Using Equilibrium Thermodynamics to Understand Metamorphism and Metamorphic Rocks

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Retrograde reaction of garnet with matrix sillimanite to form a corona involving spinel and cordierite

Metamorphic rocks, formed at elevated temperature and pressure from pre-existing rocks inside mountain belts, provide a seemingly unpromising target for the application of equilibrium thermodynamics. This is because metamorphic rocks develop their mineral assemblages along a pressure–temperature (*P–T*) path, with pressure and temperature continuously changing along the path. However, in a successful model for the formation of such rocks, involving the essential role of fluid or melt, the mineral assemblages observed at the Earth's surface can be considered to reflect a state of frozen-in equilibrium as the rocks are exhumed towards the Earth's surface. Equilibrium thermodynamics applied to such mineral assemblages allow *P–T* information to be extracted. Currently the best way to do this is via calculated phase diagrams, the most powerful being *P–T* pseudosections. These diagrams portray the variation of mineral assemblages with *P–T* for a specified rock composition. Pseudosections allow the *P–T* conditions of the frozen-in equilibrium to be estimated, and can also give information on the *P–T* path followed. Such paths are an essential input in constraining the processes involved in mountain-building and the evolution of continental crust.

KEYWORDS: equilibrium in metamorphism, pseudosections, THERMOCALC, spinel stability

INTRODUCTION

Metamorphism involves the growth of new mineral assemblages in pre-existing sedimentary, igneous and metamorphic rocks, primarily under the influence of elevated pressure and temperature, and commonly associated deformation. In continental crust, the main environment in which metamorphism occurs is in mountain belts (orogenic belts), where thermally weakened crust is thickened as a consequence of compressive far-field stresses. In the modern Earth this occurs almost exclusively at convergent plate boundaries, where sedimentary and igneous rocks, as well as pre-existing metamorphic rocks (basement), are fed into developing orogens, as in the Andes, Alps and Himalayas. Because of the buoyancy of a plate composed of thickened crust, orogens are mountainous regions exposed to erosional processes. In time the crust is thinned by erosion to a more normal thickness, revealing the deeper levels of the orogen and the metamorphic rocks which formed there.

The basement of continental crust, beneath any obscuring sedimentary basins, is made up of the remains of old orogenic belts stitched together by continental collision related to the supercontinent cycle. Understanding crustal evolution, and in particular orogenic processes, involves reading the encoded geological record represented by the metamorphic rocks that make up the basement. Metamorphic rocks collected from ancient and modern orogens have followed a more or less complicated pressure (*P*) and temperature (*T*) path, from the Earth's surface, through the bowels of the orogen and back to the Earth's surface again. Commonly the only evidence for the *P–T* path that a rock has followed comes from its mineral assemblage and the textural relationships between the minerals. Yet *P–T* paths are a critical parameterisation of orogeny, as *P* paths reflect the depth history of rocks and *T* paths the thermal state as depth changed. Establishing *P–T* paths is essential to understanding orogeny and consequently the history of the continental crust.

The main tool for establishing *P–T* paths is equilibrium thermodynamics. First we present the rationale for using equilibrium thermodynamics, then we discuss the methods employed, with a focus on calculated phase equilibria.

DEVELOPMENT OF METAMORPHIC MINERAL ASSEMBLAGES

It is not self-evident that metamorphic rocks can be studied using equilibrium thermodynamics because what is observed in them (minerals, mineral compositions, textural relationships) develop along a *P–T* path with *P* and *T* changing more or less continuously. For the approach to be applicable, it is necessary to show that a part or all of a mineral assemblage in a rock represents a state of equilibrium that is preserved from a point along the *P–T* path.

