Geol Survey

Clay Mineralogy of Pre-Pennsylvanian Sandstones and Shales of the Illinois Basin

Part I. – Relation of Permeability to Clay Mineral Suites

Thomas W. Smoot

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ABSTRACT

The clay mineral compositions of approximately 110 samples of pre-Pennsylvanian sandstones, argillaceous sandstones, and shales of the Illinois Basin have been investigated by x-ray diffractometry. Eighty percent of them are from Mississippian Chester sediments; the remainder are from the Mississippian Valmeyer Series and the Ordovician and Cambrian Systems. Ninety-five percent of the samples are from cores and the others are from outcrops.

Clay minerals from the permeable sandstones are characterized by a heterogeneous mixture of degraded illites and chlorites, kaolinite, and minor amounts of montmorillonite (if present). Those from the shales are characterized by well crystallized illite and chlorite, minor amounts of degraded illite and chlorite, and, rarely, trace amounts of kaolinite.

The differences are apparent in intimately associated sediments that came from the same general source area, and had approximately the same environments of deposition and the same originally introduced clay mineral suites. Therefore, it is concluded that the heterogeneity in the sandstone clay mineral suites has been brought about by degradation (after lithification) by circulating formation fluids, the composition of which has varied from time to time. The low permeabilities of the shales have prevented such circulation from taking place so that their clay minerals have been protected from degradation.

INTRODUCTION

This report is the first of a two-part series, both parts of which are concerned with the character of clay mineral suites, at outcrops and in core samples, of pre-Pennsylvanian sandstones and shales of the Illinois Basin. Approximately eighty percent of the samples were from the Mississippian (Chester Series); the others represent the Mississippian (Valmeyer Series), Ordovician, and Cambrian Systems.

Quade (1957), Potter and Glass (1958), Weaver (1958), and Lerbekmo (1957) have recognized that alteration of clay minerals after lithification occurs in permeable sediments, not only at surface exposures but also in the subsurface. The objectives of this study were 1) to determine the changes that take place in a weathering environment by studying outcrop and subsurface samples of comparable rocks, and 2) to study the effects of permeability in the subsurface by comparing the clay mineral suites of permeable sandstones and intimately associated impermeable shales and the clay mineral suites of comparable sandstones that exhibit various permeabilities.

This paper is based in part on the author's thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Geology in the Graduate College of the University of Illinois.

The author wishes to thank Professor R. E. Grim for his valuable suggestions and counsel throughout the course of the investigation. Thanks also are due Dr. W. F. Bradley of the Illinois State Geological Survey for his aid in some technical aspects of the study and his stimulating suggestions and criticisms.

SELECTION OF SAMPLES

In order to study the relation of permeability to the clay mineral components of sediments, I investigated many pairs of sandstone and shale samples from the Chester Series which came from the same well core and the same formation. The members of each pair were separated stratigraphically by only a few feet.

In many instances, the shale member of the pair was from a very thin shale lamella (less than a few millimeters thick) within a more massive sandstone.

In a few of the pairs very thin sandstone lamellae were interbedded in more massive shale beds, but some pairs represented sandstone and shale both of which were rather massive (more than four centimeters thick). For such closely associated sediments, it seems likely that the clay minerals in both lithologic types must have come from essentially the same source area and must have been deposited in essentially the same environment. It would seem that the differences in the environment of deposition or of source area of such sandstone and shale must have been minute and insignificant in comparison to the great degree of similarity.

Many samples of intermediate composition such as arenaceous shales or argillaceous sandstones also were investigated, some of them were from cores which could be compared with either intimately associated shales or permeable sandstones. The locations from which the samples came are shown in figure 1; the x-ray diffraction patterns of six pairs are reproduced in figure 2.

For all samples, the central portion of each core was isolated in order to minimize the effects of drilling mud contamination. For samples in which the invaded zone was greater than the radius of the core, the presence of montmorillonite generally was considered a product of drilling mud invasion and was ignored. If it was uncertain whether the montmorillonite present was from drilling mud or was an original constituent of the rock, a question mark was entered with the quantity of montmorillonite in tables 4 and 5.

Sample Preparation

Different methods were followed in preparing the shale and sandstone samples. Portions weighing 2 to 10 grams were taken from shale samples and gently crushed with a mortar and pestle, then put into an electric mixer and washed with demineralized water until the particles were dispersed. After settling, the less than 2-micron fraction was drawn off and used to prepare oriented slides.

Non-oil-bearing sandstone samples were gently crushed, using a mortar and pestle, and then further disaggregated in water using an ultrasonic generator. The samples were mixed with water in an electric mixer and washed until dispersion occurred. The less than 2-micron fraction was then drawn off after settling.

A few drops of ammonium hydroxide in approximately 250 grams of water were used as a dispersant for most samples; the same amount of sodium hexametaphosphate was used to disperse a few stubborn samples.
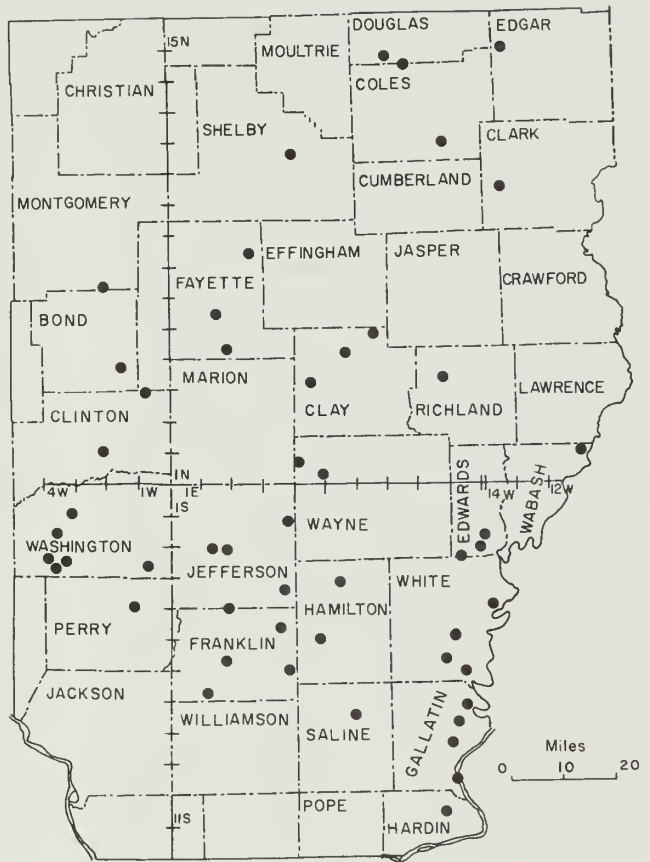

Fig. 1 - Location of Chester samples from the Illinois Basin.

