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HIGH-GRADE DIAGENESIS AND LOW-GRADE METAMORPHISM OF ILLITE IN THE PRECAMBRIAN BELT SERIES

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ABSTRACT

The response of illite to increasing stratigraphic depth was investigated in the Precambrian Belt series in western Montana and northern Idaho. The Belt series thickens from east to west and reaches a thickness of 38,000 feet, so that rocks low in the section have been under high pressure and probably relatively high temperature.

The illite of the geosynclinal Belt argillites has little or no mixed layering and contains significant iron and/or magnesium. No 2M polymorph was detected in a 4500 foot section from the Little Belt Mts., Montana, which represents the shallow eastern part of the series. In contrast to this the very thick (38,000 feet) Pend Orielle Lake, Idaho, section contains measurable 2M polymorphic material in all samples. In this latter section there is an increase in amount of 2M with depth until essentially all the dioctahedral mica is of the 2M polymorph.

Increasing temperature and pressure conditions accompanying deep burial and deformation apparently cause a transformation of the 1M*d* to the 2M polymorph in dioctahedral mica. This transformation is paralleled by textural reconstitution of the argillites to phyllite and completed at the biotite isograd.

INTRODUCTION

The changes experienced by layer silicates during the evolutionary development of geosynclines can be understood by examination of natural systems and synthesis experiments. This paper treats the changes in the dioctahedral potassium clay mineral illite, as displayed in the Precambrian Belt series, Montana and Idaho.

The micas, with which illite is often grouped, occur in several different polymorphs resulting from different stacking of the 2:1 units. Ordered rotations of the structural layers in the *a-b* crystallographic plane give rise to the various possible polymorphs. Some of the simpler rotational polymorphs have been considered by Smith and Yoder (1956).

In a study of natural muscovites and muscovite-like material Yoder and Eugster (1955) found that the commonly occurring polymorphs of natural dioctahedral micas are 2M, 1M, 1M*d*, and rarely 3T. Yoder and Eugster found that at the ideal muscovite composition the 2M structure is the stable polymorph at temperatures greater than 200°–350°C and at 15,000 psi water pressure. Below this temperature range, the 1M polymorph is apparently the stable form with metastable 1M*d* the initial phase at all temperatures and persisting at lower temperatures. These authors observed the transformation of 1M*d* to 1M to 2M upon increasing the temperature of the system and suggest that this sequence occurs in the progressive metamorphism of a sediment. They were unable to reverse the sequence. A recent phase equilibria study indicates that although 1M*d* and 1M polymorphs precede the formation of the 2M polymorph at low and moderate temperatures, 2M is the only stable form (Velde, 1965).

Burst (1959) is one of several investigators who have observed that the amount of expandable and mixed-layer clays in natural sediments diminishes as a function of geologic age. He suggests, however, that geologic age is not the controlling factor. Rather, he proposes that these transformations are a function of time, pressure, and temperature, and that the latter two can be related to depth of burial and the geothermal gradient. Further, he states that with increasing overburden, progressive metamorphism must ensue from the time of original deposition until recognizable metamorphic assemblages result. To demonstrate this, Burst describes a mineralogical trend in which montmorillonite changes first to mixed-layer illite/montmorillonite then to illite accompanied by the appearance of chlorite with increasing depth of burial in the Eocene Wilcox Formation of the Gulf Coast. The clay mineral assemblage at the maximum depth sampled (16,000 ft) was illite + chlorite ± kaolinite. Polymorphic forms of the illite are not reported.

Paleozoic shales have been shown to contain 1M*d* illite as the predominant illite polymorph in the less than one-micron size fraction (Velde and Hower, 1963). The above authors apply polymorphic stability ranges for muscovites as determined by Yoder and Eugster (1955) to the somewhat compositionally different illite and suggest that illite probably forms initially at some low temperature stage of the sedimentation-lithification process.

