Short Communication

Exhalative metasediments – clues to the real nature of regional metamorphic processes?

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Summary

Regional metamorphic petrogenetic theory is currently dominated by the hypothesis that the principal grade indicator silicates and their related minerals arise by a variety of chemical reactions. It is postulated that, with rise (or fall) in regional crustal temperature and pressure, original materials become destabilized, break down, and are dissolved in the intergranular "metamorphic fluid". The resulting ions then diffuse within, and are transported by, this fluid, in due course reacting with each other to form new minerals in equilibrium with each other and the associated fluid under the new conditions and in accordance with the Phase Rule. A second mechanism, widely accepted for such as metamorphic oxide, carbonate, sulphate and sulphide minerals, but not for the principal metamorphic silicates, is that the latter form largely by in situ solid:solid transformation plus or minus SiO_2 plus or minus H_2O, rather than by the serial process indicated above. It is now proposed that a test of the "reaction theory" is whether or not chemical equilibrium has been established on a fine (grain) scale in materials metamorphosed to high grades. On the basis of some simple – but very precise – electron microprobe measurements on some high grade, but still finely bedded, exhalative metasedimentary rocks, it appears to fail the test.

Introduction

Apart form the "porphyry coppers" the majority of the world's great ore deposits occur in the metamorphosed rocks of the Precambrian shields and of

the Palaeozoic metavolcanic and metasedimentary (palaeo-island arc) belts such as those of the Caledonides, Appalachians, Urals and the Eastern Highlands Belt of Australia. Patterns of ore occurrence within these are commonly linked to pre-metamorphic features of their host terrains, and so a capacity to see through the complexities of metamorphic change to perceive the original nature of the ore environment becomes a fundamental aspect of mineral exploration. An understanding of metamorphic processes, particularly regional metamorphic processes, is therefore of importance not only to pure science – metamorphic petrology – but also to its application in the increasingly difficult search for industrial mineral resources.

There are two principal postulated mechanisms of metamorphic silicate formation in nature.

The first, and by far the longer and more widely held as far as the principal metamorphic grade indicator minerals are concerned, is that with significant increase in temperature and pressure pre-existing materials destabilize and decompose; that they dissolve in the pore fluid and move by fluid flow and/or diffusion, the resulting simple and complex ions then reacting with each other to form a new mineral assemblage in chemical equilibrium and in accordance with the Phase Rule. This has formed the basis of much experimental work and the extensive application of thermodynamic theory.

However doubts concerning large scale movement of mineral matter in metamorphic fluids have existed for over a century (*Harker* and *Marr*, 1893). Unambiguous microstructural evidence of regional metamorphic mineral reaction is elusive (*Blackburn*, 1968; *Carmichael*, 1969; *Kwak*, 1974). As pointed out by *Yoder* (1955) some 50 years ago, there is little evidence in nature of any progression in metamorphism; that any given prograde metamorphic mineral assemblage has in fact been consumed in the production of another. With the advent of refined microanalytical techniques, examples of apparent disequilibrium mineral compositions and assemblages are appearing with increasing frequency. Related to this is the more recent recognition that metamorphism of materials containing an exhalative or subhalative "chemical" component may yield highly complex silicate assemblages in which all the standard "Barrovian" (*Barrow*, 1893) grade indicator minerals (chlorite, biotite, almandine, staurolite, kyanite, sillimanite) occur together, in prograde form, within volumes down to a small fraction of a cubic centimetre (*Stanton*, 1979).

The second, rather more recent – and, as far as the principal metamorphic silicates are concerned, more-or-less disregarded – hypothesis is that in many cases there may be little transposition of matter involved in regional metamorphic processes (*Stanton*, 1976a, 1982). Rather than a sequence of temperature-pressure induced destabilization and decomposition of the prevailing mineral assemblage $\rightarrow$ solution $\rightarrow$ ionic migration $\rightarrow$ reaction $\rightarrow$ establishment of new equilibrium mineral assemblage, the second theory holds that any transposition of matter is limited to a minute scale – that of the relevant grainsize and commonly of the order of a millimetre or less – and that apart from the variable expulsion or addition of SiO_2 and H_2O from or to developing mineral structures, chemical reaction is the exception rather than the rule. The mechanism proposed is one of solid:solid transformation, plus or minus H_2O and the variably ordered, variably hydrous, SiO_2 that is semi-ubiquitous in the regional metamorphic milieu.