X-RAY IDENTIFICATION OF THE CLAY MINERALS PRESENT

Five general groups of clay minerals are recognized in the samples investigated: illite, chlorite, kaolinite, montmorillonite groups, and a group referred to as "the mixed-layer (undifferentiated) group." Descriptions of the x-ray diffraction characteristics and subdivisions of each group follow.

Illite Group

Grim, Bray, and Bradley (1937) proposed the name "illite" as a group name for the mica-like clays, not as a species name. The wisdom of using the name as suggested has been demonstrated numerous times by many analyses which indicate that the mica-like clays vary widely in crystallization and composition.

Two subgroups of illite are recognized here. The first is well crystallized, well ordered, nonexpandable material. It is generally characterized by sharp first-order and second-order peaks which are unchanged by glycolation. The diffractometer curves of these illites are generally characteristic of those of the muscovite

type of mica crystallization, although in some of the samples from Hamilton County the low intensity 5Å peak suggests the presence of an iron-rich variety corresponding to glauconite. Where the term "illite" is used in this report, it refers to this subgroup of the illite group.

The second subgroup exhibits a first-order peak between 10Å and 11Å and always contains illite. The other constituents are degraded illite and/or other clay minerals. The first-order curve is assymetrical and generally ranges between 10Å and 12.5Å but may extend nearly to 14Å. Commonly a portion of the greater than 10Å part of the curve shifts toward lower angles after glycolation. This material is apparently intimately associated with illite and hence is referred to as "illite plus mixed-layer material." The relationship between these two subgroups is shown in figure 3.

Chlorite Group

Two general subgroups of chlorite were recognized in the samples investigated. They are referred to as "chlorite" and "chlorite plus mixed-layer material."

The chlorite is well crystallized and at room temperature commonly exhibits relatively sharp first-, second-, and third-order peaks. These reflections are little affected by glycolation, and after being heated to 575°C the mineral retains its first order peak but the second- and third-order peaks are greatly subdued. In many samples the chlorites exhibit x-ray diffraction patterns typical of the magnesium-rich crystallization (the orthochlorite type), but many others exhibit characteristics of a type intermediate between the magnesium-rich and iron-rich (leptachlorite) crystallization (Bradley, personal communication, 1958). Figure 4 presents x-ray diffraction patterns of these various chlorite types.

The chlorite plus mixed-layer material is that which is poorly ordered or poorly crystallized, and is probably randomly mixed with montmorillonite,
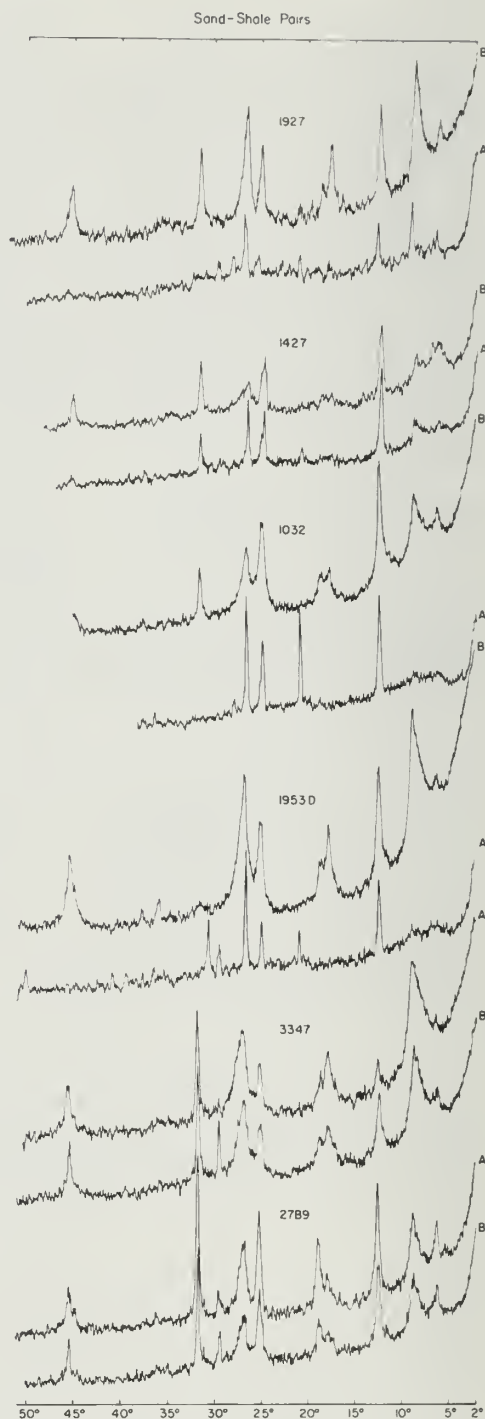

Fig. 2 – X-ray diffraction patterns of sand-shale pairs. The top curve of each pair represents shale.

vermiculite, and possibly illite. The minerals of this subgroup are in various stages of degradation. They exhibit basically a 14Å period. Their ordering is reduced by heat so that after heating to 400°C they generally exhibit subdued first-order peaks with the higher order peaks either subdued or absent. In some samples, the first-order peak was absent after heating to 400°C.

After heating to 575°C, most of the minerals of this subgroup are almost completely collapsed to 10Å, but they retain very slight "remnant" first-order peaks, suggesting that these mixed-layer chlorite assemblages contain some well ordered chlorite (fig. 5).
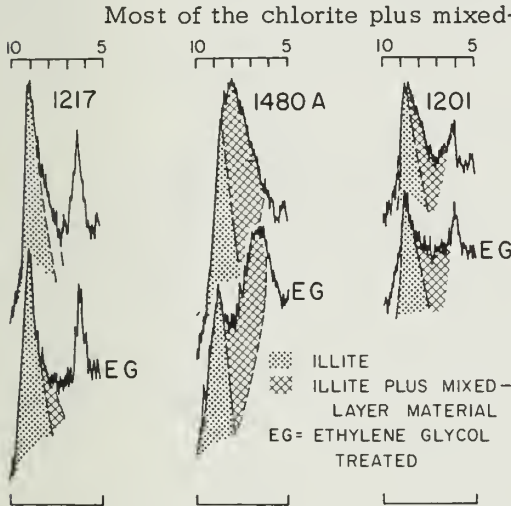
Most of the chlorite plus mixed-layer material is affected, in various degrees, by glycol saturation, suggesting that they are either mixtures containing chlorite and montmorillonite or that they are so degraded that they resemble montmorillonite. Diffraction patterns exhibiting various characteristics and variations of the subgroup chlorite plus mixed-layer material are shown in figure 5.