In contrast to these are the greenschist facies metamorphic rocks which contain the dioctahedral mica, muscovite, which appears to be exclusively of the higher temperature 2M polymorph (Turner and Verhoogen, 1960). The following analysis treats the dioctahedral micas in a sequence bridging the gap between sedimentary and metamorphic rocks.

Belt Series

The response of the dioctahedral micas to increasing depth of burial was investigated in the geosynclinal sedimentary rocks of Precambrian Belt age in western Montana and northern Idaho. This series was laid down over a wide geographic expanse; it crops out extensively in western Montana, northern Idaho, and British Columbia. The series is comprised

FIG. 1. Index map of western Montana and northern Idaho.

mainly of argillites, quartzites, and lesser amounts of carbonates. These sedimentary rocks are usually well consolidated, extremely fine grained, and have few distinctive horizons.

The thickness of the series generally increases from east to west. In the Little Belt Mountains, southeast of Great Falls, Montana, Weed (1900) reports a measured section of 4,507 feet. To the west in the Big Belt Mountains east of Helena, Mertie *et al.* (1951) report an aggregate thickness of more than 8,000 feet of Belt rocks. In Glacier National Park, Ross (1959) reports 16,000 feet plus the unmeasured Missoula Group which he believes to be between 10,000 feet and 20,000 feet. Farther west, at Pend Oreille Lake, Idaho, Harrison and Jobin (1963) report a thickness of at least 38,000 feet. Because of this extreme thickness, sedimentary rocks deep within the geosyncline have been under high pressure and probably relatively high temperature. The location of the four sections mentioned above may be seen on the accompanying index map (Fig. 1).

The lack of consensus with regard to correlation makes it necessary that the following analysis treat entire stratigraphic sections rather than individual stratigraphic units.

ANALYTICAL PROCEDURES

Sample preparations. Thirteen argillite samples from the Little Belt Mountains section, six argillite samples from the Glacier National Park section, and nineteen argillite samples from the Pend Oreille Lake Section were selected from a larger set for detailed X-ray analysis. The extreme stratigraphic thickness and structural complexity of the outcrop areas necessitated that both the Glacier Park section and Pend Oreille Lake section be composite. Only the Little Belt Mountain section is continuous.

The samples were broken in an iron mortar, and the size fraction between 12 mesh/inch and 35 mesh/inch was disaggregated with a Bendix ultrasonic generator. This was done to minimize reduction in intrinsic grain size. The resulting suspension was separated by centrifugal sedimentation into a less than 2-micron fraction and a residuum which consisted mainly of greater than 2-micron material.

X-ray diffraction. The samples were analyzed with a North American Philips diffractometer using CuKα radiation. Minerals were identified, and the shape and symmetry of the illite (001) reflections were observed without making quantitative measurements on oriented specimens (Kinter and Diamond, 1956).

The b dimensions of the unit cell were determined by measuring the (060) spacing using the quartz reflection at 1.541 Å as an internal standard. In most of the samples, additional quartz was added to the quartz already existing in the sample. The (060) spacings calibrated with quartz were measured for the less than 2-micron size, randomly oriented samples run at $\frac{1}{4}$°/min. The centers of the peaks were determined at a point about $\frac{1}{3}$ of the distance from the top of the peaks in question.

The standard deviation of the (060) spacing was measured by using six different random orientations of the less than 2-micron fraction of sample number P_60_3. The mean was 1.509 and $s=0.0005$, or 0.03 percent of the mean. Determination of this parameter on a diffraction trace run at $\frac{1}{4}$°/min rather than 1°/min is believed responsible for the high degree of precision obtained.

