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# Polytypism of chlorite in very low grade metamorphic rocks

JEFFREY R. WALKER
Department of Geology and Geography, Vassar College, Poughkeepsie, New York 12601, U.S.A.

## ABSTRACT

It has been suggested that chlorite polytype transformations may be temperature dependent, changing from type I to type II between 150 and 200 °C. The ideal sequence has not yet been documented in the field, and conditions of polytype stability have not been described completely. A compilation of reported occurrences of type I chlorite structures suggests that they are restricted to coarse-grained rocks and are rarely found in pelitic rocks.

The very low grade metasedimentary and metavolcanic rocks of Aroostook County, Maine, appear to be appropriate to document the transition from type I to type II chlorite because temperatures of metamorphism estimated in the area range between 50 and 200 °C. Polytype analyses performed on chlorite separates from these very low grade rocks, however, are exclusively type II*b*.

Detrital chlorite in sediments is usually considered to be II*b*, whereas chlorite in altered volcanic rocks may be of any polytype. Because the type II*b* polytype occurs in both metasedimentary and metavolcanic rocks of the area, the chlorite separates cannot be exclusively of detrital origin. Pore pressure and time may have been as important as temperature in controlling polytype occurrences.

## INTRODUCTION

It has been suggested that changes in chlorite stacking polytypes (Bailey and Brown, 1962) may be temperature dependent. Hayes (1970) proposed that crystallization of chlorite polytypes in the sequence

$$\mathrm{I}b_d \rightarrow \mathrm{I}b(\beta{=}97^\circ) \rightarrow \mathrm{I}b(\beta{=}90^\circ) \rightarrow \mathrm{II}b$$

represents increasing stability and progresses as temperature increases from surface temperatures to around 200 °C. Hayes further observed that structural changes seem most sensitive to temperature variations and relatively insensitive to changes in composition of the chlorite. These characteristics suggest that chlorite polytype variations could be calibrated as a geothermometer. The idealized sequence of Hayes has never been documented in the field, and physical factors governing occurrence of chlorite polytype changes have not been described completely. Results presented in this paper indicate that petrogenetic conditions in addition to temperature influence the occurrence of chlorite polytypes.

## CHLORITE POLYTYPISM

The many possible relative arrangements of the silicate and hydroxide layers of chlorite give rise to six semirandom, one-layer polytypes. Chlorite polytypes are derived by holding the silicate layer orientation constant and changing the orientation of the hydroxide layer. Since the repeating silicate layers are constrained to have the same orientation, the resulting structures are termed "one-layer" polytypes. A complete discussion of the derivation of chlorite polytypes is given by Bailey and Brown (1962).

Four of the six possible polytypes have been observed in nature. They are (in decreasing order of abundance): II*b*, I*b*($\beta$=90°), I*b*($\beta$=97°), and I*a*. The relative stability of these four polytypes is a function of superposition of tetrahedral and octahedral cations and of hydrogen-bond-length optimization (Shirozu and Bailey, 1965). The semirandom polytypes can be distinguished by their (*h*0*l*) reflections in random X-ray powder photographs. Hayes (1970) proposed a seventh polytype, I$b_d$ (disordered), for disordered type I chlorite that can be recognized by strong (00*l*) reflections and extinct (*h*0*l*) reflections.

Previous chlorite polytype studies fall into four categories: (1) theoretical polytype studies and/or ones involving new (or unusual) varieties (Bailey and Brown, 1962; Brown and Bailey, 1963; Shirozu and Bailey, 1965, 1966; Eggleton and Bailey, 1967; Lister and Bailey, 1967), (2) studies of samples from mineral collections (Bailey and Brown, 1962; Hayes, 1970), (3) one or two analyses as part of a more extensive investigation of phyllosilicates (Weaver et al., 1983; Beskin, 1984), and (4) systematic study of chlorite polytypes as part of an investigation of the diagenetic or metamorphic history of an area (Karpova, 1969; this investigation).

Studies in the first category established the determination of polytype as an important facet of the description of new chlorite varieties. Systematic study of chlorite in rock sequences has, however, lagged behind. Bailey and Brown (1962) determined the polytype of over 300 chlorite specimens from various mineral collections. Over 80% of the specimens studied were shown to be II*b*, whereas the remaining 20% were shown to be type I (*a* or *b*).

