of the FS78 data. On the other hand, much larger deviations in reproducing the PL83 experimental temperatures are ubiquitous, because most of the GB thermometers excluded the PL83 data and thus may have lost generality. This is possibly because the PL83 data might be regarded as disequilibrium data due to their method of determining equilibrium. The PL83 experiments were not reversed. The starting compositions were chosen to approach equilibrium from opposite sides of an isotherm, i.e., they used Fe-rich garnets and Mg-rich biotites as starting minerals and monitored their final compositions to see if their compositions changed significantly, and the pairs which changed the most were considered to have been at equilibrium.

If we set the standard error in reproducing the GB experimental temperatures to be less than 37 °C (Table 2, Fig. 1), we may arrive at the conclusion that the relative quality sequence as Kleemann and Reinhardt (1994) > Bhattacharya et al. (1992, Model A) \approx Perchuk and Lavrent'eva (1983) > Holdaway (2000, Model 6AV) \approx Perchuk and Aranovich (1986) \approx Perchuk (1981) \approx Kullerud (1995) > Kaneko and Miyano (2004, Model B). These eight GB thermometers are preliminarily believed to be much more reasonable than others. Still, a further test of the applicability of these thermometers should be done, using natural rocks.

However, four among the above eight GB thermometers are not considered further because: (1) The

Perchuk (1981) GB thermometer is an empirical one; (2) the Kullerud (1995) GB thermometer is based on the formalism of Perchuk and Lavrent'eva (1983) with the empirical correction of Ti^{2+} and Cl^- in natural biotite. In our collated natural metapelites almost no Cl^- in biotite is reported; (3) the Perchuk and Aranovich (1986) GB thermometer is also based on the formalism of Perchuk and Lavrent'eva (1983) and empirically considers the F^- content in biotite, but, unfortunately, F^- content in biotite in most of our collated data is not reported; and (4) the Bhattacharya et al. (1992, Model A) GB thermometer was derived by incorrectly incorporating the garnet activity model of the three-site mixing (Hackler and Wood, 1989) as a one-site mixing model. It may be inferred that the uncertainties of their GB thermometer are buried in the derived Margules parameters of biotite and the enthalpy and entropy of the GB model reaction. Thus, in the following, we compare only the four GB thermometers: Perchuk and Lavrent'eva (1983), Kleemann and Reinhardt (1994), Holdaway (2000, Model 6AV) and Kaneko and Miyano (2004, Model B).

We have noted that several thermometers actually give different formalisms of the GB thermometer (e.g., Bhattacharya et al., 1992; Holdaway, 2000; Kaneko and Miyano, 2004). However, after reproducing the experimental temperatures and applying the different formalisms to natural metapelites, we have chosen the most reliable one formula of each thermometer. We therefore preferred Model A of Bhattacharya et al. (1992), Model 6AV of Holdaway (2000) and Model B of Kaneko and Miyano (2004).

5.3. GB thermometry in prograde metamorphic sequences

5.3.1. The prograde metamorphic zones in central Menderes Massif, Turkey

An orderly prograde metamorphic zonation appears in central Menderes Massif, Turkey: the zones are garnet, staurolite, staurolite–kyanite and kyanite (Ashworth and Evirgen, 1985a,b). All of the four GB thermometers successfully discern the systematic temperature change of the prograde zones (Table 4a in Appendix A; Fig. 2a).

5.3.2. The Ryoke prograde zones, Japan

The Ryoke prograde zones in Yanai district, SW Japan, represent a low-pressure metamorphic series (Ikeda, 2004): from north to south, the metamorphic zones gradually progress through chlorite, chlorite–bi-

Table 2

Goodness and standard error (in °C) of the calculated temperatures by different garnet–biotite thermometers in reproducing the experimental temperatures (Ferry and Spear, 1978; Perchuk and Lavrent'eva, 1983; Gessmann et al., 1997)

Thermometer	R^2	Standard error	Thermometer	R^2	Standard error
T(78FS)	0.875	80.2	T(90B)	0.881	79.2
T(81LP)	0.840	37.4	T(90WG)	0.838	81.1
T(81P)	0.885	33.6	T(91D)	0.818	83.1
T(82HS)	0.869	85.4	T(91Mc)	0.804	137.5
T(82PG)	0.847	107.6	T(92BA)	0.895	31.3
T(83PL)	0.869	31.3	T(92BB)	0.821	43.7
T(84GS)	0.830	76.8	T(94KR)	0.915	29.4
T(85IMA)	0.879	73.1	T(95K)	0.825	34.1
T(85IMB)	0.876	81.2	T(97GA)	0.754	73.5
T(85P)	0.809	50.7	T(97GB)	0.779	84.2
T(86H)	0.822	140.9	T(00H)	0.898	33.2
T(86PA)	0.873	33.4	T(04A)	0.849	38.7
T(88A)	0.879	38.0	T(04B)	0.871	37.2

Thermometers in bold are considered to be reasonable.

otite, biotite, muscovite–cordierite, K-feldspar–cordierite and garnet–cordierite zones. Although a sillimanite–K-feldspar zone does not appear in the north part of this series, Ikeda (2004) suggested that the regional distribution of the three highest-grade zones is asymmetric, so the sillimanite–K-feldspar zone should be between the K-feldspar–cordierite and garnet–cordierite zones. We find that all the four GB thermometers successfully discern the systematic temperature changes of these zones (Table 4b in Appendix A; Fig. 2b), except for the lowest chlorite zone. Sample YEB5 was collected from the chlorite zone and the Mn contents of garnet (X_{Mn}) in this sample are as high as 0.60. Such a high X_{Mn} value exceeds the calibration ranges of all the GB thermometers and thus leads to erroneous temperature estimates.

5.3.3. The northern Idaho Barrovian series zones, USA

Barrovian type metamorphic zones were found in the metapelites of the Snow Peak area, northern Idaho, USA (Lang and Rice, 1985). From low to high grades, the metamorphic grade progressively changes from NE to SW and the chlorite–biotite, garnet, staurolite, transition, staurolite–kyanite and kyanite zones appear in sequence. These zones were metamorphosed in the second metamorphic event M2 and the mineral assemblages are post-kinematic. Three GB thermometers successfully discern the systematic temperature change, whereas the Perchuk and Lavrent'eva (1983) thermometer yields lower temperatures for the kyanite zone than the staurolite–kyanite zone (Table 4c in Appendix A; Fig. 2c) which seems not the case.