The polymorphic forms of dioctahedral mica were determined for the less than 2-micron size fraction. Data were obtained for material previously boiled 30 minutes in 2N HCl and consequently free from any interference from either biotite or chlorite. Both of these minerals are soluble in HCl and are removed with the above treatment. It is assumed that the polymorphic forms of dioctahedral mica from the belt series are similar in structure to the polymorphic forms of muscovite as established by Yoder and Eugster (1955). If so, the most intense unique reflections for the 2M dioctahedral mica fall between 20° and 32° 2θ for CuKα radiation. The percentage of 2M dioctahedral mica relative to total dioctahedral mica was determined by obtaining the intensity ratio of an unique 2M peak to a peak representing all the dioctahedral mica polymorphs. It was not possible to utilize the most intense unique 2M reflections because they coincide with feldspar reflections. Consequently, the lower intensity 2M $(11\bar{6})$ peak was chosen as the numerator of the desired ratio. This reflection occurs at 2.80 Å and is free from interference by feldspar. The 2.58 Å peak was used as the denominator. The 2.58 Å peak includes the

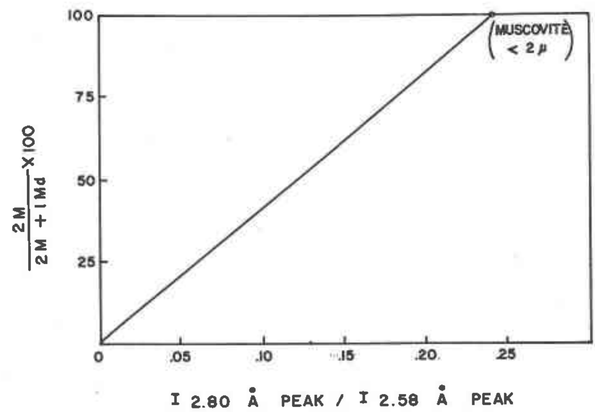

FIG. 2. Curve for determination of percent 2M dioctahedral mica.

2M $(13\bar{1})$, (116), and $(20\bar{2})$ reflections and the 1M $(13\bar{1})$ and (130) reflections. The latter reflection is also observed in 1M*d* muscovite. The 2.58 Å peak receives nearly equal intensity contributions from all the dioctahedral mica polymorphs.

The above ratio is compared to a 2.80 Å peak/2.58 Å peak intensity ratio of muscovite to determine the percentage of 2M dioctahedral mica in the Belt samples. The muscovite is assumed to be 100 percent 2M polymorph. Since the natural samples are mechanical mixtures of the different polymorphs, the X-ray effects are thought to approach linearity. Velde and Hower (1963) have determined empirical curves of $I_{3.74\,Å}/I_{2.58\,Å}$ as a function of known mixtures of 2M (muscovite) and 1M*d* (illite). In their determination, where the 3.74 Å reflection serves the same purpose as the 2.80 Å reflection in the present study, a nearly linear relationship obtained. Figure 2 shows the curve used to determine the percentage of 2M polymorph in the Belt samples. For both the standard muscovite and the Belt samples, peak areas were determined by averaging 6 planimeter measurements on traces run at $\frac{1}{4}°$/min for randomly oriented samples.

To obtain the standard deviation of the polymorph determination, the less than 2-micron size fraction of sample number P33 was used in six different preparations of randomly oriented samples. The mean value obtained for the $I_{2.80\,Å}$ peak/$I_{2.58\,Å}$ peak was 0.21 and $s=0.02$, or 10 percent of the mean. The standard deviation of 0.02 is equivalent to ± 8 percent 2M polymorph. This is thought to be fairly representative of the error of polymorph measurement in this study.

Discussion

Although this paper is concerned with the nature of the dioctahedral mica in the Belt series, it is of some interest to note the general layer silicate mineralogy of the less than 2 micron fraction for the 38 samples reported here. The assemblages and frequencies are:

dioctahedral mica (including illite)—chlorite	(29)
dioctahedral mica (including illite)	(3)
dioctahedral mica (including illite)—kaolinite	(1)
dioctahedral mica—chlorite—biotite	(3)
dioctahedral mica—biotite	(2)

The kaolinite-bearing sample occurs in the Spokane Fm. at the top of the Little Belt Mountains section. The biotite-bearing samples are metamorphic and occur in the lower portion of the Pend Oreille Lake section. The paucity of kaolinite in comparison with younger argillaceous sediments is remarkable. Its complete absence from the thick Pend Oreille Lake section may be attributed to its elimination during late diagenesis. The mineralogical and textural trends suggest a transition from high grade diagenetic to low grade metamorphic conditions.