Apparently supporting this hypothesis, the increasing refinement of microanalytical techniques over the past 30 years has led to the frequent detection of significant between-grain and non-zonal within-grain compositional variation of individual minerals over distances of 0.1 mm or less in material that has been metamorphosed to high grades (*Stanton*, 1989). Further, early microanalysis along and across beds of finely bedded metasedimentary materials containing an exhalative chemical component has shown what appears to be the preservation of delicate pre-metamorphic (i.e. chemical sedimentary) features through high (sillimanite) grade regional metamorphism (*Stanton* and *Williams*, 1978). In the across-bed sense, fine garnets in material of this kind have been found to exhibit gross concentration disequilibria over distances of the order of 1.0–0.1 mm indicating a lack of metamorphic transposition over distances greater than this.

Such observations indicate that the second proposed mechanism – one closely akin to solid–solid transformation – may well be the process involved in some cases (*Stanton*, 1976b). The present contribution therefore extends the earlier work on high metamorphic grade, but still well-bedded, metasedimentary rocks generated in the exhalative regime. The bedding – in some cases of fineness down to less than 0.1 mm – provides spatial reference, i.e. markers, against which any non-attainment of concentration equilibrium may be measured with high precision. Investigation has been directed exclusively to rocks that have undergone the highest (sillimanite) grade of metamorphism, as it is in these that chemical – concentration – equilibrium is most likely to have been achieved – and hence, by implication, that the most extensive metamorphic movement and the closest approach to chemical equilibrium is likely to have occurred. Indeed and self-evidently, if significant movement and a close approach to concentration equilibrium between adjacent grains of an individual mineral species in a common matrix has not been attained here, it is unlikely to have been attained in lower grade regimes.

Material investigated

The materials now reported upon have been sampled entirely from diamond drill core and are as follows:

1) Finely bedded quartz–garnet–(magnetite–apatite) rock, together with enclosing quartz–feldspar–muscovite–biotite–garnet–sillimanite gneiss, from the Broken Hill district, New South Wales (*Stanton*, 1976b). (Depending on the relative abundance of magnetite in the bedded rocks, these range from what are known locally as "bedded iron formation" to "garnet quartzite".)

2) Somewhat vaguely bedded quartz–feldspar–muscovite–biotite–staurolite–kyanite–sillimanite rock from the Canadian Abitibi Belt near Hemlo, on the north shore of Lake Superior, Ontario (*Johnston*, 1996).

3) Well-bedded (very sharply defined, fine bedding of the order of 2.0–0.1 mm thickness) quartz–muscovite–biotite–magnetite–garnet–staurolite rock (interbedded with kyanite- and sillimanite-bearing units) from the vicinity of the volcanic massive sulphide deposit of the Gorob Mine, Namibia (*Stanton*, 1982).

