

Chlorite-bearing polymetamorphic metapelites in the Rangeley area, Maine: Evidence for equilibrium assemblages

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ABSTRACT

This paper reviews textural and chemical aspects of chlorite in garnet-to-sillimanite-grade metapelites of the southwest Rangeley Quadrangle in Maine. Because of polymetamorphism, the textural aspects of the chlorite could lead to interpreting it as a nonequilibrium phase. However, systematic patterns of chlorite chemical composition relative to metamorphic grade, mineral assemblages, and sulfidation reactions indicate that it is in chemical equilibrium with the other minerals present. It is concluded that for petrogenetic consideration of metapelites that have been subjected to polymetamorphism, chemical evidence is the best guide for assessing approach to equilibrium by chlorite.

INTRODUCTION

Chlorite occurs in many geologic environments affected by low to moderately high temperatures and a wide range of pressures. Some of its occurrences are in metapelites, metabasalts, calc-silicates, meta-ultramafics, vein deposits, and as an alteration product in virtually all kinds of igneous or metamorphic rocks. In each environment, chlorite displays a distinctive chemical signature (Albee, 1962). Recently, Laird (1988) has comprehensively reviewed the petrologic aspects of chlorite from metamorphic environments.

Chlorite forms readily in response to both increasing and decreasing temperature. Consequently, it is often unclear whether it is in chemical equilibrium with the associated minerals. However, in using mineral parageneses to establish the *P-T* conditions at which rocks have recrystallized, it is crucial to assess the extent to which the minerals present approximate stable or equilibrium assemblages. In this context, a problem often arises for chlorite in metapelites when it appears to be an extra AFM phase. Several possible interpretations arise: (1) polymetamorphism, resulting in mineral assemblages that may or may not have reequilibrated chemically, (2) extra components in some other phase, or (3) $a_{\text{H}_2\text{O}} < 1$.

In the case of chlorite from rocks subjected to polymetamorphism, the crucial question in whether or not it has equilibrated (or reequilibrated) chemically with the associated minerals. In attempting to establish the intensive parameters at which a rock recrystallized, it makes no difference whether minerals like chlorite formed under prograde or retrograde metamorphic conditions. All that

matters is that they and the other minerals present chemically reequilibrated (or at least approached a state of equilibrium). Unfortunately, the textural features of chlorite in rocks subjected to polymetamorphism commonly lead different workers to interpret the chlorite differently with regard to its state of chemical equilibrium with the associated minerals.

In this paper, we consider chlorite only from the metapelites of the Rangeley area in western Maine. Guidotti (1970) has shown that these metapelites have been subjected to two temporally discrete metamorphism events. In some places the rocks formed during the first event have been retrograded and in other places, prograded.

In some cases, textural or chemical evidence enables recognition of chlorite in a metapelite as almost certainly not in equilibrium with other minerals present. Such an interpretation is commonly given to chlorite containing sagenitic rutile (precipitated within biotite richer in Ti) and having irregular, diffuse grain boundaries with biotite. Similarly, most workers infer from chemical evidence such as $\text{Mg/Fe (Chl)} < \text{Mg/Fe (Bt)}$ that the two minerals are not in equilibrium. Careful attention should be paid, however, to extremely Mg-enriched rocks, in which the Mg/Fe ratio of chlorite and biotite are reversed (Tewhey and Hess, 1975; Guidotti et al., 1975). Another occurrence of chlorite in metapelites routinely judged as nonequilibrium is that in the upper sillimanite zone (above staurolite stability) or in the sillimanite + potassium feldspar zone, even if the chlorite is Mg rich and appears texturally fresh. Numerous relations suggest that chlorite is unstable at such grades in metapelites.

More commonly, the equilibrium status of chlorite is equivocal and similar occurrences are differently interpreted. Common textural and chemical features of chlorite subject to equivocal interpretations include (1) chlo-

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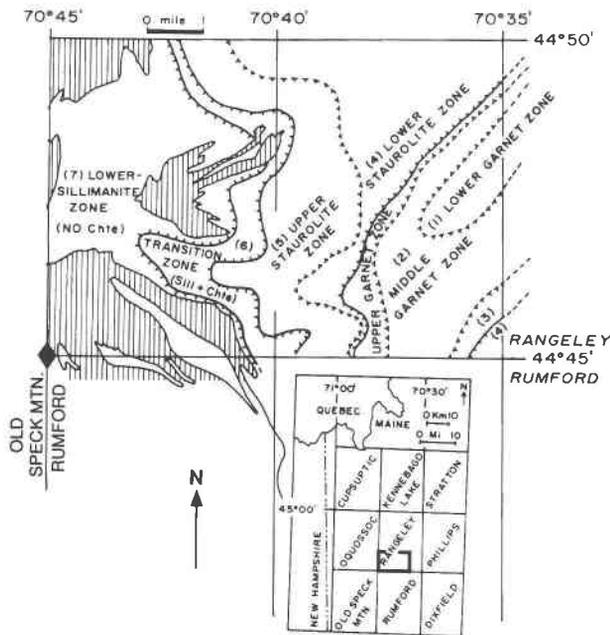


Fig. 1. Location of study area and distribution of isograds in the southwest portion of the Rangeley Quadrangle, Maine.

rite having sharp grain boundaries but partially pseudomorphing other phases such as staurolite or garnet; (2) sharply bounded, cross-cutting chlorite porphyroblasts that appear to have formed late in the growth sequence of the minerals present; (3) chlorite present in more than one distinctly different grain-size range; and (4) chlorite in assemblages not appearing to obey the mineralogical phase rule. Equilibrium interpretations for some of these cases have involved appeals to internal buffering of $a_{\text{H}_2\text{O}}$ (Guidotti, 1974; Lang and Rice, 1985) or to small scale, local equilibrium (Osberg, 1971).

In (1) through (3) the Mg-Fe partitioning between chlorite and biotite may be as expected for coexistence in chemical equilibrium. Nonetheless, the chlorite may still be interpreted as a nonequilibrium phase.

To illustrate the extent of ambiguity in the interpretation of the equilibrium status of chlorite, consider the small area of central and western Maine and northern New Hampshire. There, some workers have interpreted observations (1) through (4) to be indicative of equilibrium chlorite, others, of nonequilibrium chlorite (Osberg, 1971; Holdaway et al., 1987, 1988; Holdaway and Dutrow, 1989; Henry, 1974; Al-Mishwt, 1972; Green, 1963; Dickenson, 1988; Guidotti, 1970, 1974, 1985, 1989; Guidotti et al., 1975; Henry, 1981; Bussa, 1973; Conatore, 1974).

Despite the presence of the ambiguous features described above, the results of this study illustrate at least one case in which additional, strong arguments exist for interpreting the chlorite and other minerals as having closely approached a new state of chemical equilibrium during a second metamorphic event. The arguments in-

volve the chemistry of chlorite, element partitioning between it and other minerals, and the relationships between its chemistry and the mineral assemblage in which it occurs. Although some features indicate textural disequilibrium (Thompson, 1987), the chemical data suggest a sufficient approach to equilibrium such that one can include the chlorite data in attempts to determine values for intensive parameters that occurred during final recrystallization.

