PETROLOGICAL SIGNIFICANCE OF ILLITE POLY-MORPHISM IN PALEOZOIC SEDIMENTARY ROCKS

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ABSTRACT

An *x*-ray diffraction investigation was made of the type and abundance of the polymorphic form of illite in Paleozoic sedimentary rocks. Only the 1 Md and 2 M polymorphs were found to be present in appreciable amounts, with the 1 Md polymorph predominant in the $<1\ \mu$ size fraction. This large abundance of 1 Md polymorph illite in Paleozoic sediments indicates a predominant low-temperature origin for illite. Some data indicating relative mineral abundance changes with change in grain size in the sedimentary rocks investigated are also given.

INTRODUCTION

In this study illite is defined as a mineral group found in sediments having a primarily dioctahedral muscovite-type structure with less than 10% of expandable layers interlayered in the structure. (The average $d_{001} = 10.05$ Å, much less than 10% expandable material.) Although the illite thus defined probably contains a variety of minerals, it is difficult to determine individual structures and compositions because of their similarity. See Yoder and Eugster (1955) for a discussion of the problem. It is possible that true micas or pure muscovites are present but no conclusive chemical-structural evidence has been presented yet to demonstrate such an occurrence. Thus the definition of illite used in this study is broad enough to include all ~ 10 Å layered structure material found in sedimentary rocks. As a result, the term illite takes on a meaning similar to that of bauxite or limonite, *i.e.* a mineral group name.

Illite generally represents over half of the clay minerals that are found in sedimentary rocks (data in Weaver, 1959), and clay minerals are the most abundant group of silicates in sedimentary rocks as they are the main minerals in shales, which account for 75% of sedimentary rocks (Pettijohn, 1957). Clay minerals are also found in most other sedimentary rocks in varying amounts. Despite the fact that illite is very common, its mode (or modes) of origin is not known.

One very important difficulty arising in the investigation of the origin of illite is the structural similarity between illite and muscovite; both minerals are dioctahedral micas. The problem of the origin of illite is simple if it is just fine-grained muscovite derived by the mechanical weathering of high-grade rocks as suggested by Weaver (1959). However, many other kinds of dioctahedral micas exist (Foster, 1956, 1960). These

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micas differ in composition and in their modes of origin (in particular in terms of temperature), but their gross structures are similar. In nature, a mica of the composition of muscovite occurs in igneous and metamorphic rocks; at the other extreme, another dioctahedral mica, glauconite, forms at earth surface conditions (Burst, 1958). Thus illite could represent material from a wide variety of environments of formation.

The primary method of investigation used in the present study is x-ray diffraction. Identifications of the minerals present and in particular the structure of the illite were made. The materials used were from Paleozoic rocks of various ages and localities (Table 1). An attempt was made to include in the investigation samples of varying geographic locality, age, and tectonic environment so that a general conclusion might be drawn on the nature of the clay mineral assemblages of Paleozoic sedimentary rocks.

The type and abundance of the structural polymorphs of the illite (10 Å material) were determined in order to provide a measure of the thermal history and hence indicate the origin(s) of the illitic material in sedimentary rocks. Yoder and Eugster (1955) used synthetic and natural materials to determine the stability relations of the muscovite polymorphs. (For a description of the polymorphs see Smith and Yoder (1956).) Their work resulted in the following relationships:

1 Md is a metastable polymorph preceding the stable polymorphs as the mineral grows. The 1 M polymorph is stable below the temperature range 200°–350° C. while the 2 M polymorph is more stable above this temperature range. In the laboratory the 1 Md→1 M conversion is slow, and the 1 M→2 M conversion at 200°–350° C. is very slow.

Further work by Weaver (1958) shows that 2 M 10 Å material will weather to an expandable mineral with a 1 Md or disordered structure. Reconstitution by replacing potassium lost during weathering resulted in the structure's returning rapidly to its 2 M structure. We have found by using molten $LiNO_3$ to remove potassium from muscovites that the 2 M structure is regained when potassium is re-introduced to the structure. It is thus concluded that the three naturally occurring polymorphs of muscovite representing temperature stabilities would be 1Md, 1M, and 2M, with 1Md and 1M polymorphs representing low temperatures (< 200°–350° C.) of formation and the 2M polymorph representing the high-temperature material (> 200°–350° C.). This is the basis for our petrological interpretation of the polymorphic form of illite in Paleozoic sedimentary rocks.

As a matter of general interest the distribution of the minerals present in the Paleozoic sedimentary rocks was ascertained in a gross manner by analyzing differing particle size fractions. This was done in order to determine where the illite could be found and what relation this might have with respect to the polymorphic form of the material.