**TABLE 1.** Type I chlorite polytype occurrences by lithologic category

| Lithologic category | I*b*(β=90°) | I*b*(β=97°) | I*b*$_d$ | I*a* | Total | % of total |
|---|---|---|---|---|---|---|
| 1. Altered igneous rocks | 2 | 6 | 1 | 2 | 11 | 11 |
| 2. Iron formations | 12 | 6 | 0 | 3 | 21 | 22 |
| 3. Hydrothermal veins | 7 | 3 | 0 | 1 | 11 | 11 |
| 4. Limestones | 7 | 4 | 3 | 3 | 17 | 17 |
| 5. Sandstones | 18 | 0 | 0 | 2 | 20 | 21 |
| 6. Metamorphosed pelites | 1 | 0 | 0 | 1 | 2 | 2 |
| 7. Pegmatites | 0 | 0 | 0 | 6 | 6 | 6 |
| 8. Ultramafic rocks | 0 | 0 | 0 | 3 | 3 | 3 |
| 9. Unclassified | 0 | 1 | 1 | 5 | 7 | 7 |
| Total occurrences | 47 | 20 | 5 | 26 | 97 | 100 |
| % of type I occurrences | 48 | 20 | 5 | 27 | 100 | |

Karpova (1969) determined the polytype of chlorite formed as cement in sandstones from the Bolshoy Donbas in Russia and speculated that the transformation I*b*(β=90°) → II*b* occurred with increasing diagenesis. Karpova did not include any temperature estimates for these rocks but did indicate that the Fe content of the chlorite decreased with increasing grade. Hayes (1970) investigated chlorite extracted from a variety of sedimentary rocks and proposed that the sequence of chlorite crystallization with increasing temperature was

$$Ib_d \rightarrow Ib(\beta{=}97°) \rightarrow Ib(\beta{=}90°) \rightarrow IIb.$$

He further proposed that temperatures on the order of 200 °C are necessary to make the final transition from the I*b*(β=90°) to the II*b* structure because of the 60° or 180° hydroxide-layer rotation required by the transformation. In a detailed study of the shale-slate transition in the southern Appalachians, Weaver et al. (1983) analyzed two samples from the southern Appalachians and found that the chlorite from lower-grade rocks (high-grade diagenesis, $T_{max}$ = 250 °C) is I*a* whereas chlorite in the higher-grade sample (lower epizone, $T_{max}$ = 330 °C) is II*b*. In well samples from the Tuscaloosa Formation in the Gulf Coast, Beskin (1984) found type I*b*(β=90°) coexisting with type II*b* chlorite at a depth of 6.5 km, corresponding to a borehole temperature of approximately 190 °C.

The results of these studies imply that II*b* chlorite is the stable end-product of thermal metamorphism and that the transition from type I (*a* or *b*) to type II*b* occurs at temperatures somewhere between 150 and 250 °C. None of these studies, however, has been specific enough or detailed enough to test parameters such as temperature, composition of the circulating fluids, or nature of the protolith that might affect the transition.

A compilation was made of as many reported occurrences of type I chlorite as could be found, both published and unpublished (courtesy of Dr. S. W. Bailey), in order to describe the conditions of occurrence of type I chlorite in more detail. Occurrences of the various polytypes were subdivided by lithologic categories, and these data are summarized in Table 1; the complete listing has been placed on deposit.[1]

Rock type is used to categorize occurrences of type I chlorite because it is the only information common to the sample descriptions presented in studies of type I chlorite. Data concerning the temperature and pressure of formation of the chlorite are lacking in most cases. It is unfortunate that type II*b* chlorite occurrences, which have not been reported in such detail in the literature, could not be included in this tabulation so that the proportions of the four type I polytypes in each of the different lithologic categories could be compared to the proportion of type II chlorite. Of the 300+ samples studied by Bailey and Brown (1962), 80% were II*b*, occurring primarily in "chlorite-grade metamorphic rocks or in veins or alteration zones close to ore bodies" (S. W. Bailey, personal communication, 1986). However, few specific results have been reported.