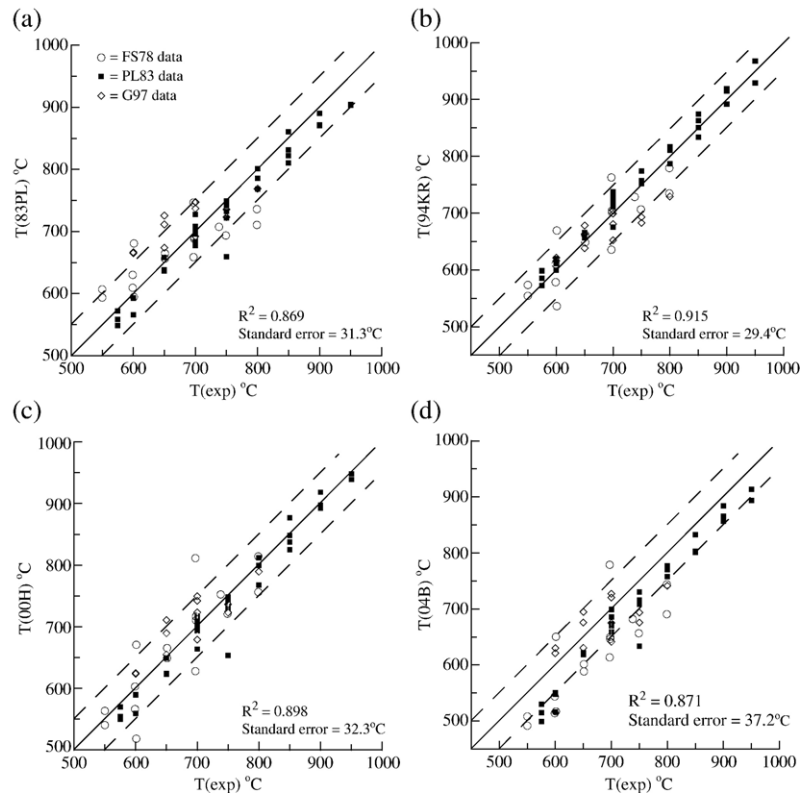


Fig. 1. Experimental vs. computed temperatures by various GB thermometers (Table 3 in Appendix A). $T(\text{exp})$ =experimental temperatures. Abbreviations of the GB thermometers are listed in Table 1. The solid line represents the 1:1 correlation, whereas the dashed lines represent the ± 50 °C deviation. Goodness and standard error of the GB thermometers in reproducing the experimental temperatures (Table 2) are labeled. Full sub-figures are deposited in the Electronic appendix section.

5.3.4. The Danba domal metamorphic terrane, China

The Danba domal metamorphic terrane within the Songpan-Garzê Orogenic Belt, northeastern Tibetan Plateau, western Sichuan, China, was developed during the regional M1b–M2 metamorphism. The dome-shaped metamorphic zonation may be classified as sericite, chlorite, muscovite, biotite, garnet, staurolite, kyanite, sillimanite, K-feldspar and migmatite zones as plutons are gradually approached (Huang et al., 2003), but contact metamorphism cannot be demonstrated. The P – T determinations of these progressive zones are listed in Table 4d in Appendix A and illustrated in Fig. 2d.

From Fig. 2d we see that the Holdaway (2000) and the Kaneko and Miyano (2004) thermometers successfully discern the systematic temperature changes of the various zones. The Perchuk and Lavrent'eva (1983) thermometer fails in discerning the temperature changes of the garnet and staurolite zones. The Kleemann and Reinhardt (1994) thermometer shows indistinguishable temperatures for the staurolite and kyanite zones.

5.4. GB thermometry in inverted metamorphic zones

5.4.1. The MCT inverted metamorphic zones in the Kishtwar-Zaskar Himalaya

A 2-km-wide, inverted metamorphic series occurs in the Main Central Thrust (MCT) zone, Kishtwar-Zaskar Himalaya (Stephenson et al., 2000). In the MCT high strain zone, garnet, staurolite and kyanite zones appear from bottom to top. Temperatures determined by the GB thermometers are summarized in Table 5a in Appendix A and illustrated in Fig. 3a. All the GB thermometers yield similar and reasonable trends of temperature change.

5.4.2. The Juneau inverted metamorphic zones

An inverted metamorphic gradient is preserved in the western metamorphic belt near Juneau, Alaska, USA. Thermal peak metamorphic conditions were seen to increase structurally upward over a distance of about 8 km (Himmelberg et al., 1991). The GB thermometers indicate similar temperatures increasing progressively from the garnet zone to the staurolite–biotite, lower

kyanite–biotite, higher kyanite–biotite and sillimanite zones (Table 5b in Appendix A; Fig. 3b). The Kleemann and Reinhardt (1994) thermometer yields unreasonably lower temperatures for the sillimanite zone than those of the higher kyanite–biotite zone, whereas the Holdaway (2000) thermometer shows undistinguishable temperatures for the garnet and staurolite–biotite zones.

5.5. GB thermometry in thermal metamorphic contact aureoles

5.5.1. Contact metamorphic zones in Eastern Rouergue, France

In the Eastern Rouergue, France, contact metamorphic zones of chlorite, biotite, garnet, staurolite and kyanite progressively developed as a result of thermal metamorphism around syntectonic granitoids (Delor et al., 1984). The GB thermometers clearly reflect the temperature changes of the garnet, staurolite and kya-

nite zones which have garnet (Table 6a in Appendix A; Fig. 4a). All GB thermometers yield similar results and the Kleemann and Reinhardt (1994) and the Holdaway (2000) thermometers yield identical temperatures.

5.5.2. Regional contact metamorphic zones in west-central Maine, USA

In west-central Maine, five regional contact metamorphic events (M1–M5) occurred in the Devonian and Carboniferous, among which the M3 and M5 are the most important two metamorphic events. Each metamorphism is closely associated with emplacement of S-type granites, such that the isograd patterns produced in the surrounding pelitic schists generally follow plutonic outlines. From north to south, grade of metamorphism varies from chlorite to sillimanite–K-feldspar–muscovite, and these zones are designated from low to high grades as Grades 3, 4, 5, 6 (M3) and Grades 6.5, 7 and 8 (M5), respectively (Holdaway et al., 1988).