Clay minerals:

I, illite
C, chlorite (plus Al-serpentine?)
M, montmorillonoid

K, kaolinite
S, Al-serpentine
K-S, kaolinite and Al-serpentine indistinguishable

M-L, mixed layered illite-montmorillonoid K?, kaolinite presence not determined

Color:

Bl, black
Gy, grey
Gr, green

dr, dark
Br, brown

Numbers are designations used in the laboratory, *e.g.*: C-8, Cambrian, sample number eight.

The symbols < and > signify: <, less than 1 μ fraction; >, greater than 1 μ and some material of the less than 1 μ fraction.

TABLE 1. EXPERIMENTAL DATA

No.	Name and Locality	Size	Clay Minerals	(060)	%2M	Color
Cambrian						
C3	Burgess sh.—B.C., Can.	<	I S	1.500	50	Gy
		>	I S	1.502	45	
C6	Gros Ventre—Wyo.	<	I S	1.505	25	Gr
		>	I C	1.503	70	
C8	Goodsir—B.C., Can.	<	I C	1.502	70	Gr
		>	I C	1.505	70	
C9	Lower Cambrian—B.C., Can.	<		1.503		Bl
		>		1.503		
C10	Trempelo lms.—Ill.	<	I	1.503	25	
		>	I	1.503	15	
C11	Eau Claire—Ill.	<	I	1.503	0	Gy-Gr
		>	I	1.502	0	
C12	Wolsey—Mont.	<	I C	1.504	0	Br
		>	I C K	1.504	0	
C13	Flathead—Mont.	<	I S-K	1.503	0	Br
		>	I C K	1.502	0	
Ordovician						
O1a	Schenectady—N. Y.	<	I C	1.503	5	dr-Gr
		>	I C	1.503	15	
O2	Utica sh.—N. Y.	<	I C K?		10	Bl
		>	I C K?	1.505	5	
O3	Sylvan sh.—Okla.	<	I	1.510	0	Gr
		>	I	1.510	10	
O5	Arbuckle lms.—Okla.	<	I	1.509	20	Br
		>	I M	1.503	15	
O6	Simpson—Okla.	<	I C M	1.502	0	
		>	I M	1.500	0	
O7	Viola lms.—Okla.	<	I	1.506	0	
		>	I	1.505	5	

TABLE 1—(*continued*)

No.	Name and Locality	Size	Clay Minerals	(060)	% 2M	Color
O8	Sylvan sh.—Okla.	<	I C	1.503	0	Br
		>	I	1.506	50	
O9	Kimmswick—Ill.	<	I		0	
		>	I	1.505	5	
O10	Tulip Creek—Okla.	<	I K S	1.508	10	Bl
		>	I K S	1.509	20	
O11	Maquoketa sh.—Ill.	<	I C	1.506	5	Gy
		>	I C		100	
O12	Mt. Hope—Ohio	<	I C	1.505	5	Bl
		>	I C	1.503	25	
O13	Normanskill sh.—N. Y.	<	I S	1.506	0	Gy
		>	I C	1.501	20	
O25	Normanskill sh.—N. Y.	<	I C	1.502	0	Bl
		>	I C	1.503	0	
O27	Normanskill sh.—N. Y.	<	I C	1.504	15	Bl
		>	I C	1.504	35	
Silurian						
S1	Rochester sh.—N. Y.	<	I S	1.498	40	Gy
		>	I C	1.506	80	
SS8113	Clinton—Penna.	<	I K C	1.505	45	Gy
		>	I C	1.504	35	
S2	Camillus sh.—N. Y.	<	I C	1.505	0	Gr-Gy
		>	I C	1.505	30	
S2b	Camillus sh.—N. Y.	<	I C	1.504	35	Gr-Gy
		>	I C	1.504	30	
S3	Chittenango sh.—N. Y.	<	I C	1.508	25	Gr-Gy
		>	I C	1.505	90	
S11	St. Clair lms.—Ill.	<	I M	1.501	40	
		>	I M	1.499	100	
S12	Niagaran—Ill.	<	I	1.501	0	
		>	I	1.503	65	
S13	Sodus sh.—N. Y.	<	I	1.501	25	Gr-Bl
		>	I	1.500	40	
S14	Rochester—N. Y.	<	I C	1.504	5	
		>	I C	1.501	30	
S15	Girardzau lms.—Ill.	<	I C	1.505	15	
		>	I C	1.505	25	
S16	Arisaig—Nova Scotia	<	I C K?	1.501	20	
		>	I C	1.499	35	
Devonian						
D58103	Marcellus—Penna.	<	I C	1.504	5	Bl
		>	I C	1.502	30	
D5	Esopus sh.—N. Y.	<	I K-S	1.501	15	Bl
		>	I C K	1.501		

