

A REVIEW OF CHEMICAL AND PETROGRAPHIC CRITERIA FOR DEFINING METAMORPHIC GRADES AND FOR RECOGNITION OF DISCONTINUOUS MINERAL REACTIONS: EXAMPLES FROM METAPELITES

CHARLES V. GUIDOTTI

Dept. of Geological Sciences, Univ. of Maine at Orono, Orono, Maine 04469 U.S.A.

ABSTRACT. — A brief review is given of the ambiguities previously encountered in defining metamorphic grades and in recognizing the specific mineralogic reactions that occur due to changes in grade. These ambiguities resulted largely from difficulties in distinguishing between mineralogic changes caused by *PT* variation and those caused by bulk composition variation. To a great extent these problems were due to petrologists describing metamorphic mineral assemblages by means of chemical systems which were not thermodynamically valid.

Some early attempts were made to circumvent this problem but no real success was achieved until about 25 years ago. However, despite the development of a conceptual framework to remove the ambiguities listed above, a surprising large number of petrologists still do not seem to be making full use of this approach or to be aware of some requirements for its use. It involves the use of observed assemblages to construct complete compatibility diagrams (designated as a *mineral facies*) which are based on chemical systems which are thermodynamically valid. A review of this approach is given which should be of particular use to field petrologists.

Introduction

All petrologists are familiar with the metamorphic facies concept which was an outgrowth of early work by BARROW (1893, 1912), ESKOLA (1920, 1939), GOLDSCHMIDT (1912) etc., and then carried forward by people like HARKER (1939) and TILLEY (1924). Application of the term isograd to metamorphic rocks, of course, brings to mind the work of TILLEY (1925).

From the very beginning, the early workers recognized that in general the minerals

present in a metamorphic rock reflected the pressure and temperature (*PT*) conditions at which the rock formed. It was also realized that the minerals present were reflecting the bulk composition present. This latter realization was shown by the fact that early definitions of metamorphic grade were discussed in terms of things like, (a) the minerals occurring in meta-basalt bulk compositions, (b) the mineral zones of Barrow in meta-pelitic bulk compositions.

Various attempts were made to distinguish between changes in mineral assemblages resulting from change in *PT* conditions vs. mineralogic changes resulting from variation of bulk composition. Removing this ambiguity was necessary in order to make comparisons of the metamorphic grades found in different areas. One of the most common approaches at avoiding this ambiguity involved proposal of chemographic systems or diagrams to show the mineralogic relations. Of these chemographic approaches, the *ACF* diagram has been used most often (see fig. 1).

Unfortunately, none of the early attempts at defining chemographic systems were overwhelmingly successful at the following: (a) removing the ambiguity in distinguishing mineral changes due to *PT* variation versus changes due to variation of bulk composition, (b) as an aid for recognizing the *actual* mineralogic reactions which occurred due to *PT* changes, and (c) as an aid in calling attention

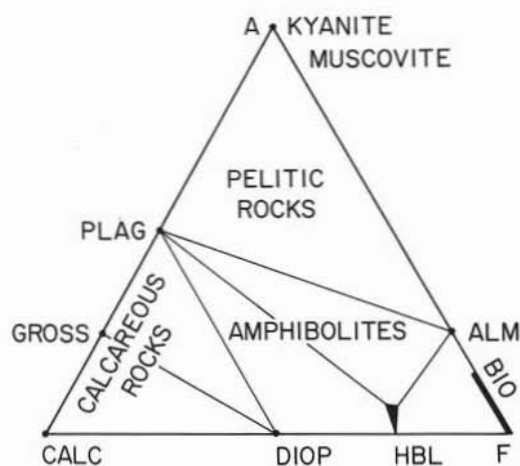


Fig. 1. — ACF diagram showing the general lithologic types to be expected in different portions of a quartz-saturated ACF system.

to the role of volatiles in influencing metamorphic reactions.

It is worth noting here, that by the 1950's petrologists (eg. YODER, 1952) had begun to focus strongly on the role of volatiles during metamorphism via the study of simplified laboratory reactions. However, inability to define precisely the reactions occurring in natural rocks prevented petrologists from discussing rigorously the role of volatiles for the reactions.