PETROLOGIC SETTING

All samples are from the Siluro-Devonian metapelites (Moench, 1971) in the southwest portion of the Rangeley Quadrangle of western Maine. They belong to a metamorphic sequence ranging from garnet zone to upper sillimanite zone (Guidotti, 1970, 1974). However, the transition zone (TZ) as defined by Guidotti (1974) (see below) marks the upper stability limit of chlorite in the study area. Figure 1 shows the distribution of grades relevant to this report. Details of the petrologic setting (reactions, mineral assemblages, etc.) are provided elsewhere (Guidotti, 1970, 1974, 1978; Henry, 1981; Henry and Guidotti, 1985; Wood, 1981; Guidotti et al., 1988; Teichmann, 1988; Lux et al., 1986) and only a few aspects need be reviewed.

Figure 2 shows AFM diagrams (based on observed assemblages) used to define the garnet, staurolite, and transition zones shown in Figure 1. The garnet zone has been subdivided into three portions, lower, middle, and upper garnet zone (LGZ, MGZ, and UGZ, respectively), and the staurolite zone has been divided into a lower and upper staurolite zone (LStZ and UStZ) (Fig. 1). These subdivisions are based on compositional variations of Ms, Bt, and Chl due to the continuous reactions given in Table 1. Thus, as seen in Table 2, the subdivisions are contours of metamorphic grade as manifested by layer silicate compositions. Isograd reactions that affect AFM topology over the range from LGZ to TZ are also given in Table 1.

The staurolite zone AFM topology shows a four-phase assemblage, St + Grt + Bt + Chl (abbreviations from Kretz, 1983), and the transition zone AFM topology shows a five-phase assemblage, Sil + St + Grt + Bt + Chl. Guidotti (1974) attributed the four-phase assemblage to the presence of significant MnO in the garnet. The five-phase assemblage, which defines the TZ, was attributed to the same extra component (MnO in garnet) plus buffering of $a_{\text{H}_2\text{O}}$ by the discontinuous reaction separating the UStZ and lower sillimanite zone (LSiZ). Most samples considered in this report are from the limiting or quasi-limiting assemblages (i.e., number of components = number of phases) Grt + Bt + Chl, St + Grt + Bt + Chl, and Sil + St + Grt + Bt + Chl. Some samples from nonlimiting assemblages are included for comparison. A few cordierite-bearing rocks are present in the study area; the reader is referred to Henry (1981) or Guidotti et al. (1988) for a discussion of their interpretations. All samples contain quartz, plagioclase (approximately An₂₀), and il-

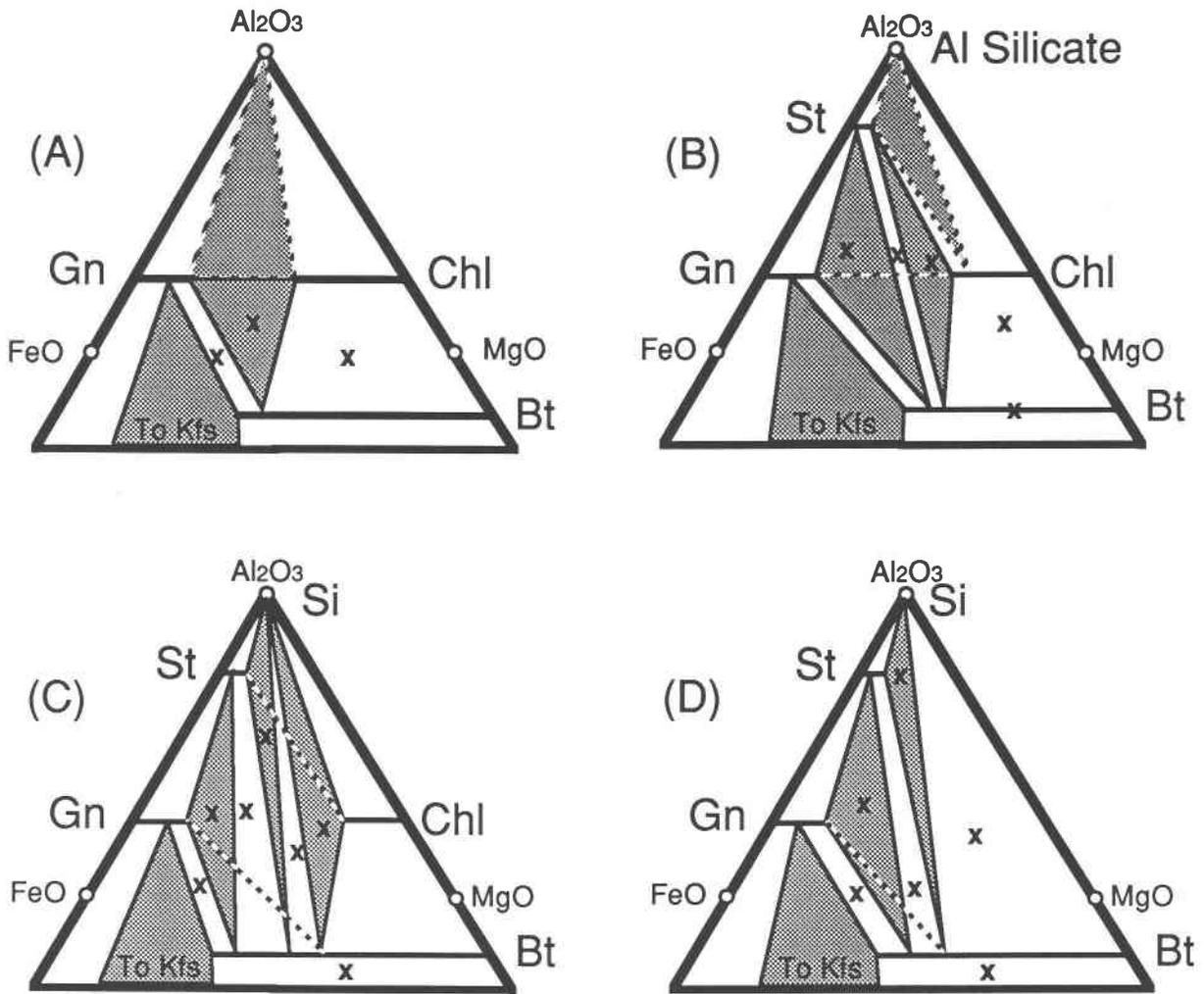


Fig. 2. Schematic AFM diagrams for the garnet zone (A), staurolite zone (B), transition zone (C), and lower sillimanite zone (D). X = observed assemblages. All assemblages are projected from muscovite, quartz, and uniform $a_{\text{H}_2\text{O}}$. Dashed, crosstie lines indicate four and five AFM phase assemblages.

menite. Most also contain graphite and 0–3 modal% pyrrhotite.

