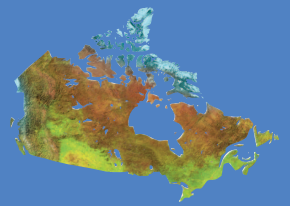




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A reaction grid for pelitic and mafic rocks

E. Froese

Geological Survey of Canada

Current Research 2010-13

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A reaction grid for pelitic and mafic rocks

E. Froese

Froese, E., 2010. A reaction grid for pelitic and mafic rocks; Geological Survey of Canada, Current Research 2010-13, 11 p.

Abstract: The compositions of minerals in pelitic and mafic rocks can be approximated in the chemical system SiO_2 , TiO_2 , Al_2O_3 , Fe_2O_3 , FeO , MgO , CaO , Na_2O , K_2O , H_2O . By rearranging the components and restricting the consideration to mineral assemblages including quartz, plagioclase of constant composition, magnetite, and ilmenite, the remaining minerals are represented in the triangular prism AKFM, with

$$K = \text{K}_2\text{O} \cdot \text{Al}_2\text{O}_3$$

$$F = \text{FeO} - (\text{Fe}_2\text{O}_3 + \text{TiO}_2)$$

$$M = \text{MgO}$$

as the base and

$$A = \text{Al}_2\text{O}_3 - (\text{CaO} + \text{Na}_2\text{O})$$

as the vertical axis. The partial Gibbs energy of H_2O is assumed to be close to that of steam at lower grades but would be reduced at higher grades.

Reactions in pelitic and mafic rocks can be combined into one grid if it is assumed that the plagioclase composition remains constant or varies smoothly with pressure and temperature. Similarly, a consistent grid would require the partial Gibbs energy of H_2O to be a smooth function of pressure and temperature.

Résumé : La composition des minéraux des roches pélitiques et mafiques peut être représentée de façon approximative dans le système chimique SiO_2 , TiO_2 , Al_2O_3 , Fe_2O_3 , FeO , MgO , CaO , Na_2O , K_2O , H_2O . En modifiant l'arrangement des composants et en limitant l'examen aux associations de minéraux comprenant du quartz, du plagioclase de composition constante, de la magnétite et de l'ilménite, les minéraux restants sont représentés dans le prisme triangulaire AKFM, dont

$$K = \text{K}_2\text{O} \cdot \text{Al}_2\text{O}_3$$

$$F = \text{FeO} - (\text{Fe}_2\text{O}_3 + \text{TiO}_2)$$

$$M = \text{MgO}$$

constituent la base et

$$A = \text{Al}_2\text{O}_3 - (\text{CaO} + \text{Na}_2\text{O})$$

représente l'axe vertical. On suppose que l'énergie partielle de Gibbs de H_2O est proche de celle de la vapeur pour les grades de métamorphisme plus faibles, mais elle serait réduite à des grades plus élevés.

Les réactions dans les roches pélitiques et mafiques peuvent être combinées en une seule grille pétrogénétique si l'on présume que la composition du plagioclase demeure constante ou varie de façon régulière en fonction de la pression et de la température. Pour s'en tenir à une même grille, il faudrait aussi que l'énergie partielle de Gibbs de H_2O soit une fonction continue de la pression et de la température.

INTRODUCTION

In many metamorphic rocks, it is possible to deduce compatible mineral assemblages by textural observation. The inference that textural equilibrium reflects chemical equilibrium provides the basis for an attempt to apply the principles of heterogeneous equilibrium to compatible mineral assemblages.

A compatible mineral assemblage has a limited stability range in terms of pressure and temperature. If several mineral assemblages share the same pressure-temperature condition, as might be inferred from their field occurrence, this pressure-temperature condition lies within the overlap of the pressure-temperature ranges of individual mineral assemblages. It is, therefore, appealing to use an association or ensemble, to use Thompson's (1957) expression, of mineral assemblages as pressure-temperature indicators. In many areas, pelitic and mafic rocks occur in close proximity, and it is advantageous to consider associations of mineral assemblages in such rocks and represent them on the same diagram.

The advantage of using mineral assemblages as indicators of relative metamorphic grade lies in the fact that this information is readily obtained from field observations and petrographic study. For some purposes, such knowledge might suffice; otherwise, it can serve as a point of departure towards more precise evaluation of metamorphic conditions based on experimental calibration of mineral assemblages and the distribution of elements among coexisting minerals.

THEORY

The consideration of associations of mineral assemblages requires a representation in a common chemical system of components. In view of their chemical complexity, some compromise is required in approximating the compositions of minerals. In dealing with minerals in pelitic and mafic rocks, at least the following components must be recognized: SiO_2 , TiO_2 , Al_2O_3 , Fe_2O_3 , FeO , MgO , CaO , Na_2O , K_2O , and H_2O .

