Mineral parageneses in low-grade metabasites at low pressures and consideration of the sub-greenschist realm

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Introduction

Many early reviews of metamorphism considered that true metamorphic conditions are only first encountered at the onset of the greenschist facies, with anything below that, in the sub-greenschist realm, not being truly metamorphic, instead being more related to diagenetic or other processes. This led to very confusing concepts such as `spilitization` and even the notion of original spilitic or keratophyric magmas.

It was the studies of Coombs and others in the late 1950`s and early 1960`s (eg Coombs et al., 1959; Coombs, 1960) that started to identify a degree of order in rocks that are now accepted as belonging to the zeolite facies, based on intermediate to silicic volcaniclastic rocks in South Island, New Zealand. These rocks contained unstable volcanic glass along with relict high temperature primary igneous minerals and these components, combined with a high porosity, made them highly reactive in the presence of low temperature fluids. The type locality for the zeolite facies is the Taringatura Hills in South Island, New Zealand, and the term `burial metamorphism` was adopted in view of the fact that the rocks have not been deformed, instead appearing to vary in mineralogy with depth of burial. Subsequent studies by Boles and Coombs (1975) in the adjacent Hokonui Hills region demonstrated that the composition of the circulating fluids had an appreciable effect on the secondary minerals present.

The presence of prehnite and pumpellyite in some of the highest grade rocks studied by Coombs (1960, 1961) led to definition of the prehnite-pumpellyite metagreywacke facies. Whilst this facies, currently known more simply as the prehnite-pumpellyite facies, is now widely recognised, as late as the early to mid 1970`s there was still debate as to whether such alteration was truly metamorphic in origin (see Amstutz, 1974).

It was the upsurge in studies of low-grade metamorphic sequences in the mid to late 1970s that led to the establishment of the various low-grade metamorphic facies that are recognised today. An important study by Kawachi (1975) extended an understanding of the pumpellyite-actinolite facies as defined earlier by Hashimoto (1966), noting also that this is the grade which sees the first development of a rock schistosity. This study also reinforced the relationship between Fe/(Fe+Mg) in whole rock and chlorite, as previously recorded by Horikoshi (1965), a relationship that was further elaborated in the study by Coombs et al. (1976) of pumpellyite-actinolite facies semi-schistose rocks of the Taveyanne Formation at Loèche, Switzerland.

In the late 1970`s to early 1980`s metabasites containing mineral assemblages characteristic of the prehnite-pumpellyite facies were recognised widely in the Caledonide sequences of Wales (Bevins, 1978; Bevins and Rowbotham, 1983), Ireland (Oliver, 1978) and Scotland (Oliver and Leggett, 1980). These studies, amongst others, precipitated an upsurge in interest in low-grade metamorphic studies in the 1990's, which led to the establishment of IGCP Project 294 *'Very low-grade metamorphism'*.

Mineral assemblages in low-grade metabasites are dominated by prehnite, pumpellyite, epidote, chlorite and actinolite, along with zeolites, especially laumontite, at slightly lower temperatures and pressures, and stilpnomelane in rocks with high whole rock Fe/Mg ratios.