Fig. 3 - X-ray diffraction patterns of illite and illite plus mixed-layer material.

Montmorillonite Group

Illite plus mixed-layer material, chlorite plus mixed-layer material, and the mixed-layer (undifferentiated) material usually contain montmorillonite as part of the mixtures, but the term "montmorillonite" as used here is reserved for that material which, upon glycol saturation, expands to 17Å. In many samples a 17Å peak appeared (after glycolation) which could not be separated from background radiation peaks. For such samples, the possible presence of a trace of montmorillonite is indicated in tables 4 and 5 by symbol Tr? under the heading "montmorillonite."

Group Composed of Undifferentiated Mixed-Layer Material

In highly permeable samples and in oil-bearing sandstones there are clay mineral mixtures that are at least partially expanded by glycol saturation and are not closely associated with 10Å, 14Å, or 17Å periods. Some of the diffraction patterns exhibit a broad shoulder held above background radiation intensity by a series of low intensity peaks between the 10Å and 16Å positions (fig. 5). The peak intensities are so low and the sample is so thoroughly mixed that separation into individual mineralogical components is extremely difficult, hence, such assemblages are here referred to as "mixed-layer (undifferentiated) material."

Kaolinite Group

The criteria used to distinguish kaolinite from chlorite are those outlined by Bradley (1954). Thus, in the absence of a 14Å peak, a 7Å peak on the diffraction patterns was interpreted as a reflection from kaolinite. In the presence of a 14Å peak, a 2.4Å peak was considered as the third-order reflection of kaolinite. Another criterion used to distinguish kaolinite from chlorite was a doublet peak

with an approximate period of 3.56Å.
Both kaolinite and chlorite give a reflec-
tion at this approximate position – kao-
linite at approximately 3.57Å and chlo-
rite at 3.55Å.

Fig. 4 – X-ray diffraction patterns
of chlorite.

Fig. 5 – X-ray diffraction patterns of
chlorite plus mixed-layer material.

All Mixed-Layer Material

It has been convenient to refer
to illite plus mixed-layer material,
chlorite plus mixed-layer material,
montmorillonite and mixed-layer (undif-
ferentiated) material as a single group
which has been designated as "all mixed-
layer material." The term is used in
the following sections of text and in the
graphs.

QUANTITATIVE ESTIMATES

The methods used to estimate
the proportions of the clay minerals in
the samples varied slightly with the
clay mineral suites and, therefore, at
least in part, with the lithology of the

samples. Two general methods were used. For shale samples, in which illite and chlorite were the dominant minerals, the procedure outlined by Johns et al. (1954) was followed. The method used to make quantitative estimates of the samples proportionately high in mixed-layer material and of those samples with small amounts of clay minerals was a modification of a method suggested by Bradley (personal communication, 1958).

Bradley supplied intensity values for first-order illite, first-order chlorite, first-order glycol-expanded montmorillonite, and third-order kaolinite.   On the scale of the diffractometer used in this investigation these values (table 1) represented approximately pure samples of the minerals. The intensities were the sum of the peak intensity of the curves plus the intensities one-half a degree on each side of the peak position.

Table 1. – Intensity Values of Clay Minerals

Mineral and order	Intensity Values
Illite, first-order	2,000
Chlorite, first-order	500
Montmorillonite, first-order, glycol-expanded	1,000
Kaolinite, third-order	200

The intensities of the various peaks from a sample were compared with the the values above (table 1). A hypothetical example is given below in table 2.

Table 2. – Calculated Parts in Ten Based on Intensity

Mineral and order	Measured intensity	Approximate "pure" intensity value	Calculated parts in ten in sample
Illite, first-order	800	2,000	800/2000 = 4/10 = 4
Chlorite, first-order	100	500	100/500 = 2/10 = 2
Montmorillonite, first-order, glycol-expanded	0	1,000	0/1000 = 0 = 0
Kaolinite, third-order	20	200	20/200 = 1/10 = 1
		Total	7/10 = 7

As the total of the calculated parts in ten is seven, the other three parts are assigned to mixed-layer material (table 2). The diffraction pattern may indicate the nature of the mixed-layer material. For example, if the illite curve is very asymmetrical, part of the mixed-layer material is probably illite plus mixed-layer material; similarly, part may be chlorite plus mixed-layer material. However, it is assumed in this hypothetical case that it is mixed-layer (undifferentiated) material.

After glycolation, the estimates were further refined with respect to the nature and amount of the mixed-layer material. For instance, if glycolation reduced the illite or chlorite intensities, I assumed the expandable portions of the mixture had expanded so that the intensities within one-half a degree of the peak intensity were shifted to the low-angle side of the range. That portion of the total intensity which was shifted was then relegated to the mixed-layer components of illite and chlorite. Using the same hypothetical case as above, the complete quantitative estimates were calculated as shown in table 3.

Table 3. – Calculated Quantitative Estimates

Mineral and order	Untreated intensity	Untreated quantita- tive estimate	Post- glycol intensity	Post- glycol quantita- tive estimate	Difference in quantity
Illite, first-order	800	4	600	3	1
Chlorite, first-order	100	2	50	1	1
Montmorillonite, first- order, glycol-expanded	0	0	0	0	0
Kaolinite, third-order	20	1	20	1	0
Mixed-layer material	–	3	–	3	0

The difference in intensity in the illite portion (table 3) is attributed to il-
lite plus mixed-layer material; in the chlorite portion it is attributed to chlorite
plus mixed-layer material. Thus, the complete refinement of the quantitative es-
timate results in a total clay mineral quantitative estimate as follows, in parts per
ten: illite, 3; chlorite, 1; kaolinite, 1; illite plus mixed-layer material, 1; chlo-
rite plus mixed-layer material, 1; mixed-layer (undifferentiated) material, 3.