The main cause for the difficulties encountered can be attributed to the fact that the chemical systems chosen to describe metamorphic mineral assemblages as a function of PT were not true thermodynamic systems in the sense of having the proper number of components to describe the equilibria or reactions of concern. The difficulties were most evident via the graphical representations used to illustrate these non-thermodynamic chemical systems. Such graphical systems would include the ACF and AKF diagrams which have been in common usage for the past sixty years.

These systems and resulting diagrams do enable one to see some important aspects of metamorphic mineral assemblages. Probably mineralogic aspects related to the bulk Al_2O_3 content are the ones best revealed by these two systems. However, two procedures in the usage of these systems caused the dif-

ficulties mentioned above: (1) thermodynamically distinct components (eg. MgO and FeO) were considered as a single component; (2) minerals containing large amounts of components not included in the ACF or AKF systems were included in the mineral equilibria shown on the ACF or AKF diagrams. Not only did these procedures make it so that the systems were not true thermodynamic systems, but they made it so that the diagrams were not true phase diagrams. In turn, this obscured the extremely important Mg-Fe distribution relations of many metamorphic reactions.

Basically, the ACF, AKF, and similar systems, are *too* general in the sense of trying to describe all rock types with one system. The first overwhelming successful attempts at avoiding the difficulties and ambiguities discussed above were those of THOMPSON (1957, 1961). However, even now, 25 years later, relatively few petrologists seem to follow or be aware of the procedures needed for unambiguous definition of metamorphic grades and recognition of the specific mineralogic reactions that occur with change in grade. Hence, in the following discussion the writer will:

- (a) consider some early attempts at avoiding the difficulties discussed above;
- (b) review the procedures required to attain the goals implied by the title of this paper.

The approach taken will largely be in the context of a field petrologist faced with the study of natural parageneses. Particular emphasis will be given to two key aspects which have not received sufficiently explicit emphasis by field or theoretical petrologists or the writers of textbooks. It is somewhat ironic that, at a time when the study of metamorphism has become very sophisticated in a thermodynamic sense, some very fundamental starting points for understanding metamorphic rocks are still not widely appreciated.

However, no claim to any conceptual originality is made by the author because all of the concepts to be discussed below are already present in the literature. Only the relative amount of specific emphasis given to certain points is unique to this paper.

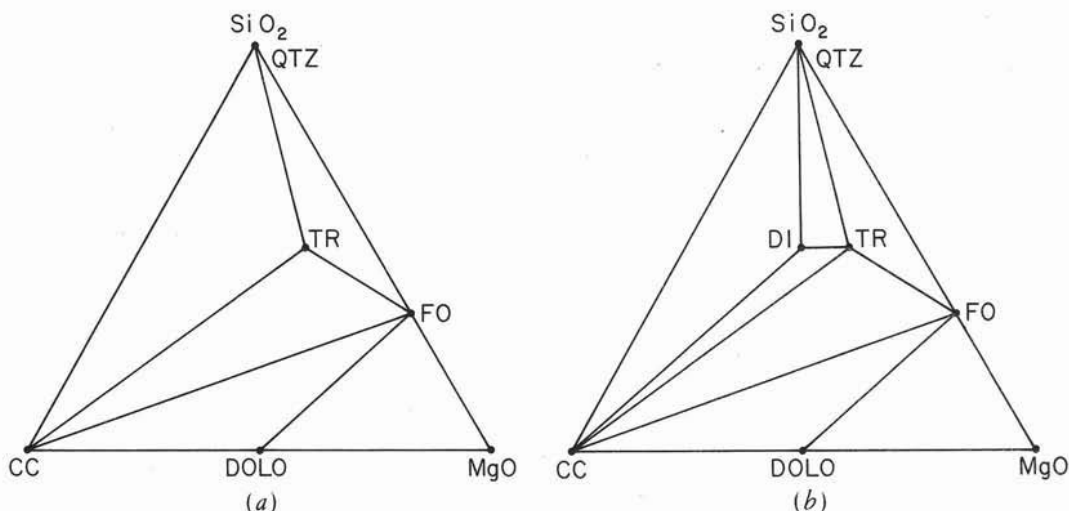


Fig. 2. — Compatibility diagrams for two different metamorphic grades in rocks of siliceous dolomite bulk composition. See text for the mineralogic reaction reflecting the change in compatibilities between (a) and (b). Based on BOWEN (1940).