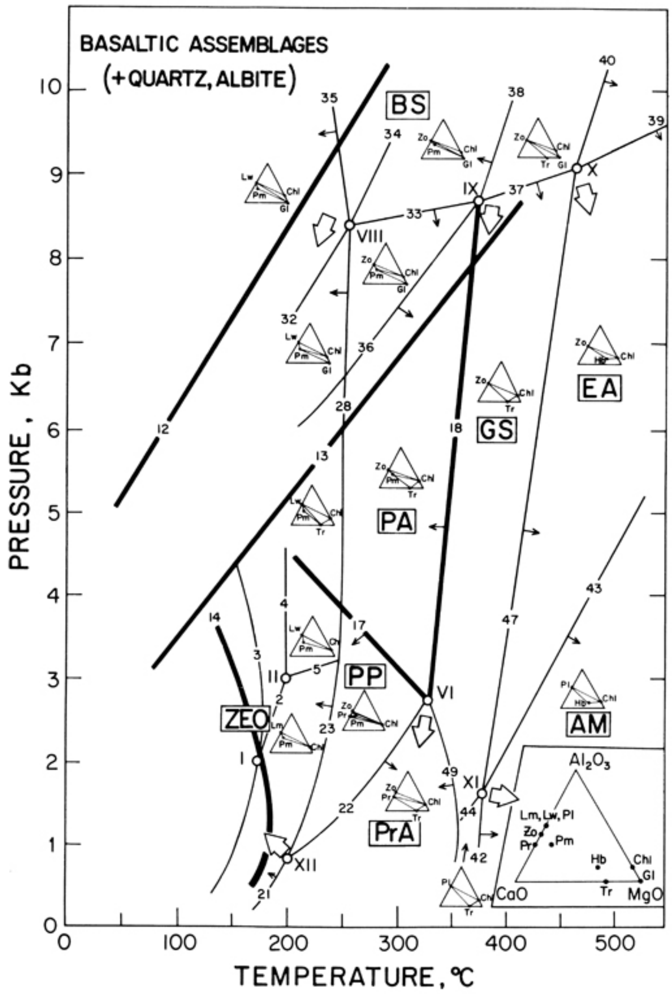

FIGURE 1. *P-T diagram showing a petrogenetic grid for low-grade metamorphic facies in the model NCMASH basaltic system. Facies abbreviations as follows: BS = blueschist facies; EA = epidote-amphibolite facies; AM = amphibolite facies; GS=greenschist facies; PA = pumpellyite-actinolite facies; PP = prehnite-pumpellyite facies; PrA = prehnite-actinolite facies; ZEO = zeolite facies. Reproduced with kind permission of the Mineralogical Society of Great Britain and Ireland from a paper by Liou et al. (1989) Mineralogical Magazine,* ***49,*** *321-333*

What is important to establish is whether the particular assemblages observed in the field represent equilibrium assemblages, and what are the relationships between different parageneses. Various methods have been used to evaluate quantitative data obtained for minerals, including the development of petrogenetic grids and chemographic projections, as detailed next.

Petrogenetic grids

An important advance in understanding equilibria amongst low-grade metamorphic parageneses was the development of a petrogenetic grid based on the NCMASH (Na_2O-CaO-MgO-Al_2O_3-SiO_2-H_2O) model system by Liou et al. (1985). In defining the relationships between the various sub-greenchist facies, Liou et al. (op.cit.) described for the first time the prehnite-actinolite facies (see Figure 1). However, this grid has drawbacks as it was based on data from experiments of end-member mineral compositions in a model system which is Fe-free. Accordingly, invariant points in the *P-T* grid become divariant if FeO or Fe_2O_3 are included and trivariant if both are included. Also recognising stable equilibria in experimental runs has proven difficult to determine. What is clear in this model system in terms of its inadequacy is the relatively small stability field occupied by prehnite+pumpellyite (+chlorite+epidote+albite+quartz+water) when in reality this is a very common sub-greenschist facies assemblage. Finally, there are certain reactions in the grid that actually contradict the relative appearances and disappearances of critical minerals in the field.

In a subsequent refinement of this grid, Frey et al. (1991) utilised internally consistent thermodynamic data of Berman (1988) and also GEOCALC software of Berman et al. (1987) to further explore equilibria in the NCMASH system. Again, however, even when the model takes account of Fe the commonly observed prehnite-pumpellyite-chlorite assemblage occupies a ±25°C *ca.* 3-4 kbar field (see Figure 2), which again conflicts with empirical observations. A critical factor in these models is that the thermodynamic properties of critical phases and solid solutions are not currently available.

One of the principal conclusions of the grid presented by Frey et al. (1991) was that the various sub-greenschist facies (zeolite, prehnite-pumpellyite, pumpellyite-actinolite and prehnite-actinolite) overlap considerably in *P-T* space. We return to this later in the paper.

Mineral projections

A variety of mineral projections have been developed since the original work of Thompson (1957) and later by Greenwood (1975). Harte and Graham (1975), and later Laird (1980), developed a projection from epidote for high grade metabasites, which was modified subsequently by Beiersdorfer and Day (1983) in their studies of low-grade metabasites from the Smartville Complex, northern Sierra Nevada, USA, and utilised further in a number of studies, including those by Springer et al. (1992), Bevins and Robinson (1994, 1995), and Beiersdorfer and Day (1995). As pointed out by Schiffman and Day (1999) the principal advantages of this projection in the study of low-grade metabasite parageneses are:

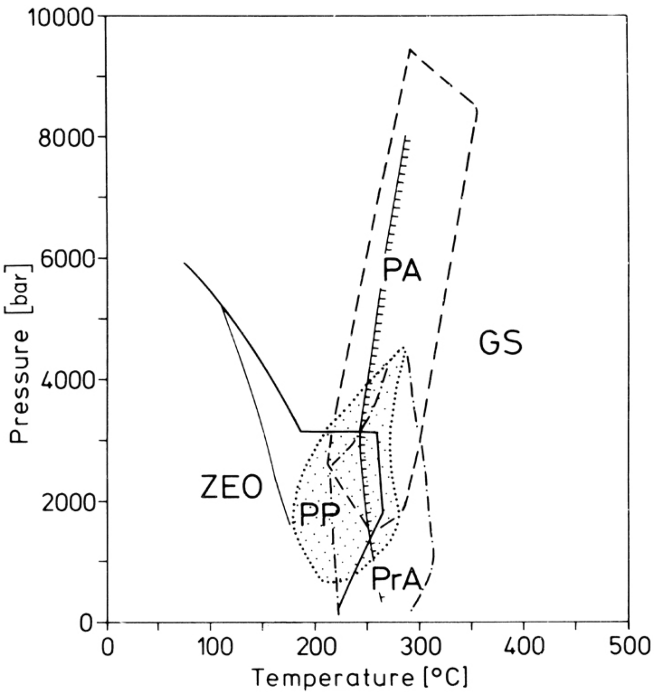

FIGURE 2. *P-T fields for various low-grade metamorphic facies as determined by Frey et al. (1991). Note the considerable overlap for the various facies in P-T space. Reproduced with the permission of Blackwell Science.*

i. it readily illustrates the $MgFe_{-1}$ substitution between key phases in the rocks;
ii. the low contents of FeO and MgO in the phases present mean that MgO/(MgO+FeO) ratios are preserved; and
iii. the projection points are invariably present in the rocks (the so-called `ubiquitous phases` listed below).