The method of Bradley as modified and that outlined by Johns et al. (1954)
were each used to determine the clay mineral quantities in many samples. Gener-
ally the results agreed within one part in twenty, but for some samples the differ-
ences were as much as one part in five. In such instances, the clay mineral suites
were composed mostly of either illite and chlorite or all mixed-layer material. Nine
samples which contained varying amounts of illite were analyzed for potassium oxide
in order to resolve the cause of the difference in results and to determine the correct
quantities.

The amount of potassium oxide in a "pure" illite sample was assumed to be
seven percent. Therefore, the actual potassium oxide content of a clay sample was
compared with the theoretical seven percent to determine the proportionate amount
of illite present (percentage of $K_2O/7.0$ x 100 = percent illite in sample). The per-
centage of illite in the nine samples was calculated by this method. The intensi-
ties of the 10Å peak of each of these samples were calculated from their respective
x-ray diffraction patterns. The results of these analyses as compared with the
respective 10Å intensity values are plotted in figure 6; it should be noted that the
approximate mean value (solid line) of the nine samples closely approaches the
theoretical values (dashed line) suggested by Bradley.

Generally, the estimates obtained using the Johns et al. (1954) method were
in closer agreement with the plotted values of the samples containing more than 30
percent illite than the estimates based on the method suggested by Bradley. How-
ever, in samples with low amounts of well crystallized clay minerals, the values
obtained using the method suggested by Bradley seemed to be closer to the plotted
values than the values obtained based on the Johns et al. (1954) method.

Guided by the above findings, the proportional estimates of the shale sam-
ples used in this investigation were derived using the Johns et al. (1954) method.
The permeable samples, containing high proportions of all mixed-layer material,
were estimated by use of the method suggested by Bradley and modified by the
author in estimating the proportions of illite plus mixed-layer material and chlorite
plus mixed-layer material.

Tables 4 and 5 present the quantitative data of all samples used in this investigation.

DISTRIBUTION OF CLAY MINERALS

Subsurface Samples

Tables 4 and 5 and figures 2 and 7 indicate that the differences between the clay mineral compositions of the permeable sandstone and impermeable shale samples are easily recognized, and that samples that should have intermediate permeabilities (argillaceous sandstones and arenaceous shales) have clay mineral suites intermediate between those of the permeable and impermeable samples (fig. 8).

The shale samples are composed dominantly of illite and chlorite with rarely more than a few parts in ten of illite plus mixed-layer material and chlorite plus mixed-layer material. Kaolinite and montmorillonite are rarely present.

The sandstone clay mineral suites are extremely variable. Average compositions indicate that no one mineral is dominant. Kaolinite, illite, chlorite, and illite plus mixed-layer material generally occur in about equal proportions; chlorite plus mixed-layer material and mixed-layer (undifferentiated) material are generally less abundant, and in some samples montmorillonite occurs as a minor constituent.
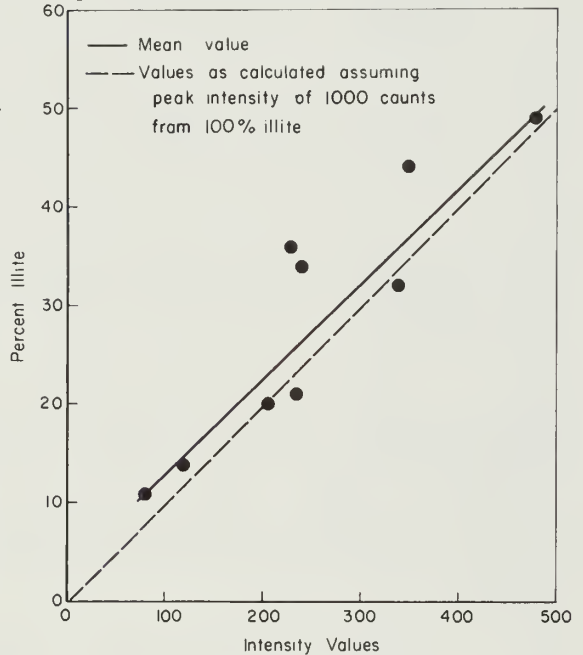
Fig. 6 – The amount of illite calculated by the amount of K_2O present and plotted against the 10Å peak intensity.

Outcrop and Core Samples

The clay mineral analyses of the outcrop samples generally do not agree with either individual core samples or the average compositions of comparable formations. The disagreement is commonly so great that in table 4, the outcrop samples are treated separately and appear below the average composition of the subsurface samples. Montmorillonite or montmorillonite-like undifferentiated mixed-layer clay minerals are the major constituents of outcrop sandstone sample suites but generally only minor constituents in comparable subsurface samples. However, the imperviousness of outcrop shale samples, composed mainly of illite and chlorite, seems to have protected their clay mineral suites, even under conditions of weathering, approximately as well as they have been protected in the subsurface. Consequently, comparison indicates that the average clay mineral suites of subsurface and outcrop shale samples are very similar.

Table 4. – Clay Mineral Composition of Chester Sandstones and Shales
(Parts in ten)

Sample number	Location	County	Illite	Chlorite	Kaolinite	Illite + mixed-layer material	Chlorite + mixed-layer material	Mont-morillonite	Mixed-layer material (undifferentiated)	Lithology
					DEGONIA					
4D	18-7S-10E	White	2	5	Tr	1	1	Tr	Tr	Mica ss
309	24-10S-9E	White	8	-	1.5	Tr	Tr	-	-	Sh
1953DA	(14-5S-14W)	Posey (Ind.)	.5	1.5	1.5	-	-	-	6.5	Ss
1953DB	(14-5S-14W)	Posey (Ind.)	4.5	4	.5	.5	.5	Tr	-	Sh
133A	12-8S-3E	Williamson	2	4	1.5	1.5	1	Tr	-	Sh, ss
133B	12-8S-3E	Williamson	1	4.5	2.5	.5	1.5	-	-	Ss
					CLORE					
133C	12-8S-3E	Williamson	1	3.5	3.5	1	1	-	-	Ss
4C	18-7S-10E	White	1	4.5	1	1+	1+	Tr	1	Ss
440	1-7S-8E	White	1	3	4	.5	.5	Tr	1	Ss
					PALESTINE					
2509	3-1S-13W	Wabash	3+	-	6+	-	-	-	.5	Ss
4P	18-7S-10E	White	1	3	2	2	2	-	Tr	Mica ss
459	15-8S-10E	Gallatin	1	1	1	1	1.5	-	4.5	Ss
487	8-8S-7E	Saline	1	1.5	3	.5	1	3	-	Mica ss
1928	26-1N-13W	Wabash	2.5	2.5	1.5	1+	2+	-	-	Sh, ss
1259	28-7S-6E	Saline	3	4.5	2	Tr	Tr	-	-	Sh sandy
133E	12-8S-3E	Williamson	1	5.5	1.5	1	Tr	-	-	Mica ss
					WALTERSBURG					
1987	8-5S-14W	White	1	2	2	1.5	2	1	.5	Ss with thin sh
3204	15-8S-7E	Saline	1	3	2.5	-	-	Tr	3.5	Ss