Several petrographic features warrant specific comment:

1. In the LGZ, chlorite averages about 12 modal% (visual estimates). It occurs as tiny groundmass laths, coarser plates associated with muscovite in pseudomorphs after staurolite, and as coarse plates forming

TABLE 1. Metamorphic zones, high-alumina limiting assemblages, and the continuous and discontinuous reactions used to define the isograds and their subdivisions

Zone	Abbrev.	Limiting assemblage*	Continuous reactions	Discontinuous reactions
Lower garnet	(LGZ)	Chl + Grt ± Po ± Gr	Fe-Chl + Ms = Grt + Bt +	
Middle garnet	(MGZ)	Chl + Grt ± Po ± Gr	Mg-Chl + Na-richer	
Upper garnet	(UGZ)	Chl + Grt ± Po ± Gr	Ms + H ₂ O	
Lower staurolite	(LStZ)	St + Chl + Grt ± Po ± Gr	Mg-Chl + K-Ms = Mg-St	Grt + Mg-Chl + Ms = St +
Upper staurolite	(UStZ)	St + Chl + Grt ± Po ± Gr	+ Mg-richer Bt + Mg-richer	Bt + Na-richer Ms +
Transition	(TZ)	St + Sil + Grt ± Chl ± Po ± Gr	Chl + Grt + Na-richer	Qtz + H ₂ O
			Ms + H ₂ O	
			Discontinuous reaction with	St + Mg-Chl + Na-Ms +
			buffered $a_{\text{H}_2\text{O}}$ so isograd is	Qtz ± (Grt?) = Bt + K-richer
			spread out into transition zone	Ms + Sil + Ab + H ₂ O

Note: From Guidotti (1970, 1974).

* All with Ms + Pl + Qtz + Bt + Ilm.

TABLE 2. Average modal amounts and major compositional aspects for the Rangeley sequence minerals from the limiting assemblages

Zones	Biotite		Chlorite		Garnet			Muscovite			Staur	Plag
	Modal %	Mg/(Mg + Fe)	Modal %	Mg/(Mg + Fe)	Modal %	Mg/(Mg + Fe)		Modal %	Mg/(Mg + Fe)	Na/(Na + K)	Modal %	An %
						Rim	Core					
LGZ	5	0.384	12	0.396	trace	0.059	0.071	29	0.486	12.69	x	15.20
MGZ	8	0.377	12	0.397	0.5	0.061	0.061	27	0.479	13.93	x	15.20
UGZ	10	0.410	7	0.428	1	0.074	0.080	30	0.523	18.72	x	15.20
LStZ	20	0.450	5	0.503	1.5	0.086	0.096	27	0.539	22.25	3	15.20
UStZ	20	0.473	2	0.504	2	0.087	0.106	26	0.543	23.52	4	17.2
TZ	22	0.468	trace	0.512	1.5	0.105	0.127	22	0.544	21.20	6	18

dodecahedral-shaped, partial pseudomorphs around euhedral garnet cores. Commonly, more than 50% of the garnet is replaced in these partial pseudomorphs. All three textural varieties of the chlorite have sharp grain boundaries and display uniform, anomalous blue birefringence colors, thereby suggesting a uniform composition in all three types. Anomalous green and brown chlorite has been observed only in the garnet-absent assemblage (Chl + Bt).

2. In the MGZ, the modal, textural, and optical properties of chlorite are similar to those observed in the LGZ, except for markedly less replacement of garnet by chlorite. In some specimens there is no replacement.

3. In the UGZ, chlorite content decreases to about 7 modal%. It occurs as groundmass laths and as coarser plates intergrown with muscovite in pseudomorphs after staurolite. In some cases, the coarse chlorite in the pseudomorphs tends to be concentrated in the outer portions of the pseudomorphs. Although both groundmass and pseudomorph chlorite are optically identical in a given specimen, birefringence colors range from blue to purplish brown, brown, and greenish brown in different samples.

4. In the LStZ, chlorite amounts to about 5 modal% and with crossed Nicols shows anomalous brown, green, and gray colors, the latter being most common in pyrrhotite-rich samples. It occurs as fine-grained, groundmass laths and medium-grained, cross-cutting plates and is associated with muscovite in partial pseudomorphs after staurolite. The outer portions of these pseudomorphs tend to have a high concentration of chlorite. In all textural types the chlorite has sharp grain boundaries. Moreover, in a given sample, all textural varieties have the same interference colors.

5. UStZ chlorite is texturally and optically quite similar to that seen in the LStZ. The main differences are that the modal amount of chlorite decreases to about 2% and that the partial pseudomorphs after staurolite contain less chlorite and more muscovite. The nature of the partial pseudomorphs also changes. In the LStZ, remnant, staurolite tends to occur as anhedral fragments near the core of the pseudomorphs. In the UStZ, staurolite commonly occurs as aggregates of small, subhedral to euhedral crystals distributed throughout the pseudomorphs and aligned

parallel to the shape of the original, larger staurolite crystals.

6. Finally, in the TZ, chlorite is present only in trace amounts as tiny matrix laths that are sharply bounded and have green to gray birefringence colors. It occurs in contact with all phases present in a given specimen, including sillimanite. Figure 2B in Guidotti (1974) shows the areal distribution of the chlorite-bearing specimens within the TZ.

In summary, some textural, assemblage, modal, and optical features of chlorite from the LGZ to the TZ are consistent with chlorite being an equilibrium phase. However, chlorite also occurs as a replacement mineral and some workers would consider this to indicate a lack of equilibrium. Moreover, the presence of different size fractions strongly suggests a lack of textural equilibrium (Thompson, 1987). To gain further insights, compositional data for these chlorites must be analyzed.

METHODS OF STUDY

The samples considered herein were also used in the following mineralogic and petrologic studies: Guidotti (1970, polymetamorphism; 1974, isogradic reactions; 1978, muscovite chemistry), Guidotti et al. (1988, biotite chemistry), Wood (1981, garnet chemistry and zoning), Henry (1981, sulfide-silicate phase relations in the UStZ). Detailed petrographic observations have been carried out on all samples as part of these studies. Hence, our observations of chlorite can be related to textural and chemical observations on most of the coexisting phases.

All analyses were performed using standard electron microprobe techniques (wavelength dispersion, Bence and Albee, 1968, oxide correction procedure). Most of the chlorite samples analyzed at the University of Maine are contained within the polished thin sections of rocks from the LGZ to LSZ used by Wood (1981). For each polished thin section, particular care was taken to analyze all of the textural varieties present. As expected from the uniformity of optical properties noted above, no differences in composition were observed among the textural varieties in a given specimen (see also Henry, 1981, p. 102 and Appendix Table 4). Plug mounts of an additional 30 samples from the UStZ and TZ were also analyzed. Some

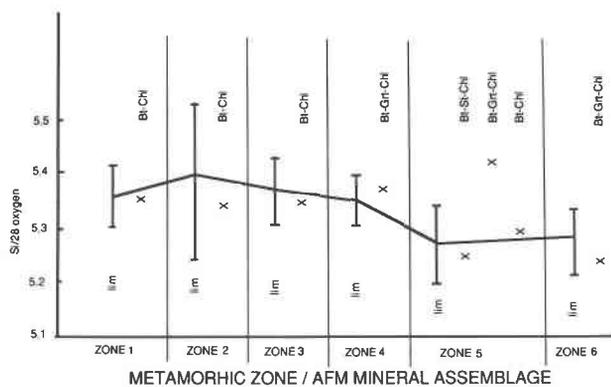


Fig. 3. Total Si contents of chlorite from the various assemblages in the metapelites of the Rangeley area. Zone numbers as in Figure 1 (lim = limiting assemblage).