Nature of the 10 Å Mineral in the Belt Argillites.

Absence of mixed-layering. A comparison of the diffraction patterns of untreated and glycol solvated samples shows no detectable expandable layers in the 10 Å mineral. The 10 Å peaks in samples from the eastern sections and upper part of the Pend Oreille Lake section are frequently asymmetrical, tailing off somewhat toward low angles. The 10 Å peaks for samples from the lower Pend Oreille Lake section tend to be more symmetrical. In addition, the diffraction patterns show a considerable amount of scattering at low angles, which decreases with increasing 2M content until it is virtually non-existent when the dioctahedral mica is near 100 percent 2M. Rather than an indication of mixed-layering or heterogeneity, this feature appears to be an intrinsic feature of illites, which in addition to being disordered are potassium deficient (without being interlayered) and have a high H_2O+content as compared with

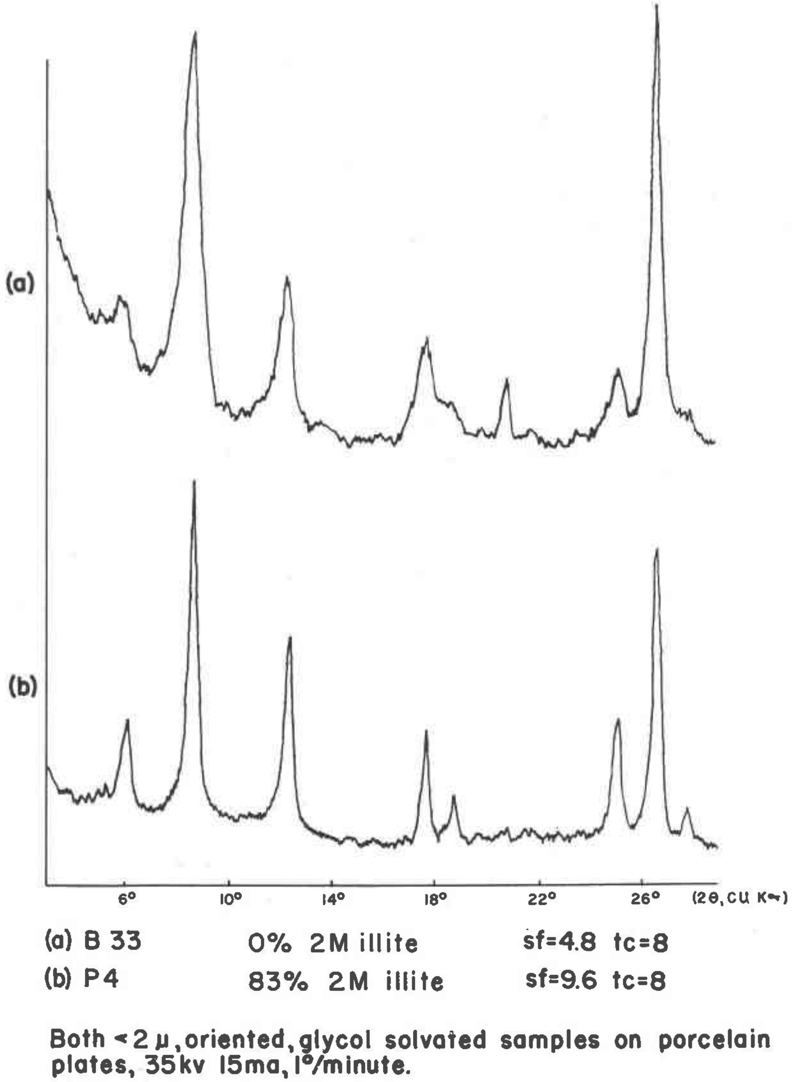

FIG. 3. X-ray diffraction traces showing 10 Å peak shape and low angle scattering intensity in a dioctahedral 10 Å mineral as a function of ordering.

true dioctahedral micas (Hower and Mowatt, 1966). Figure 3 is a comparison of the 10 Å peak shape and low angle scattering intensity in a dioctahedral 10 Å mineral which is not detectably ordered (a) with one which is essentially completely ordered (b).

