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## THE MICA IN ARGILLACEOUS SEDIMENTS†

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### INTRODUCTION

The determination of the mineral composition<sup>1</sup> of a variety of soils, clays, and shales, mostly from Illinois, has revealed in many of them a constituent belonging to the mica group of minerals and somewhat similar to muscovite, heretofore referred to as the "sericite-like" mineral. Other investigators<sup>2</sup> have noted what is apparently the same mineral in similar sediments, describing it as resembling muscovite, but as differing from it by containing less potash and more water. It has been called "hydromica" or sericite, the latter term being used because some of its physical properties appeared to be similar to those which have been associated with the mineral sericite.

Recently Endell,<sup>3</sup> Hofmann, and their co-workers have described a mineral from argillaceous sediments as "glimmerton," which has the

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<sup>1</sup> Grim, R. E., Petrology of Pennsylvanian shales and noncalcareous underclays associated with Illinois coals: *Bull. Am. Cer. Soc.*, vol. 14, pp. 113-119; 129-134; 170-176, 1935.

Bray, R. H., Grim, R. E., and Kerr, P. F., Application of clay mineral technique to Illinois clay and shale: *Bull. Geol. Soc. Am.*, vol. 46, pp. 1909-1926, 1935.

Grim, R. E., and Bray, R. H., The constitution of various ceramic clays: *Jour. Am. Cer. Soc.*, vol. 19, pp. 307-315, 1936.

Grim, R. E., Bray, R. H., and Bradley, W. F., The constitution of bond clays and its influence on bonding properties: *Trans. Am. Foundrymen's Assoc.*, vol. 7, No. 5, pp. 211-228, 1936.

<sup>2</sup> Bayley, W. S., Kaolin in North Carolina with a brief note on hydromica: *Ec. Geol.*, vol. 15, pp. 236-246, 1920.

Hickling, G., China clay, its origin and nature: *Trans. Inst. Min. Eng.*, vol. 36, pp. 10-33, 1908-9.

Galpin, S. L., Studies of flint clays and their associates: *Trans. Am. Cer. Soc.*, vol. 14, pp. 301-346, 1912.

<sup>3</sup> Endell, K., Hofmann, U., Maegdefrau, E., Über die Natur des Tonanteils in Rohstoffen der Deutschen Zement-Industrie: *Zement*, vol. 24, pp. 625-632, 1935.

same optical and x-ray properties<sup>4</sup> and mode of occurrence as the "sericite-like" mineral. It is probable that the mineral described by Ross and Kerr<sup>5</sup> as the "potassium-bearing clay mineral" is also similar to the "sericite-like" mineral. Values given by Ross and Kerr for indices of refraction and birefringence are somewhat lower than those of the material herein studied, but the presence of small amounts of impurities, such as quartz and beidellite, would account for these variations and also for the very high silica-to-alumina ratio shown by their formula approximately representing one sample.

Thus, there is no doubt that a mineral belonging to the mica group is a common constituent of argillaceous sediments. So far as is known, no attempts have been made to study this material in detail. Frequently it is found intimately mixed with other clay minerals in a very fine particle size so that analytical data concerning it are difficult or impossible to obtain. By the use of supercentrifuge fractionation technique<sup>6</sup> it has now become possible to obtain the mineral in a relatively pure state in certain fractions of colloidal size, from several shales, clays, and slightly weathered tills. The purest samples have been obtained in the fraction composed of particles varying from about 0.1 micron to about 0.06 micron in diameter,<sup>7</sup> which has been designated as the fine colloid fraction. This paper presents the results of a detailed investigation of this purified material.

#### NOMENCLATURE OF THE MICA MINERAL IN ARGILLACEOUS SEDIMENTS

The question arises whether the mica mineral in argillaceous sediments should be (1) classed with some species of mica, such as muscovite, phengite, or damourite, (2) denominated as sericite, "sericite-like," "hydromica," or "glimmerton," or (3) designated by a new name.

The data herein presented show that it is not possible to designate the mineral as muscovite, phengite, or any other recognized mica species because (1) it possesses important differences in composition from those assigned to other mica species, (2) it is formed as an authigenic mineral<sup>8</sup> in argillaceous sediments soon after deposition from the original debris and also probably as an alteration weathering product, whereas musco-

<sup>4</sup> This similarity was determined following an interchange of material by Hofmann and Grim. X-ray analyses were made on the exchanged material by Hofmann; optical analyses by Grim.

<sup>5</sup> Ross, C. S., and Kerr, P. F., The clay minerals and their identity: *Jour. Sed. Pet.*, vol. 1, p. 59, 1931.

<sup>6</sup> Bray, R. H., Grim, R. E., and Kerr, P. F., *Idem*, 1935.

<sup>7</sup> Effective diameter as calculated from Stokes' law.

<sup>8</sup> Grim, R. E., *Idem*, 1935.

vite and other species of mica have mainly a different mode of origin as igneous or hydrothermal minerals, (3) it occurs in situations different from those in which most muscovite and other mica species occur, and (4) it is found in a physical state that does not convey the usual conception of the properties of muscovite. The mineral occurs commonly in particles less than one micron in diameter, and consequently a mass of it appears unlike a mass of sheets of muscovite. Thus, it would be confusing to designate soil colloidal material by the name muscovite or some other recognized species of mica.