The key is whether a metamorphosing rock has fluid or silicate melt along its grain boundaries, because their presence strongly promotes equilibration between the minerals, either directly or by allowing recrystallization in response to, for example, stress. Consider first a rock that has not been previously metamorphosed: most likely

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hydrous minerals and fluid are present. Even in nominally dry protoliths, such as basalt, hydration is commonly largely complete by the time the rock reaches low greenschist facies conditions. In the up-*T* (prograde) part of a rock's history, progressive maintenance of equilibrium on some scale is consistent with observation. Hydrous minerals break down continuously (dehydration), and – critically – much of the fluid thus generated is lost (under the influence of gravity, possibly aided by deformation). In the down-*T* (retrograde) part of a rock's history, fluid is not there to reverse the prograde dehydration, and the mineral assemblage tends to be preserved; thus, little happens during the retrograde history (unless fluid is reintroduced into the rock). Dehydration relates to steep, but not vertical, *P–T* lines of water content in mineral assemblages. Preservation of mineral assemblages tends to occur close to, but not at, the maximum *T* on the *P–T* path. These *P–T* conditions are commonly referred to as the metamorphic peak.

In this view, the mineral assemblage observed in such a first-cycle metamorphic rock at the Earth's surface is that preserved from the metamorphic peak. Assuming this is an *equilibrium* mineral assemblage, equilibrium thermodynamics can, in principle, be used to determine the *P–T* conditions of the metamorphic peak. In practice, metamorphic rocks commonly show features that reflect the prograde history (for example, zoning of minerals and inclusions that have survived by being armoured from further reaction by their host) and retrograde history (for example, retrogressive replacement of peak minerals). On the one hand, these features may give information about the *P–T path*; on the other, with appropriately nuanced petrographic interpretation, a "main" mineral assemblage may be identified and be the subject of phase equilibria modelling.

The identification of a "main" metamorphic mineral assemblage, and its use in phase equilibria modelling, can be extended to metamorphism that involved partial melting and to metamorphic reworking (metamorphism of previously metamorphosed rocks), with the presence or absence of fluid (or melt) being central. In the case of reworking, rocks tend to lack a fluid and may not respond at all to changing *P–T* conditions, unless fluid addition and/or recrystallization are involved. Reworking may be restricted to shear zones.

The appropriateness of treating metamorphic mineral assemblages as if they reflect a preserved equilibrium has underpinned much of metamorphic geology in the last half-century. Initially, the recognition of the recurrence of mineral assemblage associations (the metamorphic facies concept) suggested an equilibrium explanation and militated against the control of mineral assemblages by dominantly kinetic processes. More recently the equilibrium assumption has been largely implicit, but recent discussions in a calculated phase equilibria context include Guiraud et al. (2001), White and Powell (2002), Powell et al. (2005) and Powell and Holland (2008). The success of using calculated phase equilibria to study metamorphic rocks provides, at least in part, an *a posteriori* justification of the assumption.

APPLYING EQUILIBRIUM THERMODYNAMICS TO METAMORPHIC ROCKS

Given a justification for using equilibrium thermodynamics to consider mineral assemblages in metamorphic rocks, there is still a considerable step before being able to do useful calculations on them. Assuming that the rocks themselves are well understood – in terms of field relationships, relative timings via the deformation history, petrography, mineral-chemistry maps, absolute timing via radiometric methods, and so on – various inputs are needed for calculations.

The essential input is thermodynamic descriptions of the phases (minerals, fluids, silicate melt) involved in the development of the mineral assemblage in a rock. Such descriptions can be considered in terms of the thermodynamic properties of the end-members of phases, and then the energetics of the way these end-members are combined to form the phases (the activity–composition, or *a–x*, relationships). It was recognised in the 1970s that calorimetric data alone were not precise enough to constrain the thermodynamic properties of end-members for rock calculations because their uncertainties were too large. Out of this grew the idea of internally consistent thermodynamic datasets which aimed to combine, and make consistent, calorimetric data with constraints from experimentally determined phase equilibria, primarily from "reversed" reactions in simple systems (e.g. Helgeson et al. 1978).