Table 1 indicates that certain lithologic categories, notably altered igneous rocks, iron formations, and limestone, have a nearly equal distribution of the four type I structures whereas the other rock types contain only one or two polytypes. Particularly interesting is the observation that type I chlorite in sandstones is almost exclusively I*b*(β=90°). The relationship between polytype and geologic occurrence indicates some of the important variables affecting the stability of type I chlorite: (1) Type I chlorite forms preferentially in rock types that afford open space into which the mineral can crystallize, as shown by the common occurrence of type I chlorite as vug and vein fillings and by its preference for standstones and granular limestones (as opposed to pelites or micritic limestones). (2) Type I chlorite forms preferentially in an environment with a high pH as evidenced by the occurrence of all four type I structures in limestones. (3) Type I chlorite forms preferentially at lower temperatures than II*b* chlorite, as

[1] A copy of the listing of various polytype occurrences may be ordered as Document AM-89-413 from the Business Office, Mineralogical Society of America, 1625 I Street, N.W., Suite 414, Washington, D.C. 20006, U.S.A. Please remit $5.00 in advance for the microfiche.

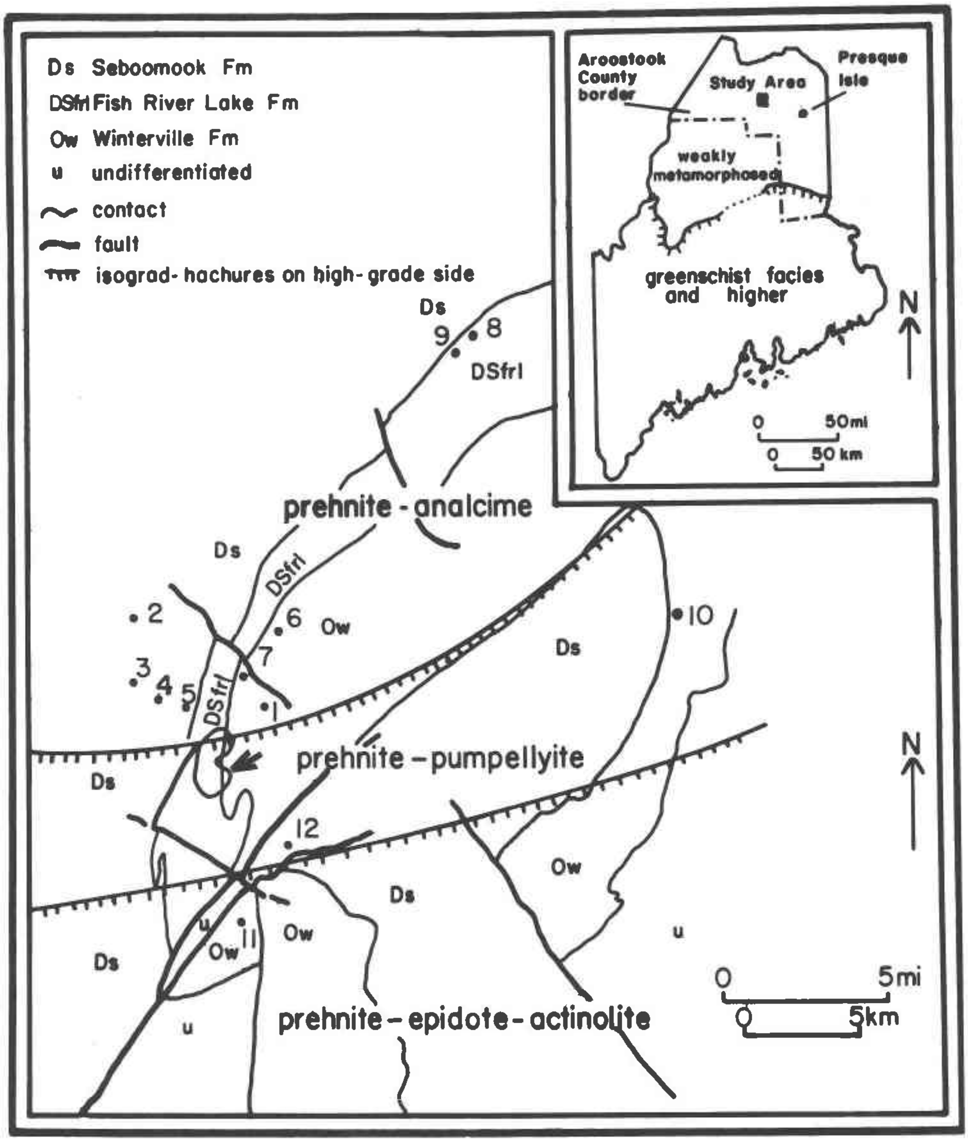


Fig. 1. Generalized geology of the Fish River Lake area after Boone (1958, 1979) and Horodyski (1968). Dots represent sample locations referred to in the text and in Table 2. The small, kidney-shaped area in the east-central part of the map (indicated by an arrow) is Fish River Lake. Inset: Location map of study area. Metamorphic facies after Osberg et al. (1985).

demonstrated by the distribution of chlorite polytypes within hydrothermal vein systems: type II*b* chlorite is more common in the higher-temperature portions of a vein system, and type I chlorite is more common in the lower-temperature portions (S. W. Bailey, personal communication, 1986).

The purpose of the present investigation is to study chlorite polytypes in a very low grade metamorphic terrane in an attempt to document variations with changing grade. Most type I chlorites that have been described previously occur in sedimentary rocks and low-temperature hydrothermal veins. Most metamorphic chlorites previously analyzed have the II*b* structure. Assuming that type II metamorphic chlorite forms at least in part from previously crystallized type I chlorite, the low-grade metamorphic terrane in northern Maine, which covers a temperature range between 100 and 250 °C, was chosen to study the transition from type I to type II chlorite.