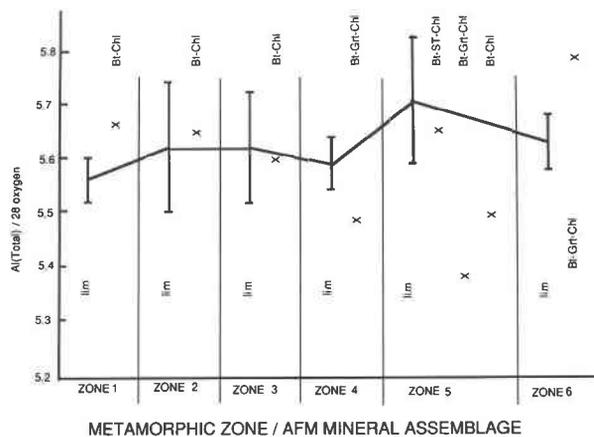


Fig. 4. Total Al contents of chlorite from the various assemblages in the metapelites of the Rangeley area. Zone numbers as in Figure 1 (lim = limiting assemblage).

analyses of the latter chlorite samples were reported previously (Guidotti, 1974). Comparison of the new and old data shows them to be similar within the limits of analytical error.

Henry's (1981) data were obtained using the University of Wisconsin electron microprobe. His data for UStZ chlorite samples are very similar to the data for UStZ chlorite of Teichmann (1988) but will be treated separately for discussion of the effects of sulfide-silicate phase relations.

ANALYTICAL RESULTS

The compositions of Rangeley chlorite samples are considered in terms of (1) mineral assemblages within a given grade, (2) metamorphic grade, and (3) degree of sulfidation reactions. Two additional data sets are discussed: one almost certainly illustrates nonequilibrium chlorite, the other involves either nonequilibrium chlorite or chlorite intergrown with micas on a very fine scale. The data are presented as averages of groups of samples from a given assemblage (Tables 3 and 4), in graphical form, or both. Analyses of individual samples can be obtained upon request from C.V.G.

Chlorite composition as a function of mineral assemblage

Here, we include only Teichmann's (1988) data. These data are constrained by three factors: (1) Based on mineral compositions and assemblages, all chlorite is inferred to have formed in rocks spanning only a relatively small range of bulk composition. (2) Virtually all coexists with graphite, ilmenite, and only minor (0–3 modal%) pyrrhotite. (3) Because of the assemblages present (see Table 5 and the AFM topologies for each grade, Fig. 2), chlorite generally should be saturated with Si, Al, and Ti, and thus should contain relatively uniform amounts of these elements. The only exception might be the Al in chlorite from the garnet zone as no truly Al-saturating phase is present.

Figures 3 and 4 and Table 6 summarize information correlating the effects of mineral assemblage with chlorite composition. Within the error bars for the chlorite of the limiting assemblages (the largest sample group for each grade) it is evident that Si and Al_{tot} contents are fairly uniform for all assemblages over all grades. In some staurolite zone cases, the average Si or Al_{tot} of chlorite from the Bt + Grt + Chl assemblage lies somewhat outside the error bars, but no significance can be made of this because of the small number of samples analyzed (see Table 5). The overriding feature is the general uniformity of the Si and Al_{tot} contents, regardless of the assemblage. Because the Ti contents of chlorite are so low, no discussion is warranted.

The other major cations (Mg, Fe, and Mn; Table 6) show several interesting patterns. As expected from the AFM diagrams for the garnet zone, the average Mg/(Mg + Fe) ratio of chlorite in the Bt + Chl assemblage is higher than that of chlorite from the assemblage Grt + Bt + Chl. In the three higher grades, the differences in the Mg/(Mg + Fe) ratios of chlorite in the St + Grt + Bt + Chl and Grt + Bt + Chl assemblages are small and somewhat ambiguous because of lack of data on the Mn content of the garnet of the three-phase assemblage. (See Albee, 1968, for a discussion of the effects of garnet manganese content on the Mg/(Mg + Fe) ratios of coexisting minerals.) On the other hand, for the UStZ, the average Mg/(Mg + Fe) ratios of chlorite from the assemblages St + Bt + Chl and Bt + Chl are markedly higher than those ratios for the chlorite of the garnet-bearing rocks. The absence of garnet plus Mg enrichment of the chlorite result from sulfidation reactions (see below). Finally, it is notable that similar patterns were found in the coexisting biotite (Guidotti et al., 1988) and muscovite (Guidotti, 1978).

Mn contents of the chlorite display several systematic patterns. In the LGZ and MGZ, the Mn content is quite similar in both assemblages. It should be noted that, in

TABLE 3. Average chlorite analyses for lower garnet zone to transition zone specimens, Rangeley area, Maine

Zone	Lower garnet zone			Middle garnet zone			Upper garnet zone			Lower staurolite zone		
	(8)	(2)	(1)	(4)	(7)	(3)	(6)	(2)	(8)	(2)	(2)	
No. in Ave.	std*	std	std	std	std	std	std	std	std	std	std	
Oxide wt% Si	24.81	25.03	24.78	25.12	25.23	25.13	25.44	25.66	24.81	25.44	25.66	
Ti	0.09	0.06	0.07	0.07	0.07	0.07	0.10	0.08	0.09	0.10	0.08	
Al	21.85	22.49	22.22	22.19	22.42	22.34	22.54	22.22	21.85	22.54	22.22	
Cr	0.05	0.03	0.07	0.05	0.04	0.05	0.04	0.04	0.05	0.04	0.04	
Fe	29.63	28.64	28.61	29.20	28.18	26.52	24.99	26.17	29.63	24.99	26.17	
Mn	0.29	0.27	0.25	0.25	0.10	0.27	0.15	0.04	0.29	0.15	0.04	
Mg	10.88	11.25	11.15	10.77	11.84	12.93	14.18	14.06	10.88	14.18	14.06	
K	x.x	0.01	0.02	x.x	x.x	0.02	x.x	0.00	x.x	x.x	0.00	
Total	87.57	87.78	87.21	87.56	87.87	87.35	87.33	88.40	87.57	87.33	88.40	
H ₂ O	12.43	12.22	12.79	12.44	12.13	12.65	12.68	11.60	12.43	12.68	11.60	
Formula units—based on 28 O atoms												
Si	5.36	5.35	5.35	5.40	5.37	5.35	5.35	5.36	5.36	5.35	5.36	
¹⁴ Al	2.64	2.65	2.66	2.60	2.63	2.65	2.65	2.63	2.64	2.65	2.63	
¹⁴ Total	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00	
¹⁶ Al	2.92	3.02	2.99	3.02	2.99	2.95	2.94	2.86	2.92	2.94	2.86	
Cr	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
Fe	5.35	5.12	5.16	5.25	5.02	4.72	4.40	4.58	5.35	4.40	4.58	
Mn	0.05	0.01	0.05	0.05	0.02	0.05	0.03	0.01	0.05	0.03	0.01	
Mg	3.50	3.59	3.59	3.45	3.76	4.10	4.45	4.39	3.50	4.45	4.39	
Ti	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
¹⁶ Total	11.85	11.80	11.80	11.78	11.80	11.84	11.83	11.87	11.85	11.83	11.87	
K	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
Total	19.85	19.80	19.82	19.78	19.81	19.84	19.83	19.87	19.85	19.83	19.87	
Al _{tot}	5.56	5.67	5.65	5.62	5.63	5.60	5.59	5.48	5.56	5.59	5.48	
¹⁴ Al/Si	0.494	0.494	0.497	0.484	0.490	0.497	0.495	0.489	0.494	0.495	0.489	
Mg/Fe	0.655	0.700	0.695	0.659	0.750	0.871	1.015	0.959	0.655	1.015	0.959	
Mg/(Mg + Fe)	0.396	0.412	0.410	0.397	0.428	0.465	0.503	0.489	0.396	0.503	0.489	
Mineral assemblage	Bt + Grt	Bt	Bt	Bt + Grt	Bt + Grt	Bt	Bt + St + Grt	Bt + Grt	Bt + Grt	Bt + St + Grt	Bt + Grt	