Results

1. Between- vis á vis within-bed compositional features

Some 25 years ago the author, with K. L. Williams (*Stanton* and *Williams*, 1978), showed that the compositions of garnets of some of the finely bedded garnet quartzites of Broken Hill were extremely uniform along beds but liable to substantial variation from one bed to the next. The investigation now reported confirms this earlier finding. Figure 1 shows the result of a more comprehensive and refined series of analyses than that attempted by Stanton and Williams and involving two beds of essentially unzoned, fine garnets set 1.0–1.5 mm apart and sharing the one quartz matrix (Fig. 2a). Within-bed compositional variation in each case is barely more than analytical precision. However mean MnO of the garnets of Bed 1 is 16.66 wt% (S.D. = 0.75) and of Bed 2, 10.84 wt% (S.D. = 0.60) – a difference of 54% of the lesser figure – and the two populations are unambiguously distinct. The length of each analysed bed (i.e. as these appear within the confines of the relevant polished thin section) is ca. 2 cm – approximately 15–20 times the mean distance between the two beds. In the earlier investigation the authors conceded that the constancy of composition of the garnets along beds might well be interpreted as indicating essentially unrestricted diffusion and consequent compositional equilibration within the relevant layers. However they noted that this constancy prevailed within beds through distances over which there were marked differences between beds. They considered that as some of the analysed garnet beds were in direct contact (being delineated from each other only by grainsize of more subtle features of appearance) it could not be argued that whereas transposition of matter might occur unimpeded along a mono-mineralic bed, it might be impeded between beds separated by other mineral species. Thus one was confronted with the proposition
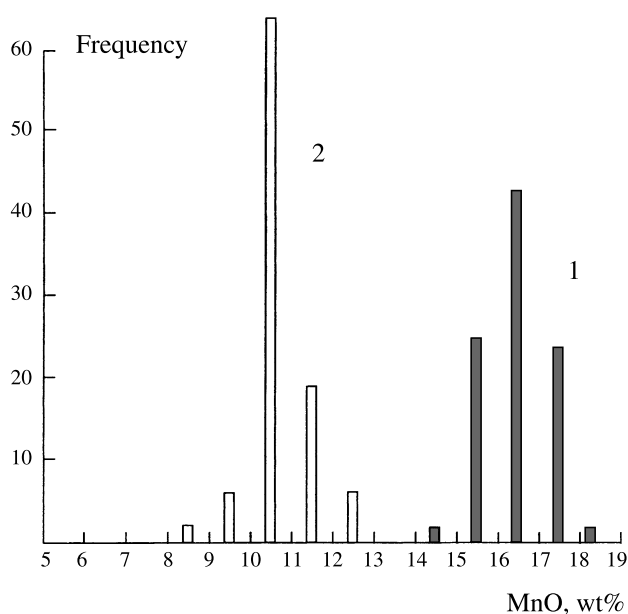

Fig. 1. MnO contents of (1) 95 garnets analysed along the length (one to two grain thickness) of bed 1 and (2) 92 garnets analysed similarly along bed 2 of the quartz–garnet–magnetite–apatite exhalative metasediment (banded iron formation-garnet quartzite) of Fig. 2a
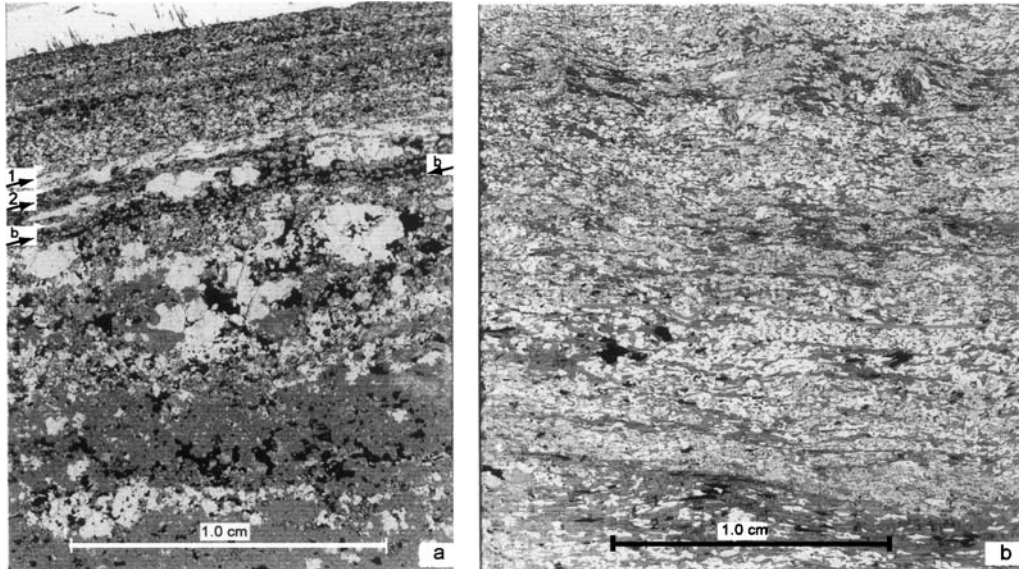
Fig. 2. **a** Macrophotograph of bedded "banded iron formation-garnet quartzite" (upper third of photograph) with juxtaposed quartz–feldspar–muscovite–garnet–magnetite gneiss (lower two thirds of photograph) in diamond drill core from Broken Hill, New South Wales; "b" indicates approximate boundary between the two lithologies, 1 and 2 the beds along which the garnets of Fig. 2 have been analysed **b** Macrophotograph of layered quartz–feldspar–chlorite–muscovite–biotite–garnet–staurolite–kyanite–sillimanite rock in diamond drill core from the Hemlo area, Ontario. Populations 1 and 2 of Fig. 3a and b represent the biotites of the domains above and below the blue line respectively