At constant pressure and temperature and at stable equilibrium, a particular composition within the chemical system (thermodynamic system) is characterized by a minimum Gibbs energy, which is given by the summation

$$\mathbf{G} = \sum \left(\frac{\partial \mathbf{G}}{\partial n_i} \right) n_i \quad (1)$$

where \mathbf{G} is the total Gibbs energy of the thermodynamic system, n_i is the number of moles of component i , and \hat{n}_i is the number of moles of all components other than i . The partial derivative is the partial Gibbs energy \bar{G}_i of a component. In this summation, one can choose either n or \bar{G} for

each component as the independent variable (Korzhinskii, 1959). If one distinguishes j -components as those having independent n , and k -components as those having independent \bar{G} , one can write

$$\mathbf{G} = \sum \bar{G}_j n_j + \sum \bar{G}_k n_k \quad (2)$$

A function \mathbf{L} can be defined as

$$\mathbf{L} = (\mathbf{G} - \sum \bar{G}_k n_k) \quad (3)$$

Since \mathbf{G} is at a minimum, if all values of \bar{G}_k are kept constant, \mathbf{L} also is at a minimum and equal to

$$\mathbf{L} = \sum \bar{G}_j n_j \quad (4)$$

Dividing by $\sum n_j$ gives

$$L = \sum \bar{G}_j X_j \quad (5)$$

where L is the molar value of the function \mathbf{L} , and X_j is the mole fraction of a j -component. The function $(\mathbf{G} - \sum \bar{G}_k n_k)$

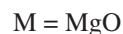
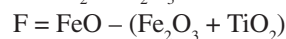
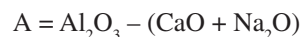
was derived by Korzhinskii (1959) and has been discussed by Thompson (1970), who used the designation \mathbf{L} and the grouping into j - and k -components.

The minimum value of L can be plotted as a surface in space defined by axes L and the mole fractions X of the j -components. A projection of the L surface onto the composition base expressed in terms of the mole fractions X of the j -components results in a phase diagram.

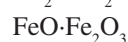
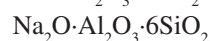
GRAPHIC REPRESENTATION

In view of these relations, it is advantageous to recast the components to be used as follows:

j -components



k -components



FeO·TiO₂
H₂O

One way to keep \bar{G}_k constant, at constant pressure and temperature, is to specify the presence of a phase with a composition equal to that of the component. Thus, the presence of quartz, magnetite, and ilmenite would keep \bar{G}_k of SiO₂, FeO·Fe₂O₃, and FeO·TiO₂ constant. The presence of plagioclase of constant composition would insure the constancy of \bar{G}_k of CaO·Al₂O₃·2SiO₂ and Na₂O·Al₂O₃·6SiO₂. A phase of approximately pure H₂O may be present at lower grades but, at higher grades, one has to assume that \bar{G}_{H_2O} is some function of pressure and temperature.

Minerals, in terms of j-components, can be plotted in a prism with a triangular base of

$$K = K_2O \cdot Al_2O_3$$

$$F = FeO - (Fe_2O_3 + TiO_2)$$

$$M = MgO$$

and a vertical axis of

$$A = Al_2O_3 - (CaO + Na_2O)$$

Non-potassic minerals can be plotted on the AFM face of this prism. Mineral assemblages lying inside the prism, with one potassic mineral, coexist with three minerals on the AFM face. Two potassic minerals coexist with two minerals on the AFM face connected by a tie line, and three potassic minerals coexist with one AFM mineral having a fixed composition. As a consequence, it is possible to indicate areas on an AFM diagram that coexist with a particular potassic mineral. For example, Figure 1 is an AFM diagram that corresponds to a metamorphic grade above the biotite-sillimanite-almandine isograd just to the left of invariant point 9 (Fig. 2), with superimposed fields of muscovite and biotite. This method of dealing graphically with a fourth component has been used previously by Albee (1965).

THE GRID

A thermodynamic system has $c + 2$ independent variables, where c is the number of components and 2 stands for pressure and temperature. The choice of either n or \bar{G} as the independent variable of a component is subject to the requirement of choosing at least one extensive variable, i.e. n , for each phase. Thus the maximum number of independent intensive variables, P , T , and \bar{G} 's, is given by the Gibbs phase rule

$$f = c + 2 - p$$

where f is the variance and p is the number of phases.

In the phase diagram of the AFM chemical system (Fig. 1), there are nine components. For a thermodynamic system with three coexisting phases, extensive variables (n 's) for at least three components must be chosen. This has been done for the three j-components by stipulating that

$\sum n_j = 1$ and stating any two mole fractions of the j-com-

ponents. The variance of this system is eight, given by the values of P , T , and the \bar{G} 's of the six k-components. If it is assumed, as an additional restriction, that the \bar{G} 's of the k-components are functions of P and T , the system is bivariant in P and T . In order to determine the state of the system, values of P and T must be chosen within the range over which the phases are stable.