TABLE 1—(*continued*)

No.	Name and Locality	Size	Clay Minerals	(060)	% 2M	Color
D10	Bakoven sh.—N. Y.	<	I C	1.504	0	dr-Br
		>	I C	1.501	50	
D11	Hamilton—N. Y.	<	I C	1.504	5	dr-Gr
		>	I C	1.502	50	
D12	Hamilton—N. Y.	<	I C	1.501	30	Bl
		>	I C	1.502	50	
D19	Kiskatom redbeds—N. Y.	<	I C	1.504	95	Red
		>	I C	1.504	100	
D20	Middlesex sh.—N. Y.	<	I S	1.504	10	Bl
		>	I C	1.504	15	
D21	Cardiff sh.—N. Y.	<	I C K?	1.501	0	Bl
		>	I C K?	1.502	35	
D22	Moscow sh.—N. Y.	<	I C		0	Bl
		>	I C	1.501	35	
D24	Mottville sh.—N. Y.	<	I C K?	1.501	50	Bl
		>	I C	1.501	30	
D25	Upper Chittenango—N. Y.	<	I C K?	1.501	0	Bl
		>	I C	1.502	30	
D26	Geneseo—N. Y.	<	I C K?	1.501	5	Bl
		>	I C K	1.500	5	
D27	Cardiff—N. Y.	<	I C	1.505	40	Bl
		>	I C	1.506	85	
D28	Cardiff—N. Y.	<	I C	1.503	40	Bl
		>	I C	1.505	65	
D29	Cardiff—N. Y.	<	I C	1.501	25	Bl
		>	I C	1.503	85	
D30	Cardiff—N. Y.	<	I C	1.500	30	Bl
		>	I C	1.499	90	
D31	Mottville sh.—N. Y.	<	I C	1.500	0	Bl
		>	I C	1.504	30	
D33	Haragan marl—Okla.	<	I	1.503	10	Bl
		>	I	1.503	65	
D34	New Albany—Ill.	<	I C	1.501	40	Bl
		>	I C	1.501	55	
D36	Backbone lms.—Ill.	>	I	1.509	10	
		>	I	1.497?	55	
D37	Ohio sh.—Ohio	<	I	1.500	45	dr-Br
		>	I C K?	1.503	55	
D38	New Albany sh.—Ind.	<	I S	1.502	35	Bl
		>	I K-S	1.502	70	
D39	Ludlowville sh.—N. Y.	<	I C	1.502	30	
		>	I C	1.499	100	
D1a	Duvernay—Alta, Can.	<	I	1.503	0	Br
		>	I	1.504	0	
D2a	Duvernay—Alta, Can.	<	I	1.502	20	Br
		>	I	1.504	25	

TABLE 1—*(continued)*

No.	Name and Locality	Size	Clay Minerals	(060)	% 2M	Color
D3a	Duvernay—Alta. Can.	<	I	1.503	5	Br
		>	I	1.504	35	
D4a	Duvernay—Alta, Can.	<	I			Br
		>	I	1.500	5	
D5a	Duvernay—Alta, Can.	<	I K	1.502	5	Br
		>	I K C	1.500	15	
D7a	Duvernay—Alta, Can.	<	I	1.505	10	Br
		>	I	1.506	10	
Mississippian						
M3	Banff—Alta, Can.	<	I C K		10	Bl
		>	I C K	1.504	30	
M4	Woodford—Okla.	<	I	1.509	0	Bl
		>	I	1.505	30	
M5	Burlington lms.—Mo.	<	I K-S	1.505	0	
		>	I K-S	1.510	30	
M6	Madison lms.—Mont.	<	I	1.501	65	
		>	I K-S	1.498	85	
Pennsylvanian						
M1	Morris illite—Ill.	<	I	1.504	0	**Br**
		>	I	1.503	50	
P1	Amsden—Mont.	<	I	1.503	0	Red
		>	I	1.505	60	
P2	Allegheney—Penna.	<	I C	1.501	5	Bl
		>	I C	1.505	5	
P4	Buckhorn lms.—Alta, Can.	<	I M		40	Br
		>	I M	1.504	100	
P5$_s$	Springer (sh.)—Okla.	<	I M-L	1.501	0	Gy
		>	I M-L	1.501	30	
P5$_1$	Springer (lms.)—Okla.	<	I M-L	1.499	0	
		>	I M-L	1.503	5	
F1	Fithian illite—Ill.	<	I C	1.507	0	Br
		>	I C	1.505	30	
P6	Wea sh.—Mo.	<	I C	1.50	20	Br
		>	I C K	1.504	100	
P7	Lane sh.—Mo.	<	I C	1.501	15	Br
		>	I C	1.505	30	
P8	Allegheney—Penna	<	I C	1.506	25	Bl
		>	I C	1.502	30	