## Field setting, Aroostook County, Maine

The study area is located near Fish River Lake in central Aroostook County (Fig. 1, inset), approximately 40 km northwest and west of Presque Isle. The region is characterized as "weakly metamorphosed" (Osberg et al., 1985), well below the greenschist and higher levels of metamorphism in southern Maine. Detailed geologic mapping of this area by Boone (1958, 1979) and by Horodyski (1968) has been generalized in Figure 1.

Three principal units crop out within the field area: (1) Ordovician volcanic rocks of the Winterville Formation; (2) Siluro-Devonian pelitic rocks of the Fish River Lake Formation; and (3) Devonian slates of the Seboomook Formation. Minerals representative of prehnite-pumpellyite–facies metamorphism have been reported from the Winterville Formation. Metamorphism increases from north to south and is believed to have occurred in the Devonian during the Acadian orogeny (Coombs et al., 1970).

Richter and Roy (1976) mapped zonation within the prehnite-pumpellyite facies and concluded from the absence of high-temperature zeolites that temperatures during metamorphism were moderate (150–200 °C). A preliminary study of conodont color-alteration indices indicates an increase in temperature from 50 °C in the north to 250 °C in the south [Anita Harris, personal communication, cited in Roy (1980)].

## Experimental techniques and results

Polytype analyses were made on chlorite and muscovite from nine pelitic and three volcanic samples from the Fish River Lake vicinity. Samples were collected primarily in the lower grade (prehnite-analcime) region of the study area (Fig. 1) and are assumed to represent peak metamorphic conditions.

Chlorite samples that contained a high percentage of white mica were purified using high-gradient magnetic separation (Walker, 1986a) until the proportion of mica was less than 25%. Chlorite polytype analyses were performed on spindles of randomly oriented powder using a Gandolfi camera (Fe$K\alpha$; 114.6-mm diameter). Polytypes were identified according to Bailey (1984, Table 1.22). Physical mixtures of two or more chlorite polytypes are difficult to identify if one of the phases is present in small amounts (<10%). Mica polytypes were identified using the method of Maxwell and Hower (1967), which gives the proportion of $2M_1$ muscovite to total muscovite ($2M_1$ + $1M_d$).

Results of polytype analyses are presented in Table 2. Chlorite is uniformly the II*b* polytype in the pelitic and volcanic rocks analyzed from northern Maine. The volcanic rock samples also contain significant percentages of an interstratified chlorite/smectite phase along with discrete chlorite (Walker, 1986b). In sample no. 6, chlorite/smectite is the dominant trioctahedral clay, and a broad band is observed at $d = 2.55$ Å, indicative of turbostratic layer-stacking of the smectite.