However, there are certain drawbacks, including:

i. Al_2O_3 and Fe_2O_3 are combined as a single component and hence $AlFe_{-1}$ substitutions are not considered (a substitution which is important in pumpellyite); and
ii. pumpellyite and tremolite lie a long way from the image plane so minor variations in composition result in considerable shifts in plotted positions. This is especially the case for pumpellyite; actinolite shows more consistent compositions in low-grade metabasites and hence shows relatively little in the way of variations in plotting position.

Low-grade metabasite mineral assemblages typically contain the ubiquitous phases albite+quartz+epidote+chlorite+H_2O, along with two or more of pumpellyite, prehnite and actinolite. In addition the fluid phase present is considered to be essentially pure H_2O, given the low carbonate contents in rocks. When CO_2 contents are high the non-diagnostic assemblage calcite-quartz-chlorite-develops, as modelled by Cho and Liou (1987). Stilpnomelane develops as an additional phase in rocks with high Fe/Mg ratios. Ignoring stilpnomelane, the rocks in the epidote projection are considered in terms of an eight component system, namely SiO_2-(Al_2O_3+Fe_2O_3)-FeO-MgO-CaO-Na_2O-TiO_2-H_2O (NCFMASH). Taking account of the phase rule, if two of the additional phases (pumpellyite, prehnite, actinolite) are present then there would be three degrees of freedom, which Bevins and Robinson (1993) considered to be pressure, temperature and whole rock composition. In the case of three phases ie prehnite, pumpellyite and actinolite then the system is fully defined mineral compositions should be invariant. Hence in this system with small changes in whole rock composition there should be no change in mineral composition. Before exploring metabasite mineral parageneses in the epidote projection in more detail it is useful to review the character of mafic phyllosilicates in these low-grade rocks.

Mafic phyllosilicates in low grade metabasites

Low-grade metabasic rocks were commonly referred to as `greenstones` in the early literature, due in large part to the abundance of a mafic phyllosilicate. For rocks belonging to the upper (laumontite-bearing) zeolite facies and through the various other sub-greenschist facies the mafic phyllosilicate is usually chlorite, with a near end member clinochlore composition when plotted on a NIC (Non-Interlayer Cations) versus total Al diagram. At lower grades, however, studies have shown the mafic phyllosilicate to be smectitic with high X_{MgO} ratios (>*ca.* 0.8). Aguirre et al. (2000) and Robinson et al. (2004) recorded a progression from smectite (saponite) with no interlayers, through interlayered smectite/chlorite, to chlorite with chlorite layers in excess of 75% in a Mesozoic sequence in the Andes of Chile which passes from the zeolite through to the prehnite-pumpellyite facies (Figure 3).

As noted earlier, Kawachi (1975) noted a high degree of correlation between whole rock and chlorite X_{MgO} ratios, a relationship also established by Bevins et al. (1991a), utilising data from Wales and Greenland (see Figure 4). In contrast, collating data from the North Shore Volcanic Group reported by Schmidt and Robinson (1997), Robinson and Bevins (1999) were able to show that at the lowest grades, where smectite is present, there is no relation between whole rock

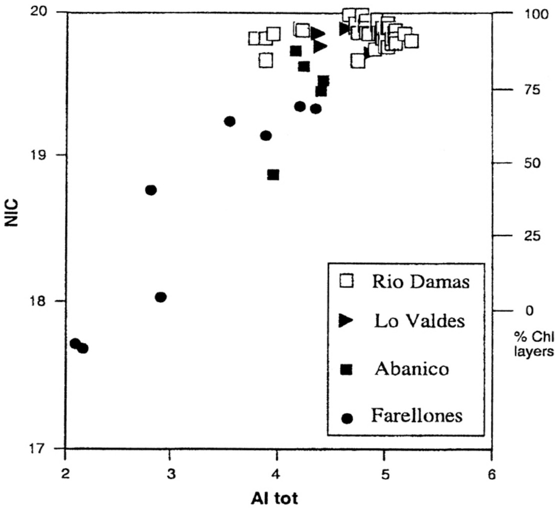

FIGURE 3. *Plot of sum of non-interlayer cations (NIC:Si+Al+Fe+Mg+Mn+Ti) against total Al in mafic phyllosilicates from Mesozoic to Cenozoic mafic volcanic rocks in the Andean orogenic belt to the east of Santiago, Chile. Reproduced with permission of Springer-Verlag, from Robinson et al. (2004*