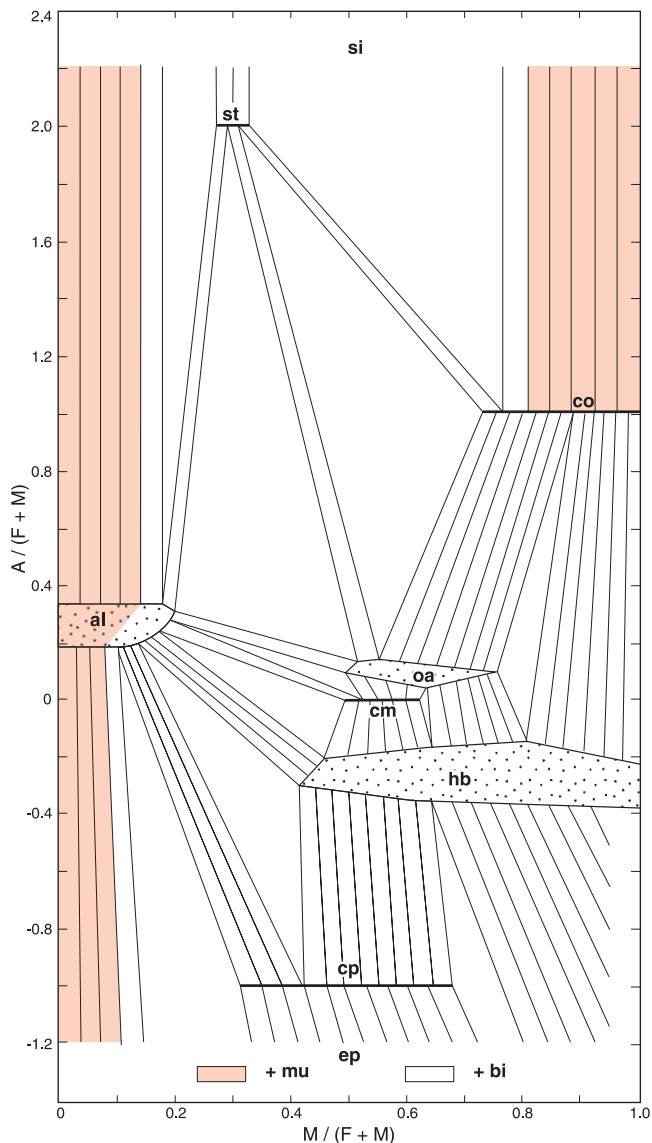


Figure 1. AFM diagram of mineral assemblages above the biotite-sillimanite-almandine isograd, just to the left of invariant point 9 (Froese, 2010; modified to show the association cordierite-hornblende). j-components: $A = Al_2O_3 - (CaO + Na_2O)$; $F = FeO - (Fe_2O_3 + TiO_2)$; $M = MgO$. k-components: \bar{G} of SiO₂ = G of quartz; \bar{G} of CaO·Al₂O₃·2SiO₂ = \bar{G} of CaAl₂Si₂O₈ in plagioclase of constant composition; \bar{G} of Na₂O·Al₂O₃·6SiO₂ = \bar{G} of 2NaAlSi₃O₈ in plagioclase of constant composition; \bar{G} of FeO·Fe₂O₃ = G of magnetite; \bar{G} of FeO·TiO₂ = G of ilmenite; \bar{G} of H₂O = function of P and T . For mineral abbreviations see Figure 2.