Pre-1957 approaches to the problem

Several direct or indirect attempts were made before that of THOMPSON (1957) to avoid the difficulties discussed above. These early attempts bear direct similarities to those described below but unfortunately were largely unrecognized by petrologists.

THOMPSON (1957) has pointed out that BARTH (1936) and OSBERG (1952) proposed systems and graphical representations which avoided the problems resulting from considering $\text{FeO} + \text{MgO}$ as a single component. BOWEN (1940) in his classic paper on « *Progressive metamorphism of siliceous limestone and dolomite* » came even closer to treating metamorphic assemblages in a fashion similar to that presented below. The main similarities were:

(1) he used the combinations of mineral assemblages as illustrated on compatibility diagrams to designate grades of metamorphism. Of particular importance is the fact that he attempted to do this in the context of thermodynamically valid systems (in the sense of Phase Rule components). Hence, his compatibility diagrams could be considered as true phase diagrams;

(2) he emphasized the need to use a number of assemblages, the mineral of which could be almost completely described by the

chosen system, to characterize a grade — not just the presence of a single phase or a single assemblage;

(3) he used the change in the configuration of tie lines of the compatibility diagrams as a guide for postulating specific mineral reactions occurring as a function of grade change;

(4) he emphasized the need for using a wide range of bulk composition *within* the chosen system in order to recognize a sequence of grade changes.

Fig. 2 shows two mineral compatibility diagrams appropriate to the metamorphism of siliceous dolomites. The reaction to express the change in compatibilities from *F2A* to *F2B* can readily be deduced from the change in the configuration of the lines as:

Unfortunately, the general implications of Bowen's paper went largely unrecognized. As will be evident from the following discussion, much of what he said is intrinsic to any unambiguous designation of metamorphic grade or recognition of the mineralogic reactions relating two grades. To some extent similar comments can be made with respect to the treatment by NIGGLI (1954) of mineral equilibria in metamorphic rocks.

Unambiguous designation of metamorphic grades

The method of unambiguous designation of a metamorphic grade will be discussed here as a series of procedural steps. It should be noted that these are conceptual steps rather than a sequence which a field petrologist would follow specifically. They might be thought of as conceptual guides to be followed after initial field collecting and initial thin section petrography has indicated whether one is dealing with a region containing meta-pelites, siliceous dolomites, or metabasalts, etc.

Step § 1

One must choose a set of components which form a true thermodynamic system so that it will completely describe the phase relations in a thermodynamic or phase rule sense. Obviously, this would exclude systems which consider ($\text{Na}_2\text{O} + \text{K}_2\text{O}$) or ($\text{MgO} + \text{FeO}$) as single components.

In the simplest cases these would be systems like Al_2SiO_5 for the aluminum silicate phase relations or SiO_2 for the phase relations among quartz-tridymite-cristobalite etc. However, it is obvious that one would like to use systems which describe the phase relations among more than just the aluminum silicates or the SiO_2 phases. Fig. 3 shows a number of systems which can be used to describe *completely* groups of minerals that field petrologists commonly want to consider. The first two systems of fig. 3 (the *AFM* and *AKNa* Systems) are now presented in most of the textbooks but have received little use by most field petrologists and only superficial use by many specialists in the study of metamorphism. Moreover, as developed below, few of the presently available textbooks discuss several important aspects required for effective use of these systems.