SAMPLE PREPARATION

The shales were broken into fragments of about 1 cm diameter in a mortar, the smaller sizes being discarded. This material was then dis-aggregated in distilled water with a Bendix type UG-3B-1 ultrasonic

cleaner. This disaggregation procedure appears to result in a minimum of breaking down of the intrinsically large particles. The disaggregated shale sample or the insoluble residue of limestone samples (dissolved in 1N HCl) was put into suspension in a pH 9.5 $NaCO_3$ solution and then centrifuged to obtain the various particle sizes. No apparent change in the illite structure was observed during a treatment with boiling 6N HCl. It is thus assumed that 1N HCl did not significantly affect the illite. After each run the samples were again placed in the ultrasonic generator to insure complete disaggregation of the material. Normally three or four runs were made to separate each size fraction of the sample. The time and rpm data for the centrifuge were computed by the formula:

$$Tc = Kc \frac{Log \frac{R}{S}}{N^2D^2}$$

$Kc = 2.52$
$N = 1800$ rpm
$D =$ diameter of particle
$S = 10$ cm
$R = 18$ cm

as found in Jackson (1956, p. 130).

The times are as follows for an International Centrifuge model SVB, size 1:

$0.2\ \mu$ 82 min.
$1\ \mu$ 3.3 min.

$10\ \mu$ separations were obtained by settling the material in a 10 cm water suspension for 15 minutes (Krumbein and Pettijohn, 1938, p. 166). This resulted in four size fractions: less than $0.2\ \mu$, greater than $0.2\ \mu$ but less than $1\ \mu$, greater than $1\ \mu$ but less than $10\ \mu$, and greater than $10\ \mu$. The major portion of the material in the samples studied was in the 0.2 to $10\ \mu$ range. The two size fractions separated in this range usually contained about equal volumes of material.

Oriented samples of the layer silicate minerals were prepared on porous porcelain plates after the method of Kinter and Diamond (1956). These plates were run on a Philips-Norelco x-ray diffraction unit at 1° 2θ per minute, using filtered CuKα radiation. Randomly oriented samples were prepared as described below.

IDENTIFICATION OF CLAY MINERALS

The criteria used to identify the clay minerals present in the parallel oriented specimens were as follows: 10 Å material, (001)≅10 Å, was identified as illite. Reflections tailing to high d values were interpreted to indicate disordered, mixed-layer material present. When the 10 Å peak had a slightly higher d value, the material was considered random mixed-

layered. The d values seldom exceeded 10.4 Å, about 20% expandable layers (one water layer) according to the curves of Brown and MacEwan (1951). Chlorite was identified by the 14 Å reflection, together with a 7 Å reflection of about four times the intensity of the 14 Å reflection. The 7 Å reflection could be due in part to aluminous serpentine (see below). The 14 Å and 4.7 Å chlorite reflections were of about equal intensity. Montmorillonite was identified by a 14 Å reflection shifted to approximately 17 Å upon glycolation. The 14 Å reflection of montmorillonite was usually rather diffuse and far more intense than the 7 Å peak. Sharpness of the 14 Å and 7 Å peaks was often diagnostic in distinguishing between chlorite and montmorillonite, the montmorillonite having more diffuse peaks. Kaolinite was determined to be present in the samples when chlorite was also present by observing the (131) kaolinite reflection at 2.33 Å. The iron-rich chlorite (006) reflection is at about 2.37 Å (Grim, 1953). The determinations to distinguish kaolinite were made on randomly oriented samples.

The material with a primary basal spacing of 7 Å presented a problem. Intensities of 7 Å and 3.5 Å reflections were generally equal to one another as is typical of kaolinite. However, some of the materials would form a phase with an approximately 13 Å spacing when heated to 500° C. The 7 Å material found in the sedimentary rocks showed only (00l) reflections when x-ray mounts that produced parallel orientation of the grains were run. This indicates a platy morphology as was found in synthetic aluminous serpentines by Yoder (1952). A 7 Å material with the properties described above has been called *septechlorite* by Nelson and Roy (1958). However, aluminous serpentine would probably be a better term on structural grounds (Yoder, 1952). Generally, the material was only slightly affected by NaOH, making a distinction between Al-serpentine and kaolinite difficult. It was concluded that some Al-serpentine was present in the samples with 7 Å reflections, but relative amounts could not be determined. Results of the identifications are shown in Tables 1 and 2.