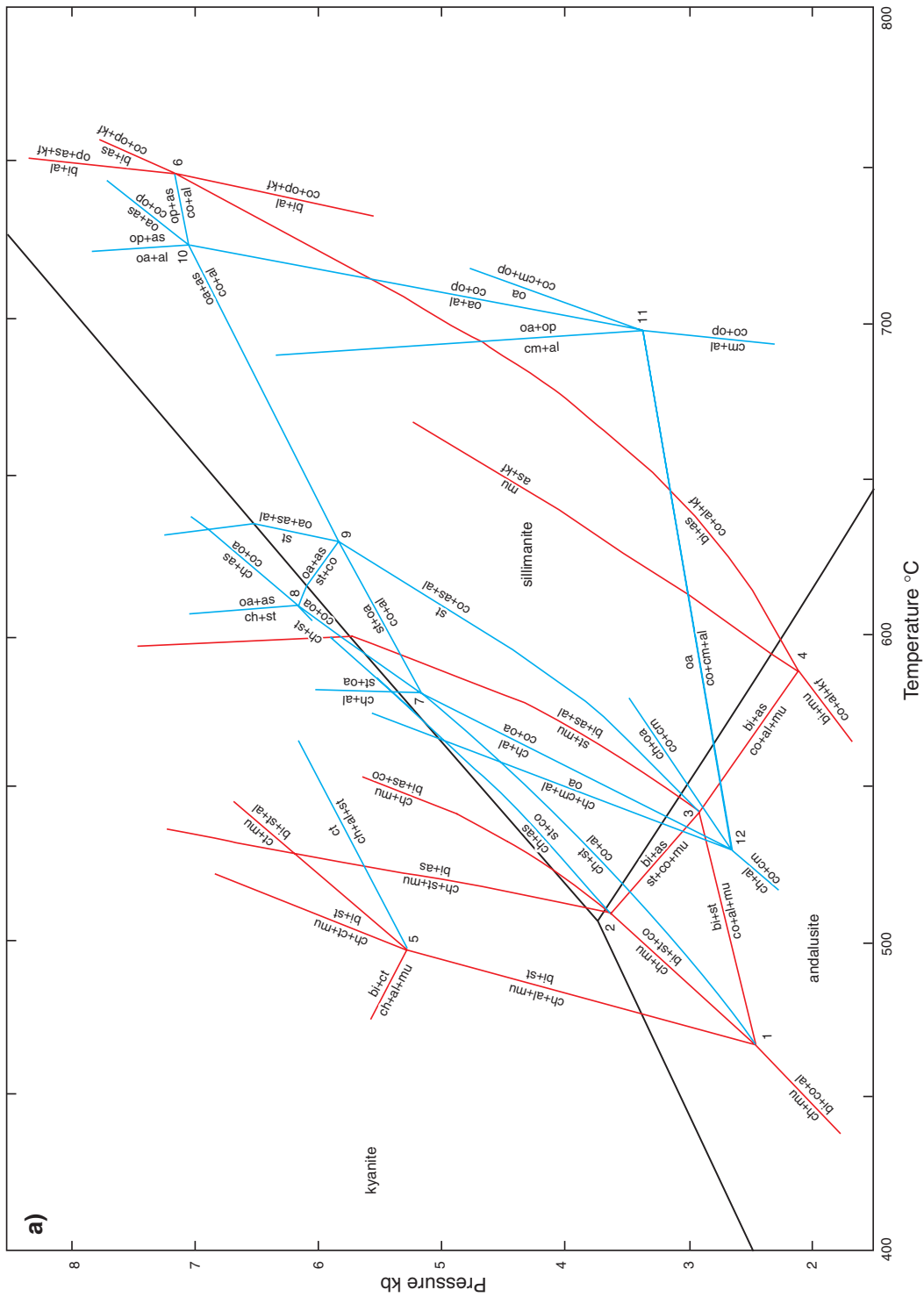


Figure 2. Mineral reactions in the prism AKFM (non-calcic minerals in red; calcic minerals in purple) and on the AFM face (non-calcic minerals in blue; calcic minerals in green). Reactions also involve k-components not indicated on the labels. Mineral abbreviations: as, aluminum silicate; st, staurolite; ct, cordierite; co, cordierite; al, almandine; ch, chlorite; oa, orthoamphibole; op, orthopyroxene; cm, cummingtonite; hb, hornblende; ep, clinopyroxene; ep, epidote; mu, muscovite; bi, biotite; kt, K-feldspar. **a)** Reactions in pelitic and mafic rocks.