At any rate, it is evident that a number of systems are available for describing a wide range of bulk compositions. However, it should be stressed that one would consider the equilibria *among only* the phases which can be completely (at least ideally) described by the components chosen for the system. Hence, it may be necessary to ignore some phases that contain too much of some com-

SOME THERMODYNAMIC SYSTEMS FOR DESCRIBING METAMORPHIC ROCKS

$\text{Al}_2\text{O}_3 - \text{FeO} - \text{MgO} - \text{K}_2\text{O} - \text{SiO}_2 - \text{H}_2\text{O} :$
PELITES -- Thompson (1957)
$\text{Al}_2\text{O}_3 - \text{K}_2\text{O} - \text{Na}_2\text{O} - \text{SiO}_2 - \text{H}_2\text{O} :$
PELITES -- Thompson (1961)
$\text{CaO} - \text{MgO} - \text{SiO}_2 - \text{CO}_2 :$
SILICEOUS DOLOMITES -- BOWEN (1940)
$\text{CaO} - \text{Al}_2\text{O}_3 - (\text{Fe}_2\text{O}_3) - \text{FeO} - \text{MgO} -$
$\text{Na}_2\text{O} - \text{SiO}_2 - \text{CO}_2 - \text{H}_2\text{O} :$
METABASITES -- Harte + Graham (1974)

Fig. 3. — Examples of true thermodynamic systems for describing metamorphic mineral parageneses.

ponent which is extraneous to the chosen system. For example, one would exclude biotite from consideration with the *AKNa* system. THOMPSON (1957) has discussed some of the implications of small amounts of components which are extraneous to the chosen system entering some of the phases of interest (e.g. MnO in almandine-pyrope garnet). For detailed work which involves a quantitative, thermodynamic treatment, the mineralogic equilibria can be treated via activity models to account for the deviation of some of the phases from being completely described by means of the chosen system. However, such approaches tend to be more detailed than necessary for the purposes of many field petrologists.

Step § 2

One must develop ways to show the chosen system diagrammatically so that the phase relations can be seen graphically. Moreover, these graphical approaches must be such that the resulting diagrams are true phase diagrams in the sense of a Phase Rule interpretation (figs. 4 *a* and 4 *b*).

GREENWOOD (1975), among others, has discussed this point in his paper on thermodynamically valid projections. In actual practice, a field petrologist will usually be able to choose for his particular study from among a number of previously defined systems and graphical representations of these systems — e.g. the *AFM* or *AKNa* systems for metapelites.

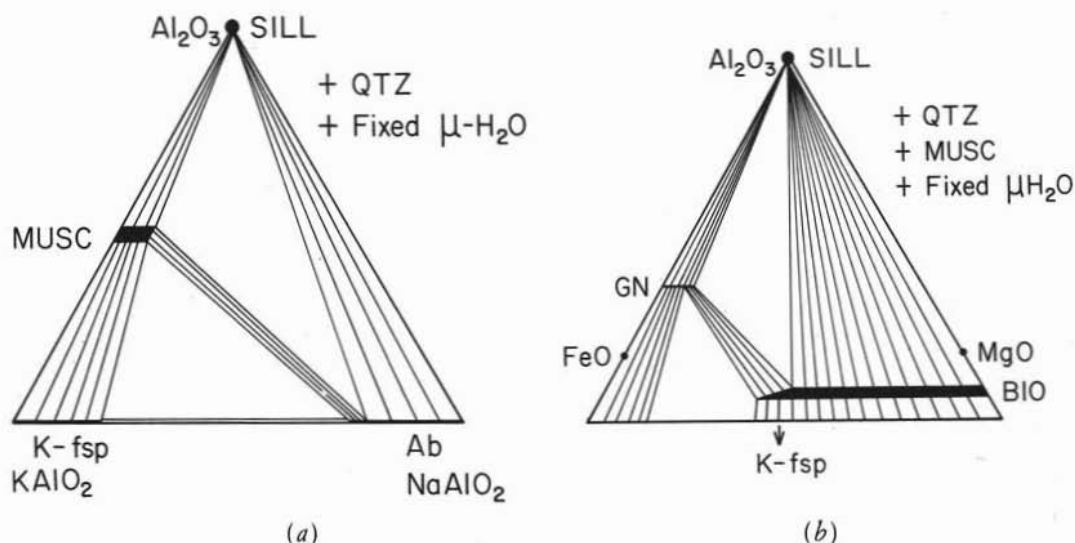


Fig. 4. — (a) Compatibility (phase) diagram for sillimanite zone pelitic schists based upon the *AKNa* projection of the system $\text{Al}_2\text{O}_3\text{-K}_2\text{O-Na}_2\text{O-SiO}_2\text{-H}_2\text{O}$ (THOMPSON, 1961). (b) Compatibility (phase) diagram for the higher grade portion of the sillimanite zone based upon the *AFM* projection of the system $\text{Al}_2\text{O}_3\text{-FeO-MgO-K}_2\text{O-SiO}_2\text{-H}_2\text{O}$ (THOMPSON, 1957).