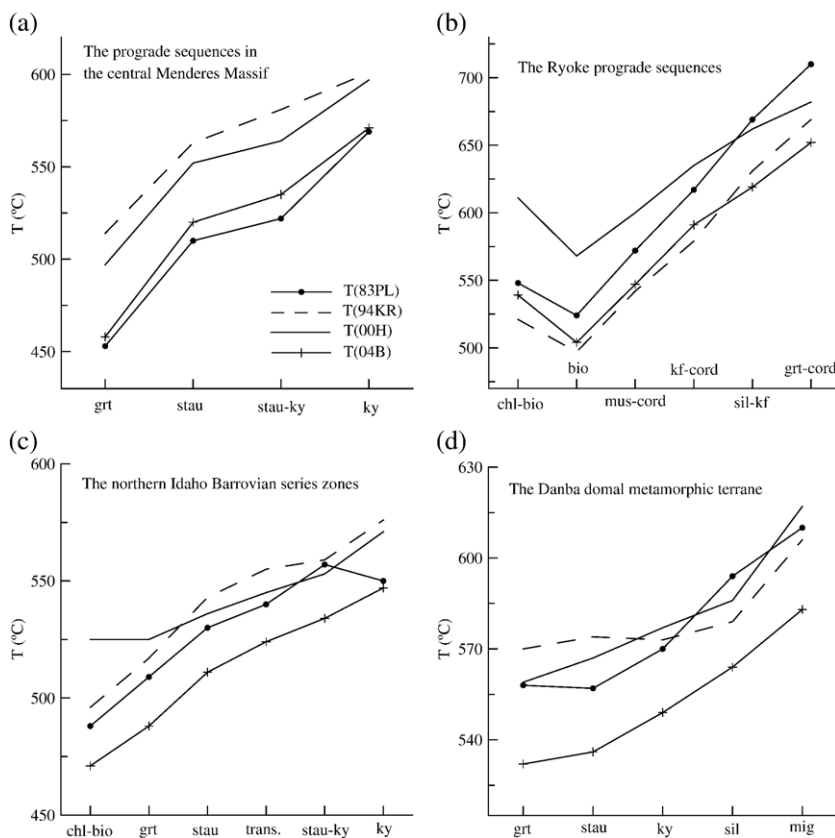


Fig. 2. GB thermometers in prograde metamorphic sequences (Table 4 in Appendix A). Solid line=Holdaway (2000), solid line with black dots=Perchuk and Lavrent'eva (1983), dashed line=Kleemann and Reinhardt (1994), and solid line with crosses=Kaneko and Miyano (2004, Model B). (a) Prograde metamorphic sequences in the central Menderes Massif, Turkey (Ashworth and Evirgen, 1985a,b); (b) Ryoke prograde sequences, Japan (Ikeda, 2004); (c) northern Idaho Barrovian series zones (Lang and Rice, 1985); and (d) Danba domal metamorphic terrane, China (Huang et al., 2003).

The four GB thermometers show perfect systematic temperature changes for all of the progressive series (Table 6b in Appendix A; Fig. 4b).

5.5.3. Contact metamorphic zones of the Coast Belt of the North American Cordillera

In the Ruby Range, Coast Belt of the North American Cordillera, contact metamorphic zones developed in the contact of the Ruby Range Batholith and other intrusive bodies (Mezger et al., 2001). The mineral isograds are parallel to the contact of the intrusive bodies and their hotter side is consistently towards the intrusive contact, showing that they result from contact metamorphism. This contact aureole is in size comparable to the regional contact metamorphic zones in west-central Maine (Holdaway et al., 1988). From far to near the contact, garnet, staurolite and cordierite zones gradually appear. The garnet zone may be of regional metamorphic origin, predating the intrusion (Mezger et al., 2001).

Except for Kaneko and Miyano (2004) and Holdaway (2000), the other two GB thermometers yield indistinguishable temperatures between the garnet and staurolite zones (Table 6c in Appendix A; Fig. 4c). This is because of the relatively higher Mn contents in garnet of these two zones.

5.6. Valid GB thermometers in medium- to high-grade metapelites

There seems no acceptable reason for choosing preferentially any one experimental data set over the others, arising from their different experimental approaches. Thus we believe that all the three sets of Fe–Mg exchange experiments (Ferry and Spear, 1978; Perchuk and Lavrent'eva, 1983; Gessmann et al., 1997) are

somewhat equally reliable, and they may act as the fundamental bases in testing the accuracy of the various GB thermometers.

From the above comparisons it may be concluded that, although the four thermometers (Perchuk and Lavrent'eva, 1983; Kleemann and Reinhardt, 1994; Holdaway, 2000; Kaneko and Miyano, 2004) yield dissimilar temperatures in reproducing the experimental temperatures, they all have relatively small standard errors (29–37 °C) in reproducing the experimental temperatures. From this regard we may primarily conclude that they are nearly equally valid.

However, when applying these GB thermometers to natural metapelites, these four thermometers generally show similar, although not completely the same, temperature changes for the prograde sequences, inverted metamorphic zones and thermal contact aureoles. When incorporating their absolute errors they can be regarded to be almost equally accurate. Still, we find that the Holdaway (2000) calibration shows relatively the most concordant temperature changes for the different zones within each metamorphic terrane.

The complete failure of many of the earlier GB calibrations results mainly from the failure to take into account, or the over-correction of, the nonideality in biotite, although some of these thermometers are still being used by recent authors.