that if inter-garnet diffusion had in fact occurred, it had done so in one direction with respect to bedding but not the other. This they considered to be untenable, leading to the inescapable inference that metamorphic diffusion had been restricted to distances less than their intergrain measurements, and that the layered compositional pattern now observed must be a direct reflection of original bedding.

The data of Fig. 1 would appear – at first sight, at least – to confirm these earlier prognostications, and to demonstrate the relevant principle at an even higher level of certainty.

However more recent work involving hydrodynamic modelling of fluid flow in variably permeable media has been put forward as indicating that individual beds of a sedimentary sequence may each be characterized by an intergranular solution of distinctive composition, separate and distinct from the compositions of the intergranular solutions of adjacent and nearby beds. Thus the metamorphic mineral assemblage of any individual bed is postulated to develop in equilibrium with the particular intergranular solution that permeates that particular bed – not necessarily in equilibrium with the developing metamorphic mineral assemblage of an adjacent bed or with the intergranular solution permeating the latter. The concept of metamorphic equilibrium is in this way considered to be upheld: in any given lithology characterized by more than one distinct compositional domain, metamorphic equilibrium pertains – but only within each domain, not between them.

This gives formality to a general view held widely for at least most of the 20th century.

However, can such a postulate really account for the data of Fig. 1? The two garnet layers represented in the histogram and shown in Fig. 2a are not components of two adjacent but distinct mineral assemblages as, for example, noted by *Turner* and *Verhoogen* (1960, p 456); on the contrary, they are simply contiguous parts of a single assemblage quartz–garnet–(magnetite–apatite). Despite this their garnet MnO, although extremely constant within each layer, differs by more than 50% between layers. As the two layers effectively share a common matrix, such a gross and clearly-defined compositional difference seems to indicate – quite inescapably – within-domain disequilibrium that cannot be attributed to synmetamorphic hydrodynamic factors.

Such a deduction seems to be confirmed by consideration of the thickness and continuity of many of the garnet layers associated with the two beds of Figs. 1 and 2a. Some of these are not more than one grain thick, and in such cases are commonly discontinuous on a fine scale; beds "tail out" into a string of small garnets isolated from each other along the "horizon" of the layer concerned and then, within a centimetre or so, resume continuity. That a fluid might maintain a distinctive composition along a layer a single grain and no more than 0.1 mm thick and, here and there, of no thickness at all, seems impossible to sustain.

We may in fact consider the "no thickness at all" case a little more closely by examining material in which substantial change in mineral chemistry occurs unaccompanied by any visible change in the mineralogical nature of the rock concerned. An example of this comes from a diamond drill core intersection of quartz–chlorite–muscovite–biotite–feldspar–almandine–staurolite–kyanite–sillimanite (all of these occur within a few centimetres of each other) rock from the environs of the Hemlo gold mine, situated in the Precambrian Abitibi belt on the north shore of Lake Superior in Canada. The material subjected to microprobe analysis is characterised by a rather poorly-defined layering that, in spite of its somewhat vague definition, is relict bedding.