TABLE 3—Continued

Zone	Upper staurolite zone				Transition zone			
	(28)	(4)	(2)	(1)	(6)	(1)	(1)	(1)
No. in Ave.								
Oxide wt% Si	25.11	25.53	26.26	25.66	25.08	0.49	24.99	0.49
Ti	0.09	0.10	0.11	0.12	0.10	0.04	0.13	0.04
Al	23.10	23.33	21.97	22.63	22.68	0.17	23.47	0.17
Cr	0.04	0.04	0.07	0.06	0.04	0.02	0.00	0.02
Fe	24.93	21.97	25.63	23.51	24.63	1.33	25.16	1.33
Mn	0.11	0.22	0.27	0.33	0.16	0.05	0.19	0.05
Mg	14.20	16.76	14.38	16.12	14.50	0.98	13.94	0.98
K	0.02	0.02	0.03	0.04	0.02	0.02	0.00	0.02
Total	87.59	87.96	88.74	88.50	87.18	0.37	87.88	0.37
H ₂ O	12.41	12.04	11.27	11.50	12.82	0.37	12.12	0.37
Formula units—based on 28 O atoms								
Si	5.27	5.25	5.46	5.30	5.29	0.05	5.24	0.05
⁴¹ Al	2.73	2.75	2.54	2.70	2.71	0.05	2.77	0.05
⁴⁷ Total	8.00	8.00	8.00	8.00	8.00	0.00	8.00	0.00
⁶⁰ Al	2.99	2.91	2.84	2.81	2.92	0.03	3.03	0.03
Cr	0.01	0.01	0.01	0.01	0.01	0.00	0.00	0.00
Fe	4.38	3.78	4.45	4.06	4.35	0.28	4.41	0.28
Mn	0.02	0.04	0.05	0.06	0.03	0.01	0.03	0.01
Mg	4.44	5.14	4.46	4.96	4.55	0.26	4.35	0.26
Ti	0.01	0.02	0.02	0.02	0.02	0.01	0.02	0.01
⁶⁷ Total	11.85	11.90	11.82	11.92	11.87	0.03	11.85	0.03
K	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.01
Total	19.86	19.90	19.83	19.93	19.88	0.03	19.85	0.03
Al	5.72	5.66	5.38	5.51	5.64	0.05	5.80	0.05
⁴¹ Al/Si	0.518	0.523	0.466	0.510	0.513	0.014	0.528	0.014
Mg/Fe	1.023	1.363	1.000	1.222	1.056	0.130	0.988	0.130
Mg/(Mg + Fe)	0.504	0.576	0.500	0.550	0.512	0.030	0.497	0.030
Mineral assemblage	Bt + St + Grt	Bt + St	Bt + Grt	Bt	Bt + St Grt + Sil		Bt + Grt Sil	

* Standard deviations.

TABLE 4. Other groups of chlorite analyses from the Rangeley area, Maine

Group:	USiZ retrograde chlorite				USTz and TZ chlorite with high K and Ti values						
	Sample	M-426	Ra-b41	avg(2)	std*	Ra-c71	Ra-a98	Ra-a91	Ra-b59	Ra-c9	Ra-c63
Oxide wt%	Si	25.56	28.52	27.04	1.48	28.48	28.24	29.57	29.86	28.14	26.33
	Ti	0.02	0.11	0.07	1.05	0.28	0.26	0.17	0.11	1.91	0.17
	Al	20.78	18.72	19.75	1.03	18.73	18.24	19.07	19.26	17.81	19.64
	Cr	0.00	0.05	0.03	0.03	0.00	0.08	0.06	0.05	0.10	0.02
	Fe	28.20	27.86	28.03	0.17	24.82	26.86	26.81	26.14	27.71	27.37
	Mn	0.18	0.06	0.12	0.06	0.13	0.04	0.04	0.16	0.10	0.15
	Mg	12.13	12.12	12.13	0.01	13.52	12.74	11.53	10.18	10.52	12.32
	K	0.02	0.30	0.16	0.14	1.24	0.98	0.79	0.46	0.40	0.23
Total		86.89	87.74	87.32	0.43	87.20	87.44	88.04	86.22	86.69	86.23
	H ₂ O	13.11	12.26	12.69	0.43	12.80	12.56	11.96	13.78	13.31	13.77
Formula units—based on 28 O atoms											
Si	5.52	6.05	5.78	0.27	6.03	6.02	6.21	6.36	6.06	5.71	
¹⁴¹ Al	2.48	1.95	2.22	0.27	1.97	1.98	1.79	1.64	1.94	2.29	
¹⁴¹ Total	8.00	8.00	8.00	0.00	8.00	8.00	8.00	8.00	8.00	8.00	
¹⁶¹ Al	2.80	2.73	2.77	0.04	2.70	2.60	2.93	3.20	2.59	2.72	
Cr	0.00	0.01	0.00	0.00	0.00	0.01	0.01	0.01	0.02	0.00	
Fe	5.09	4.94	5.02	0.07	4.39	4.79	4.71	4.66	4.99	4.96	
Mn	0.03	0.01	0.02	0.01	0.02	0.01	0.01	0.03	0.02	0.03	
Mg	3.90	3.83	3.87	0.03	4.27	4.05	3.61	3.23	3.38	3.98	
Ti	0.00	0.02	0.01	0.01	0.05	0.04	0.03	0.02	0.31	0.03	
¹⁸¹ Total	11.83	11.55	11.69	0.14	11.43	11.50	11.29	11.14	11.30	11.73	
K	0.01	0.08	0.04	0.04	0.34	0.27	0.21	0.13	0.11	0.06	
Total	19.84	19.63	19.73	0.11	19.76	19.77	19.50	19.26	19.41	19.79	
Al _{tot}	5.29	4.68	4.98	0.30	4.67	4.58	4.72	4.84	4.52	5.02	
¹⁴¹ Al/Si	0.450	0.322	0.386	0.064	0.327	0.329	0.288	0.258	0.319	0.402	
Mg/Fe	0.767	0.776	0.771	0.004	0.971	0.845	0.767	0.694	0.677	0.802	
Mg/(Mg + Fe)	0.434	0.437	0.435	0.001	0.493	0.458	0.434	0.410	0.404	0.445	
Mineral assemblage	Bt + St Grt + Sil	Bt + St Grt + Sil									