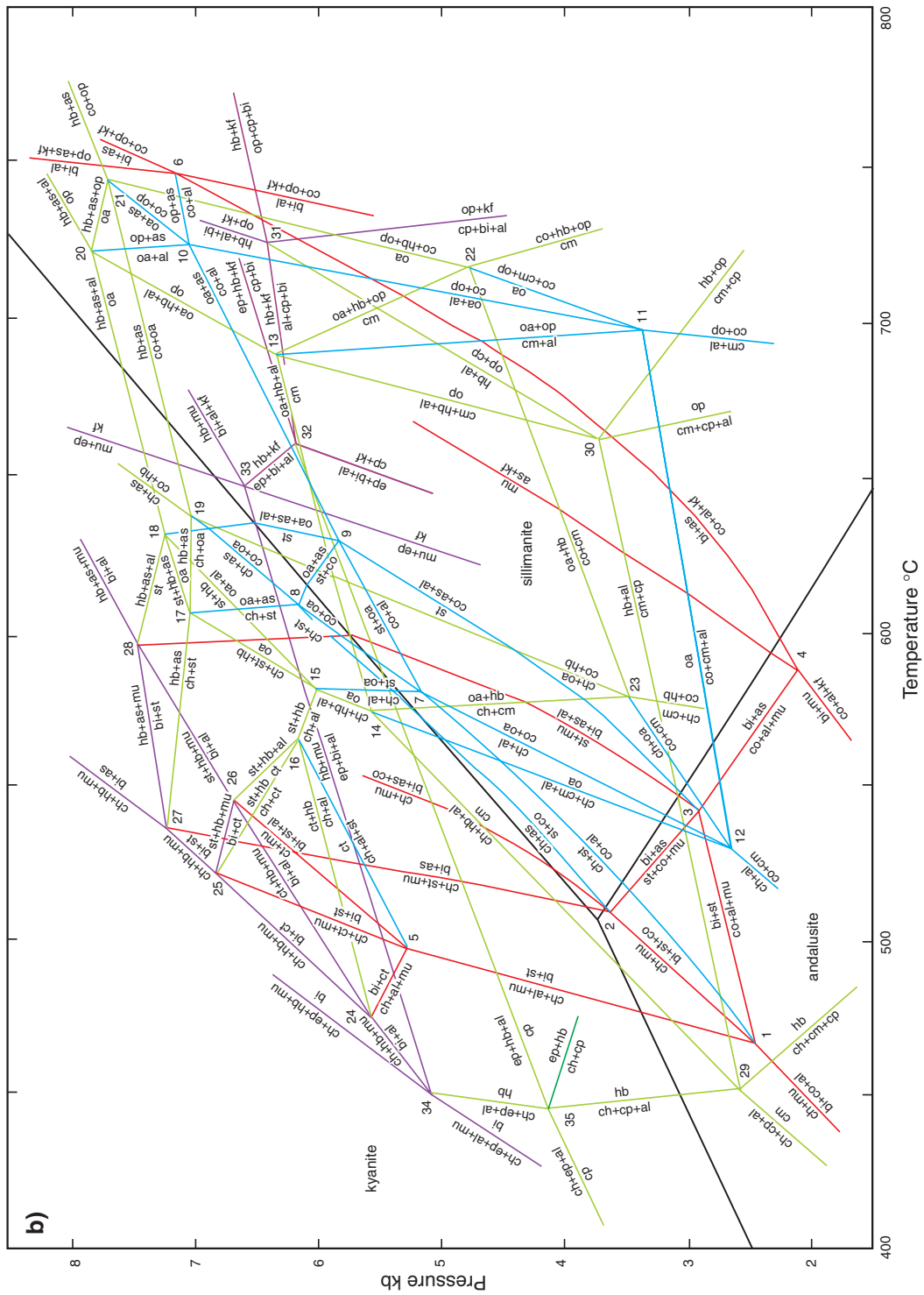


Figure 2. (cont.)

With four phases in stable equilibrium, there is one fewer independent intensive variable, and the variance of the thermodynamic system is reduced by one. Retaining the restriction that the \bar{G} 's of the k-components are functions of P and T , the system is univariant in P and T . Only one can be freely chosen and the stability of a univariant four-phase assemblage can be plotted as a line on a P - T diagram. At any point along this univariant line, there is a particular composition that can be expressed by alternative combinations of phases to achieve the same minimum value of L . Graphically this relationship can be shown either by crossing tie lines or by tie lines joining one phase to three surrounding phases. Such relationship can also be expressed by a univariant reaction involving the four minerals and some amounts of k-components, e.g.



which divides the P - T diagram into two stability fields of the alternative phase assemblages. These univariant reactions can be combined into a grid and intersect at invariant points, with five stable phases and five univariant reactions extending from each point.

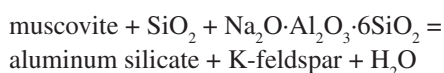
Within the AKFM system, univariant reactions are expressed by five phases and some amounts of k-components. There are six stable phases at invariant points and six univariant reactions extending from each invariant point. Reactions are designated by one of the six phases not participating in the reaction. However, the biotite-absent reaction is not shown in Figure 2 in order to provide more room on the grid, thus reducing the number of reactions extending from an invariant point to five. Reactions in pelitic rocks are shown in Figure 2a and reactions in both pelitic and mafic rocks are shown in Figure 2b.

The sequence of reactions around an invariant point must be consistent with Schreinemakers' rules (Zen, 1966). Lindsley et al. (1968) suggested the following convenient procedure to check the consistency of the arrangement of reactions around an invariant point. Reaction equations are written onto the reaction boundary such that reactants and products straddle the boundary. If a reaction is designated by the absent phase, its metastable extension must fall into a sector in which the "half-reactions" include the absent phase.

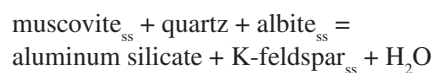
SOURCES

In Figure 2, the stability fields of the aluminum silicates have been calculated from the thermodynamic database of Berman (1988).

The reaction shown as



is the reaction



where ss refers to a solid solution of K and Na end members, taken from Chatterjee and Froese (1975).

All invariant points selected in the preparation of the grid shown in Figure 2 have been identified in earlier studies. As discussed below, the positions of some have been accepted here; the positions of others have been changed to ensure an overall conformity with Schreinemakers' rules.

The grid of Hess (1969) included invariant points 1–4. Invariant point 5 is taken from the grid of Korikovskii (1969). Invariant point 6 is part of a grid given by Korikovskii (1979). Kepezhinskas and Khlestov (1977) showed invariant points 1–3, in each case with one reaction not involving muscovite. These reactions form part of Korikovskii's (1970) grid for K_2O -poor rocks, which included invariant points 7–10. Invariant point 9 had been proposed previously by Robinson and Jaffe (1969). Trzcieski (1971) deduced invariant point 7 from field evidence; *see also* Carmichael et al. (1978).

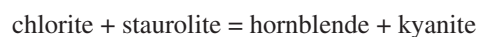
D.M. Carmichael (*in* Davidson et al., 1990) combined invariant points 2, 3, and 6–10 into one grid. The locations of these invariant points in Figure 2 have been taken from this grid, with minor modifications. All other invariant points are shown at estimated positions.