Similarly, it is not desirable to class the mineral as sericite, "sericite-like," "hydromica," or "glimmerton." The use of the term sericite has been varied. As a mineral name, sericite has been loosely used for material differing from muscovite only in its physical characteristics. Some material designated as sericite has been shown to differ from muscovite by containing less potash and more water,<sup>9</sup> or by variations<sup>10</sup> in other components. Rogers<sup>11</sup> has suggested that the term be restricted to a low temperature hydrothermal mineral. Sericite may indicate, therefore, a white mica with certain attributes of physical properties, chemical composition or origin, depending upon the user of the term. Unavoidably such diverse usage has led to confusion so that at the present time a precise concept is not conveyed when a mineral is designated as sericite. It means only a white mica which the user of the term has not wished to class as muscovite for some reason which is usually not stated. If the term sericite were to be used for the mica in argillaceous sediments in any definite sense it would require a specific new definition on the basis of composition and occurrence which would eliminate some material previously included under it. The present confusion regarding the term sericite would be increased by placing the mica herein considered under this term.

The terms "sericite-like," "hydromica," and "glimmerton" (clay mica) are not satisfactory means of designating the material. "Sericite-like" is undesirable because of the vagueness of the significance of sericite, as discussed above. "Hydromica" implies any mica more highly hydrated than muscovite, and although the mica herein considered is more highly hydrated than muscovite, its composition differs from muscovite and all other micas in other important ways, as will be shown. Moreover, other micas differing from it in occurrence or in some phase of

<sup>9</sup> Shannon, E. V., The minerals of Idaho, *U. S. Nat. Mus., Bull.* **131**, 1926.

<sup>10</sup> Niggli, P., Zur Zusammensetzung und Bildung der Sericite: *Schweiz. Miner. u. Petrog. Mitt.*, vol. **13**, pp. 84-90, 1933.

<sup>11</sup> Rogers, A. F., Sericite, a low temperature hydrothermal mineral: *Ec. Geol.*, vol. **11**, pp. 118-150, 1910.

composition may be more highly hydrated than muscovite and are, therefore, "hydromicas." The term "glimmerton" implies nothing of composition or relation to other micas, but only a mica occurring in clays. A more specific term is needed—to indicate for example, that the clay mineral mica in argillaceous sediments is distinctive from previously named species.

There remains only the alternative of giving a new name to the mica occurring in argillaceous sediments, and the term *illite*, taken from the State of Illinois, is here proposed. It is not proposed as a specific mineral name, but as a general term for the clay mineral constituent of argillaceous sediments belonging to the mica group. Many additional analyses must accumulate before the limits of variation of illite are known, and therefore, before it can be specifically named. The writers favor the use of a new name at this time before all limits of variations are known for the following reasons. (1) Because of differences from other mica minerals in composition, occurrence, and origin, a new name or new names for the clay mineral mica in argillaceous sediments will be required. It will cause no confusion to apply now a new general term to this material although future analytical data may make it possible to differentiate what is here included in illite into separate species with specific names, in which case the term illite may be utilized as a group or family name. (2) Petrographic analyses of clays, soils, and shales so urgently require a name to describe this common constituent that it is believed that a new name is fully justified at this time. The additional analyses necessary for specific naming will require a considerable interval of time to accumulate. The least confusion in solving the problem will arise if the required analytical data can be gathered under a definite term.

It is probable that mica similar to that herein described may occur in argillaceous material formed as a weathering product. Although such material may not be found in sediments, the writers favor calling it illite if the material cannot be classed with recognized mica species and if it is similar to illite.

#### ANALYTICAL DATA OF ILLITE PURIFIED FROM ILLINOIS MATERIAL

The following analytical data are representative examples of the data obtained from a very large number of samples of purified illite from slightly weathered tills, clays and shales from Illinois which have been studied. X-ray and microscopic study with high magnification (1500 $\times$ ) indicate that samples obtained from the Maquoketa shale from Calhoun County, and from the Pennsylvanian underclay from Vermilion County (Nos. 1 and 2 of Tables 1 and 3) are the purest illite

samples investigated. The data from them are, therefore, most significant. Samples 3 and 5 (Tables 1 and 3), containing admixed pigmentary limonitic material, and sample 4 containing a small amount of quartz, are included to represent the character of the information obtained from less pure material.

Some analytical properties of certain clay minerals vary with the exchangeable bases which the minerals may contain. Thus, in giving analytical data it is desirable to state the exchangeable bases contained by the analyzed material. The samples herein analyzed contain ammonia as the exchangeable base.

The very small size of the individual particles in the fine colloid fractions has made it impossible to determine all physical properties. The mineral is gray, light green, or light yellow-brown and is micaceous in habit.

By carefully drying aqueous suspensions containing the mineral, it has been possible to obtain aggregates in which the individual particles all have about the same crystallographic orientation.<sup>12</sup> Optical and x-ray measurements have been made on these aggregates which can be studied like large single flakes.