Until the early 1990s, the main use of thermodynamic calculations was in so-called conventional thermobarometry (determining the *P–T* of formation), in which an observed mineral assemblage *and* its mineral compositions would be combined with reactions written between the mineral end-members to constrain *P–T*. In some studies the focus was on individual, specially calibrated, reactions (e.g. Ferry and Spear 1978), whereas in others it involved combining information from as many reactions as possible (e.g. the average *P–T* approach of Powell and Holland 1988, 1994). Various problems are involved in such *inverse modelling*, not least the dependence on mineral composition, an aspect of observations that is harder to defend in terms of a preserved equilibrium. As thermodynamic datasets improved in scope, it became possible to envisage *forward modelling* of phase equilibria, for example, for specified rock compositions (so-called pseudosections) that would provide thermobarometric information without suffering from this limitation, and also supply various other benefits (as summarised in Powell and Holland 2008).

It was obvious that internally consistent thermodynamic datasets would have to be augmented with less-well-constrained data – for example, from natural assemblage information – if forward calculations even in relatively simple systems were to be undertaken. For example, for calculations in the system KFMASH (K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O), given that essential minerals involve not only the $FeMg_{-1}$ substitution but also the Tschermaks ($MgSiAl_{-1}Al_{-1}$) substitution, data are needed for more than the experimentally well-constrained magnesian end-members. In 1990 came the first dataset that allowed calculations relating to rocks (Holland and Powell 1990), admittedly in fairly simple systems like KFMASH (e.g. Powell and Holland 1990). A significant update of this dataset is given in Holland and Powell (1998), and it is currently the *de facto* standard for metamorphic phase equilibria calculations.

In the 1990s, modelling metamorphic rocks using equilibrium thermodynamics was restricted to relatively simple systems by the thermodynamic descriptions of phases available then. At that stage, *P–T* projections (or petrogenetic grids), compatibility diagrams and pseudosections were used more or less equally. As summarised in Powell et al. (1998), *P–T* projections show all reactions and intersections of reactions for a chemical system, compatibility diagrams are constant *P–T* sections

showing mineral assemblage and mineral composition information (like AFM: Thompson 1957), and P–T pseudosections show the mineral assemblages for a given rock composition.

Pseudosections are currently the most powerful way of modelling (Powell and Holland 2008). A P–T pseudosection is effectively a P–T "map" of mineral assemblages for the specified chemical bulk composition. A T–x pseudosection is a T "map" of mineral assemblages, at a constant P, for a chemical composition vector, x. A P–x pseudosection is at a constant T. The composition vector can be used to investigate mineral assemblages as a function of poorly constrained variables such as H_2O or O_2, or to span a range of actual rock compositions. Choosing a chemical composition to use in a pseudosection requires care and an understanding of the likely scale of equilibration in the rock being considered. Accepting this, the particular advantage of pseudosections in modelling mineral assemblages and thermobarometry is that the results do not depend on the observed mineral compositions; as well, pseudosections provide a context for the observed mineral assemblages in terms of alternatives as the P–T conditions vary (Powell and Holland 2008). Box A illustrates the various modelling approaches.

The preeminence of pseudosections over other types of calculated phase diagrams is a consequence of the larger chemical systems that are now used in modelling. It is rarely possible to reduce the effective size of a larger system in order to draw compatibility diagrams. And P–T projections, in showing only reaction (invariant and univariant) information for a chemical system, are of little direct use because the great majority of rock compositions involve equilibria with less minerals than univariants, as observed on pseudosections. P–T projections in smaller systems may help in envisaging the topology of pseudosections in a larger system, particularly if they effectively constitute a "backbone" for that system (as KFMASH does for metapelites). But using smaller, petrologically unrepresentative systems for thermo-barometry will generally give unreliable or biased results.

Since 1990, the pseudosection approach has gained momentum, driving and being driven by advances in a–x models for the phases involved in metamorphism. A key step in a–x model development and the implementation of such models in software was provided by Powell and Holland (1993) and Holland and Powell (1996a, b). As new models have been developed and old models improved, the power and the range of applicability of the pseudosection approach have grown. For example, with the haplogranitic model of Holland and Powell (2001), and various extensions since then, modelling of pelitic and greywacke-composition granulite facies rocks, in which partial melting plays a key role, has become possible (e.g. White and Powell 2002). Using the framework provided by Evans and Powell (2006), pseudosections involving mixed-solvent aqueous fluids can be calculated. The new general model for the amphiboles of Diener et al. (2007) now allows amphibole-bearing rocks to be modelled. And the progress continues.