6. Valid GASP geobarometry in aluminosilicate-bearing metapelites

6.1. Brief history of GASP barometry

Kretz (1959) and Kepezhinskas (1973) recognized the pressure sensitivity of the ratio of calcium content in coexisting garnet and plagioclase as a potential geoba-

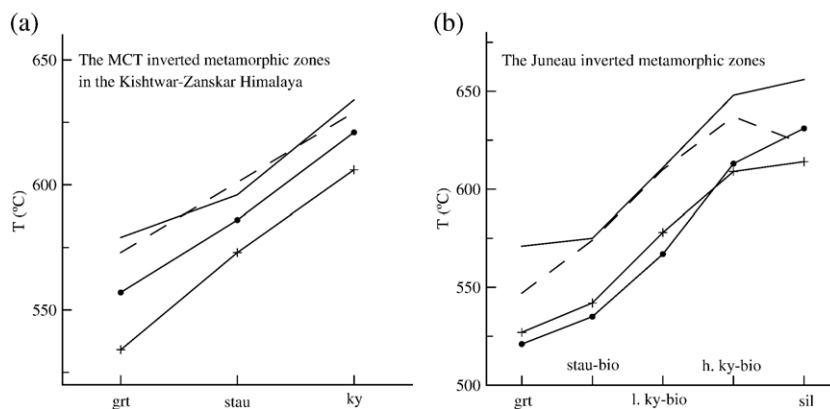


Fig. 3. GB thermometers in inverted metamorphic zones (Table 5 in Appendix A). Lines are the same as in Fig. 2. (a) MCT inverted metamorphic zones in the Kishtwar-Zaskar Himalaya (Stephenson et al., 2000); and (b) Juneau inverted metamorphic zones (Himmelberg et al., 1991).

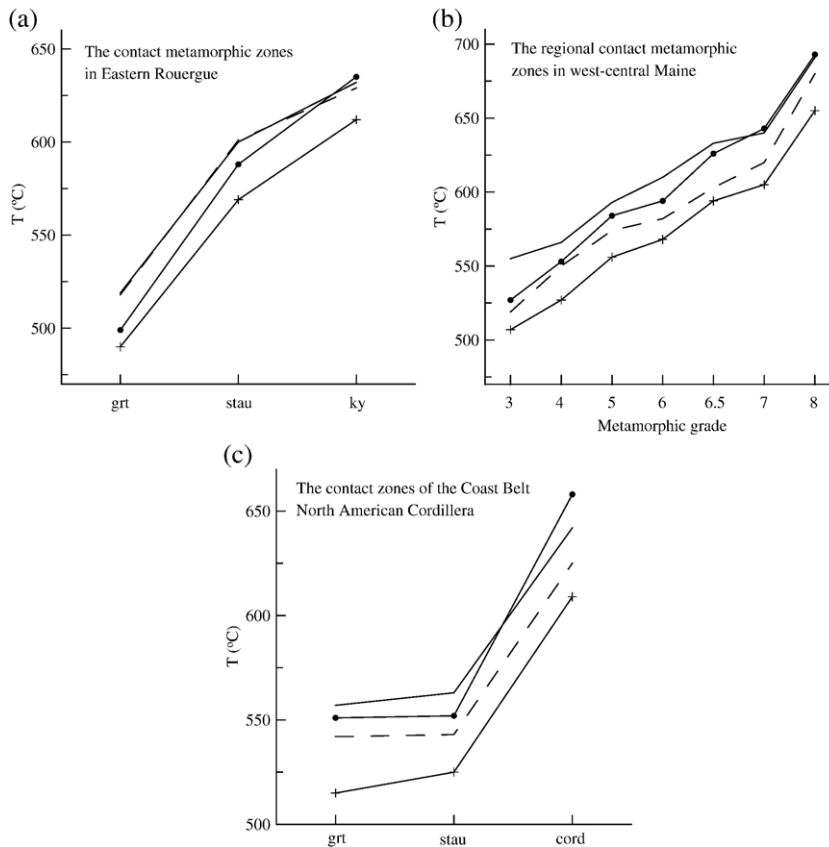


Fig. 4. GB thermometers in thermal contact metamorphic zones (Table 6 in Appendix A). Lines are the same as in Fig. 2. (a) Contact metamorphic zones in Eastern Rouergue, France (Delor et al., 1984); (b) regional contact metamorphic zones in west-central Maine, USA (Holdaway et al., 1988); and (c) contact metamorphic zones of the Coast Belt of the North American Cordillera (Mezger et al., 2001).

rometer. Ghent (1976) first devised three GASP barometer formalisms. Later, Ghent et al. (1979) derived an empirical GASP barometer.

The GASP barometer of Newton and Haselton (1981) is based on the GASP experiments of Goldsmith (1980) and the kyanite=sillimanite transition boundary of Holdaway (1971) and adopted the activity model of garnet of Newton et al. (1977) and plagioclase activity model of Newton et al. (1980).

Later, Hodges and Spear (1982) developed a new GASP barometer for sillimanite-bearing metapelites. Ganguly and Saxena (1984) proposed another GASP barometer. They assumed complete Al–Si ordering in plagioclase and the anorthite activity coefficient γ_{An} was arbitrarily fixed to be 2.0.

Perchuk et al. (1985) devised a new GASP barometer based on the GASP experimental data of Schmid et al. (1978) and Goldsmith (1980) and the Newton et al. (1980) activity model of plagioclase. Their GB thermometer and GASP barometer are thermodynamically consistent.

Koziol and Newton (1988) reversed the GASP experiments in the range 900–1250 °C and 19–28 kbar and combined an experimental bracket (650 °C, 12.8–14.14 kbar). They devised two new formulations of the GASP barometer using the same method as Newton and Haselton (1981).

Meanwhile, McKenna and Hodges (1988) re-evaluated the kyanite-forming GASP experimental data available excluding that of Schmid et al. (1978) using statistical treatment and gave a new P – T equation of this phase transition reaction but did not give any suggestions of activity models in applying this barometer.

Berman (1990) derived new quaternary solid solution models of garnet from the available experiments and gave new GASP barometry using the thermodynamic data set of Berman (1988) and the plagioclase activity model of Fuhrman and Lindsley (1988).

Holdaway (2001) coupled the existing GASP experimental data and 76 kyanite-and/or sillimanite-bearing metapelitic samples from 11 localities all over the world

to constrain the standard enthalpy and entropy values of grossular. The end-member GASP line was constrained to pass through the best-known point on the experimental calibrations. With the four garnet activity models, two plagioclase activity models and proper grossular properties derived in his calibration, Holdaway (2001) yielded eight versions of the GASP barometer among which the calibration using the averaged garnet activity model and the Fuhrman and Lindsley (1988) plagioclase activity model was preferred. Meanwhile, he applied this GASP barometry to the additional 63 samples from the Swiss Lepontine Alps and thus confirmed its validity. This GASP barometry is thermodynamically consistent with Holdaway's (2000) GB thermometry.