The proportion of $2M_1$ muscovite ranges from 18 to 66% (Table 2). Samples analyzed for both chlorite and muscovite contain between 33 and 66% $2M_1$ mica.

## Discussion

Exclusive occurrence of II*b* chlorite in northern Maine is surprising in light of the fact that the maximum temperature estimated for the field area is on the order of 150 to 200 °C, which brackets the temperature range suggested by Hayes (1970) for the transition from type I to type II structures. Two explanations for this are possible: (1) the chlorite may be primarily detrital, and (2) conditions during metamorphism may have been such that type I chlorite never formed or was changed to type II sometime after formation.

**Table 2.** Polytype determinations—northern Maine

| Sample | Chlorite polytype* | Mica polytype** | Rock type |
|---|---|---|---|
| 1 | II*b* | n.d. | volcanic |
| 2 | II*b* | n.d. | pelite |
| 3 | II*b* | 66% | pelite |
| 4 | n.d. | 50% | pelite |
| 5 | II*b* | n.d. | pelite |
| 6 | II*b* + turbostratic | n.d. | volcanic |
| 7 | II*b* | n.d. | volcanic |
| 8 | II*b* | 66% | pelite |
| 9 | II*b* | 33% | pelite |
| 10 | II*b* | n.d. | pelite |
| 11 | n.d. | 18% | pelite |
| 12 | n.d. | 33% | pelite |

*Note:* n.d. = not determined.
* Bailey (1984).
** % $2M_1$ of total mica; Maxwell and Hower (1967).

### Detrital influence

Previous studies have shown that the II*b* polytype is the most common chlorite structure, especially in metamorphic terranes. On the basis of this observation, it has been inferred that the II*b* polytype is also the most stable and that detrital chlorite consists primarily of the II*b* structure (Hayes, 1970). Occurrence of II*b* chlorite in meta-igneous rocks of northern Maine argues against the hypothesis that the chlorite in all of the samples studied is detrital. The data do not preclude, however, that some of the premetamorphic chlorite in the metasedimentary rocks was detrital and that newly crystallized metamorphic chlorite formed as epitaxial overgrowths on detrital precursors, adopting the II*b* structure.

Because the Acadian orogeny is assumed to have been the principal metamorphic event in the area (Osberg et al., 1985), however, the Winterville volcanic rocks (which constitute the probable source terrane for the pre-Acadian units) would not have been metamorphosed prior to erosion, and detrital chlorite would, therefore, not be expected as a common constituent in the clastic material making up the Fish River Lake and Seboomook Formations. In addition, pelitic rocks in the field area have undergone extensive recrystallization as evidenced by a redistribution of Fe between octahedral sites in the chlorite structure (total Fe remains constant) with increasing grade and by the strong degree of preferred orientation of chlorite grains parallel to the plane of foliation (Walker, 1987). It is expected, therefore, that detrital grains are rare, and that the chlorite analyzed in this study grew during diagenesis and Acadian metamorphism, equilibrating with the maximum temperatures achieved.

### Petrogenetic control

Metamorphic conditions that could affect the stability of chlorite polytypes include temperature, pressure (lithostatic and hydrostatic), and time. It is likely that a combination of these factors is important in determining the chlorite polytypes found in the rocks of northern Maine.

Metamorphic temperatures in the study area have been estimated by previous investigators using two independent paleothermal indicators: conodont alteration colors (Roy, 1980) and equilibrium mineral assemblages (Richter and Roy, 1976). These estimates agree that the maximum temperature achieved in the area was no more than 250 °C. Minimum temperature estimates range from 50 °C from conodont colors to 150 °C from mineral assemblages. The occurrence of 50/50 ordered interstratified chlorite/smectite in the volcanic rocks of the study area (Walker, 1986b) further corroborates these temperature ranges because chlorite/smectite has been shown to be a stable phase between 70 and 200 °C (Chang et al., 1986; Tomasson and Kristmannsdottir, 1972). The 150 °C minimum temperature estimated from mineral assemblages is at the lower end of the proposed temperature range for the type I to type II transition (150 to 200 °C; Hayes, 1970), and so if temperature is the primary control on the polytype transformation, type I chlorite formed during metamorphism might be expected in those areas that experienced the lowest temperatures. However, no evidence has been found of type I chlorite. Mica polytypes are variable within the study area, and in no sample is pure $2M_1$ mica found. Occurrence of mixed $2M_1$ and $1M$ micas suggests that temperatures were not high enough to form the most stable mica structure. A similar relationship between chlorite and mica polytypes is present in fine-grained rocks of the Salton Basin, California, where chlorite is exclusively II*b* and illite varies systematically from $1M_d$ to $1M + 2M_1$ over the temperature interval 135 to 275 °C (Walker and Thompson, 1990).