POLYMORPH DETERMINATIONS

The determination of the approximate percentage of 2M material in the sample was carried out in the following manner: the dried, sized material was ground briefly in an agate mortar to break any re-aggregated material. The sample was mounted in a welled sample holder and card was taken to reduce parallelism of the flakes to a minimum. The sample was then run on a Philips-Norelco x-ray diffraction unit through 38° to 20° 2θ at a scanning rate of 1° per minute, using CuKα radiation. This is the critical range of 2θ for identifying the characteristic reflections for the

TABLE 2. SIZE SEPARATION DATA

No.	Name	Size in microns	I_{10}/I_7Å	% 2M	Q	F
C6	Gros Ventre	<0.2	25.0	0	0	0
		<1	8.6	30	0.6	0
		<10	5.8	20	1.2	5.1
		>10	8.1	100	2.7	7.3
C8	Goodsir	<0.2	no material present			
		<1	2.7	25		
		<10	0.7	100		
		>10	0.4	100		
Ola	Schenectady	<0.2	3.8	0	0	0
		<1	1.3	5	0	0
		<10	1.2	65	4.2	2.3
		>10	1.0	100	5.9	3.8
O11	Maquoketa	<0.2	5.6	0	0.3	0
		<1	2.0	25	1.4	0.6
		<10	1.4	100	5.4	1.0
		>10	1.1	100	7.0	3.0
O25	Normanskill	<0.2	3.3	0	0	0
		<1	2.0	40	0.4	0.5
		<10	1.3	45	3.2	2.3
		>10	1.5	100	6.2	3.8
S3	Chittenango	<0.2	3.3	0	0	0
		<1	1.7	5	1.0	0
		<10	1.0	30	4.8	1.1
		<10	1.2	55	8.3	1.7
D10	Bakoven	<0.2	10.5	0	7	4
		<1	3.3	50	23	8
		<10	1.6	70	65	23
		>10	1.0	75	60	40
D12	Hamilton	<0.2	3.6	30	1.0	0.3
		<1	1.2	45	0.8	0.2
		<10	0.8	65	4.3	1.1
		>10	0.8	100	7.5	2.5
D19	Kiskatom redbeds	<0.2	2.8	100	1.6	1.0
		<1	2.6	100	1.6	0.7
		<10	1.4	100	2.8	1.0
		>10	1.4	100	7.8	2.2
D27	Cardiff	<0.2	3.5	0	0.9	0.2
		<1	2.1	25	0.9	0.2
		<10	1.2	30	8.9	1.1
		>10	0.7	100	8.9	1.1
D58103	Marcellus fm.	<0.2	3.1	25	1.8	0.8
		<1	1.5	40	1.3	0.8
		<10	0.7	35	5.0	1.6
		>10	0.6	80	8.0	2.0
P7	Lane sh.	<0.2	6.4	0	0	0
		<1	1.0	0	0.3	0
		<10	1.0	20	2.8	1.4
		>10	0.9	100	5.8	4.6

various polymorphs of dioctahedral mica. The identification of poly-morphs was based on diffraction patterns resulting from this procedure.

The diffraction patterns were further used for a semiquantitative analysis of the amount of 2M polymorph in each sample. The only polymorphs that appeared with significant frequency were 2M and 1Md. The reflections used for the semiquantitative analysis were the 2M, 3.74 Å (023) reflection and the 2M, $\sim$2.58 Å (13$\bar{1}$) (116) (20$\bar{2}$), and 1Md, $\cong$2.58 Å (130) (200) multiple reflections. These particular reflections were used because they were not much affected by the presence of
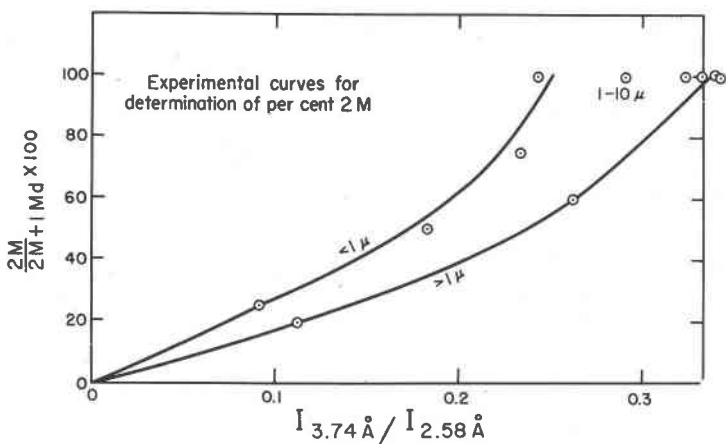
FIG. 1. Experimental curves for determination of per cent 2M.