Sample	Location	County								Description
133F	12-8S-3E	Williamson	1	5	1	Tr	.5	-	2	Sh, silty
359	13-5S-2E	Williamson	2	1	-	.5	1.5	5	-	Sh
1244	15-10S-6E	Saline	1	6.5	1	Tr	-	1+	-	Ss, argil.
1716	(23-5S-14W)	Posey (Ind.)	1	4	Tr	-	-	-	4.5	Ss
1692	30-2S-11E	Edwards	1	3	3	Tr	Tr	?	2	Ss, shaly
308	30-1N-13W	Wabash	4	3	2	1	-	-	-	Sh, calc.
						TAR SPRINGS				
1974	22-3N-7E	Clay	Tr	6	Tr	3	-	Tr	.5	Ss, argil.
1853A	24-9S-9E	Gallatin	2.5+	Tr	3	3.5+	-	-	Tr	Ss, argil.
1853B	24-9S-9E	Gallatin	7	-	2.5	.5	-	-	-	Mica ss
1472	16-4S-6E	Hamilton	1	2	4	1	1.5	-	.5+	Ss
1000	2-10S-6E	Saline	1.5	1.5	5	1	1	-	-	Ss, argil.
						CYPRESS				
101.1	26-4N-9E	Richland	2	4+	1.5	-	-	Tr	1.5	Ss
133.1	33-4S-14W	White	1	2	4	2	1	-	-	Ss
2508	15-5S-10E	White	1	3-	2+	2	2+	-	-	Ss
2621	25-9S-9E	Gallatin	2	2	1+	2	2	-	1	Ss
KL1	13-14N-7E	Douglas	1	6	1.5	-	-	-	1.5	Ss
295	9-5N-7E	Clay	2	5	1	1.5	5	-	-	Ss
296	24-5S-8E	White	1.5+	-	3+	2.5	2.5	-	-	Ss
298	32-8S-10E	Gallatin	5.5	2	Tr	1	1	Tr	-	Ss, clayey
738	26-4N-9E	Richland	2	6+	1	-	-	-	1	Ss, clayey, porous
1987C	8-5S-14W	White	.5	2	1	2.5	2	-	-	Ss
1201	13-3S-10E	Edwards	2	3.5	1.5	2	Tr	Tr	-	Ss
1480A	31-7N-2W	Bond	2	3	Tr	-	-	-	5	Clay, silty, sandy
1480B	31-7N-2W	Bond	1.5+	2	1.5	.5+	4	-	-	Ss, clayey
2196	31-5N-4E	Fayette	4.5	3	-	1	1.5	-	-	Ss, clayey
						PAINT CREEK				
2661	10-3S-2E	Jefferson	4.5	1	Tr	2.5	1	-	-	Ss, clayey
2578	17-11S-9E	Hardin	8	-	-	.5	-	1.5	-	Ss, argil.

Table 4. – Continued

Sample number	Location	County	Illite	Chlorite	Kaolin-ite	Illite + mixed-layer material	Chlorite + mixed-layer material	Mont-moril-lonite	Mixed-layer material (un-differentiated)	Lithology
					PAINT CREEK (continued)					
1036	18-1N-5E	Wayne	3	3	3	Tr	.5	–	–	Ss, argil.
1339	4-4S-10E	White	1.5	3	4.5	Tr	Tr	–	–	Ss, argil.
1618	28-6N-2E	Hamilton	4.5	Tr	–	4.5	Tr	–	–	Sh
					BETHEL					
133.2	33-4S-14W	White	1.5	5.5	2	–	–	–	1	Sh ?
2507	25-4S-9E	White	3.5	6	–	1	–	–	.5	Sh, sandy
2589	4-3N-1W	Clinton	4	Tr	3	1	2	–	–	Sh, sandy
2603	15-9S-8E	Gallatin	4	–	4	–	–	–	2	Ss, calc.
2634	30-5N-3E	Fayette	1.5	7	Tr	1	Tr	–	–	Sh
2605	17-4N-2W	Bond	.5	2	2.5	2.5	2.5	–	–	Ss
2661	10-3S-2E	Jefferson	1	4.5	2	1	1	–	–	Ss, sh
414	2-13N-7E	Coles	1	2 ?	4	1	2 ?	–	–	Ss
1343B	11-7S-4W	Jackson	Medium	Weak	Weak	Weak	–	–	Weak	Ss
401	33-7S-8E	Saline	3	–	–	7	–	–	–	Mica ss
					RENAULT					
2365	12-7S-4E	Franklin	8.5	–	–	1	Tr	–	Tr	Sh
2518R	25-4S-4E	Jefferson	2.5	6.5	Tr	1	–	–	–	Mica ss
2750	16-4S-7E	Hamilton	4.5	5.5	–	–	–	–	Tr	Sh
2578	17-11S-9E	Hardin	5	–	–	1.5	–	–	3.5	Ss, argil.
					AUX VASES					
136.1	1-4S-7E	Hamilton	4	4+	1	–	–	Tr	–	Sh
2634	30-5N-3E	Fayette	2+	4.5	2+	–	–	–	1	Ss, argil.
1987	8-5S-14W	White	1.5	4	2-	2.5+	Tr	Tr	–	Ss, argil.
2789A	34-6S-9E	White	2+	4.5+	Tr	2-	Tr	1	–	Sh, sandy