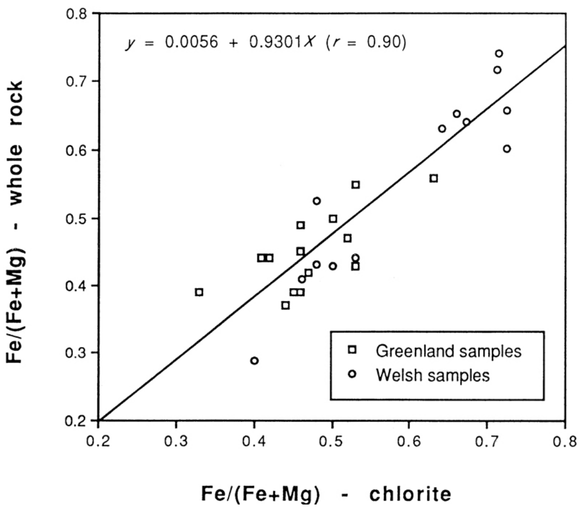

FIGURE 4. *Whole-rock Fe/(Fe+Mg) versus chlorite Fe/(Fe+Mg) for samples from Wales and eastern North Greenland. Note the strong positive correlation (r=0.90) between the two variables. Reproduced from Bevins et al. (1991) with the permission of Blackwell Science.*

and mafic phyllosilicate X_{MgO} ratios. The significance of this is that the epidote projection only becomes of value in the upper part of the zeolite facies, where epidote makes its first appearance and the mafic phyllosilicate acquires true chloritic compositions.

Relationships between whole rock compositions, mineral compositions and mineral parageneses

Horikoshi (1965), in his study of the Sanbagawa schists, was probably one of the earliest workers to report a relationship between Fe^{2+}-Mg substitution in chlorite and whole rock $Fe^{2+}/(Fe^{2+}+Mg)$ values, a conclusion also reached by Kawachi (1975) for pumpellyite-actinolite metabasites from Upper Wakatipu. A similar finding was reported by Bevins and Rowbotham (1983) for metabasites from the Caledonides of Wales. However, the real significance of this relationship in terms of influence on secondary mineral assemblages in metabasites was only fully realised some years later, in a study of the Tal y Fan intrusion, in North Wales by Bevins and Merriman (1988). They noted that in those areas of the intrusion with high MgO/(MgO+FeO) ratios in chlorite and whole rock the assemblage contained, in addition to the ubiquitous phases, actinolite and prehnite, while in those areas with lower MgO/(MgO+FeO) ratios in chlorite and whole rock, pumpellyite was present with prehnite and no actinolite. Taken at face value, this could infer that different parts of the intrusion had experienced different grades of metamorphism (namely prehnite-actinolite and prehnite-pumpellyite facies conditions respectively). A more realistic conclusion, however, is that the intrusion has been uniformly affected by a single set of intensive parameters, and that the variations in mineral parageneses are controlled by variations in whole rock composition. A similar relationship has subsequently been identified in the St David's Head Intrusion in South Wales, which shows a greater range in whole rock Fe-Mg compositions than the Tal y Fan Intrusion (see Bevins and Robinson, 1993).

These findings serve to explain why Frey et al. (1991) reported considerable overlap between the various low-grade metamorphic facies in their petrogenetic grid based on internally-consistent thermodynamic data for the NCMASH system (see Figure 2). Given that chlorite

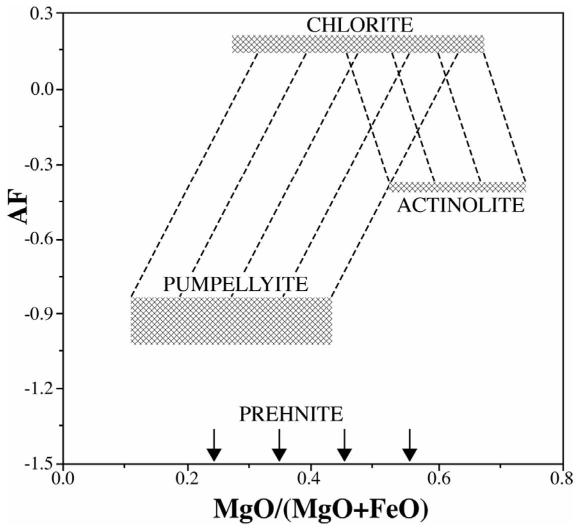

FIGURE 5. *Schematic relationships between pumpellyite, chlorite, actinolite and prehnite in the epidote projection for metabasites from the sub-greenschist and greenschist facies. AF*=100(Al_2O_3+Fe_2O_3-0.75CaO-Na_2O+0.75TiO_2/(Al_2O_3+Fe_2O_3-0.75CaO-Na_2O+0.75TiO_2+FeO+MgO). Reproduced with permission from the European Journal of Mineralogy, from Bevins and Robinson (1993).*

MgO/(MgO+FeO) ratios can be taken as a reasonably reliable proxy for whole rock MgO/(MgO+FeO) ratios, it is possible to explore further the influence of whole rock composition on low-grade metabasite mineral parageneses by examining mineral relationships in the epidote projection. This is because chlorite is a ubiquitous phase at these grades and is one of the phases which can be plotted in the epidote projection (see Figure 5 for a schematic plot). This is explored further in the next section.