It should be noted that in the GASP barometer there are two different competing plagioclase activity models, and the model of Fuhrman and Lindsley (1988) does better than that of Elkins and Grove (1990).

In this paper, the pre-1981 GASP barometers are not considered. In the following, GASP pressure determination is based on its accompanying GB thermometer in order to maintain internal thermodynamic consistency. However, no GB thermometer is suggested by the Newton and Haselton (1981) or the Koziol and Newton (1988) barometer. For these cases we choose the appropriate thermometer(s) which yield the most reasonable P – T plots when combined with any one of these GASP barometers. We found that the Kleemann and Reinhardt (1994) GB thermometer is most suitable for the Newton and Haselton (1981) and Koziol and Newton (1988) barometers. In this paper, natural data used to test the applicability of the various GASP barometers are collated in Tables 4–10 in Appendix A and the quality of these GASP barometers is comparable in Table 11 in Appendix A.

6.2. GASP barometry versus aluminosilicate phase relations

6.2.1. Metapelites containing one kind of aluminosilicate polymorph

Table 7 in Appendix A lists 335 metapelitic samples containing one kind of aluminosilicate polymorphs, among which 22 samples contain only andalusite, 180 samples contain only sillimanite and the other 133 samples contain only kyanite. The temperature and pressure ranges of these rocks are 450–880 °C and 0.8–11.9 kbar, respectively. The P – T plots are depicted in Fig. 5. Also labeled in the right corner of each sub-figure are the percentage and standard deviation of

pressures from the phase boundaries of the samples which have been misplaced, for example, a kyanite-bearing sample has been plotted in the sillimanite stability field. Table 11a in Appendix A also lists the percentage and average pressure deviation from the phase boundaries of the misplaced one aluminosilicate-bearing metapelites.

From Fig. 5 and Table 11a in Appendix A we find that the Hodges and Spear (1982), the Ganguly and Saxena (1984) and the Berman (1990) GB thermobarometers yield too many geologically unreasonable high temperatures for the sillimanite-bearing samples, although few of these samples violate the kyanite=sillimanite phase transition boundary. The Ganguly and Saxena (1984) barometer, however, yields obviously low pressures, especially many sillimanite-bearing metapelites have been misplaced in the andalusite stability field.

Other GASP barometers seem to give similar results. The order of validity may be tentatively concluded to be Holdaway (2001) \approx Newton and Haselton (1981) > Perchuk et al. (1985) \approx Koziol and Newton (1988) > Berman (1990) > Hodges and Spear (1982) \approx Ganguly and Saxena (1984).

6.2.2. Metapelites containing two kinds of aluminosilicate polymorphs

A much more sensitive test than placing a given sample within one stability field can be applied to rocks that contain two or more aluminosilicate polymorphs (Berman, 1990).

Table 8 in Appendix A lists 33 metapelitic samples which contain two kinds of aluminosilicate polymorphs, among which 10 contain andalusite and sillimanite and the other 23 contain kyanite and sillimanite. They cover a temperature range of 520–740 °C and a pressure range of 2.0–8.7 kbar, respectively. The P – T determinations are depicted in Fig. 6. Also labeled in each sub-figure are the standard deviations of the pressure determinations from the phase transition boundary lines And=Sil and Sil=Ky, respectively. Table 11a in Appendix A also lists the average deviation of pressures from the phase transition boundaries for the two aluminosilicate-bearing metapelites.

From Fig. 6 and Table 11a in Appendix A we see that the relative accuracy order may be determined as Newton and Haselton (1981) \approx Perchuk et al. (1985) \approx Holdaway (2001) > Berman (1990) > Koziol and Newton (1988) > Ganguly and Saxena (1984) > Hodges and Spear (1982).

However, options differ concerning the aluminosilicates present in metapelites. In most metamorphic

occurrences we can make a good case that the higher temperature form replaces the lower temperature form, and we are seeing an indication of progressive metamorphism. For example, when kyanite and sillimanite occur together, sillimanite usually replaces kyanite. We know that there was some degree of thermal overstepping in the progressive formation of sillimanite from kyanite and sillimanite from andalusite. In some cases, especially andalusite to sillimanite, overstepping can lead to andalusite samples lying in the sillimanite field. This is probably due to the very small free energy of this transition. The same observation applies to the occurrences of any two aluminum silicates together. In most cases, we can be reasonably sure that when two occur together, the assemblage has surpassed, but does not lie on, the appropriate equilibrium boundary (Holdaway, 2005, personal communication). For this reason, two aluminosilicate-bearing metapelites may not provide unambiguous criteria.

6.3. GASP barometry in limited geographic areas

6.3.1. The Baltimore metapelites, USA

Lang (1991) collected thirteen samples from the two metamorphic mineral assemblages, a staurolite assemblage (staurolite + garnet + biotite + quartz + muscovite + plagioclase + ilmenite + magnetite), and a staurolite–kyanite assemblage (kyanite + staurolite + garnet + biotite + quartz + muscovite + plagioclase + ilmenite + rutile + pyrrhotite). These rocks are randomly distributed along a 0.5-km-long exposure at the Hunt Valley Mall, north of Baltimore, Maryland, USA. Among these samples, six rocks contain kyanite and the

rest are aluminosilicate-free. P – T determinations are listed in Table 9 in Appendix A and Fig. 7.

Fig. 7 also presents the standard deviation of these nearby kyanite-bearing rocks. From Fig. 7 and Table 11b in Appendix A we see that the relative accuracy order may be determined as Perchuk et al. (1985) \approx Holdaway (2001) \approx Newton and Haselton (1981) $>$ Koziol and Newton (1988) $>$ Berman (1990) $>$ Ganguly and Saxena (1984).