Step § 3

This brings us to an aspect commonly ignored by most field petrologists, many specialists, and also by the most commonly used textbooks. Maybe this neglect is a result of using the *AFM* system and diagram much as has been done over the years with the *ACF* diagram — i.e. merely for plotting the phase assemblage of the most common rock type in an area. Ironically, this aspect was emphasized very strongly by THOMPSON (1957, p. 855).

It involves: (a) purposely collecting at a given locality of as wide a range of assemblages (i.e. bulk compositions) as possible — the assemblages being ones which can be represented quite fully by the chosen system; (b) plotting these assemblages graphically in order to show the configuration of tie lines or compatibilities over the whole composition range of the system (see figs. 5 a - 5 c). In the case of the *AFM* and *AKNa* diagrams, such plotting can be done in a qualitative way based on the now fairly well known composition ranges of the solid solution phases — e.g. the Mg/Fe ratio of biotite is always greater than that of garnet.

Of particular importance is that it is this configuration of tie lines or the compatibility diagram which unambiguously defines a me-

tamorphic grade. This is a direct result of the fact that the configuration of tie lines is a function of the intensive parameters — (e.g. *P*, *T*, etc.). THOMPSON (1957) referred to such an array of assemblages as shown on a compatibility diagram as a *Mineral Facies*. Numerous subsequent theoretical papers like those of HESS (1969), ALBEE (1965 a) etc. on petrogenetic grids have made ample use of this approach. And of course, some field-oriented petrologic studies on pelitic schists used this approach soon after THOMPSON (1957) defined the *AFM* system — e.g. GREEN (1963) and ALBEE (1965 b).

However, to a very great extent workers dealing with the field occurrences of metamorphic rocks have not taken this approach in the sense of constructing complete compatibility diagrams. Indeed, the writer made a survey of 61 reprints (post 1960) in his collection of reprints on field oriented studies of medium grade pelitic schists and found that only 19 used the procedures described above. Indeed as early as 1968 (THOMPSON and NORTON, 1968, p. 321) attention was called to the fact that relatively few petrologists were using the above approach to define metamorphic grades in meta-pelites.

From a field petrologist's point of view, several things should be noted about constructing complete compatibility diagrams.

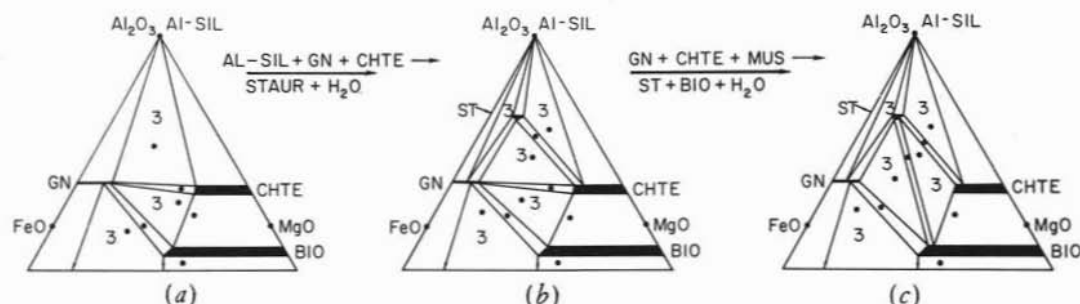


Fig. 5 *a, b, c.* — Representative AFM compatibility diagrams for common mineral facies of medium to high grade meta-pelites. Large dots are to represent the assemblages a petrologist might observe in order to construct the given diagram. The indicated mineralogic reactions are based upon the indicated changes in tie line configurations. The 3's are to designate the assemblages with 3 AFM phases. All assemblages with muscovite + quartz present.

(*a*) The possibility of bulk composition factors controlling the presence or absence of a given mineral or mineral assemblage is eliminated.