Review of epidote projection data from Wales and comparative regions

For this review we have drawn on published and unpublished mineral data from Wales and comparative terranes worldwide, all showing mineral assemblages characteristic of the prehnite-pumpellyite to greenschist facies at low pressures.

Metamorphism in the Caledonide region of Wales has been previously described by Bevins and Rowbotham (1983) and Bevins and Robinson (1993), who determined a low pressure setting. Figure 6 shows all of the data available for Wales, which represents 35 individual samples. When all the data are presented simultaneously meaningful patterns are difficult to discern; however, more sense is made when the samples are considered in relation to the variations in grade across Wales established by Robinson and Bevins (1986) on the basis of illite crystallinity studies.

Robinson and Bevins (1986) determined that the lowest grades of metamorphism occurred principally in areas marginal to the main depocentre of the Welsh Basin, namely in the Welsh Borderland and on the Llŷn Peninsula, where diagenetic to anchizone IC values were determined. In these areas metabasites show the presence of prehnite and pumpellyite, along locally with zeolites. Figure 7 shows that pumpellyite-chlorite tie lines for analysed samples from these areas show a broad `fan`, ranging from negative slopes ($K_D^{Mg\text{-}Fe}$ =1.53) to positive slopes with a range of $K_D^{Mg\text{-}Fe}$ values from 0.27-0.58, with chlorite showing a range of X_{Mg} [MgO/(MgO+FeO)] values (from 0.20-0.65), the assemblages noteably lacking actinolite, even in samples showing the highest X_{Mg} values.

As reported by Bevins and Robinson (1993), most metabasites in areas showing epizone illite crystallinity values contain prehnite, pumpellyite +/- actinolite, with actinolite appearing in rocks

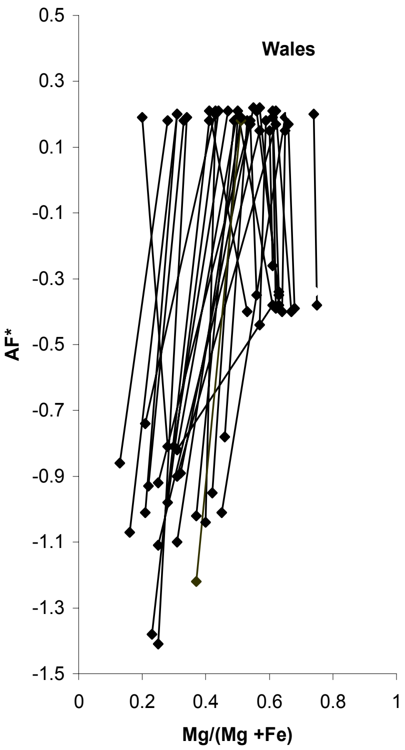

FIGURE 6. *Epidote projection for all samples analysed from Wales. Total number of samples = 35. AF* formula presented in caption to Figure 5.*

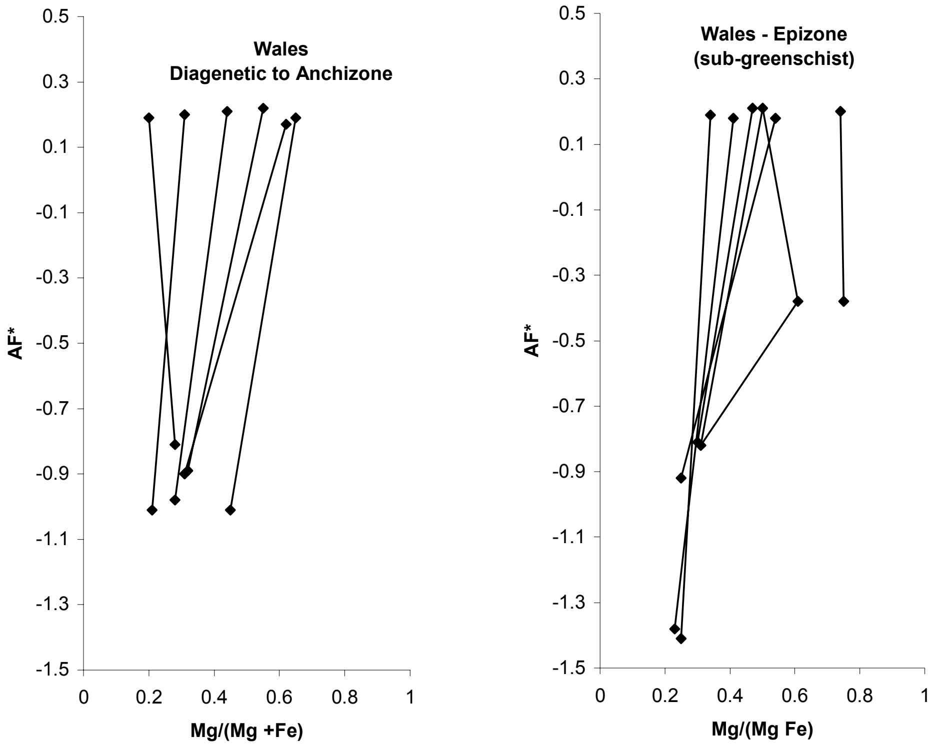

FIGURE 7. *Epidote projection for sub-greenschist metabasites from Wales. Left: Samples analysed from the diagenetic to anchizone in Wales. Right: Samples analysed from the epizone in Wales. AF* formula presented in caption to Figure 5.*