6.3.2. The File Lake metapelites, Canada

Six metapelitic rocks within 800 m of each other that straddle the sillimanite–biotite isograd in the File Lake area, Manitoba, Canada were collected by Gordon et al. (1991). Among these, one sample (20–27) contains sillimanite but no garnet, two samples (2026-2 and 2040-2) contain sillimanite and garnet, and the other three samples (1001, 2025A and 2038) contain garnet but no aluminosilicate. P – T determinations for samples 2026-2 and 2040-2 are also listed in Table 9 in Appendix A and Fig. 7.

From Fig. 7 and Table 11b in Appendix A we find that, the Newton and Haselton (1981), the Perchuk et al. (1985) and the Koziol and Newton (1988) thermobarometers do not misplace these two sillimanite-bearing samples into andalusite or kyanite stability fields. The Hodges and Spear (1982), the Berman (1990) and the Holdaway (2000, 2001) thermobarometers put sample 2026-2 into the andalusite stability field but slightly deviate from the And=Sil transition boundary. The Ganguly and Saxena (1984) thermobarometer completely misplaces these samples in the andalusite field.

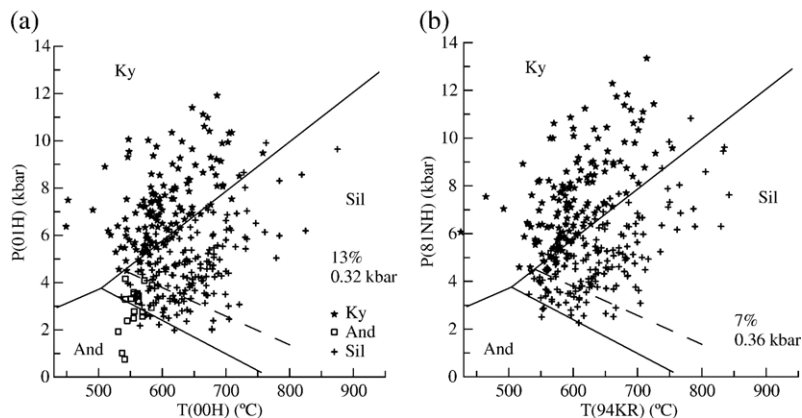


Fig. 5. P – T plots of the 335 metapelitic samples containing only one kind of aluminosilicate polymorph (Table 7 in Appendix A). The solid lines are from Holdaway and Mukhopadhyay (1993) and the dashed line is from Pattison (1992). \square = samples containing only andalusite. $+$ = samples containing only sillimanite. \star = samples containing only kyanite. Percentage and averaged pressure deviation from the phase boundary lines of the misplaced samples are labeled in the right area in each sub-figure. Full sub-figures are deposited in the Electronic appendix section.

6.4. GASP barometry in thermal metamorphic contact aureoles

6.4.1. Regional contact metamorphic zones in west-central Maine, USA

The west-central Maine regional contact metamorphic zones cover a large area. Although Holdaway et al. (1988) suggested that M3 metamorphism occurred at 3.1 kbar and M5 at 3.8 kbar, the various GASP barometers show no obvious pressure difference more than 0.5 kbar between M3 and M5 and they can be regarded as identical, within error. The P – T determinations are depicted in Table 10a in Appendix A and Fig. 8. Also depicted in Fig. 8 is the standard deviation of pressure determination for each barometer.

Fig. 8 presents the goodness of the convergence of the pressure clusters of these metapelites in the contact aureoles. From Fig. 8 and Table 11b in Appendix A we find a relative accuracy sequence of the GASP barometers as Holdaway (2001) \approx Perchuk et al. (1985) > Newton and Haselton (1981) > Hodges and Spear (1982) > Berman (1990) > Ganguly and Saxena (1984) > Koziol and Newton (1988).

6.4.2. Mafic complex contact aureole, Ivrea Zone, northern Italy

In northern Italy, near the Italian–Swiss border, is exposed a Late Carboniferous–Early Permian mafic intrusive complex. Emplacement of a major part of the mafic complex occurred during decompression from ambient pressures at the thermal maximum during the regional granulite facies episode. Final emplace-

ment caused anatexis and metamorphism only within a narrow (2–3 km) aureole in the proximal supracrustal rocks. These events overprint the regional amphibolite to granulite facies prograde metamorphic zonation (Barboza and Bergantz, 2000).

Most of the GASP barometers for the metapelites collected from within the narrow contact aureole (Table 10b in Appendix A; Fig. 9) show that, except for sample 80596-3, the contact metamorphism occurred at nearly constant pressure. With the exception of the Ganguly and Saxena (1984) barometer that yields unrealistic lower pressure estimates violating the sillimanite stability field, the other barometers generally do not. Comparing the standard deviation of pressure estimates, the relative quality order of the GASP barometers is evaluated as Perchuk et al. (1985) \approx Holdaway (2001) > Newton and Haselton (1981) > Koziol and Newton (1988) > Hodges and Spear (1982) > Berman (1990) > Ganguly and Saxena (1984), as depicted in Fig. 9 and Table 11b in Appendix A.

6.5. Valid GASP barometers in aluminosilicate-bearing metapelites

The accuracy order of the GASP barometers is not so easily determined because the GASP barometers are generally based on the GB thermometers. Based on the above comparative studies we may arrive at the conclusion that the Newton and Haselton (1981), the Perchuk et al. (1985) and the Holdaway (2001) barometers have almost the same accuracy and thus are nearly equally valid.

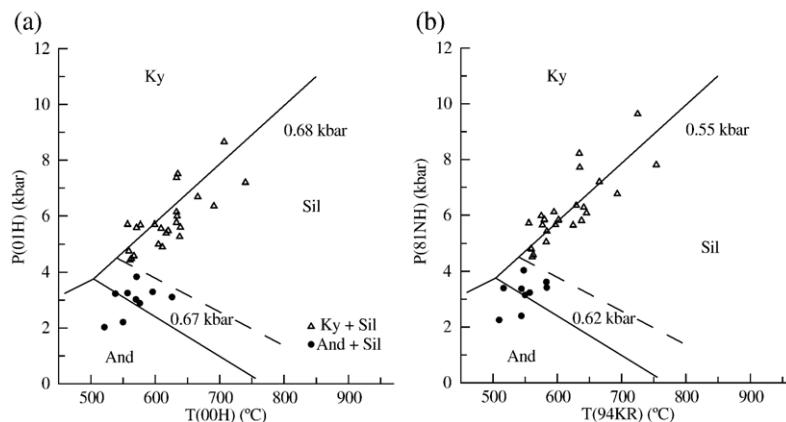


Fig. 6. P – T plots of the 33 metapelites containing two kinds of aluminosilicate polymorphs (Table 8 in Appendix A). Lines are the same as in Fig. 5. ● = samples containing andalusite and sillimanite. ▲ = samples containing kyanite and sillimanite. Standard deviations of pressures from the phase transition boundaries are labeled adjacent to the respective lines. Full sub-figures are deposited in the Electronic appendix section.