One hundred and seventeen biotite flakes in a zone dominated by quartz–biotite were analysed by the electron microprobe using energy dispersive spectrometry. Although there were no visual features (apart from a very subtle colour difference discerned only following analysis) to indicate the fact, biotite compositions generated two domains separated by the blue line in Fig. 2b, and illustrated by the TiO_2 and MgO frequency distributions of Fig. 3. There is no visible discontinuity along this line and, apart from the change in composition of the biotites, nothing to indicate a now-invisible one. As far as the writer has been able to discern, there is no indication of any feature along this line that might have been conducive to the preservation of two juxtaposed intergranular solutions with which, respectively, two compositionally distinct biotite populations might have grown in equilibrium. If this is the case the biotite of the two domains – two domains composed of identical mineral assemblages, with identical grainsizes and microstructures, in immediate contact, and with no apparent physical feature delineating one from the other – have formed and remained in a state of chemical disequilibrium with each other in material metamorphosed to sillimanite grade.
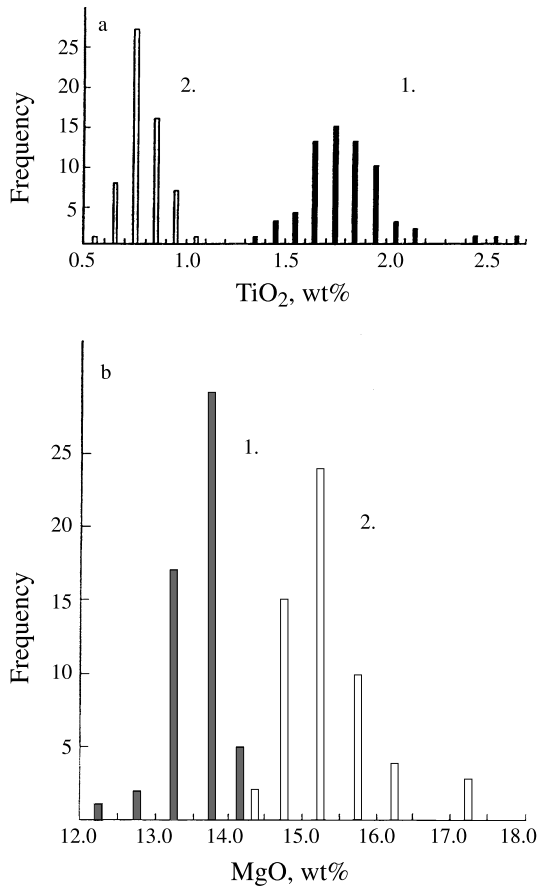
Fig. 3. Frequency distributions of **a** TiO$_2$ contents of (1) 54 biotite flakes of the domain above, and (2) 63 biotite flakes of the domain below the blue line of the material of Fig. 2b, and **b** MgO contents of the same biotite flakes

The fine, semi-continuous layering of the Broken Hill garnet quartzites may – with the exercise of considerable credulity – justify a flimsy case for the separation of intergranular solution and the preservation of distinctive small scale domains of equilibrium, but the Hemlo material does not. The latter seems to indicate that individual grains of a single mineral, with essentially nil separation, may grow – or persist – out of equilibrium with each other under highest grade regional meta-morphic conditions.

If this be accepted, the clear implication is that pronounced concentration dis-equilibria may be initiated and preserved over minute (less than 0.1 mm) or nil distances through prolonged, high (sillimanite) grade regional metamorphic epi-sodes. In such cases it inevitably follows that ionic or elemental movement – by diffusion or fluid flow – has been limited, in the examples concerned, to distances of the order of 0.1 mm, or has not occurred at all.

If however this is not conceded there are two further, more powerful ways of attacking the problem; the examination of subtle, grain-by-grain compositional variation of a single mineral species along a single layer, and of variation of elemental partition coefficients between contacting grains of a given mineral pair along a single layer.