Sample	Location	County								Description
2789B	34-6S-9E	White	2.5	4+	1	Tr	1	1	–	Sh
2798	25-8S-6E	Saline	5.5	4	–	–	–	Tr	.5	Sh
2652	35-5N-6E	Clay	1.5	5.5	Tr	1-	1+	1-	–	Sh, sandy
3231	18-2N-8E	Clay	5.5	4	–	.5	Tr	–	–	Sh
3339	35-5S-4E	Franklin	1.5	4+	–	2+	1.5	.5+	–	Sh, calc.
3347A	15-11N-4E	Shelby	5-	3-	–	–	–	–	2.5	Sh
3347B	15-11N-4E	Shelby	3.5	2.5	1	3	–	–	–	Sh, sandy
3354	11-8S-5E	Saline	7+	1+	–	–	–	–	1.5+	Sh
2588	14-2S-4E	Jefferson	5+	2	1.5	–	–	–	1	Ss
297	36-7S-1E	Franklin	Tr	7	Tr	2.5	–	Tr	–	Mica ss
2503	23-1N-5E	Wayne	4	2	Tr	3	Tr	–	–	Sh, calc.
290	8-4N-8E	Clay	1.5+	6-	–	.5	2-	Tr	–	Sh
1217	35-14N-7E	Coles	2.5	7	Tr	Tr	–	–	–	Sh
2336	24-11S-3E	Johnson	8	1	–	Tr	Tr	–	–	Sh
992	32-2S-4W	Washington	–	5.5	–	2	2.5	Tr	–	Mica ss
2632	35-5N-6E	Clay	3	6	–	.5	.5	–	–	Ss, argil.
Chester averages:		Ss, porous	1.3	2.8	2.2	1.2	1	.2	1.3	
		Sh	4	3.6	.5	.9	.5	.2	.3	
		Ss, argil.	2.8	3	1.5	1.1	.8	.2	.6	
DEGONIA OUTCROP										
T10S	19-2S-4E	Marion	–	–	4	–	–	4	2	Ss, porous
T17SA	15-11S-2W	Union	7+	–	2+	–	–	–	–	Sh
T17SB	15-11S-2W	Union	3	–	3	2	–	–	2	Ss, clayey
WALTERSBURG OUTCROP										
O.C.W.	8-13S-5E	Pope	–	–	2	–	–	–	8	Ss, porous
CYPRESS OUTCROP										
T11SA	32-13S-3E	Johnson	1	–	1	2	4	2	–	Ss, slightly shaly
PAINT CREEK OUTCROP										
T11SB	32-13S-3E	Johnson	1.5	2.5	5	.5	.5	–	–	Ss, very argil.

Table 5. – Clay Mineral Analyses of Pre-Chester Sandstones and Shales
(Parts in ten)

Sample number	Location	County	Illite	Chlorite	Kaolinite	Illite + mixed-layer material	Chlorite + mixed-layer material	Montmorillonite	Mixed-layer material (undifferentiated)	Lithology
					MISSISSIPPIAN-VALMEYER					
					Rosiclare					
3361	20-14N-8E	Coles	5	-	-	3	1	-	1	Sh, calc.
2956	33-3S-1W	Washington	7	-	-	2.5	-	-	.5	Sh, calc.
1547	15-2S-5E	Wayne	6	2.5	-	1	.5	-	-	Ls, argil.
KL2	24-14N-7E	Douglas	4	3.5	.5	-	-	Tr?	1.5	Ss, dolo.
2578R	17-11S-9E	Hardin	7	-	Tr	1	-	?	1.5	Ss, calc.
					Osage Group					
12	6-15N-13W	Edgar	5+	3+	-	.5+	.5+	-	-	Sh, silty
3168	17-5N-10E	Jasper	6	2	-	2	Tr	-	-	Sh, silty
2487	25-10N-14W	Clark	4	2.5	Tr	2	1	-	-	Sh, silty
1927A	22-12N-7E	Coles	5.5	3	-	.5	1	-	-	Ss, argil.
1927B	22-12N-7E	Coles	7	1	-	1.5	.5	-	-	Sh
					ORDOVICIAN					
					Thebes					
1343T	11-7S-4W	Jackson	1.5	Tr	7	1	Tr	-	-	Sh, silty
3533A	4-11N-9E	Coles	4	-	Tr	1.5	4	-	-	Ss, argil.
3533B	4-11N-9E	Coles	5	-	-	3.5	1.5	-	-	Mica ss
3533C	4-11N-9E	Coles	5	3	-	1	1	-	-	Ss, argil.
					St. Peter					
2239	29-3S-4W	Washington	7	-	-	3	-	-	-	Siltstone, argil.
3500	4-4S-4W	Pike	4.5	1	2.5	2	-	-	-	Ss

2512	4-4S-4W	Pike	7+	–	Tr	2+	–	–	–	Mica ss
3200A	8-2S-8W	St. Clair	4	–	Tr	6	–	–	–	Ss
3200B	8-2S-8W	St. Clair	8	–	Tr	1.5	Tr?	–	–	Sh
970	21-8N-3E	Fayette	7+	–	–	2+	–	–	–	Mica ss
					Everton					
2512B	4-4S-4W	Pike	6	–	Tr	3	–	–	–	Sh partings, fine ss
KL3A	32-30N-10E	Kankakee	3	–	3	2	2	–	–	Mica ss
					CAMBRIAN Eau Claire					
KL3B	32-30N-10E	Kankakee	6	2	–	1+	1+	–	–	Sh partings, fine ss
					Mt. Simon					
3534	19-13N-4W	Mercer	7+	–	Tr?	2+	–	–	–	Mica ss
Averages:	Sediments, very argil.		6	1.2	.7	1.7	.5		.1	
	Mica ss, and ss, porous		5.2	.8	1	2	.7	–	.2	
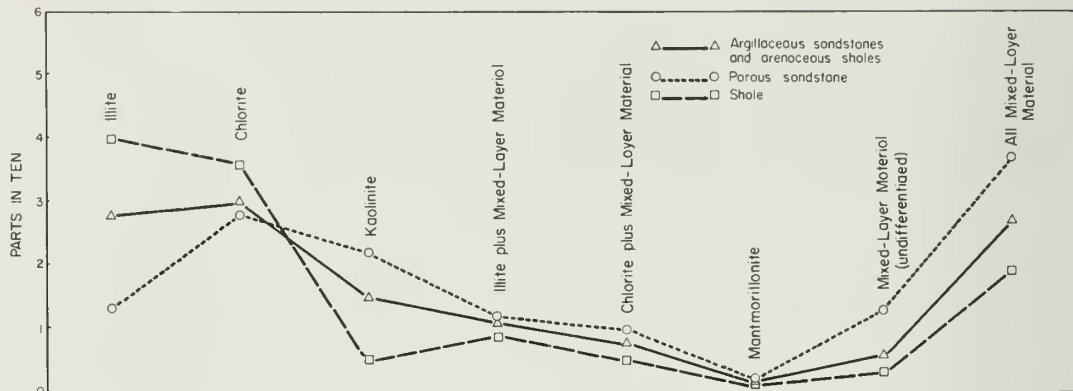
Fig. 7 – Average composition of the three lithologic types investigated.