For modelling most metamorphic rocks, the model system of choice is currently NCKFMASHTO (Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–O), with CO_2 added for considering carbonate-bearing rocks, and Cl (and now S) for considering aqueous solutions. The idea is to use a chemical system that is as close as feasible to that of rocks, with the obvious constraint that thermodynamic data must exist for the end-members of the phases in that system. Another obvious candidate for inclusion is MnO, on account of its role in stabilising garnet, but the data for many Mn end-members of minerals are currently not up to date (they are currently being revised, however).

Among the software used to calculate pseudosections, THERMOCALC (Powell and Holland 1988; Powell et al. 1998) is based on a non-linear equation solver and calculates the lines and points of pseudosections directly; the coordinates are then manually assembled for drawing (by DRAWPD). Other software, for example, Perplex (Connolly 1990), are Gibbs energy minimisers and can be good at locating fields in pseudosections, but lines and points have to be found by interpolation, with small fields being easily missed. Petrologically it is the lines and points (and those small fields) that are significant, so THERMOCALC is better suited for pseudosection calculation, particularly in bigger systems like NCKFMASHTO. Box B illustrates the method of calculation in inverse and forward modelling.

PSEUDOSECTION CALCULATIONS: AN EXAMPLE

A metamorphic geology example is used to illustrate some of the ideas relating to phase equilibria calculations on rocks. Whereas pseudosections can be used for thermobarometry, they are additionally a powerful way of understanding the dependence of mineral assemblage on P–T and on chemical aspects of rocks that are difficult or impossible to observe, for example, oxidation state. In the example below, the mineral assemblage of this typical rock composition is a rather subtle function of P–T and ferric iron content.

In the transition from the amphibolite facies to the granulite facies, hercynitic spinel (sp) may occur in quartz-saturated metapelitic rocks, with for example cordierite – biotite – sillimanite (cd + bi + sill), and alkali feldspar, plagioclase, quartz and, at metamorphic conditions, silicate melt (ksp + pl + q + liq), and one or more Fe–Ti oxides. In simple modelling, for example in AFM (i.e. KFMASH + ksp + q + liq), cd + bi + sill is a readily understood mineral assemblage, and, as a result of the reaction bi + sill = g + cd with increasing temperature, such an assemblage makes

way for g + cd assemblages. Until the role of ferric iron was included in the modelling, the presence of sp + bi was difficult to put in context.

A calculated P–T pseudosection in the NCKFMASHTO system for an aluminous metapelite is shown in Figure 1. For this composition, a small proportion of all-Fe-as-FeO [(FeO$_{total}$) = 6.66] is considered to be Fe$_2$O$_3$, denoted by O = 0.07; thus the actual FeO = 6.66 – 2(0.07) = 6.52, and Fe$_2$O$_3$ = 0.07. All-Fe-as-ferric iron corresponds to O = 3.33, so O = 0.07 is x(O) = 0.07/3.33 = 0.021. Note the characteristic feature of pseudosections: boundary lines mark where a mineral appears or disappears (the blue line marks the limit of stability of spinel). Mineral modes (proportions) change continuously with P–T in such diagrams, and a boundary line occurs where a particular mode becomes zero. THERMOCALC calculates the lines, and the points where lines intersect, in terms of what modes are zero (to see how to do this, go to www.metamorph.geo.uni-mainz.de/thermocalc and hit the "elements" button). The results for an example calculation are shown in Box C.

In Figure 1, with x(O) = 0.021, there is indeed a stability field for sp + bi (field outlined in red), the mineral assemblage observed in low-P transitional granulites. The phase relationships at lower x(O) (<0.025) are shown by black dashed lines: spinel is not stable in this P–T range, in line with modelling in simpler systems, like KFMASH and NCKFMASH.