Mixed-layered illite-montmorillonite is a ubiquitous component of sedimentary rocks (Weaver, 1956). The absence of expandable clays and of mixed-layering in the illite indicates, as did the absence of kaolinite, that the whole Belt sequence has been subjected to high grade diagenetic conditions. In fact, except for disordering in the dioctahedral 10 Å mineral and its apparent potassium deficiency, the dominant mineralogy of the Belt argillites is that of the greenschist facies.

Chemical composition. Measurement of the (060) spacing of the 10 Å mineral indicates that it contains appreciable octahedral magnesium and iron. Yoder and Eugster (1955) have reported (060) values of 1.499 Å for both synthetic 1M and 2M muscovite. This represents the lower limit of (060) spacing in potassium dioctahedral micas. With the introduction of large ions such as Fe^{3+}, Fe^{2+}, and Mg^{2+} in place of Al^{3+} in the octahedral positions, this spacing increases. The specific relationships, however, are not well known (Ernst, 1963). A rough estimate of the magnesium plus iron content can be derived from Figure 4, which is the relationship between $d_{(060)}$ and the number of octahedral (Mg+Fe) ions per $O_{10}(OH)_2$ for illites and illite/montmorillonites. The plot was made from data reported by Hower and Mowatt (1966). The (060) spacings of the Belt illites range from 1.501 to 1.509, with most being 1.503 or over. This indicates that from a quarter to a third of the octahedral cations in the illites are magnesium and iron.

Polymorphic changes. In the Belt series, dioctahedral mica including illite, trioctahedral mica, and chlorite make up 99 percent of the layer silicates in the samples analyzed. In the 4507-foot section from the Little Belt Mountains, Montana, representing the shallow eastern part of the series, there is no measurable 2M dioctahedral mica in any of the samples. The occurrence of very small amounts of 1M in a few samples is questionable. 1M*d* is the dominant polymorph. In contrast to this is the 38,000-foot Pend Oreille Lake, Idaho, section which contains measurable 2M material in all samples. No 1M polymorph was identified in this section. Dioctahedral mica which is not 2M is thought to be 1M*d*. The Glacier Park, Montana, section of intermediate thickness shows values of 2M material which in general are intermediate between those of the two sections above (Table 1). It is apparent that illite polymorphs trend from the low temperature 1M*d* form in the thin eastern part of the series to increased amounts of the high temperature 2M polymorph occurring westward in the thicker part of the series. This can be seen in the X-ray traces of Belt samples run at 1°/min at the intermediate and low angles. These are for untreated samples and are not the traces used to make the polymorph measurements. Rather, these traces give a more general picture of the mineralogy in the less than 2 micron size fractions. The top trace (a)