other materials in the samples. Also, they appeared with the greatest con-sistency in the unoriented sample patterns. Empirical working curves were calculated by determining the intensity ratio of the 3.74/2.58 Å re-flections for polymorph mixtures of known amounts of 2M (muscovite) and 1Md (illite) in the two size ranges, $<1\mu$ and $>1\mu$ (Fig. 1). Mont-morillonite, chlorite, and kaolinite also have reflections near 2.58 Å. However, no significant correlation could be made between the abun-dance of these minerals and the % 2M polymorph in the 10 Å material. Examples of patterns showing various amounts of 2M polymorph ma-terial are shown in Fig. 2. Intensities were determined by measurement of the area under the peaks of the reflections with a Keuffel and Esser planimeter. A determination of the reproducibility of the method gave a standard deviation of $\pm 15\%$ 2M for the range of 3.74/2.58 Å reflection intensity ratios of 0.05 to 0.31 or 15 to 85% 2M in the greater than 10 μ size range. Greater deviations occur at the ends of the curve. Determina-tions at the lower percentages of 2M are less precise because the 2M,

3.74 Å peak has a low intensity compared with the 2M–1Md, 2.58 Å peak. The 1M polymorph has a reflection at ~2.58 Å (130) (131) (200) also but was rarely present and could be ignored in most cases.

It was found that the peak ratios for 100% 2M material would change with a change in particle size. Thus two curves, one for the less than 1 μ and one for the greater than 1 μ material were determined. The size fractions of 1–0.2 μ and less than 0.2 μ seemed to have approximately equal
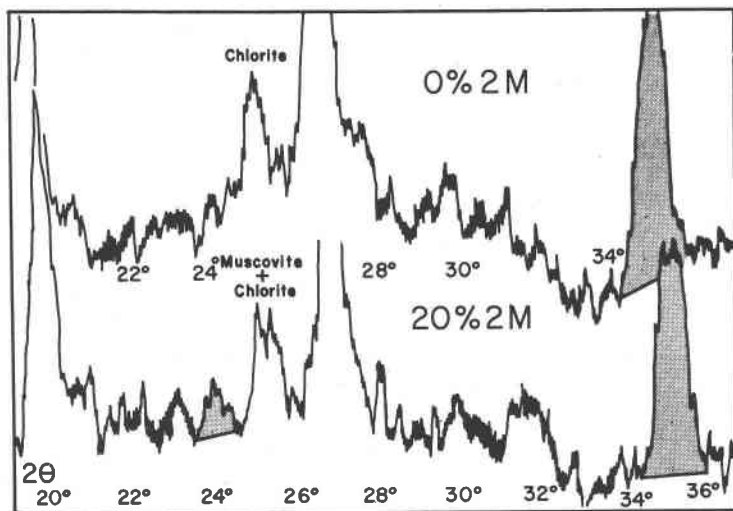
FIG. 2. X-ray diffraction patterns of 1Md and 2M polymorph mixtures. Blackened peaks are those used for percentages of 2M determinations.

ratios or apparent per cent of 100% 2M mica. The 1–10 μ muscovite had an intermediate ratio of 0.28 and the 10–128 μ fraction had a ratio of about 0.33.

Yoder and Eugster (1955) discussed the effects of grinding on the structure of muscovite. One important result is the unequal decrease in intensity of the (114) and (002) reflections with grinding time. This indicates that for the small grain sizes only the (00l) reflections are prominent. In the present study the intensity of the 2M (023) (3.74 Å) reflection is compared with the intensity of a multiple peak which is a (13$\bar{1}$) (116) (20$\bar{2}$) or (hkl) reflection (~2.58 Å). The intensity ratio is independent of the extinction of hkl's relative to 00l's shown to be due to small grain size in the grinding experiments. There is a variation in relative intensity with grain size as is shown in Fig. 1, but it is believed that when this is taken into account the presence of the 1Md polymorph can be assessed.

Seventy-four samples were separated into two size fractions of less than 1 μ and greater than 1 μ plus some residual fines ($\approx 25\%$). The percentage of 2M polymorph present was determined by measuring the 3.74 Å/2.58 Å intensity ratio and picking it off the working curve (Fig. 1). Further size separations were made on twelve of the above 74 samples (Table 2). These were split into the <0.2 μ, 0.2–1 μ, 1–10 μ, and >10 μ fractions. The amounts of 1Md and 2M material were interpolated from values on the curves (Fig. 1) and values for 2M muscovite of each size fraction. It must be noted that this is a semi-quantitative method.