Another sort of pseudosection can be used to study the dependence of phase relationships on x(O): a T–x pseudosection, where the x is x(O) (Fig. 2). The example shown is calculated for 4.5 kbar, for the lower range of x(O). The sp + bi field (bounded by the red line) extends over a wide range of x(O). In Figure 2, note how magnetite is replaced by spinel as a carrier of ferric iron as temperature increases at a particular x(O). Note also that magnetite replaces spinel as a carrier of ferric iron at high T with increasing x(O). Indeed at much higher x(O) magnetite itself is no longer stable, because ilmenite, as it becomes more hematite-rich, can accommodate the ferric iron in a more energetically favourable way than magnetite.

An interesting way to consider the phase relationships for the relatively narrow range of P–T conditions in which the assemblage bi + sp is stable is via a compatibility diagram. The starting point is to say that cd + bi + sill with ksp + pl + q + liq is an expected mineral assemblage, and then to say what additional minerals can be involved with them. With all these phases "in excess", the phase relationships can be represented as in Figure 3, which is part of the Fe-Ti oxides triangle, FeO–Fe$_2$O$_3$–TiO$_2$ (personal communication K. Taylor-Jones). The focus is on the Fe-Ti oxides involved (ilmenite and magnetite), but it is also on spinel and garnet, which project onto this triangle. This shows that the "standard" cd + bi + sill assemblage can be joined by various combinations of ilmenite, magnetite, spinel and garnet, depending on the oxidation state and TiO$_2$ content. At slightly higher pressure, the crossing-tie-lines reaction sp + ilm = g + mt effectively precludes spinel from mineral assemblages in rocks, as is indeed reflected by observations in nature.