Invariant points 11 and 12 led to the inclusion of cummingtonite in the grid. Invariant point 11 was deduced by Marakushev (1965) and by Percival et al. (1982). Here the topologies of some reactions have been changed to be consistent with the reaction



extending from invariant point 10 (Korikovskii, 1970). Invariant point 12, with gedrite as the orthoamphibole is taken from Spear and Rumble (1986); anthophyllite has been ignored.

Invariant point 13 was deduced by Froese and Goetz (1981) and forms part of a grid (invariant points 13–23), which shows the coexistence of hornblende with aluminous minerals (Froese and Hall, 1983). Invariant point 14 is supported by the assemblage chlorite-hornblende-almandine-cummingtonite-orthoamphibole (Hall, 1980; Froese and Hall, 1983) and invariant 15 by the assemblage chlorite-almandine-staurolite-orthoamphibole-hornblende (Spear, 1982). In the present paper, it has been accepted that the composition of cummingtonite lies on the F side of the tie line orthoamphibole-hornblende (Robinson and Jaffe, 1969; James et al., 1978). As a consequence, the topologies of some reactions have been changed. Selverstone et al. (1984) described the assemblage of the reaction



which extends from invariant point 17. Frey (1969, 1974) reported the assemblage of the reaction

staurolite = hornblende + kyanite + almandine

extending from invariant point 18. The assemblage orthoamphibole-cordierite-hornblende (Schumacher and Robinson, 1987) indicates that the reaction

chlorite + orthoamphibole = cordierite + hornblende

extending from invariant point 19 has gone to the right.

Thompson and Leclair (1987) extended the grid accommodating the coexistence of hornblende with aluminous minerals by proposing invariant points 24–28. They recorded the assemblage of invariant point 24. The coexistence of chloritoid-hornblende is also mentioned by Fox (1975). Klaper and Bucher-Nurminen (1987) listed the assemblages of two reactions

biotite + staurolite = hornblende + kyanite + muscovite and

biotite + almandine = hornblende + kyanite + muscovite

extending from invariant point 28.

Invariant points 29 (Froese, 1980) and 30 and 31 (Froese and Jen, 1979), extend the grid to include clinopyroxene. In the present paper, it has been assumed that the composition of hornblende lies on the M side of the tie line cummingtonite-clinopyroxene. This provides a better accommodation of the association hornblende-cummingtonite-clinopyroxene common in low-pressure metamorphism (Miyashiro, 1973). The topologies of reactions around invariant 31 are based on mineral analyses in Jen (1975), also given in Jen and Kretz (1981).

Invariant points 32–35, taken from Froese (1980), introduce epidote. If the composition of epidote is used rather than that of clinozoisite, clinopyroxene lies on the F side of the tie line hornblende-epidote, again leading to a change in the topologies of some reactions. The topologies of two reactions extending from invariant point 34 have been changed to correct an error.

AN EXAMPLE OF MAPPED ISOGRADS

In southeastern Ontario, isograds have been mapped on the basis of some reactions that form part of the grid. In Figure 3, the isograds have been taken from Carmichael (1970) and Carmichael et al. (1978) with some modifications and additional information.

From Bishop Corners and Lake of Islands, Thompson and Leclair (1987) reported the mineral assemblage of invariant point 24; it includes the assemblage of the reaction

chlorite + almandine + muscovite = biotite + chloritoid

On the basis of mineral assemblages given by Thompson (1972) and Carmichael et al. (1978), two isograds may be inferred:

chlorite + chloritoid + muscovite = biotite + staurolite and

chloritoid + muscovite = biotite + staurolite + almandine

These three reactions involving chloritoid are also discussed in recent work by Ford (2002, 2006), and the first and third reactions are shown as isograds. According to F.D. Ford (pers. comm., 2010), between Bishop Corners and Myers Cave, at a metamorphic grade above the third isograd, there is an occurrence of chloritoid-staurolite-hornblende (locality 92-2 in Ford, 2002) and of staurolite-hornblende (locality 92-24 in Ford, 2002). The other isograds shown between Bishop Corners and Ardoch are based on mineral assemblages given by Hounslow and Moore (1967) and Carmichael et al. (1978).

In the Whetstone Lake area, all isograds have been taken from Carmichael (1970). The assemblage biotite-staurolite-almandine was stabilized, presumably at a pressure slightly lower than that at Bishop Corners, by the reaction

chlorite + almandine + muscovite = biotite + staurolite

Above the biotite-kyanite isograd, the assemblage hornblende-almandine-cummingtonite (Carmichael et al., 1978) indicates that the reaction

chlorite + hornblende + almandine = cummingtonite

has gone to the right. To the north of Whetstone Lake, at Brinklow, the mineral assemblage of invariant point 14 has been recorded (Hall, 1980; Froese and Hall, 1983); it includes the assemblage of the last reaction. Also at Whetstone Lake, Trzcieski (1971) has documented two reactions extending from invariant point 7; these are:

chlorite + staurolite = cordierite + orthoamphibole and

chlorite + staurolite = cordierite + almandine

Reinhardt and Skippen (1970) presented a diagram showing mineral assemblages in pelitic and mafic rocks from the Westport area. This diagram has been redrawn here as Figure 4; on the grid, it would be represented by a point to the right of the reaction

hornblende + almandine + biotite = orthopyroxene + K-feldspar

extending from invariant point 31.