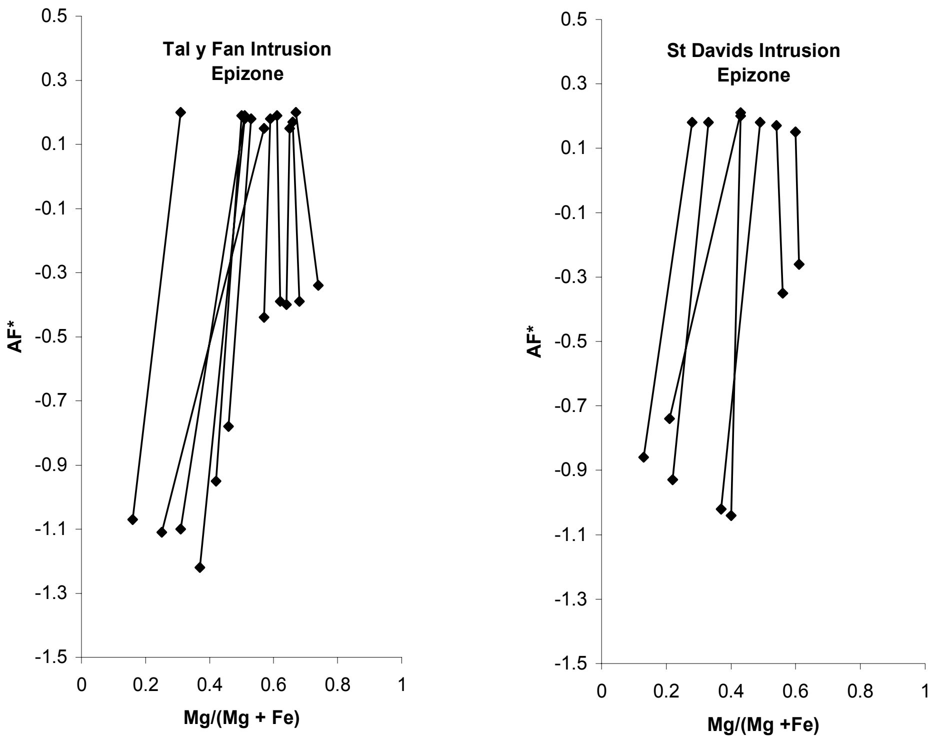

FIGURE 8. *Epidote projection for sub-greenschist (epizone) metabasites from two intrusions in Wales. Left: Samples analysed from the Tal y Fan Intrusion. Right: Samples analysed from the St Davids Head Intrusion. AF* formula presented in caption to Figure 5.*

with high X_{Mg} values (see Figure 7). This is highlighted by samples from two intrusions, namely the Tal y Fan Intrusion, in North Wales (see Figure 8) and the St Davids Intrusion, in South Wales (see Figure 6e). Pumpellyite-chlorite tie lines in samples from these intrusions are all positive, showing quite a broad range in of $K_D^{Mg\text{-}Fe}$ values from 0.26 to 0.80, with a tendency perhaps for samples at higher X_{Mg} values to show a low K_D value, and hence crossing tie lines.

In all of the samples investigated from Wales (many more samples than reported on here have been investigated petrographically) only a single sample has been found to contain prehnite, pumpellyite and actinolite. This sample, from the Presceli region in South Wales, shows this assemblage at a X_{Mg} value in chlorite of 0.54 (see Figure 7). The actinolite-chlorite $K_D^{Mg\text{-}Fe}$ ratio in this sample is 1.56. In South Wales, samples with X_{Mg} values >0.54 have assemblages containing prehnite and actinolite but lacking pumpellyite (see Figure 9). Actinolite-chlorite $K_D^{Mg\text{-}Fe}$ values for these samples average 1.10. Samples at this grade in North Wales show the transition to prehnite-actinolite dominated assemblages at a slightly higher chlorite X_{Mg} values in the range 0.57-0.59, although actinolite-chlorite $K_D^{Mg\text{-}Fe}$ average values are noticeably similar to those in the South Wales samples, at a mean of 1.08 (see Figure 9).