DISCUSSION AND CONCLUSIONS

Thermodynamic modelling of metamorphic rocks is currently in a vibrant state, but much still needs to be done. Obviously the quality of modelling can only be as good as the petrography, and this needs to be done with a phase equilibrium mindset so that the potential meaning of textural relationships can be properly ascribed. Advances
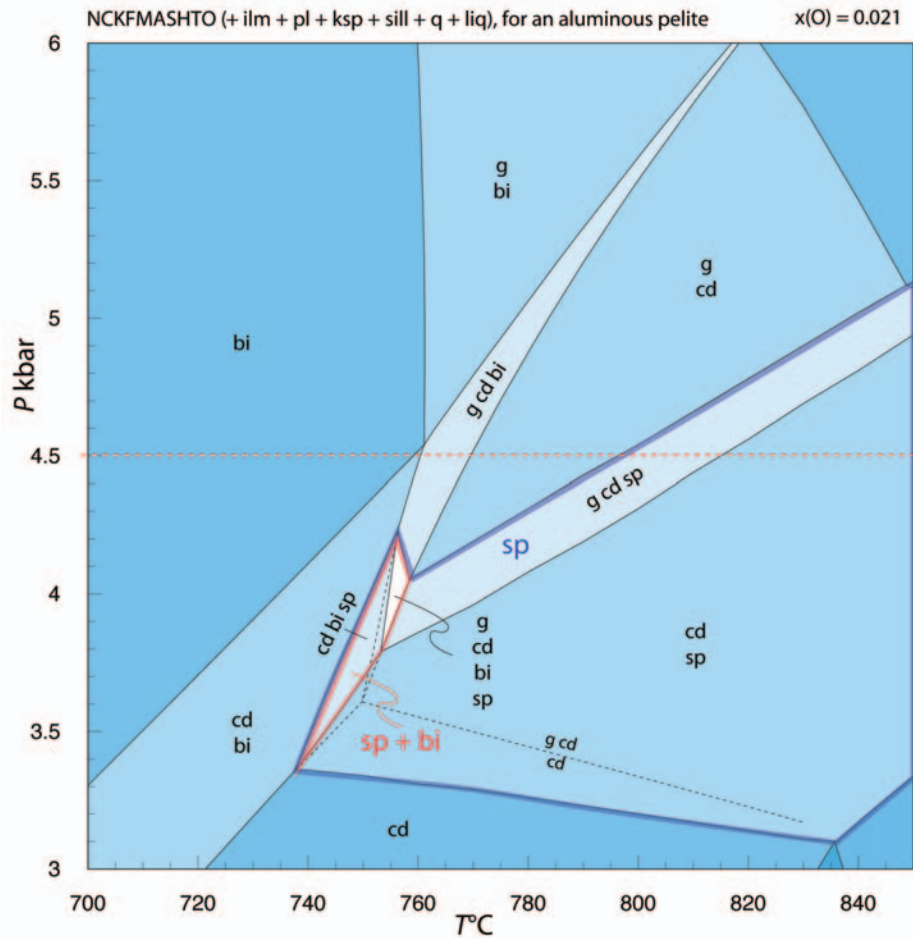
FIGURE 1 A calculated *P–T* pseudosection in the system NCKFMASHTO for the same bulk rock composition (aluminous pelite) as used by Powell and Holland (2008, Fig. 1): in mole %, SiO_2 = 65.47, TiO_2 = 0.65, Al_2O_3 = 14.49, CaO = 0.49, MgO = 2.55, FeO_{total} = 6.66, K_2O = 2.8, Na_2O = 1.17, O = 0.07, and H_2O = 5.65. In this composition, the O corresponds to a small proportion of Fe_2O_3, meaning that actual FeO = 6.66 – 2(0.07) = 6.52, and Fe_2O_3 = 0.07. The blue line marks the stability limit of spinel and the red line marks the stability field of spinel and biotite. Abbreviations: g = garnet, cd = cordierite, bi = biotite, sill = sillimanite, sp = hercynitic spinel, ilm = ilmenite, mt = magnetite, pl = plagioclase, ksp = alkali feldspar, q = quartz, liq = silicate melt. The red dashed line at *P* = 4.5 kbar corresponds to Figure 2. See text for discussion.
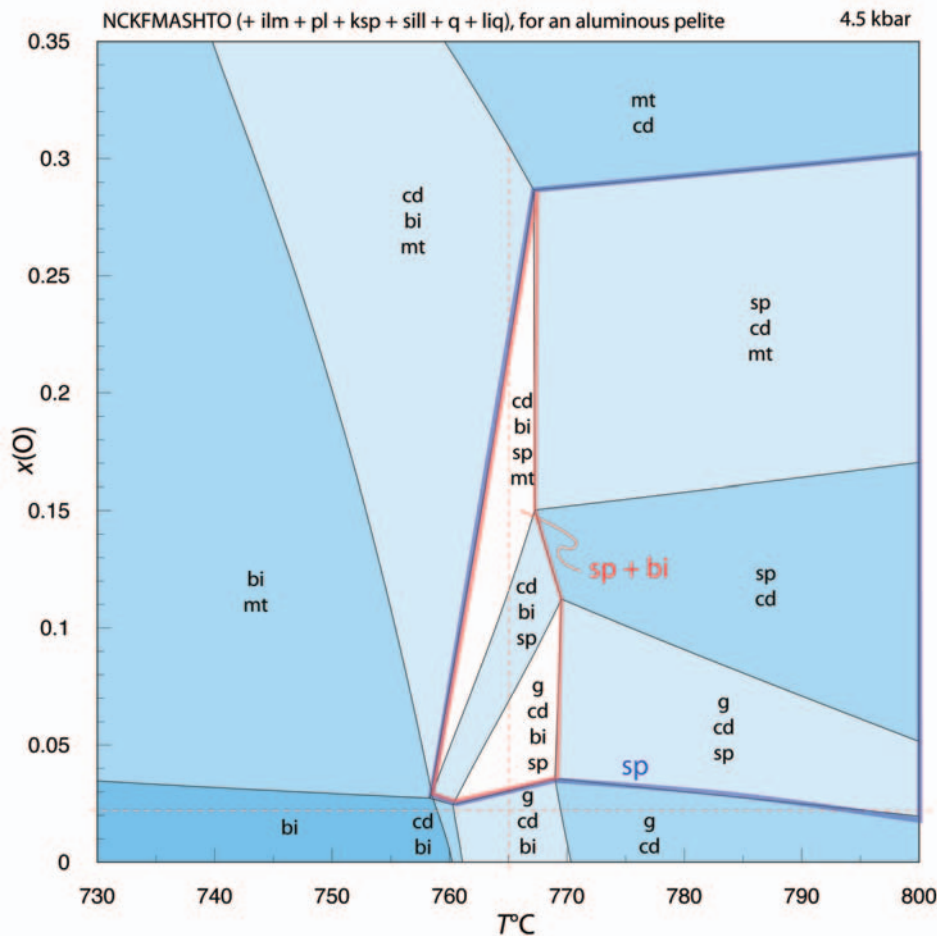