(*b*) It avoids defining metamorphic grades on the basis of a single phase. For example, staurolite can be present in several different compatibility diagrams, but in all cases the grade is defined by the particular configuration of the tie lines — not the mere presence of the staurolite.

(*c*) It enables a much finer definition of metamorphic grades.

Points (*a*) - (*c*) will be illustrated further below by means of an example.

Recognition of discontinuous mineralogic reactions relating different mineral facies

It is important to realize that it is the change in the configuration of tie lines (i.e. the compatibilities) which unambiguously defines a *change* of metamorphic grade. Recognition of such changes of tie line configurations obviously depends upon having as complete compatibility diagrams as possible — as a result of collecting a wide range of assemblages as discussed above.

Two things of considerable significance which arise directly from an unambiguous definition of a change in tie line configuration — (i.e. a change in mineral facies) are:

(*a*) recognition of the specific mineralogic reaction occurring as a result of the change in metamorphic grade (see fig. 5);

(*b*) from the nature of this mineralogic reaction, one can rationally assess the role that fluid (e.g. PH_2O or XH_2O) has on controlling the change in grade.

In essence, one can quickly and correctly comprehend the parameters controlling the mineralogic reaction leading from one *mineral facies* to another. Hence, one can start relating (in terms of P, T, X_{fl}) the metamorphism of a given area with the petrogenetic grid which applies to the chemographic system used to describe the rocks. Fig. 6 is a schematic P, T, X_{fl} diagram or petrogenetic grid. On it, P, T, X_{fl} reaction surfaces separate each mineral facies from other mineral facies. Fig. 7 illustrates how reaction surfaces will appear on a map to form isograds separating different mineral facies.

Finally, it should be noted that because the above procedure enables recognition of the actual reactions which occur in rocks (and in terms of the correct components) it should serve as a useful guide for experimental and/or theoretical petrologists. In essence, it provides them with information on which reactions apply in nature and which components to use.

Summary

The approach which has been discussed above is not new inasmuch as some aspects were developed by BOWEN (1940). More specifically, the above discussion has been a review of a powerful approach available to petrologists and field workers since the paper by THOMPSON (1957). Subsequent

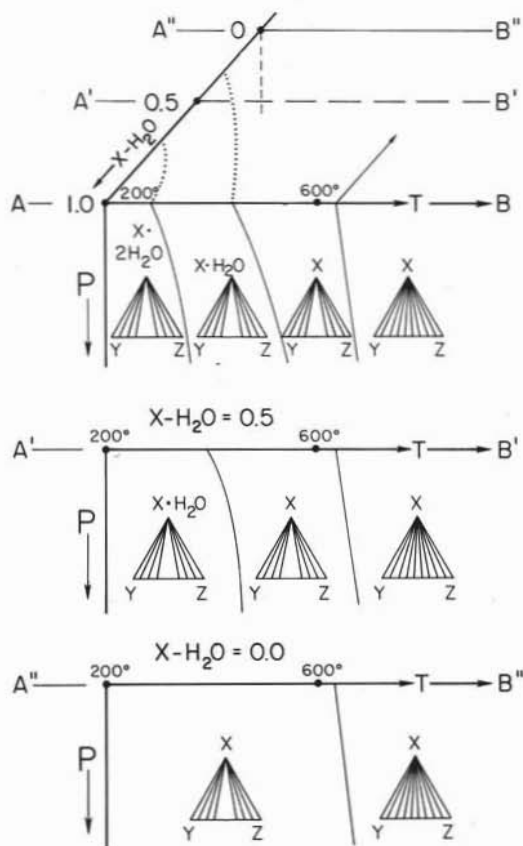


Fig. 6. — Schematic P, T, XH_2O petrogenetic grid showing hypothetical mineral facies diagrams separated by divariant reaction surfaces. Section AB and $A''B''$ below, show how the divariant reaction surfaces move to lower T with decrease of XH_2O in the fluid present during metamorphism.

papers by other workers have made this approach applicable to rock types other than only metapelites.