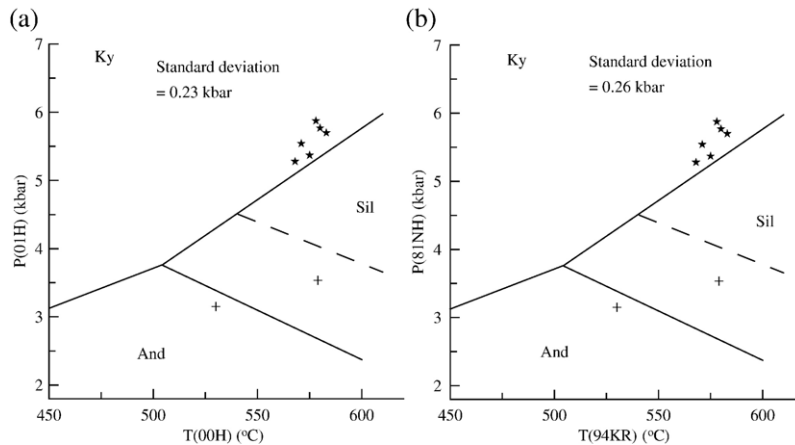


Fig. 7. P – T plots in limited geographic areas (Table 9 in Appendix A). Lines are the same as in Fig. 5. ★=closely distributed kyanite-bearing metapelites from Hunt Valley Mall, north of Baltimore, Maryland, USA (Lang, 1991). +=closely distributed sillimanite-bearing metapelites from the File Lake area, Manitoba, Canada (Gordon et al., 1991). Standard pressure deviations of the Hunt Valley Mall kyanite-bearing metapelites are depicted. Full sub-figures are deposited in the Electronic appendix section.

Unfortunately, the Perchuk et al. (1985) GB thermometer shows some failures in discerning the systematic temperature changes of the prograde sequences (Ashworth and Evirgen, 1985a,b; Lang and Rice, 1985; Huang et al., 2003), the inverted metamorphic sequence (Himmelberg et al., 1991) and the thermal contact aureole (Novak and Holdaway, 1981), thus their GB thermometer and GASP barometer cannot be accepted as accurate thermobarometers, although they seem to be reasonable in plotting the natural metapelites in the correct areas (Figs. 5–9).

Thus we may conclude that the most accurate two GASP barometers are those of Holdaway (2001) and Newton and Haselton (1981; based on Kleemann and Reinhardt (1994) thermometer).

7. Discussion

The GB thermometry and the GASP barometry have long histories. In general, the most recent calibrations do better in accounting for the nonidealities in garnet, biotite and plagioclase. This is because the successive calibrations consider much more reasonably the effects of the minor components such as Al^{VI} and Ti in biotite and Ca and Mn in garnet for thermobarometry.

After applying these thermometers to both experimental data and natural metapelites, however, we find that the four GB thermometers (Perchuk and Lavrent'eva, 1983; Kleemann and Reinhardt, 1994; Holdaway, 2000, Model 6AV; Kaneko and Miyano, 2004, Model B) are the most valid and reliable of this type.

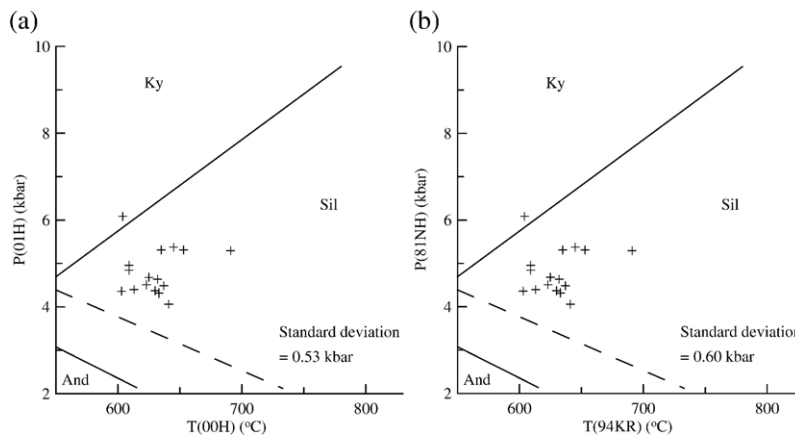


Fig. 8. P – T plots of the regional contact metamorphic zones in west-central Maine, USA (Table 10a in Appendix A; Holdaway et al., 1988). Lines are the same as in Fig. 5. Standard pressure deviations are depicted. Full sub-figures are deposited in the Electronic appendix section.

More specifically, we prefer the Holdaway (2000) and the Kleemann and Reinhardt (1994) calibrations due to their small errors in reproducing the experimental temperatures and good accuracy in successfully discerning the systematic temperature changes of the different sequences. We have also noticed that the Holdaway (2000) thermometer usually yields 5–20 °C higher temperatures than other GB thermometers.

Perhaps the most frequently cited GB thermometers in the literature are the Ferry and Spear (1978), Hodges and Spear (1982) and Ganguly and Saxena (1984) calibrations. Unfortunately, these three calibrations have severe difficulties that make them unreliable thermometers. The Ferry and Spear (1978) calibration was based on 12 experimental runs of extremely iron-rich garnets ($X_{\text{Fe}}=0.90$) over a small temperature range (600–800 °C), and ideal mixing in both garnet and biotite was assumed. Thus, when we apply this thermometer to the PL83 and G97 experimental data, large deviations appear. The Hodges and Spear (1982) and Ganguly and Spear (1984) thermometers are also based only on the FS78 data. Furthermore, these two thermometers have yielded geologically unreasonable high temperatures for many natural metapelites (Fig. 5).