DISCUSSION

Much of the difference between the clay mineral suites exhibited in various subsurface samples is apparently controlled by the permeabilities of the rocks. Because it seems probable that only minute changes occurred in the source area or environments of deposition during the deposition of the various sand-shale pairs, and only minor changes occurred in these parameters in comparable samples from the same formations within relatively short distances, it therefore seems probable that the clay mineral suites of both lithologies were originally the same in any given pair.

Evidences of the origin of the clay minerals (to be discussed in a later paper) suggest that the clay mineral suites in both lithologies had been, in many instances, altered from the clay mineral suites that were originally deposited.

It must suffice here, however, to say that at some time, just before or during
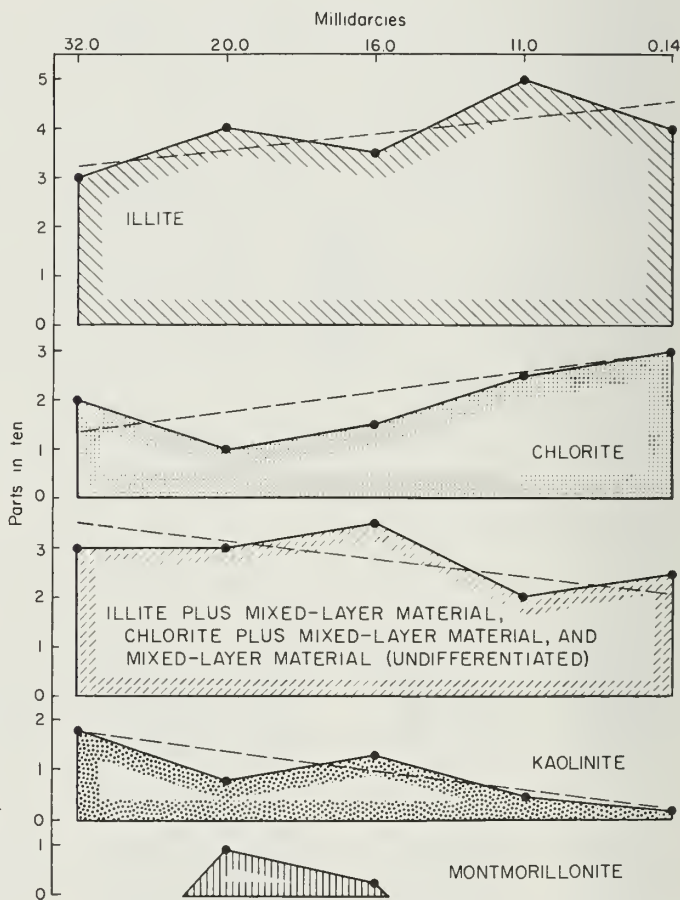
Fig. 8 – Clay mineral analyses of sandstones that have different permeabilities.

lithification, the clay mineral suites of both sandstones and closely associated shales were probably essentially the same.

It therefore appears that the considerable differences between the clay mineral suites of any given pair must have been controlled by some process that had affected the clay mineral suites of the different lithologies in various degrees since the beginning of the lithification processes. There are two major differences in the lithologic characteristics of sandstones and shales: 1) differences in com- pactability and 2) differences in permeability.

If the difference in compaction were the major factor, it would seem rea- sonable to assume that the changes taking place in the shales were reactions sim- ilar to low-grade metamorphism that formed illite and chlorite from the mixed-layer assemblage plus kaolinite of the sandstone-type suite of clay minerals. This se- quence seems unlikely because it is known (from laboratory experiments) that tem- perature, not pressure, is the most important factor in metamorphic type reactions, and it is unlikely that there were any temperature differences between the sand- stones and closely associated shales. Furthermore, if the amount of compaction was the controlling factor, it would seem that argillaceous sandstones should ex- hibit clay mineral suites more like those of sandstones than those of shales be- cause the compaction ratios of argillaceous sandstones should be more like that of sandstones than shales unless there is enough clay in the rock to enable it to act like a shale. The samples investigated suggest that the reverse is true (sam- ples 133, 1427, 1480, 2789, and 3347 of table 4). On the evidence of these and similar samples, it does not appear that variations in compaction can adequately explain the different clay mineral suites.

Because the permeabilities of shales are very low, it seems unlikely that the formation fluids could have changed much in composition since the time of lithification and therefore are true connate water. Thus, if it can be assumed that the clay minerals in shales are at equilibrium with their environment and that the environment has not changed essentially since the time of deepest burial, it seems reasonable that the clay minerals in shales have been essentially unaltered since the time of deepest burial when the clay minerals probably attained stability. This would be true especially in samples from the subsurface which probably have been little affected by weathering processes.

In contrast, the permeability of sandstones would have allowed almost con- tinuous changes in environment because the composition of the formation fluid would have changed almost continuously, creating constant disequilibrium. Meents et al. (1952) and Gorrell (1958) show, by analyses of subsurface oil field brine sam- ples, that considerable variation is present in brine compositions in the same for- mations over lateral distances and within different formations at the same locations.

Meents et al. (1952) show many maps which leave little doubt that in a sin- gle formation in the Illinois Basin the major factor controlling variations in its brine concentration is the distance from its outcrop area. This is apparent not only in relation to the present outcrop areas, especially of the Mississippian Chester for- mations, but also in the northern portion of the basin, where the Chester formations cropped out in pre-Pennsylvanian time. The variations shown in the northern por- tion are strikingly similar to a pre-Pennsylvanian paleogeologic map (for example, that of Workman, 1940). From this, it seems probably that compositions of forma- tion fluids in permeable sandstones must have been changed or altered many times during the history of the rock.

It would seem that if permeability is the controlling factor which causes differences in the clay mineral suites of sandstones and shales, then argillaceous

sandstones, with their intermediate permeabilities, ought to exhibit clay mineral suites that are intermediate between those of permeable sandstones and impermeable shales – a condition that was found during this investigation.