Beiersdorfer (1993) recorded a similar relationship in metabasites from the Smartville Complex, of the Western Belt of the Sierra Nevada. With increasing chlorite X_{Mg} prehnite-pumpellyite facies assemblages give way to prehnite-actinolite bearing metabasites at a transitional X_{Mg} value in the range 0.63-0.65, with a mean actinolite-chlorite $K_D^{Mg\text{-}Fe}$ value of 1.06 (see Figure 10). A similar relationship is also seen in upper zeolite to prehnite-pumpellyite facies metabasites of the Mesozoic Abanico Formation, in the Andes of Chile (see Figure 10), as reported by Robinson et al. (2004). The variations in X_{Mg} values in chlorite marking the

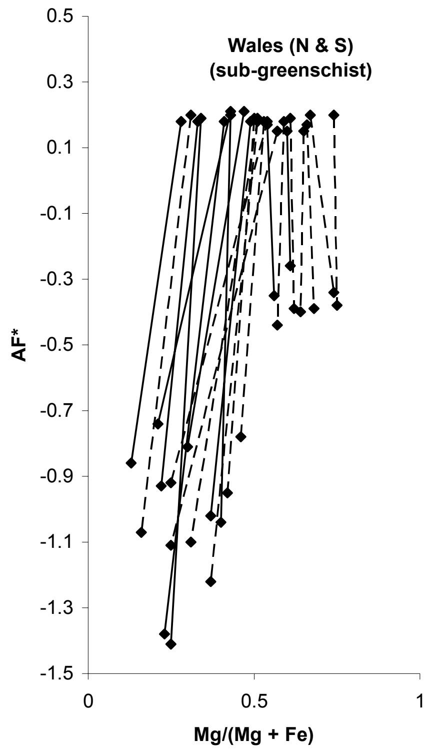

FIGURE 9. *Epidote projection for all samples analysed from North Wales (dashed lines) and South Wales (solid lines). AF* formula presented in caption to Figure 5.*

disappearance of pumpellyite in these different suites may reflect a slight variation in grade between those areas.

Finally, a small number of samples analysed from Cadair Idris, in Central Wales in the epizone area, contain actinolite + epidote in the absence of prehnite and pumpellyite. These samples are considered to reflect greenschist facies assemblages. Actinolite-chlorite $K_D^{Mg\text{-}Fe/}$ values average 1.34, which reflects markedly shallower negative slopes in the epidote projection (see Figure 11) for these samples compared with other samples from Wales. A similar relationship is seen in metabasites showing the transition from the prehnite-pumpellyite facies to the greenschist facies, from Peary Land, eastern North Greenland (Bevins and Rowbotham, 1984; Bevins et al., 1991b), and the Smartville Complex (Springer et al., 1992), as illustrated in Figure 12. Average actinolite-chlorite $K_D^{Mg\text{-}Fe}$ values are noteably similar to those from Cadair Idris, at 1.41 and 1.36 respectively.

What emerges from this review of the transition from the prehnite-pumpellyite facies to the greenschist facies is that there appear to be regular transitions in parageneses linked to changes not only in intensive parameters but also in whole rock compositions.

Conclusions

The examples detailed above reveal a relatively consistent change in mineral parageneses as the greenschist facies is approached. However, the disappearance and appearance of critical phases can clearly be seen to be influenced not only by changes in intensive parameters but also by whole rock compositions. This leads to examples of individual rock bodies with internal compositional variations that result in consistent parageneses developing that are characteristic of contrasting facies. These facies, namely the prehnite-pumpellyite, prehnite-actinolite and the

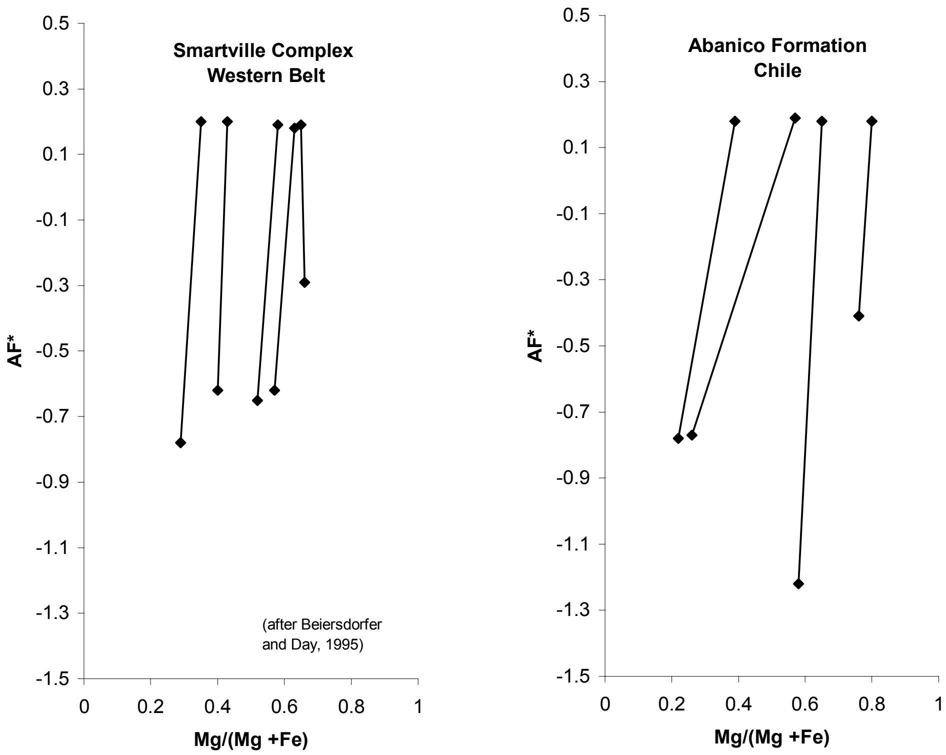

FIGURE 10. *Epidote projection for samples analysed from (left) the Smartville Complex, Western Belt, Sierra Nevada (presented by Beiersdorfer and Day, 1995) and (right) from the Abanico Formation, Chile (presented by Robinson et al., 2004). AF* formula presented in caption to Figure 5.*