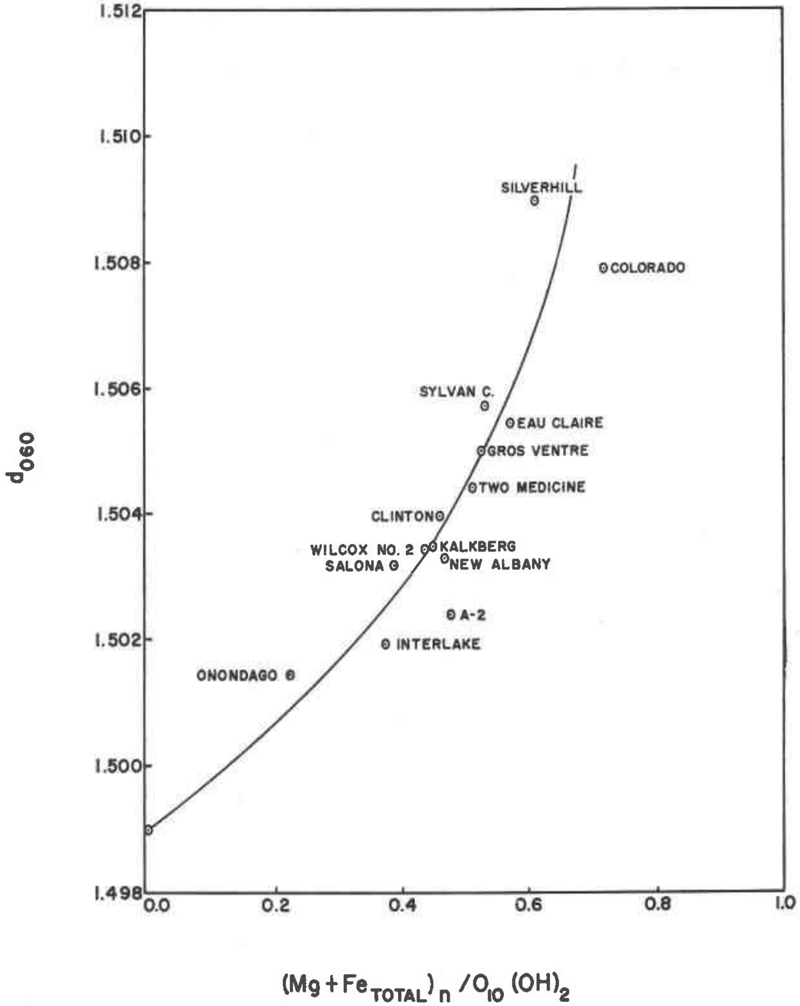

FIG. 4. Magnesium plus iron in dioctahedral minerals as a function of d_{060}.

is for a sample from the Little Belt Mountains, the middle trace (b) is from the top of the Pend Oreille Lake, Idaho, section at about 2000–4000 feet depth, and the bottom trace (c) is from the Pend Oreille Lake, Idaho, section at about 20,000–21,000 feet depth (Figure 5). Trace (b) also shows the principal hematite reflection at 2.69 Å.

Of even greater significance is the vertical trend within the Pend Oreille Lake, Idaho, section. There is an increase in amount of the 2M poly-

TABLE 1. EXPERIMENTAL DATA

Pend Oreille Lake Section

Sample no.	Formation	Generalized section (Harrison & Jobin, 1963)	$I_{2.80}/I_{2.58}$	% 2M in total illite	d_{060} dioctahedral mica
P62	Libby$_3$		0.06	24	1.504
P69	L. Libby		.10	41	1.504
P30	Striped Peak$_4$		.08	33	1.504
P26	Striped Peak$_4$		.09	37	1.505
P19	L. Striped Peak (∼S. P. 2)		.08	33	1.503
		3,780′			
P8	Wallace$_5$		.15	62	1.505
P51	Wallace$_4$		.13	54	1.506
P49	Wallace$_2$		.18	75	1.503
P6	Wallace-St. Regis contact		.19	79	1.507
		14,060′			
P33	St. Regis		.21	87	1.508
P35	L. St. Regis		.18	75	1.509
P60$_3$	Burke		.20	83	1.509
P57	Burke		.17	71	1.509
		20,260′			
P4	Pritchard$_3$		.20	83	1.504
P1	Pritchard$_3$		.22	92	1.502
P104	Pritchard$_1$		.21	87	1.501
P106	Pritchard$_1$		.24	100	1.502?
P103	Pritchard$_1$		.21	87	1.503
P102	Pritchard$_1$		.20	83	1.503
		38,260′			

Glacier National Park Section

Sample no.	Formation	Maximum thickness (Ross, 1959)	$I_{2.80}/I_{2.58}$	% 2M in total illite	d_{060} dioctahedral mica
G6	Missoula group	} unmeasured	0.10	41	1.505
G12	Missoula group		.15	62	1.507
		0′			
		5,000′			
G25	Grinnell		.12	50	1.507
G31	Grinnell		.11	45	1.506
G40	Appekunny		.12	50	1.505
G47	Appekunny		.15	62	1.508
		16,000′±			