FIGURE 2 A calculated *T–x* pseudosection in the system NCKFMASHTO at *P* = 4.5 kbar for the same bulk composition as in Figure 1 but with O varying from 0 to 3.33, corresponding to *x*(O) = 0 to 1. The horizontal dashed line corresponds to the *x*(O) of Figure 1, and the vertical line relates to Figure 3. See caption of Figure 1 for abbreviations. See text for discussion.
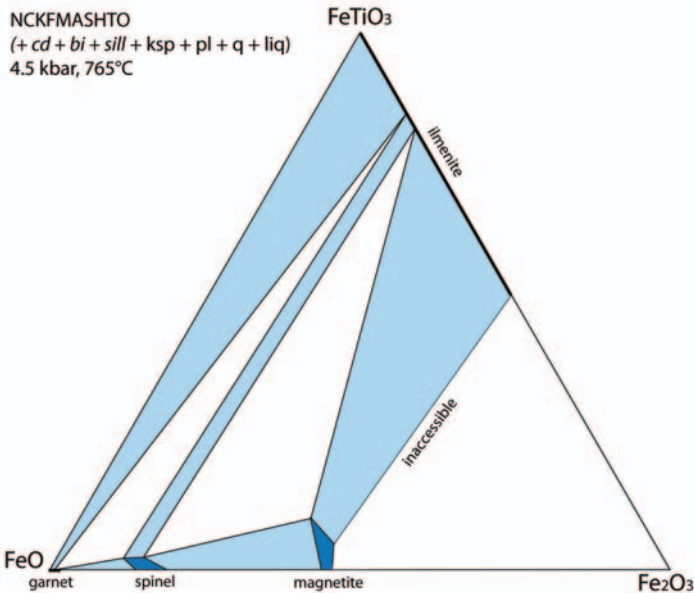
NCKFMASHTO
(+ cd + bi + sill + ksp + pl + q + liq)
4.5 kbar, 765°C

FeTiO₃

ilmenite

inaccessible

FeO

garnet   spinel   magnetite   Fe₂O₃

FIGURE 3 A calculated $FeO–Fe_2O_3–FeTiO_3$ compatibility diagram at 4.5 kbar and 765 °C in the system NCKFMASHTO, in projection from the calculated compositions of bi + cd + sill + pl + ksp + q + liq. See caption of Figure 1 for abbreviations. See text for discussion.

particular, mineral compositions evolve during metamorphism, and are then preserved. In this way it is hoped that the subtle effects preserved from the prograde history and superimposed during the retrograde history can be used to extract more reliable *P–T* path information, in addition to the *P–T* of the metamorphic peak provided by the "main" mineral assemblage.

Such a view of metamorphic rocks is resolutely a *P–T–x* one. However, depending on the processes operating during metamorphism, volume (*V)* rather than *P* might be the more appropriate variable, for example. In the context of such conjugate pairs of variables, more useful phase diagrams may be calculated (Powell et al. 2005). A particular aspect currently being developed involves considering chemical potentials rather than compositions. This procedure is useful in studying textural relationships among minerals in which, for example, equilibration was in progress when the relationships were preserved, as in corona development during cooling (White et al. 2008; see also the photomicrograph on page 309).). In combination with quantitative modelling of diffusion, such an approach has the capacity to inform understanding of equilibration and indeed, how mineral assemblages evolve and are preserved (Carlson 2002).

In the thermodynamic modelling itself, better thermodynamic descriptions (both end-member properties and activity–composition relationships) of phases are needed, extending also into chemical systems not currently available to study. At least some of these advances will come via natural mineral assemblage calibration, for example, where calibration by experiment is impossible or prohibitively slow. ◾

in mineral–chemical imaging have the capacity to help greatly in this. As more becomes known, progress will be made in understanding how mineral assemblages and, in

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