RESULTS

The clay mineralogy of the samples is given in Table 1. It should be noted that the mineralogy is simple and relatively uniform for all the samples. A general clay mineralogy could be given as follows: illite is the dominant clay mineral (over half) with chlorite (plus Al-serpentine?) second in abundance. Kaolinite is minor in occurrence when present.

The polymorph of the illite in the material separated into <1 μ and >1 μ plus residual material is a mixture of 1Md and 2M in most cases. Little 1M was present; indications of 1M were found in only six samples and it was never a major polymorph. The frequencies of per cent 2M in the samples are shown in Figs. 3 and 4. There is an increase of 1Md material in the small size range over the larger size range of particles.

The distribution of 2M polymorph, quartz, and feldspar in the ma-
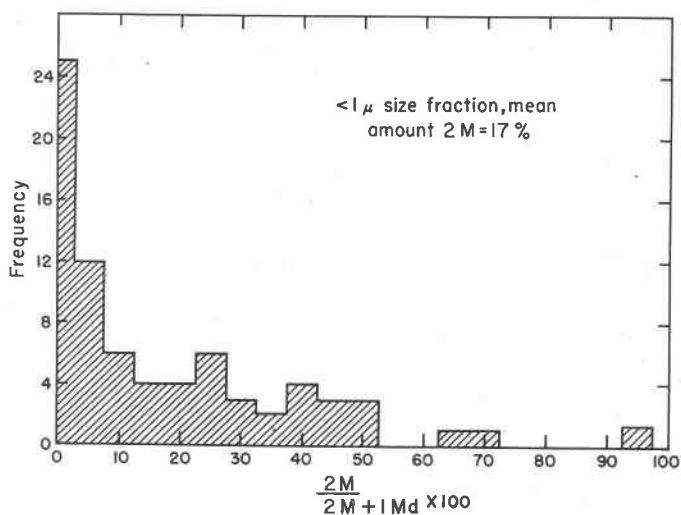
FIG. 3. Per cent 2M in <1 μ size fraction.
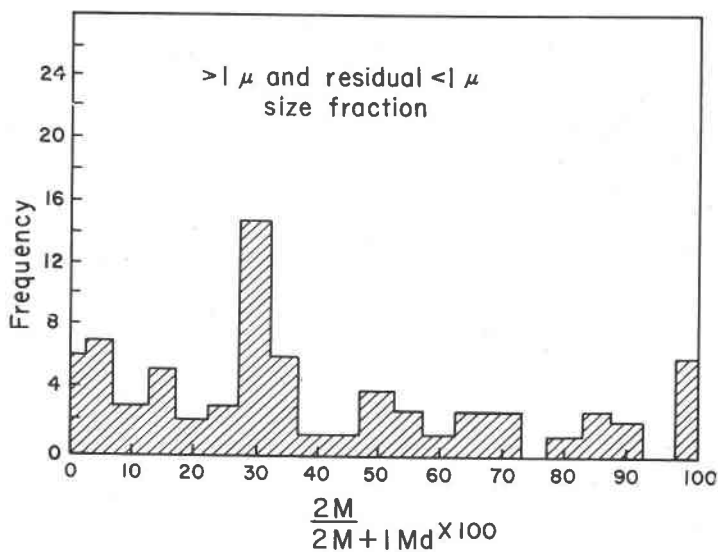
FIG. 4. Per cent 2M in $>1\ \mu$ plus residual $<1\ \mu$ fraction.
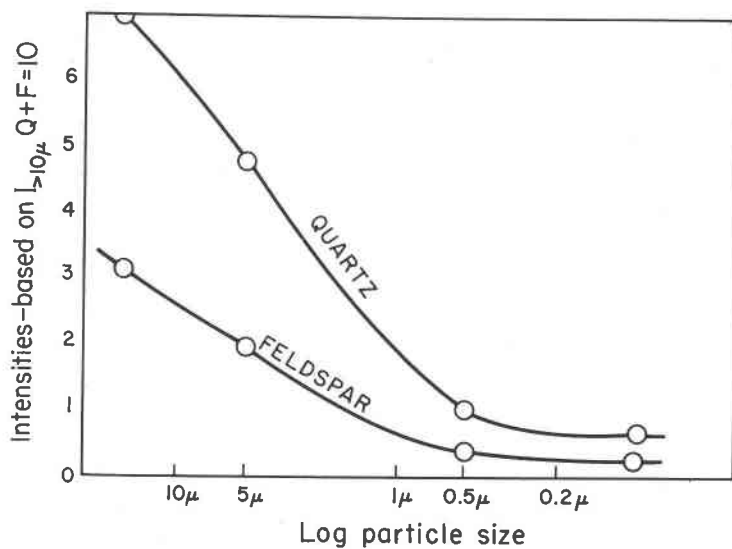
FIG. 5. Average relative quantities of quartz and feldspar for twelve size samples. Cumulative intensity of quartz 4.27 Å and feldspar 3.21 Å peak in the $>1\ \mu$ fraction equals 10 units.
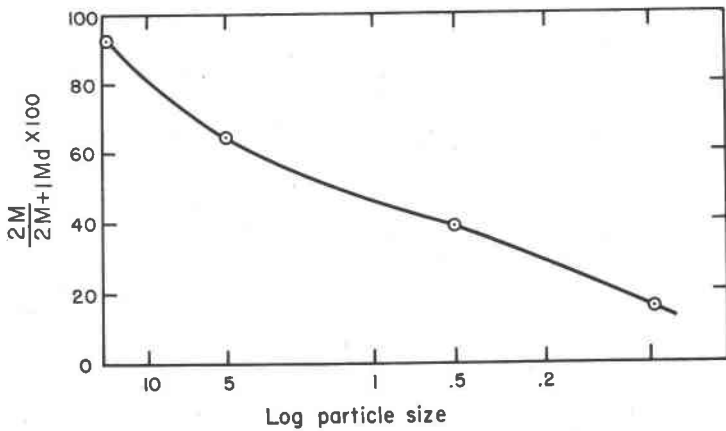
Fig. 6. Per cent 2M *vs.* particle size; 12 sample average.