* Standard deviation.

both grades, the modal amount of garnet is quite small (see Table 2) and the garnet is partially pseudomorphed by chlorite (see above). Moreover, the data of Wood (1981) show that such garnet has relatively Mn-enriched

TABLE 5. Chlorite-bearing AFM assemblages and number of samples used in this study

Assemblages in low to moderate sulfide content rocks (southwest Rangeley area)		*	**
Metapelitic schists			
Lower garnet zone	Bt + Grt + Chl†	8	1
Lower garnet zone	Bt + Chl	2	
Middle garnet zone	Bt + Grt + Chl†	4	2
Middle garnet zone	Bt + Chl	1	
Upper garnet zone	Bt + Grt + Chl†	7	3
Upper garnet zone	Bt + Chl	3	
Lower staurolite zone	Bt + Grt + St + Chl‡	6	4
Lower staurolite zone	Bt + Grt + Chl†	2	
Upper staurolite zone	Bt + Grt + St + Chl‡	28	5
Upper staurolite zone	Bt + St + Chl‡	4	
Upper staurolite zone	Bt + Grt + Chl†	2	
Upper staurolite zone	Bt + Chl	1	
Transition zone	Bt + Grt + St + Chl + Sil‡	6	6
Transition zone	Bt + Grt + Chl + Sil‡	1	
Lower sillimanite zone	Bt + Grt + St + Sil + Chl§	2	7
Miscellaneous			
Chlorite with high K values		7	

Note: From Teichmann (1988).

* Number of samples with indicated assemblage.

** Number code on graphs of Figure 5.

† Quasi AFM limiting assemblage.

‡ True AFM limiting assemblage.

§ Retrograde chlorite.

rims. Garnet from the UGZ occurs in greater amounts, is unpseudomorphed, and contains less Mn. The coexisting chlorite clearly shows a markedly smaller Mn content than that in the LGZ and MGZ. In contrast, chlorite from the garnet-free rocks of the UGZ contains Mn at levels similar to those in the LGZ and MGZ.

Similar patterns of Mn values in the coexisting biotite were described by Guidotti et al. (1988). They considered the patterns to reflect the extent to which Mn preferentially partitions into garnet relative to biotite and chlorite.

In the three higher grades, the pattern of Mn content in chlorite is less clear than for the lower grades. This reflects the small number of samples in some assemblages (see Table 5). Nonetheless, in some assemblages the relative Mn concentration in the chlorite fits well with the model of Guidotti et al. (1988) discussed above. Specifically, in the UStZ the Mn contents of chlorite from the garnet-free rocks are relatively high for chlorite of that grade (see Table 6). This behavior is analogous to that of the coexisting biotite (Guidotti et al., 1988).

Chlorite composition as a function of metamorphic grade

Here, we discuss only chlorite from the limiting assemblage in each grade. Only in such assemblages will mineral compositions be functions solely of the intensive parameters (P , and especially T or $a_{\text{H}_2\text{O}}$, or both, being most relevant in the present case). These compositional data are most easily shown graphically (Fig. 5).

Considering the size of the error bars, it is evident that

TABLE 6. Effects of mineral assemblage on the average Mg/(Mg + Fe) ratio and the average Mn values of chlorite from the southwest Rangeley sequence (LGZ to TZ)

Zone	Mg/(Mg + Fe)				Mn			
	Bt + Grt + Chl	Bt + Chl	Bt + St + Chl	Bt + Chl	Bt + Grt + Chl	Bt + Chl	Bt + St + Chl	Bt + Chl
LGZ	0.396	0.412			0.053	0.049		
MGZ	0.397	0.410			0.045	0.046		
UGZ	0.428	0.465			0.018	0.048		
LStZ	0.503	0.489			0.026	0.024		
UStZ	0.504	0.500	0.576	0.55	0.020	0.038	0.038	0.058
TZ	0.512	0.497			0.029	0.034		

Si, Al_{tot}, ⁴Al, and ⁶Al are little affected by metamorphic grade. At most, there is a weak suggestion of an increase of Al and decrease of Si in the UStZ and TZ. Velde and Rumble (1977) made a similar observation regarding constant Al in chlorite from metapelites for a similar range of grades.

The only major elements exhibiting large, systematic changes as a function of metamorphic grade are Fe and

Mg. Here, we assume that the Fe is largely Fe²⁺ because most of the rocks contain graphite and, as suggested by Henry (1981), the coexisting ilmenite contains only small amounts of Fe³⁺ (<3% hematite component). This assumption is being tested by a Mössbauer study now underway at the University of Oregon (M. D. Dyar, in preparation).