2. Along-layer compositional variation among individual grains
of a single mineral species

While we have seen that there is a strong tendency to uniformity of composition
of a particular mineral along bedding, even the sharply defined populations of
Fig. 1 involve some degree of within-bed variation (it is the distinctiveness of
the populations there that is significant; not that the populations themselves are
invariant).

  Three examples of significant along-layer variability are shown in Fig. 4.

a) Trace barium in 40 well-developed muscovite grains analysed by wavelength
   dispersive spectrometry along approximately 4.0 cm of a sharply defined
   quartz–muscovite–biotite–almandine–staurolite-bed in the Gorob Mine se-
   quence, Namibia (mean distance between grains approximately 0.1 mm). This
   material is closely associated with kyanite-sillimanite bearing units in the relevant
   diamond drill core. Within the analysed bed length, barium varied from 2441 to
   5517 plus or minus 100 ppm, as shown in the frequency distribution of values of
   Fig. 4a. The spread of values is 3076 ppm, or 126% of the lower value.
b) Trace iron in 132 well-developed kyanite grains analysed along approximately
   2.5 cm of a conspicuous kyanite-rich bed (represented by three serial sections in
   order to expose sufficient kyanite grains) within a quartz–muscovite–biotite–
   staurolite–garnet–kyanite–sillimanite assemblage from the environs of the
   Hemlo Mine referred to above (mean distance between grains approximately
   0.56 mm). Measured Fe values ranged from 1024 to 7766 plus or minus
   100 ppm as shown in Fig. 4b. The full spread of values is thus 6742 ppm or
   some 6.5× the lower value. If the extreme value of 7766 ppm be arbitrarily
   discarded (though confirmed by repeat analyses) the spread becomes
   3880 ppm, or approximately 4.8× the lesser value.
c) Trace iron in 100 idiomorphic cross-sections of sillimanite crystals (cores
   and rims) in a single gneissic swathe of this mineral, approximately 2.5 cm
   long, in a quartz–feldspar–muscovite–biotite–garnet–sillimanite gneiss from
   Broken Hill, NSW. Measured Fe values ranged from 837 to 2378 plus or minus
   80 ppm, as shown in the frequency distribution of Figure 4c. The spread of
   values is thus 1541 ppm, or approximately 1.8× the lesser value.

As proportions of the trace element abundances concerned, the variations involved
in these three examples are gross, indicating the non-attainment of along-bed
concentration equilibrium between individual grains of a single mineral over
distances of 0.05–0.5 mm.