terial separated into four size fractions shows that these minerals are concentrated in the more coarse materials (Figs. 5 and 6). (The feldspar-quartz reflection intensities (Fig. 5) are relative to each other and do not represent quantities relative to the other minerals of the samples. The intensities are compared with a base of the quartz plus feldspar intensities of the $> 10 \mu$ fraction for each sample equalling 10 units.) Also the relative intensities of illite and chlorite-kaolinite show that illite is concentrated or more abundant in the fine grain sizes (Fig. 7). The figures represent averages for the twelve sized samples; each sample showed the same trend that appears in the averaged data.

SUMMARY

A major assumption used in the petrological interpretation of the data
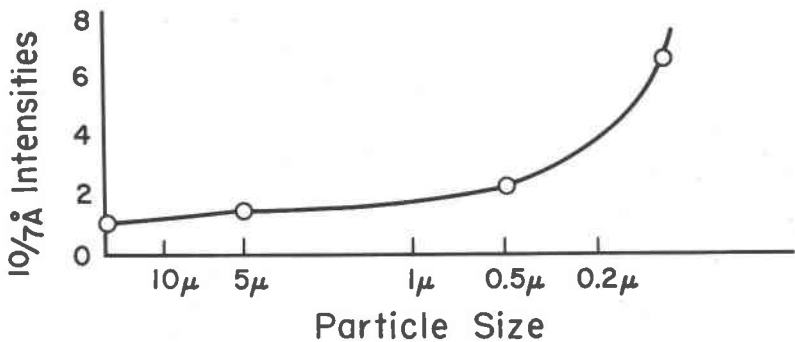
Fig. 7. Relative amounts of 10 and 7+14 Å material *vs.* particle size compared by 10 and 7 Å peak intensities; 12 sample average.

shown above is that the polymorphic types and stability ranges obtained at the ideal muscovite composition (substantiated with a few examples of illite specimens) determined by Yoder and Eugster apply to illites. Although illite has compositional differences from muscovite—less potassium and aluminum and more iron and magnesium (Grim, 1953, p. 372)—it is believed here that the stability ranges of the muscovite polymorphs hold the same significance for illite with the possibility of a shift in the critical temperature range 200°–350° C. at which the polymorphs invert. However, because the muscovite and illite structures and compositions are similar, this would probably not be a major change in temperature.

Assuming that the phase relations determined by Yoder and Eugster are valid for illite, it can be seen in Fig. 3, 4 and 6 that the 2M polymorph is greatly concentrated in the coarse size fractions and is in far less abundance than the 1Md polymorph. The 10 Å material in Paleozoic sedimentary rocks is a mixture of 2M and 1Md, with 1Md being present in considerably greater abundance. Thus, as has been concluded before (Yoder and Eugster, 1955; Kerr *et al.*, 1950; Jackson *et al.*, 1952) illite is a mixture of materials. Assuming that any disordered (1Md) material which was derived from the degradation during weathering of a high-temperature (2M) progenitor would reconvert to 2M by potassium fixation in the marine environment, the 1Md material in the rocks investigated represent structures formed at low temperatures ($<200°$–$350°$ C.). Granting the assumptions made above, it appears that the majority of illitic material is of a low-temperature origin, probably having been formed at some stage of the sedimentation-lithification processes.

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