TABLE 1—(*continued*)

Little Belt Mountains Section

Sample no.	Formation	Measured section (Weed, 1900)	$\frac{I_{2.80}}{I_{2.58}}$	% 2M in total illite	d_{060} dioctahedral mica
B68	Spokane		0	0	1.502
B65	Spokane		0	0	
		210′			
B63	Greyson		0	0	1.503
B60	Greyson		0	0	1.504
B59	Greyson				1.505
		1,165′			
B44	Newland		0	0	1.504
		1,727′			
B40	Chamberlain		0	0	1.503
B33	Chamberlain		0	0	1.503
B29	Chamberlain		0	0	1.501
B24	Chamberlain		0	0	1.504
B15	Chamberlain		0	0	1.502
		3,805′			
B11	Neihart		0	0	1.501
B6	Neihart		0	0	1.501
		4,507′			

morph with depth until essentially all of the dioctahedral mica is of the 2M polymorph (Fig. 6). At this stage biotite and porphyroblastic chlorite appear.

In considering these data, one should bear in mind that 2M illite was never detected in amounts less than 24 percent of total illite. Since the unique 2M illite peak at 2.80 Å is considerably less intense than the peak at 2.58 Å representing total illite, it seems likely that 24 percent is the lower limit of detection. Consequently, the values obtained for the Little Belt Mountains do not entirely preclude the presence of 2M material, but, if present, it is significantly less than the amounts found in the thicker sections to the west.

Textural changes. Petrographic examination of the argillites shows that significant textural changes take place during recrystallization of the 1M*d* illite to 2M dioctahedral mica. There is a general trend of increasing grain size with depth in the Pend Oreille Lake section. In addition, the fabric changes from one showing excellent foliation parallel to the bed-