DISCUSSION

The construction of the grid involves approximations and simplifying assumptions leading to discrepancies between the grid and observations in nature. The main difficulty stems from the decision to keep the number of j-components to four in order to allow graphic representation of mineral assemblages in the chemical system AKFM.

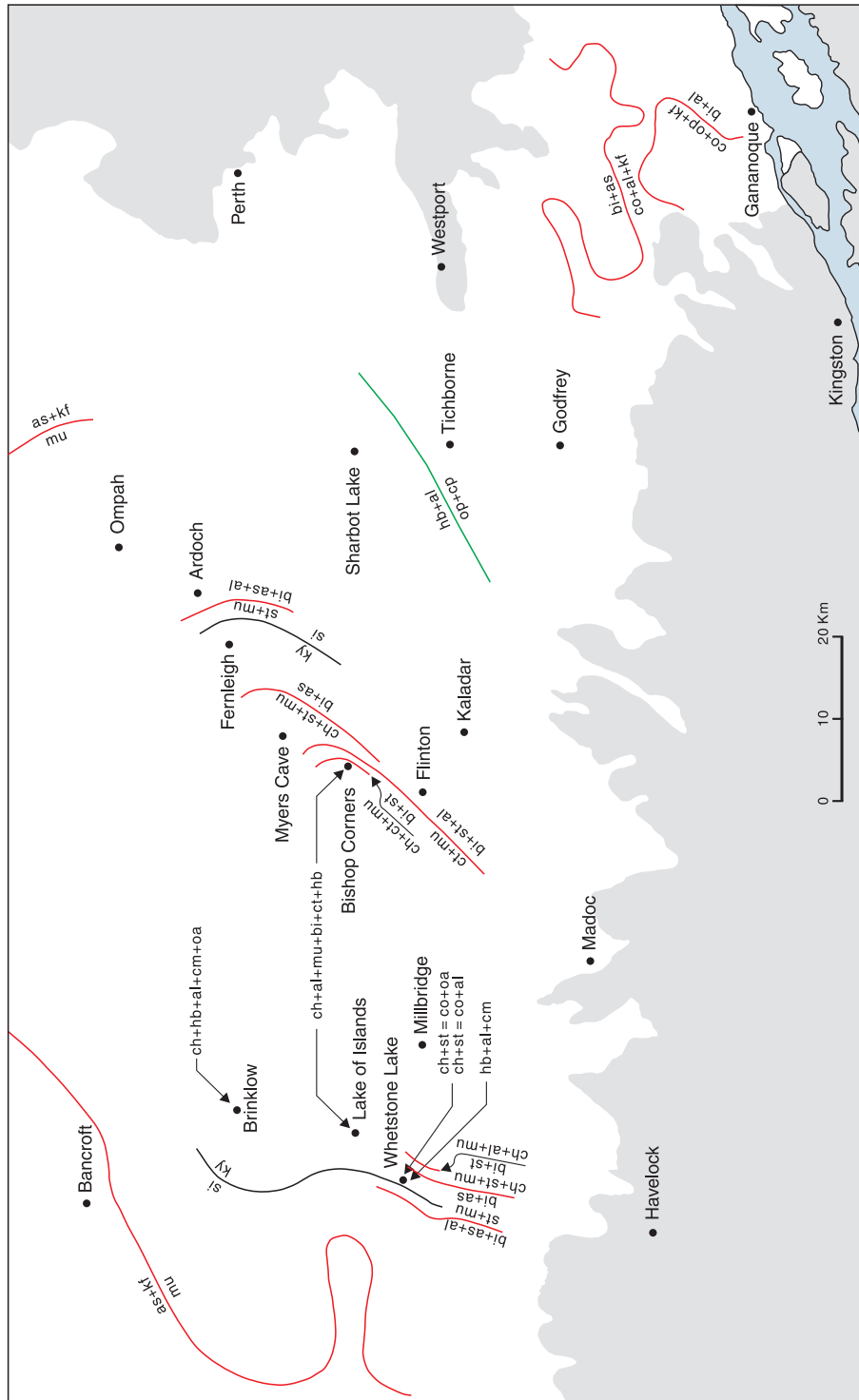


Figure 3. An example of mapped isograds from eastern Ontario.

Considering Mn, which has been neglected, as an additional j-component, would turn garnet-bearing assemblages appearing as univariant reactions in Figure 2 into bivariant assemblages stable over a P - T range.