The variations of Fe, Mg, and Mg/(Mg + Fe) shown

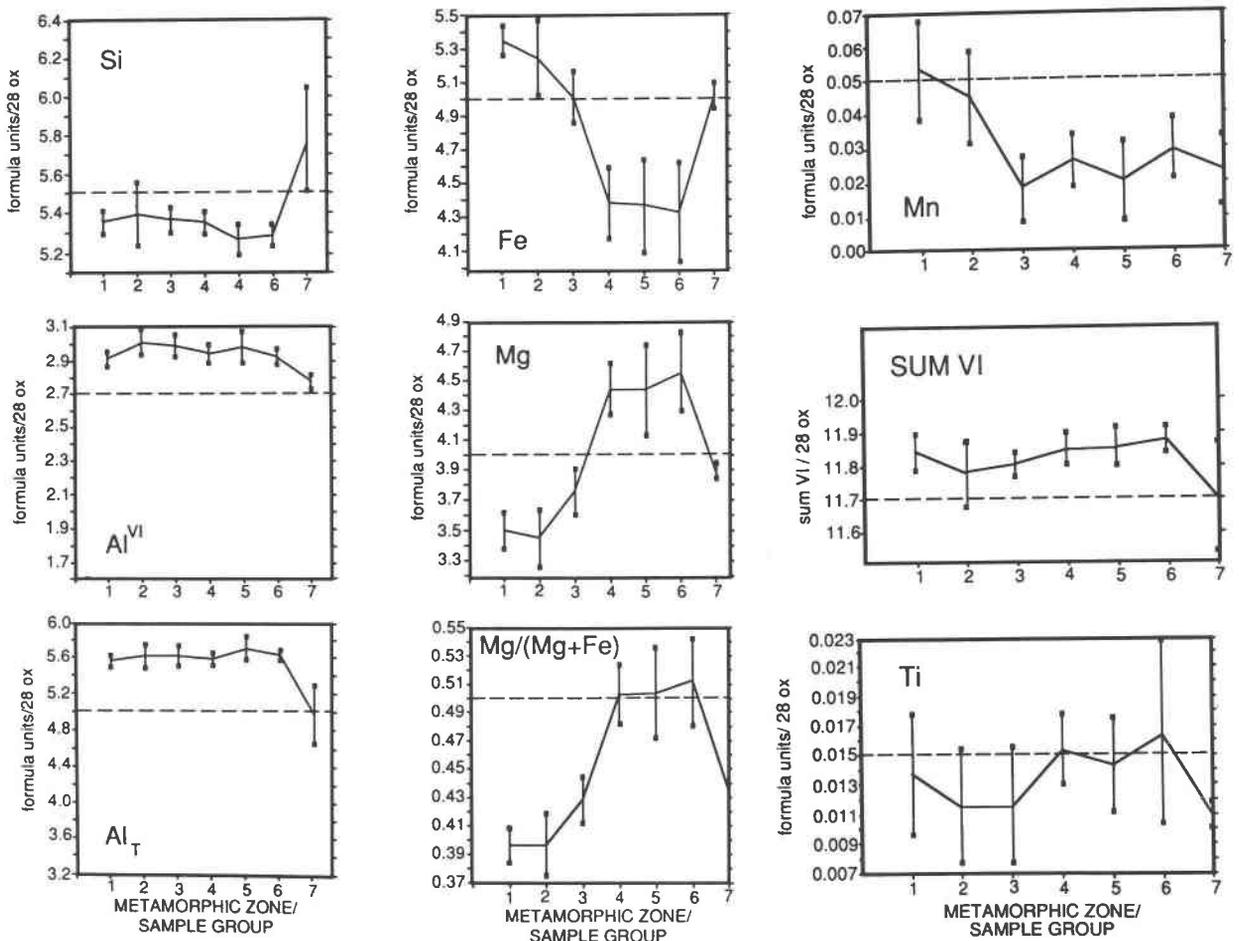


Fig. 5. Variation of important compositional parameters of chlorite from the limiting assemblages in the metapelites of the Rangeley area. Zone numbers from Figure 1. In Zone 7, chlorite is not considered an equilibrium phase.

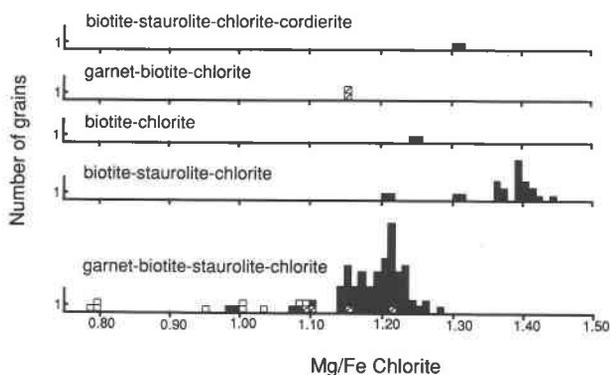


Fig. 6. Ranges of Mg/Fe ratios of chlorite for the various AFM assemblages. (From Henry, 1981). Individual chlorite analyses are grouped as follows: open box = samples without pyrrhotite, box with slash = samples with a trace to 1% modal pyrrhotite, and closed box = samples with greater than 1% modal pyrrhotite.

in Figure 5 are consistent with those anticipated from the continuous and discontinuous reactions shown in Table 1. They are also closely mirrored by similar changes in coexisting muscovite (Guidotti, 1978), biotite (Guidotti et al., 1988), and garnet (Wood, 1981). Distinct differences in the Mg, Fe, and Mg/(Mg + Fe) values are found on opposite sides of the one discontinuous reaction that occurs within the range of grades considered. Elsewhere, the variation is more gradual and continuous. Evans and Guidotti (1966) have described a similar, marked change for the Na/(Na + K) ratio of muscovite across a discontinuous reaction.

The variations of Mn and Ti contents with grade are also shown in Figure 5. The ranges of the error bars suggest that the variation of Mn may be meaningful. Moreover, the relation advanced above, that chlorite manganese content depends on the modal amount of garnet present, would also be expected to operate in response to a change in metamorphic grade. Finally, coexisting garnet (Wood, 1981) and biotite (Guidotti et al., 1988) show the same pattern of Mn variation with grade.

The pattern of Ti variation is mirrored by that of the coexisting muscovite (Guidotti, 1978) and biotite (Guidotti et al., 1988). However, the small amounts of Ti in chlorite, and the extensive overlap of the error bars makes uncertain any conclusions about the effects of grade on the Ti content of chlorite.

Chlorite composition as a function of sulfidation reactions

Although data from Teichmann (1988) and Guidotti (1974) all show the effects of sulfidation reactions on the composition of chlorite from the UStZ, Henry's (1981) large, self-consistent data base was analyzed specifically to assess the effects of sulfidation reactions. Working in the context of the general model for sulfidation reactions developed by Thompson (1972), Guidotti et al. (1988)

discussed how biotite responds to sulfidation effects. As shown below, chlorite behaves similarly.

Figure 6 shows the ranges of Mg/Fe ratios of chlorite for the various AFM assemblages present in the UStZ. The analyses are keyed to the modal amount of pyrrhotite present in the samples. For the assemblages with a large number of chlorite analyses available, Grt + Bt + St + Chl and Bt + St + Chl, the Mg/Fe ratios of the chlorite are systematically related to the modal amount of pyrrhotite in the sample. More specifically, Henry (1981, p. 106) found that in the Grt + Bt + St + Chl assemblage, "samples without pyrrhotite have chlorite Mg/Fe ratios of <1.1, whereas samples with a trace to 1% pyrrhotite have Mg/Fe ratios of 1.1 to 1.2. Samples with more than 1% pyrrhotite generally have Mg/Fe ratios of 1.1 to 1.3." Moreover, although not shown on the diagram, the garnet-absent assemblage is usually more pyrrhotite rich than is the garnet-bearing one.

Henry (1981, p. 106) also noted that "for samples in which chlorite coexists with garnet there is a positive correlation between chlorite Mn/Fe ratios and the modal amounts of pyrrhotite. However, in samples not containing garnet the chlorite Mn/Fe ratios are substantially higher than those containing garnet." In the former assemblage, the Mn/Fe ratio of the garnet rims shows the same effects from the sulfidation reactions. These observations agree with those of Guidotti (1974) and Teichmann (1988), and with these for coexisting biotite of Guidotti et al. (1988).

Other chlorite from the Rangeley area

Two other groups of chlorite samples from the study area deserve discussion. One, and possibly both, involve nonequilibrium chlorite and provide a useful contrast with the chlorite discussed above.