However, an extremely important aspect of this approach appears to have not been appreciated by most field petrologists and even many specialists in metamorphic petrology. The unappreciated aspect (i.e. establishing complete compatibility diagrams based on thin section study of rocks with a wide range of bulk composition within the system of concern) provides a means for establishing an unambiguous, rigorous framework of the metamorphic petrology of an area. It provides most of the essential information about the petrology of a group of rocks. Indeed, even in cases where the thin

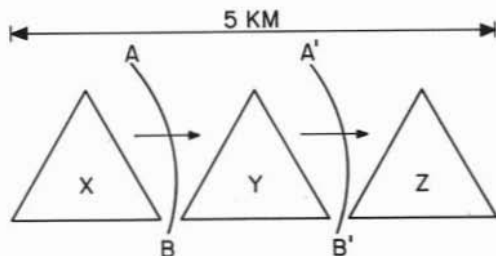


Fig. 7. — Schematic relationship of mineral facies diagrams with metamorphic isograds. The isograds represent earth-surface slices through divariant reaction surfaces as they exist in three dimensional space within the crust of the earth. Although intimately related to the divariant reaction surfaces shown in fig. 6, the isograds shown in fig. 7 should not be considered as simple slices through fig. 6. $A-B$ and $A'-B'$ are isograds separating topology X from Y from Z respectively.

section assemblages can not be plotted to form consistent compatibility phase diagrams, the approach leads to physio-chemically based suggestions of alternative possibilities such as: (a) lack of equilibrium, (b) highly variable Xf on a local scale, (c) the presence in some phases of significant amounts of components which are not part of the chemographic system being used, etc.

It should be noted that virtually everything discussed above can be done merely by thin section petrography — no elaborate equipment is required. In fact, complete compatibility diagrams should be established before attempting any sophisticated thermodynamic calculations etc. because it will, (a) enable definition of the appropriate components, (b) enable some assessment on whether equilibrium was achieved, and (c) enable defining the continuous and discontinuous reactions. Moreover, commonly the simple fact of establishing complete compatibility diagrams for a region provides most of the geologically relevant petrologic information. In many cases, the subsequent, more sophisticated approaches involving thermodynamic calculations etc., provide details which do not increase the geologic relevance of the data.

Considering the fact that a petrographic microscope will suffice for establishing complete mineral facies diagrams, it is unfortunate that so few field petrologists (e.g. workers who describe the metamorphism of

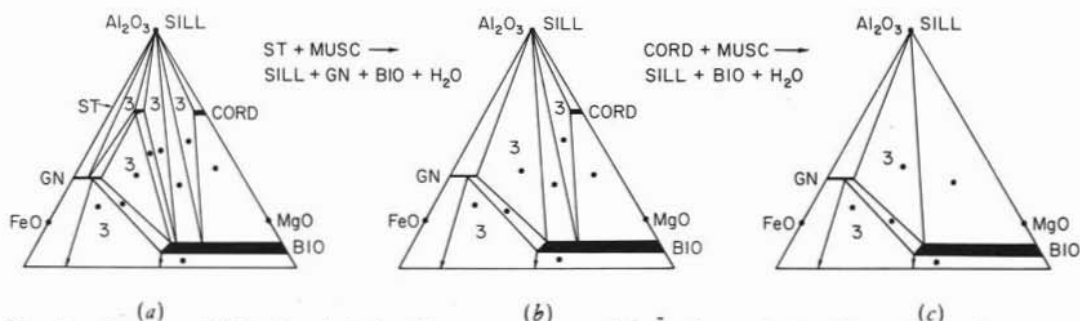


Fig. 8 *a, b, c*. — *AFM* mineral facies diagrams constructed from *observed* mineral assemblages in western Maine. Dots indicate *observed* assemblages. The 3's are to indicate 3 coexisting *AFM* phases in the particular triangular shaped area. Muscovite and quartz are present in all assemblages.

(a) Area 1.

(b) Area 2.