Treating $\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$ and $\text{Na}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 6\text{SiO}_2$ as k-components demands that the composition of plagioclase remain constant or is a function of only P and T . This is an approximation at best and, if the composition varies independently of P and T , the two compositions should be regarded as j-components. As a consequence, plagioclase-bearing assemblages, shown as univariant lines, would be

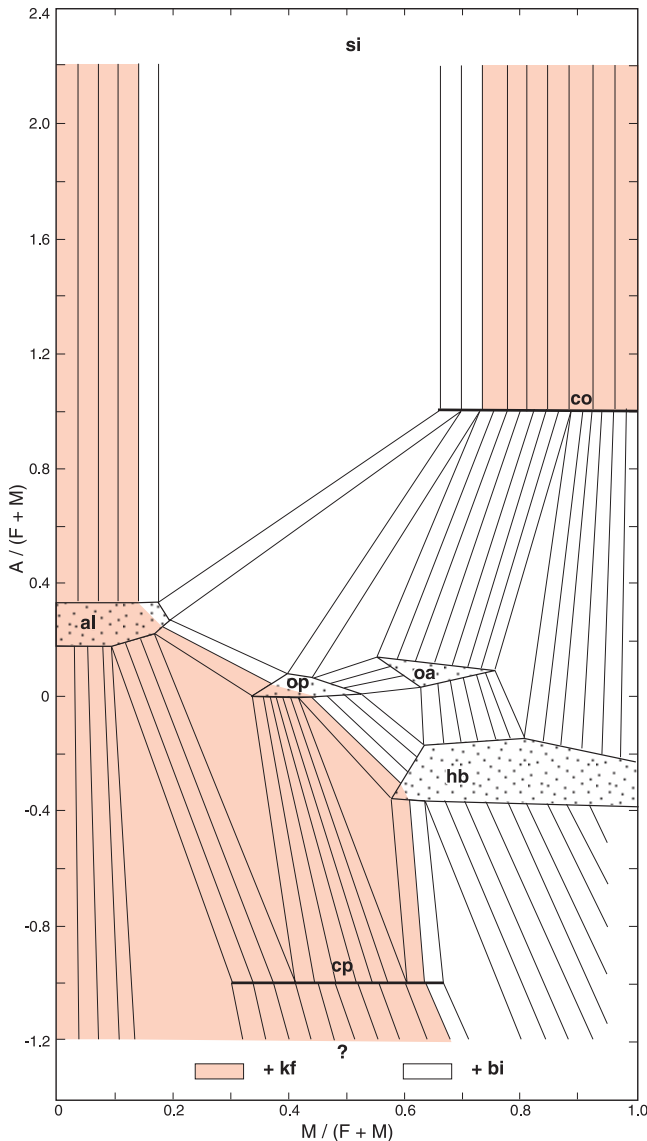
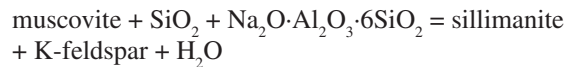


Figure 4. AFM diagram of mineral assemblages from the vicinity of Clear Lake, Westport area (from Reinhardt and Skippen, 1970). j-components: $A = \text{Al}_2\text{O}_3 - (\text{CaO} + \text{Na}_2\text{O})$; $F = \text{FeO} - (\text{Fe}_2\text{O}_3 + \text{TiO}_2)$; $M = \text{MgO}$. k-components: \bar{G} of $\text{SiO}_2 = G$ of quartz; \bar{G} of $\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2 = \bar{G}$ of $\text{CaAl}_2\text{Si}_2\text{O}_8$ in plagioclase of constant composition; \bar{G} of $\text{Na}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 6\text{SiO}_2 = \bar{G}$ of $2\text{NaAlSi}_3\text{O}_8$ in plagioclase of constant composition; \bar{G} of $\text{FeO}\cdot\text{Fe}_2\text{O}_3 = G$ of magnetite; \bar{G} of $\text{FeO}\cdot\text{TiO}_2 = G$ of ilmenite; \bar{G} of $\text{H}_2\text{O} = \text{function of } P \text{ and } T$. For mineral abbreviations see Figure 2.

stable over a P - T range. In the absence of plagioclase in some cordierite-orthoamphibole rocks, $\text{Na}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 6\text{SiO}_2$ should be regarded as a j-component.

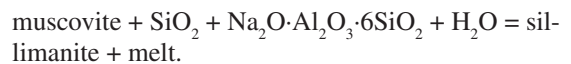
The absence of magnetite and ilmenite, the compositions of which have been taken as k-components, would mean that Fe_2O_3 and TiO_2 should be regarded as j-components.

The assemblage muscovite-sillimanite-K-feldspar has been used to define the univariant reaction



However, this assemblage occurs in some areas, e.g. central Massachusetts (Tracy and Robinson, 1983), over a zone several kilometres in width. This suggests that the assemblage is bivariant, which could be caused by

1. retrograde metamorphism of sillimanite-K-feldspar assemblages with different amounts of the fluid phase to produce the dry assemblage muscovite-sillimanite-K-feldspar;
2. dilution of the fluid phase to various degrees with gas species other than H_2O ; or
3. formation of an undersaturated melt by the reaction



The assemblage muscovite-sillimanite-undersaturated melt is stable within a P - T interval and subsequent crystallization of the melt produces quartz, plagioclase, and K-feldspar.

In spite of these limitations, the grid provides a coherent, even though only approximate, overview of the associations of mineral assemblages in pelitic and mafic rocks.

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