It is striking that type I chlorite has rarely been reported from fine-grained, low-grade pelitic rocks (Table 1). At the outset of this investigation, it was assumed that this fact was due primarily to the difficulties inherent in separating chlorite from interfering phases in a fine-grained sample. The results of the study of chlorite from northern Maine imply, however, that type I chlorite may be rare in pelitic rocks even at very low metamorphic grades. Growth of type I chlorite may be suppressed in fine-grained rocks by elevated pore pressures. In coarse-grained rocks, such as sandstones or granular limestones, pore pressures are expected to be predominantly hydrostatic and lower than total lithostatic pressure. In fine-grained rocks, on the other hand, pore pressures are likely be higher and to approach lithostatic pressure. The relationship between chlorite polytypes and fine-grained rocks seen in northern Maine and in the Salton Basin (Walker and Thompson, 1990) suggests that $P_{H_2O}$ may have to be less than $P_{total}$ for type I chlorite to form.

The effects of time on the transition from type I to type II chlorite are not well known. Type I chlorite has been reported from Precambrian iron formations dated at 1900 Ma and from Phanerozoic localities as well (Bailey and Brown, 1962; Hayes, 1970). Conversely, authigenic II*b* chlorite has been reported from rocks as young as Pliocene-Pleistocene (Walker and Thompson, 1990). Although the effects of the Acadian orogeny were not felt over an extended period of time in northern Maine, it may be that thermal input was sufficient to provide a strong start to the type I to type II reaction and that the time elapsed between the Devonian and the present allowed for a complete transformation of structures.

Knowledge of the stability of chlorite polytypes is limited. Occurrence of II*b* chlorite exclusively in the rocks of northern Maine may be explained by a combination of temperature, pressure, and time effects. The picture becomes complicated when one tries to reconcile these results with those from the Salton Basin where II*b* is the only polytype detected in young, low-temperature rocks. Careful experiments are required to clarify which, if any, of the chlorite polytypes are stable under controlled conditions. It is possible that type I chlorite forms as a metastable phase that slowly transforms to the stable II*b* polytype given sufficient time under elevated temperature and pressure conditions. Thermodynamic data necessary to evaluate this hypothesis, however, are not available.

## Conclusions

Results of this study have shown that the II*b* polytype is the dominant chlorite structure in very low grade pelitic and volcanic rocks in northern Maine. This homogeneity could result from a combination of effects. A detrital source for all of the chlorite is ruled out; furthermore, a significant amount of recrystallization has occurred during metamorphism. Pore pressure and time, however, may have been as important as temperature in controlling polytype occurrences; this possibility suggests that the crystallization sequence proposed by Hayes (1970) is not applicable to sedimentary and metamorphic rocks in general but is limited to particular rock types such as sandstones and granular limestones.

Polytype studies to date have not focused on the environmental factors governing the occurrence of type I and type II chlorite. Results compiled in this study are too general to allow detailed characterization of the conditions under which different chlorite polytypes are formed. Type I chlorites comprise only about 20% of the naturally occurring chlorites that have been studied to date, yet they potentially contain valuable information about the environment in which they formed if structural variations are related to the ambient conditions of their formation. Occurrences of type I chlorites, therefore, could yield important petrogenetic information about their host rocks once the relative stabilities of type I and type II chlorites are characterized in detail.

*Note added in proof:* For a more complete description of the high-gradient magnetic separation procedure, see the recently published paper by Tellier, K. E., Hluchy, M. M., Walker, J. R., and Reynolds R. C., Jr. (1988): Application of high-gradient magnetic separation (HGMS)

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