(c) Area 3.

an area as part of a geologic report) use this approach. This problem may arise because many field petrologists are not specialists in the details of metamorphism and so work in the context of metamorphic petrology as presented in the advanced textbooks. A survey of a number of post 1968 textbooks on metamorphic petrology (including the most commonly cited ones, WINKLER (1976, 1979), TURNER (1968, 1981), and MIYASHIRO (1973)), reveals that none of them develop fully the aspects emphasized above. This, despite the fact that all of them do go through a derivation of the *AFM* projection. It should be noted that WINKLER (1979, pp. 32-34) does briefly introduce the idea of a mineral paragenesis diagram in terms of a system which is thermodynamically valid. Moreover, he also discusses the use of changes in a compatibility diagram for deducing mineralogic reactions. However, no mention is made of the importance of constructing such compatibility diagrams based on natural assemblages. Of the various texts surveyed by the writer, only that of BROWNLOW (1979) (*a Geochemistry text!*) develops and emphasizes the concept of a mineral facies diagram based on *observed* assemblages. And although papers by specialists make fairly common use of the mineral facies concept, they naturally do not elaborate on the conceptual framework of the approach.

Finally it can be noted that no consideration has been given to continuous reactions and the compositional variation of solid solution minerals within the grade characterized by a given mineral facies diagram. Mineral facies diagrams are, of

course, essential for a meaningful understanding of continuous reactions. These topics are reviewed by SASSI (1983) in this volume.

An example of use of mineral facies diagrams

The example discussed here is based on work (GUIDOTTI and CHENEY, in preparation) in western Maine. It involves subdivision of what may be broadly termed as sillimanite zone. Fig. 8 shows the mineral facies diagrams which define three different grades of metamorphism and also the reactions that reflect the changes in mineral compatibilities. Recognition of these three different mineral facies was possible solely because a wide range of bulk compositions were sampled. For example, the Sill + Cord + Bio field on a quantitatively correct plot is very thin and lies very close to the Al_2O_3 -MgO side of the projection.

The example shown in fig. 8 illustrates several of the features discussed earlier in this preview.

(I) The mineral facies defined are independent of bulk composition.

(II) A much finer scale subdivision of metamorphic grade is obtained via mineral facies diagrams than would be obtained from a designation based on metamorphic facies or index minerals. All of the grades shown on fig. 8 would be sillimanite zone based on index minerals and amphibolite facies based on metamorphic facies.

(III) The nature of the isograd mineral

reactions, which relate the different mineral facies, can be read off directly from the tie line changes.

(IV) The general role of H_2O for each reaction is immediately apparent.

Suggestions for obtaining the required range of assemblages

It is evident that a wide range of bulk composition (within the system of concern) is required in order to obtain the assemblages required to define a mineral facies diagram. THOMPSON (1957) noted that many metapelites have sufficient compositional heterogeneity to enable collection of a considerable number of assemblages from a single outcrop. For example, many metapelites reflect original bedding which is graded and so ranges from quartzo-feldspathic at the bottom (i.e. relatively low Al_2O_3) to highly micaceous at the top (i.e. relatively high Al_2O_3). Similarly, different beds in a given outcrop will usually show some range in $Mg/Mg+Fe$ ratio and thereby increase the number of assemblages that can be obtained.

However, it may be useful to consider the following suggestions in order to obtain an even wider range of bulk compositions.

(1) Because increased oxidation state influences the amount of Fe that will go into oxide phases as Fe^{+3} , it also influences the bulk Fe^{+2} that is available to enter silicate minerals. If a large proportion of the Fe in a rock is « tied up » in oxides such as magnetite, the remaining silicate bulk compo-

sition will be relatively enriched in MgO and Al_2O_3 . CHINNER (1960) and HOUNSLOW and MOORE (1967) have described mineral assemblages reflecting this effort.

(2) In a similar fashion, increased sulfidation (indicated by increasing amounts of pyrrhotite or pyrite) causes a relative increase in the MgO and Al_2O_3 content of the silicate bulk composition. In the examples discussed above (fig. 8), the Sill + Cord + Bio assemblages occurs only in very sulfide-rich rocks. The effect of sulfide-silicate reactions on silicate bulk composition has been discussed by GUIDOTTI et al. (1977).

(3) Make use of unusual bulk compositions such as meta-bentonites.

Conclusions

The approach reviewed in this paper is one which is basically quite simply conceptually relevant information which can be obtained from metamorphic rocks. It also should be used to establish the framework within which more sophisticated procedures (e.g. geothermometry, geobarometry etc.) are carried out.

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