The Hodges and Spear (1982) GB thermometer and GASP barometer were developed on the basis of their seven sillimanite- and sillimanite–andalusite-bearing metapelitic samples from Mt. Moosilauke, New Hampshire, USA which was believed to be very near the aluminosilicate triple point (501 ± 20 °C, 3.76 ± 0.3 kbar) of Holdaway (1971). In their calibration, ideal mixing of Fe–Mg in biotite was assumed. Their data span narrow compositional ranges of minerals, which may not adequately and accurately describe the mixing properties of garnet and biotite, thus it seems that these

thermobarometers lose generality in depicting the mixing properties of these mineral phases.

Ganguly and Saxena (1984) derived an asymmetric activity model for garnet from experimental and natural data that yielded a new GB thermometer incorporating only the FS78 data. Thus, their thermometer shows similar deviations to that of Hodges and Spear (1982). Jackson (1989) has stated that there is an error in Ganguly and Saxena's (1984) garnet activity model.

We have also noted that the very recent calibration, the Kaneko and Miyano (2004) thermometer, appears to be not as good as several earlier calibrations in reproducing the experimental temperatures. This may have resulted from their use of mixed Margules parameters for garnet. They adopted the garnet Margules parameters of the averaged values of Berman and Aranovich (1996), Ganguly et al. (1996) and Mukhopadhyay et al. (1997) for the binaries except those concerning Mn, i.e., asymmetric binary parameters of Fe–Mn (Ganguly et al., 1996), Ca–Mn (Ganguly and Cheng, 1994) and Mn–Mg (Holdaway et al., 1997). The Margules parameters of the binaries are not thermodynamically consistent.

The GASP barometer is somewhat difficult to evaluate. However, after various comparisons, we conclude that the Holdaway (2001) and the Newton and Haselton (1981) calibrations are the most reliable and thus valid GASP barometers.

The GB thermometers were calibrated experimentally using nearly Al^{VI} - and Ti-absent biotites and nearly Ca- and Mn-absent garnets, which makes it difficult to accurately describe the mixing properties of these minerals, and thus limits the application of this thermometer to assemblages with high- Al^{VI} and -Ti biotites and/or high-Ca and -Mn garnets. For example, for the high-Mn garnets of the Yanai district, SW Japan (Ikeda,

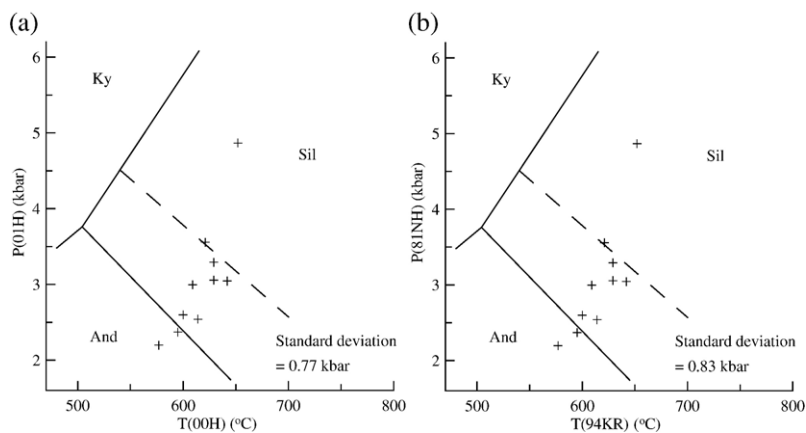


Fig. 9. P - T plots of the mafic complex contact aureole, Ivrea Zone, northern Italy (Table 10b in Appendix A; Barboza and Bergantz, 2000) Lines are the same as in Fig. 5. Standard pressure deviations are depicted. Full sub-figures are deposited in the Electronic appendix section.

2004), X_{Mn} in garnet may exceed 30%. Under such circumstances none of the available GB thermometers is applicable.

The GASP barometers were experimentally calibrated using nearly pure anorthite and nearly pure grossular at high pressures (>12.3 kbar) and high temperatures (>900 °C), except for run 98 conducted at 600 °C (Hariya and Kennedy, 1968). When we extrapolate the univariant GASP reaction lines to lower pressures, larger errors may exist, although at present such inherent errors are hard to evaluate. Moreover, applying these pure-phase barometers to natural minerals of complex chemical systems, may introduce some systematic uncertainties which are hard to evaluate at present.

8. Conclusion

On the basis of applying the various versions of the GB thermometers to experimental data, as well as to metapelitic rocks of various prograde sequences, inverted metamorphic zones and thermal contact aureoles, we conclude that the four GB thermometers (Perchuk and Lavrent'eva, 1983; Kleemann and Reinhardt, 1994; Holdaway, 2000; Kaneko and Miyano, 2004) are nearly equally valid. Here we prefer the Holdaway (2000) and the Kleemann and Reinhardt (1994) calibrations because they not only do better in reproducing the experimental temperatures but also successfully reflect the systematic temperature changes of the typical metamorphic sequences.

After applying the GASP barometers to a large number of natural metapelites containing one kind or two kinds of aluminosilicate, as well as rocks in limited geographic areas and thermal contact aureoles, it is proposed that the calibrations of Holdaway (2001) and Newton and Haselton (1981; based on Kleemann and Reinhardt (1994) thermometer) are the most reliable and valid GASP barometers.

It is necessary to do further experimental work to calibrate both the GB thermometer and GASP barometer at low to high temperatures and at low to high pressures, using garnet, biotite and plagioclase with chemical compositions comparable to natural minerals. However, many experiments have already failed to achieve reversal data at low temperatures because of slow kinetics and at high temperatures because of the loss of degeneracy. It is equally important to improve our knowledge of the various chemical interactions within minerals such as biotite, garnet and plagioclase, so that we can improve the activity models for these minerals.

It is also suggested that the ongoing papers in calibrating geothermobarometry be constructed in a way

that is friendly to readers, that is, the formalisms be explicitly expressed (e.g., Perchuk et al., 1985; Wu et al., 2004). A writing style in which no clear expression of a thermometer or a barometer is presented should be avoided in order to provide an easy way for geologists, especially field geologists, to use the geothermobarometers conveniently.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.lithos.2005.09.002.

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