3. Along-layer variation in trace element partitioning between
individual pairs of grains of a given mineral pairing

The example presented here (Fig. 5) is the partitioning of trace barium between
biotite and the muscovite of Figure 4a. The relevant bed exhibits abundant biotite–
muscovite pairings involving relatively large, clear, grains in sharp, clearly-defined
contact with each other. Each pair of 40 paired analyses were carried out
at approximately 2–3 microns on either side of each muscovite–biotite mutual
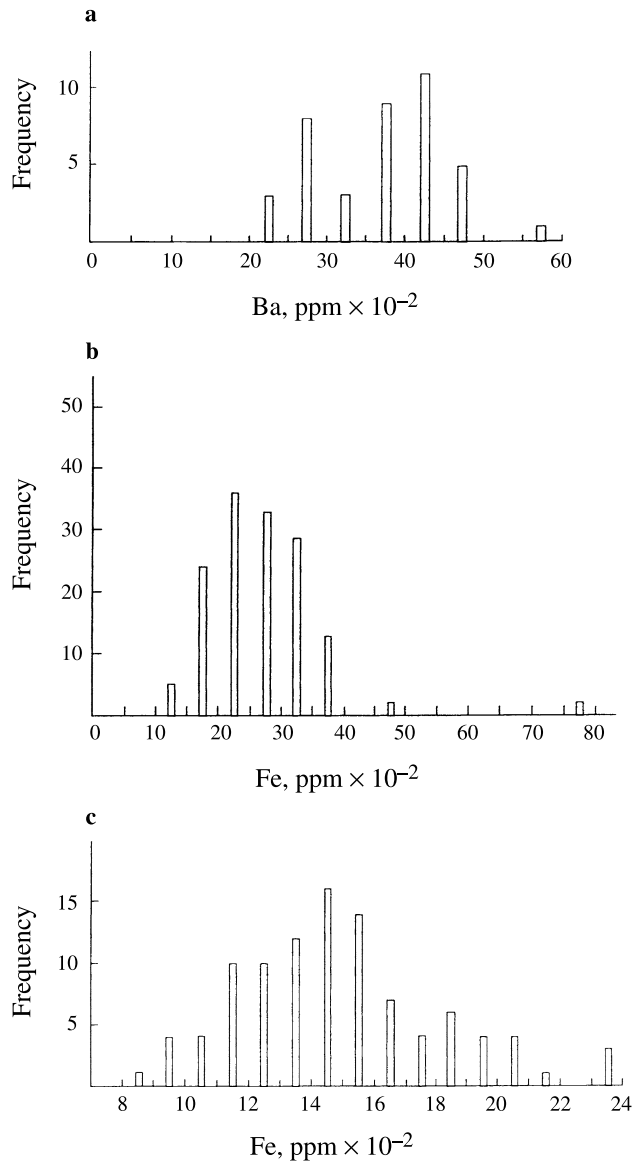
Fig. 4. **a** Frequency distribution of barium abundances in 40 muscovite grains analysed along a 4.0 cm bed length in finely bedded quartz–chlorite–muscovite–biotite–garnet–staurolite–kyanite rock in diamond drill core from the environs of the Gorob Mine, Namibia **b** frequency distribution of the abundances in 132 kyanite grains analysed along a 2.5 cm bed length of material approximately 2.0 cm from that of Figs. 2b and 3 in diamond drill core from Hemlo, Ontario and **c** frequency distribution of iron in 100 idiomorphic cross-sections of sillimanite in diamond drill core from Broken Hill, New South Wales

boundary, and at approximately 0.1 mm intervals along the bed from one end of the polished thin section to the other.

Values of K_D muscovite/biotite with respect to barium ranged from 1.69 to 4.68 and their frequency distribution is as shown in Fig. 5. The muscovite:biotite pairs immediately adjacent to that yielding $K_D = 4.68$ (i.e. at approximately 0.1 mm on either side of it) gave K_D of 1.95 and 2.71; thus the pairings $K_D = 4.68$ and 1.95 involve a variation in partition coefficients of 1.4× the lesser figure over a distance of 0.1 mm. Analogous and much more comprehensive investigations (to be reported elsewhere) that include consideration of along-layer variability in the partitioning of Ti, Mn, Cr and Zn between contacting
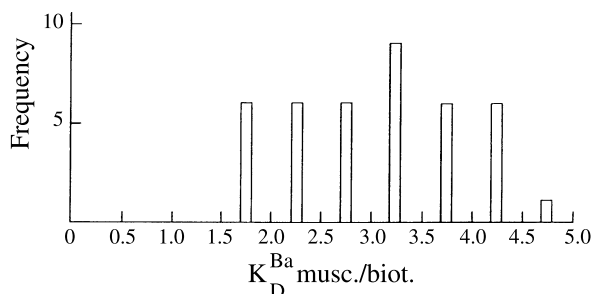
Fig. 5. Frequency distribution of values of K_D muscovite/biotite with respect to barium in 40 contacting muscovite–biotite pairs along the 4.0 cm bed length of the quartz–muscovite–biotite–garnet–staurolite bed of Fig. 4a

metamorphic hornblende:biotite pairs, and of Zn between garnet:magnetite and garnet:chlorite pairs in upper amphibolite grade materials, have given analogous results.

Such a marked variability of partition coefficients, involving pairs of analyses carried out no more than 4–5 microns apart, confirms in principle the results of 2. above.