Table 4 presents chemical analyses of the chlorite from these two groups of samples. The first group involves fine-grained chlorite from a few LSiz specimens. In some cases this chlorite contains sagenitic rutile inclusions and is gradational with biotite, but in others it occurs as distinct laths. Invariably, it has anomalous blue to purple birefringence. As seen in Table 4 and Figure 5, the Mg/(Mg + Fe) ratio is in the range observed in garnet zone chlorites. Clearly, the trend toward progressive Mg enrichment seen throughout all of the lower grades is not followed. The Al and Si contents of this chlorite also show no systematic relationship to those in the chlorite over the rest of the metamorphic range. Finally, the relative partitioning of Mg and Fe between chlorite and biotite in these rocks is reversed from that in the UStZ and Tz (Guidotti, 1974). In aggregate, these observations epitomize the case for chlorite out of equilibrium with the other minerals of a metapelite.

The equilibrium status of the other group of chlorite for which analyses are given in Table 4 is more subtle. Texturally and optically, these UStZ and TZ chlorites are similar to other samples in these zones, but their compositions show several unusual features (Table 4). They

contain significant amounts of K, an element for which chlorite has no structural site, as well as significant amounts of Ti. It is notable that the large group of chlorites for which the averages given in Table 3 are based contain little Ti, although they are Ti saturated by virtue of the presence of ilmenite. Finally, their Si and Al contents are systematically higher and lower, respectively, than the other chlorite of the Rangeley area.

These analyses may represent partially altered biotite or chlorite intergrown with biotite on a scale too fine to resolve with the electron beam. Anomalously high Ti and K values are compatible with either of these hypotheses. Regardless of which is the better interpretation, it is clearly unreasonable to integrate these analyses with those obtained for the previous groups. Laird (1988) has also emphasized the need to be suspicious of chlorite containing significant K.

DISCUSSION

Comparison of data for coexisting chlorite, biotite (Guidotti et al., 1988), and muscovite (Guidotti, 1978) shows that site substitutions in chlorite are simpler and more constrained than in the micas. On the other hand, chlorite from metapelites, like muscovite and biotite, shows significant compositional response to both mineral assemblage and metamorphic conditions (especially T and fluid composition). Hence, like the micas, it has the potential to be a useful petrogenetic indicator mineral. Our chlorite data should be particularly useful for studies that attempt to model quantitatively the equilibria in pelitic schists (e.g., Spear and Cheney, 1989).

Our data show that the Al content of chlorite is independent of T (and variation of fluid composition), at least over the range of grades affecting our rocks. Reports suggesting an increase of Al as grade increases (Frost, 1975; Ernst et al., 1970) involve assemblages not initially Al saturated, and hence, the Al enrichment is driven by metamorphic reactions and not inherent crystallochemical factors. A general discussion of such reactions in metapelites is given by Thompson (1979).

The case for Ti enrichment in chlorite as a function of T is ambiguous at best because of the small amount of Ti that occurs in our chlorite. In contrast, micas exhibit saturation levels for Ti substitution that clearly change as a function of T , and in the case of biotite as a function of the Mg/(Mg + Fe) ratio also (Guidotti et al., 1977).

From the above discussion it is clear that in a petrologic context chlorite has much in common with the micas. However, its equilibrium status is more frequently questioned, primarily because of its textural attributes. The most important conclusion of this study is that a strong case for close approach to chemical equilibrium between chlorite and other minerals can be made despite the presence of features indicating a lack of textural equilibrium for chlorite.

Some petrographic features do suggest an approach to equilibrium for the chlorite in this study. For example, its modal percent varies systematically with grade, and

in individual specimens it exhibits sharp grain boundaries and uniform optical properties, even though as many as three textural varieties can be present. The strongest arguments for a close approach of chlorite to chemical equilibrium with the other phases from the LGZ to the TZ are, however, based mainly on mineral compositions. They involve the Mg/(Mg + Fe) ratio and Mn content, and they include the following.

1. In a given sample, chlorite composition is uniform from grain to grain and within a given grain, regardless of its textural appearance or size fraction (Teichmann, 1988).

2. At a given grade, chlorite composition shows the expected response to variation of mineral assemblage or to the extent of sulfidation reactions.

3. The chlorite varies in composition in response to metamorphic grade as expected across both continuous and discontinuous reactions. Because the sulfidation reactions cause a decrease in $a_{\text{H}_2\text{O}}$, they simulate some aspects of the effects of the prograde continuous reactions but at constant grade (i.e., T). For example, in the limiting assemblages they cause a Mg enrichment of Mg-Fe phases like chlorite. However, they also have the net effect of displacing the effective bulk composition to more Mg-rich portions of composition space. Consequently, they result in a model decrease of minerals like garnet, and this, through the Mn-concentration effect noted above, results in an increase in the Mn/Fe ratio of all of the Mg-Fe phases.

4. Chlorite compositional variations mirror those shown for the coexisting muscovite, biotite, and garnet, even for a relatively minor constituent like Mn. Mg, Fe, and Mn are systematically partitioned among all four phases and also are systematically related to metamorphic grade, sulfidation reactions, and mineral assemblage.

5. Deviation from these systematic chemical patterns provides the strongest argument that the chlorite present in a few LSiZ specimens is not part of an equilibrium assemblage. (See Fig. 5 and compare Tables 3 and 4.)

The textural features of chlorite in this study are common in many areas and indicate that complete equilibrium was not attained. In this case they result from the rocks having a polymetamorphic history (Guidotti, 1970, 1989; Holdaway et al., 1982). The cause of the polymetamorphism has been discussed by Lux et al. (1986) and DeYoreo et al. (1989). The last metamorphism caused recrystallization of the earlier mineral assemblages to the extent that chemical equilibrium among the minerals was closely approached, but not to the extent that textural equilibrium was attained. In spite of textural disequilibrium, the mineral composition data can be used to ascertain the intensive parameters that prevailed during the last metamorphic event to affect these rocks.

As noted by Thompson (1987, p. 169), "Disequilibrium textural features are of special interest because each carries with it a historical message." In the present case, disequilibrium textures of chlorite and other minerals provide much of the evidence for the polymetamorphic

history of the rocks. Had complete equilibrium been attained (chemical, textural, etc.), we would see no evidence for the polymetamorphism. In fact, in the higher grade rocks of western Maine (upper sillimanite zone and sillimanite + potassium feldspar zones), the rocks are so strongly recrystallized that textural equilibrium has been closely approached, and the rocks no longer retain the evidence for a polymetamorphic history. In essence, some of their history has been erased.

In conclusion, it appears that the assessment of whether or not chlorite in pelitic schists has approached equilibrium to the extent that it can be used for geothermometry and other petrologic analysis should be based primarily on mineral compositions. This should be done in terms of the compositions of all coexisting phases and their response to the overall petrologic setting (grade, assemblage, sulfidation reaction, etc.). The textural appearance of chlorite should be used to assess its approach to chemical equilibrium only as a last resort.

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