The Sohio Petroleum Company provided five sandstone samples, from the lower part of the Chester Series, that came from the same core but that have varying permeabilities and amounts of clay. The air permeability for each sample is known. Figure 8 represents the clay mineral analyses of these samples in relation to their permeabilities. With decreasing permeability there is a relative increase in illite and chlorite accompanied by corresponding decreases in kaolinite, montmorillonite, illite plus mixed-layer material, chlorite plus mixed-layer material, and mixed-layer (undifferentiated) material.

Available evidence favors permeability as a major factor controlling the differences of the clay mineral suites that are so apparent between sandstones and shales. It also appears that the clay mineral suites of shales have been nearly unchanged since at least the time of their maximum burial. Furthermore, because the original clay mineral suites of the shales must have been approximately the same as those in closely associated sandstones, it must be concluded that the post-lithification alterations are due to degradation of the clay mineral suites in the sandstones by the action of formation fluids whose compositions changed after the sediments were buried and lithified.

Another possible major difference, in addition to their present environments, between the outcrop samples and the subsurface samples is their original environments of deposition. All of the outcrop samples had to be collected tens of miles away from the nearest subsurface sample. Therefore, owing to the great lateral variability of facies in the Chester Series, the outcrop samples should show different clay mineral suites, not only because of different present environments but also because of different environments of deposition.

For example, sample T17SA (an outcrop shale sample, table 4) has a much higher illite content than 1953 DB (a subsurface shale sample, table 4). It seems unlikely that the higher illite content in the outcrop sample reflects a difference in the original environments of deposition of the two samples (a facies change) rather than being due to weathering.

In connection with another study, the author analyzed a group of outcrop and correlative subsurface shale samples from the Ordovician Cincinnatian Series of southwestern Ohio which showed essentially no differences in their clay mineral suites. This study further suggests that the clay mineral suites of impermeable shales are only slightly affected by surface weathering. However, just as it seems that the clay mineral suites of permeable sandstones from subsurface samples have been altered by post-lithification changes, outcrop sandstone samples probably exhibit clay mineral suites that have been altered even more by post-lithification changes because weathering processes are stronger than in the subsurface alterations. The degrading agents that have acted on the clay minerals of permeable sandstones in the subsurface are very similar to those acting on the surface, the main difference being one of degree. From this, it can be concluded that the clay mineral suites of shales, either from surface or subsurface samples, are those that had been formed about the time of lithification or deepest burial and have been little altered either by weathering at the surface or by circulating formation fluids in the subsurface.

It is strongly implied, conversely, that the clay mineral suites found in permeable sandstones are controlled in large part by post-lithification alterations caused by circulating formation fluids in the subsurface and by weathering in the

outcrops. Thus, the clay mineral suites of permeable sandstones may reflect post-lithification changes and bear little resemblance to the clay mineral suites present in the original deposits or those present soon after lithification.

Manner of Degradation

The degradational changes may be characterized as follows: illite is altered to illite plus mixed-layer material, mixed-layer (undifferentiated) material, and possibly montmorillonite; chlorite is altered to chlorite plus mixed-layer material, mixed-layer (undifferentiated) material, and possibly to montmorillonite. It seems possible that, coincidental with these changes, the authigenic "books" or "worms" of kaolinite, reported by Potter and Siever (1956) and observed during the course of this investigation, may have been formed by precipitation.

Because chlorite is rarely present in permeable sandstones and illite is commonly a minor constituent, it seems probably that chlorite is more susceptible to degradation than illite. The data collected here suggest that the degradation of chlorite first forms a mixture of vermiculite and chlorite, a suggestion based on 1) the rarity of the mixture and 2) its occurrence apparently being limited to argillaceous sandstones that are very low in permeability. Once vermiculite is formed in the mixture, it becomes comparatively easy for the magnesium atom to be removed and replaced by a different cation, giving rise to chlorite plux mixed-layer material which would be at least partially expandable. If the magnesium positions in the interlayer areas were replaced by calcium or sodium, it seems very probable that there would be no appreciable difference between the degraded chlorite and the montmorillonite. Hence, chlorite plus mixed-layer material is probably composed, at least in part, of a mixture of chlorite and montmorillonite.

Such degradation probably would not take place similtaneously throughout any one particular chlorite particle. For example, assume a chlorite particle composed of 100 alternating brucite and biotite layers. Probably not all 50 of either the brucite or biotite layers would be equally ordered. Say that two of them might be less ordered than the others and would, therefore, be weaker and thus more subject to degradation. It is supposed that these weaker layers would be the first to lose their brucite components. Then, at this period in the degradation history, only one twenty-fifth of the particle is degraded. With time and stronger degrading conditions, more and more of that particle would be degraded until all of the brucite is destroyed. But at any one time, all three variations might be represented in that one particle, that is, unaltered layers, partially degraded layers, and completely degraded layers.

It has been suggested by Grim (personal communication, 1958) that some of the slightly expandable chloritic material may be particles that have been degraded only along the edges. From this suggestion I assume that such particles would have frayed edges that could be penetrated by ethylene glycol and water. Although this hypothesis would be difficult to prove, it is logical. It seems inconceivable that one brucite layer, covering the area of one chlorite crystal, could be degraded (or hydrated) all at the same time. It seems more logical that if the crystal were to be exposed to a degrading environment, the edges would be the first areas degraded and that degradation would progress toward the center of the crystal. From the beginning of the degrading process until its completion, the central part of the crystal would probably be bound tightly together, prohibiting complete invasion by interlayer water or glycol. Only at the edges would such invasion take place.

The degradation of illite is probably very similar to that of chlorite. As in the case of a chlorite particle, it seems probable that in an illite particle, composed of many individual platelets, not all of the platelets would be equally well ordered. The potassium atoms between layers with inherent internal structural disorders would probably be held with less strength than comparable potassium atoms between well ordered platelets, and the potassium atoms would be more susceptible to replacement by other ions.

The edges of an illite particle (as proposed for chlorite) would be most susceptible and the first portions degraded. If all of the potassium atoms were removed from an interlayer position and replaced by atoms with less bonding strength, expansion in water could take place. This could result in a mixture of illite and a montmorillonite-like mineral. Therefore, a particle of illite plus mixed-layer material could be composed of illite, partially degraded illite with frayed edges, and a montmorillonite-like mineral.

Although montmorillonite is rarely present in shale samples (and if present, it is in trace amounts), it is commonly a minor clay mineral constituent in permeable sandstones. This suggests that most of the montmorillonite present can be attributed to the authigenic degradation of other three-layer, mica-like clay minerals.

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