Discussion

Whether, here and there, regional metamorphism has been isochemical, or whether it has involved the transposition of material over significant distances, has constituted a long-standing controversy in petrology. Its resolution has been impeded by a lack of sharply defined markers, against which movement of constituents might be measured, in most metapelitic rocks. Such resolution is important not only for the improvement of scientific understanding and all that follows from that, but also because the very large technical resources employed in mineral exploration are so heavily dependent on a proper understanding of regional metamorphism and its effects on primary ore environments.

The present investigation has attacked the problem by examining elemental concentrations – and the preservation of composition contrasts – in finely layered metapelites occurring in what are accepted as "high grade" regional metamorphic terrains. High grade materials are, clearly, those in which metamorphic transposition of matter and the consequent attainment of concentration equilibrium is most likely to have occurred. The fine layering provides accurate spatial control for the measurement of distances over which such concentration equilibrium has or has not been attained.

It is apparent from the evidence now presented that concentration equilibria of the elements in the materials concerned has not been attained down to a scale of the order of 1.0–0.1 mm. That is, there has been insufficient metamorphic transposition of matter to achieve equilibrum over such distances, minute thought those these are. But, as already emphasized, the materials investigated have been subjected to what in Barrovian terms are the highest grades of regional metamorphism. If

significant elemental or ionic movement has not occurred under these conditions, does it occur at all? The inevitable answer is that any supposition of large scale movement (centimetre to metre and greater) as a regional metamorphic process must be seriously questioned. That is, the process that is presumed to facilitate metamorphic reaction and the production of new metamorphic minerals may not in fact take place.

If this is the case, can reaction be the real process leading to new regional metamorphic mineral formation, or are we to accept what seems the inescapable implication of the "isochemical" metamorphism so frequently deduced by metamorphic petrologists – that, plus or minus H_2O and the semi-ubiquitous free SiO_2 of most metamorphosing materials, the new minerals arise in situ by processes of solid–solid transformation, grain boundary migration and consequent grain growth. Such a mechanism has been accepted as self-evident in the case of regional metamorphic oxides (including quartz), carbonates, sulphates and sulphides. The data presented here indicate that essentially similar processes play an important role in the development of the principal regional metamorphic silicates.

On such a basis the chemical and mineralogical constitutions of regionally metamorphosed rocks must reflect very closely the original nature of the metamorphosed materials down to a fine scale. Recognition of this should facilitate much more refined interpretation of metamorphosed terrains – the detailed nature of original materials, and, in the case of sedimentary and volcanic-sedimentary milieu, determination of sedimentological conditions, and perhaps, of provenance, palaeogeography and palaeoclimate. The indentification of the "palaeoclimates" of seafloor exhalative ore environments should be one of the simpler problems to confront the metamorphic petrologist and mineral explorer, and should lead to much greater effectiveness and accuracy in the general recognition of primary lithological environments as guides to ore occurrence. Could it be that both science and industry would benefit if the problems of regional metamorphic petrology were less in the purview of the metamorphic petrologists and more in that of the sedimentologists – particularly those with a special interest in sediments with a significant chemical and volcanic component?

It must of course be accepted, indeed emphasized, that such prognostications are based on very limited sampling. Although obtained from three continents, the material constituting the basis of the present investigation is no more than a minute sampling of the localities concerned, and an almost infinitesimally small sampling of the world's shields and associated younger metamorphic belts. Nonetheless the results reported do seem to call for the much wider application of more precise methods of testing and evaluating the approach to equilibrium in what have been identified as high grade metamorphic terrains.

Finally, if much regional metamorphism does indeed arise by essentially isochemical solid–solid transformation processes, what of Barrow's metamorphic zones? Are they principally a manifestation of a regional thermal gradient as he thought, or do they reflect a broad but cryptic clay mineral facies pattern developed during Dalradian sedimentation and diagenesis. Or do they, in that particular case, result from a combination of the two?

Acknowledgements

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