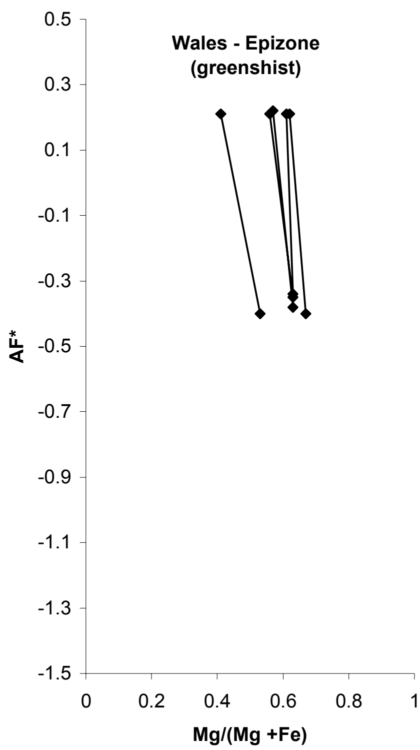

FIGURE 11. *Epidote projection for greenschist facies samples analysed from the Cadair Idris region in Central Wales. AF* formula presented in caption to Figure 5.*

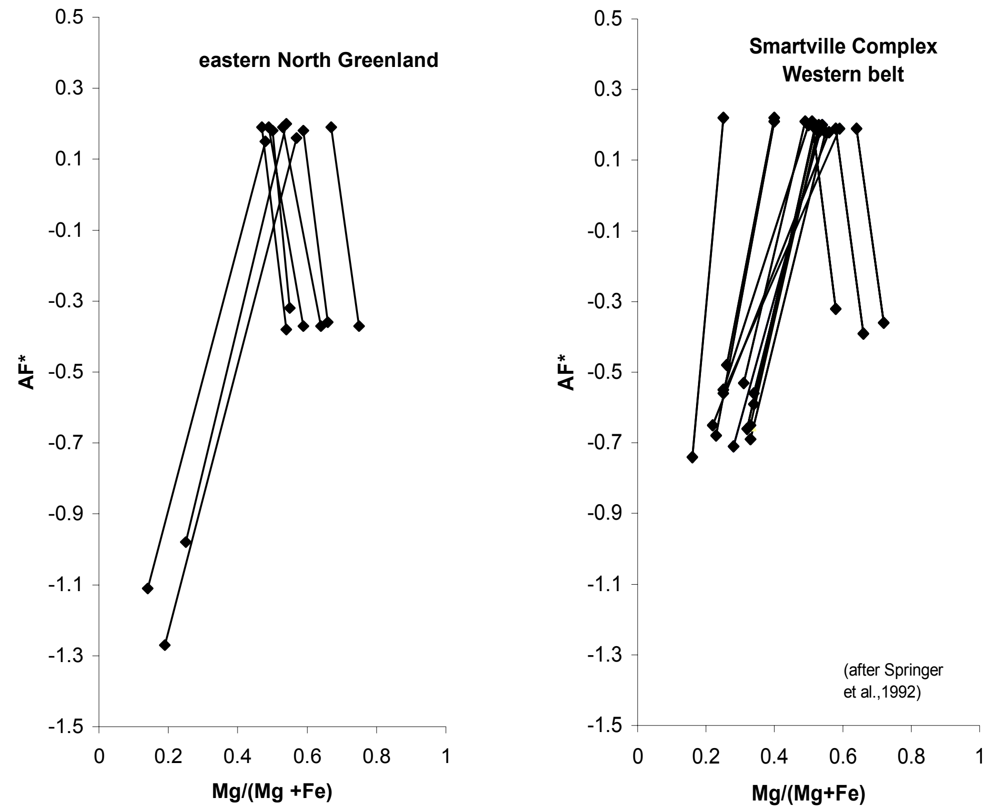

FIGURE 12. *Epidote projection for sub-greenschist and greenschist facies samples analysed from (right) eastern North Greenland (presented by Bevins and Rowbotham, 1984; Bevins et al., 1991b) and (left) the Smartville Complex, Western Belt, Sierra Nevada (presented by Springer et al., 1992). AF* formula presented in caption to Figure 5.*

pumpellyite-actinolite, were defined before the understanding of the effect of mineral compositional variation (in particular (Fe/Mg) in controlling the parageneses proposed as diagnostic of these facies. This whole rock control is almost certainly the reason why petrogenetic modelling reveals mineral relationships which show facies assemblages largely overlapping in *P-T* space. Indeed, on the present understanding it is thus preferable to designate these contrasting assemblages as diagnostic of a single subgreenschist facies.

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