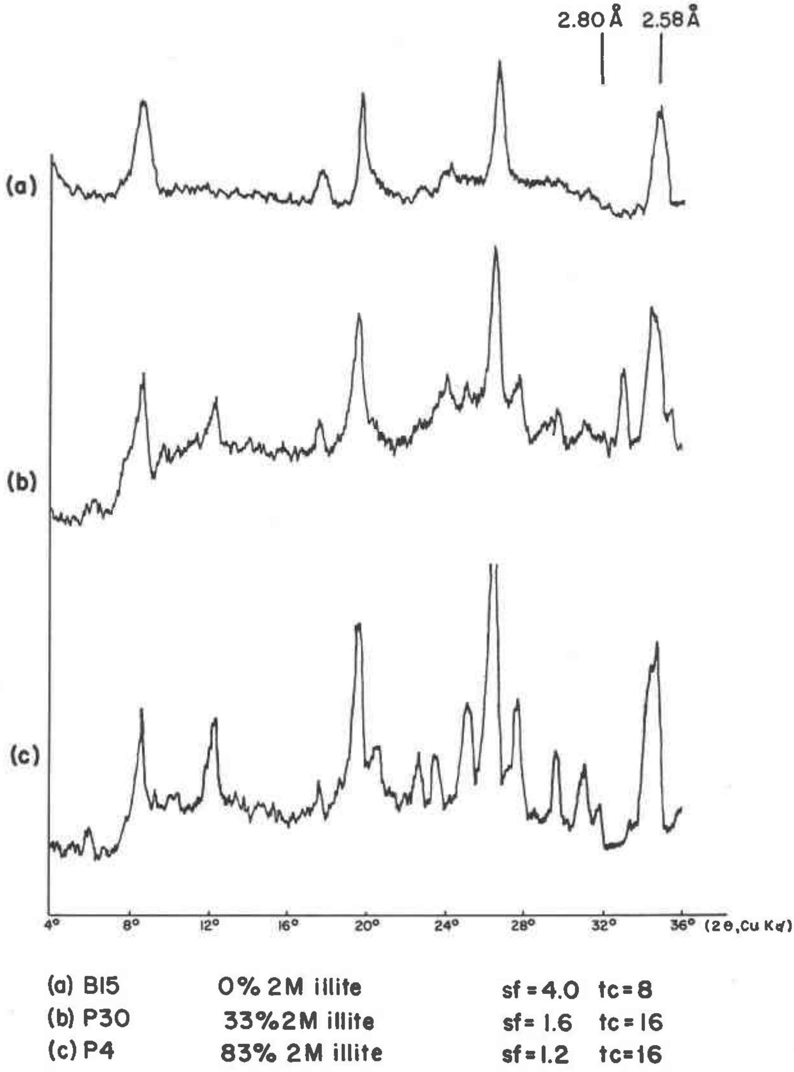

FIG. 5. X-ray diffraction traces showing % 2M illite in total illite as a function of $I_{2.80Å}/I_{2.58Å}$.

ding at the top of the section to one showing an increasing disorientation of the layer silicate crystals at the bottom of the section. The excellent foliation probably reflects early mechanical orientation of the layer silicate grains. Recrystallization at depth under conditions approximating hydrostatic pressure could account for the lesser orientation low in the section.

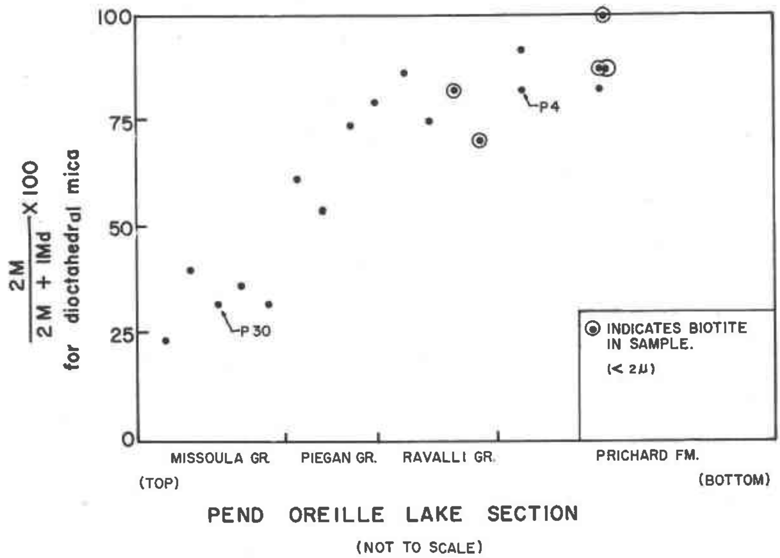

FIG. 6. Percentage 2M dioctahedral mica for the Pend Oreille Lake section.

SUMMARY

The dioctahedral mica in the Precambrian geosynclinal Belt argillites is an illite with little or no mixed layering. The illite composition differs from that of muscovite in that it contains significant iron and/or magnesium.

The 1M*d* illite in the Belt series was inherited from an early stage of diagenesis. This polymorph is abundant and is the dominant illite polymorph in the thin eastern part of the series and in the upper part of the thick western series. In the thick western part of the series, the percentage of 1M*d* illite in total illite decreases with stratigraphic depth. The 2M polymorph becomes abundant and finally dominant as depth increases. It appears that increasing temperature and pressure conditions accompanying deep burial, perhaps aided by an increased geothermal gradient associated with batholithic intrusion, cause a transformation of the 1M*d* to the 2M polymorph. This transformation is completed at the biotite isograd.

This material apparently represents a further advance in the diagenetic metamorphic continuum than the mixed-layer illites of Cambrian to Recent age described by Weaver (1956) or the 1M*d* Paleozoic illite described by Velde and Hower (1963). Actually, the dioctahedral micas of the Belt series have been under sufficiently rigorous conditions to elevate them to the greenschist facies of metamorphism at greatest depth.

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