#### *Optical data.*

The optical data are incomplete because the aggregates on which the measurements were made provide scant information concerning the crystal form and orientation of the mineral. Aggregates were found permitting the determination that  $2V$  is equal to about  $5^\circ$ . An occasional anomalous uniaxial figure can be found.  $B_{xa}(=X)$  is nearly  $=c$ .

The indices of refraction in Table 1 were made using liquids which are mixtures<sup>13</sup> of medium Government oil ( $C_{10}H_{22}$ ;  $n = 1.466$ ) and monochlor-naphthalene ( $C_{10}H_7Cl$ ;  $n = 1.633$ ). Marshall<sup>14</sup> suggested, and Correns and Mehmel<sup>15</sup> and von Baren<sup>16</sup> have shown, that the indices of some clay minerals vary for different immersion liquids. Sample 1 was mounted in bromobenzene plus iodobenzene ( $n = 1.609$ ), iodobenzene ( $n = 1.616$ ), and quinoline ( $n = 1.625$ ). According to von Baren, these liquids are most active in influencing optical values. In the first two

<sup>12</sup> Bray, R. H., Grim, R. E., and Kerr, P. F., *Idem*, 1935.

<sup>13</sup> Glass, Jewell J., Standardization of index liquids: *Am. Mineral.*, vol. 19, pp. 459-465, 1934.

<sup>14</sup> Marshall, C. E., The orientation of anisotropic particles in an electric field: *Trans. Faraday Soc.*, vol. 26, pp. 173-189, 1930.

<sup>15</sup> Correns, C. W., Mehmel M., Über den optischen und röntgenographische Nachweis von Kaolinit, Halloysit, und Montmorillonit; *Zeits. Krist.*, vol. 94, pp. 337-348, 1936.

<sup>16</sup> von Baren, F. A., Über den Einfluss verschiedener Flüssigkeiten auf den Brechungsindex von Tonmineralien: *Zeits. Krist.*, vol. 95, pp. 464-469, 1936.

liquids, the indices of refraction remained unchanged, in quinoline they were raised slightly. It can be concluded that illite's optical properties may be slightly influenced by a few liquids. This is in agreement with the concept that the optical properties of clay minerals with relatively low base exchange capacity (see pages for exchange capacity values) are influenced to the smallest degree by immersion liquids.

TABLE 1. OPTICAL PROPERTIES OF ILLITE AS DETERMINED ON AGGREGATES OF PURIFIED MATERIAL AT ROOM TEMPERATURE

Sample No.	1	2	3	4	5
$\gamma$	1.598	1.588	1.605	1.588	1.610
$\gamma - \alpha$	.033	.033	.035	.033	.035
sign	(-)	(-)	(-)	(-)	(-)
2V	5° ±	5° ±	small	small	small

1. Fine colloid fraction. Maquoketa (Ordovician) shale near Gilead, Calhoun Co., Illinois.

2. Fine colloid fraction. Pennsylvanian underclay, near Fithian, Vermilion Co., Illinois.

3. Fine colloid fraction. Slightly weathered till, IVb horizon Clarence soil, Ford Co., Illinois.

4. Fine colloid fraction. Cretaceous shale, near Thebes, Alexander Co., Illinois.

5. Fine colloid fraction. Pennsylvanian shale near Petersburg, Menard Co., Illinois.

It is well known that certain anomalous optical characteristics ("Formdoppelbrechung" and "Eigendoppelbrechung") are obtained from aggregates of uniformly oriented minute rod-shaped or flake-shaped isotropic particles when they are placed in liquids. According to Ambronn and Frey,<sup>17</sup> variable double refraction and uniaxial interference figures are produced when such aggregates are mounted in varied index of refraction liquids, and when the size of individual particles is small in relation to the magnitude of the wave length of light used. Biaxial figures and variable double refraction are obtained under similar conditions with anisotropic particles. As the double refraction characteristics of the illite aggregates were found to be the same when determined in liquids with  $n$  ranging from 1.50 to 1.70, it can be concluded that the optical values are not influenced by "Formdoppelbrechung" or "Eigendoppelbrechung."

It has been shown<sup>18</sup> that the indices of refraction of certain clay minerals vary with loss of water as the temperature is raised to about 200°C. Figure 1 shows that the indices of refraction of illite increase on heating to low temperatures. The values on which Fig. 1 are based were made by

<sup>17</sup> Ambronn, H., and Frey, A., *Das Polarisationsmikroskop*. Leipzig, 1926.

<sup>18</sup> Correns, C. W., Mehmel, M., *Idem.*, 1936.

allowing small fragments of the material placed on a glass slide to remain in an oven at a given temperature for several hours. The slide and specimen were then removed and the index was measured immediately by the immersion method. Because of the tendency of the clay minerals to take up water rapidly, and because of the influence of temperature on index liquids, the values have no high degree of accuracy. The errors would tend to reduce the observed values below the true values. Figure 1 shows also that the indices of muscovite ground to  $-1\mu$  increase less than those of illite when heated to the same temperature. Only aggregates of the  $-1\mu$  muscovite showing random aggregate orientation were available so that mean values alone could be determined.

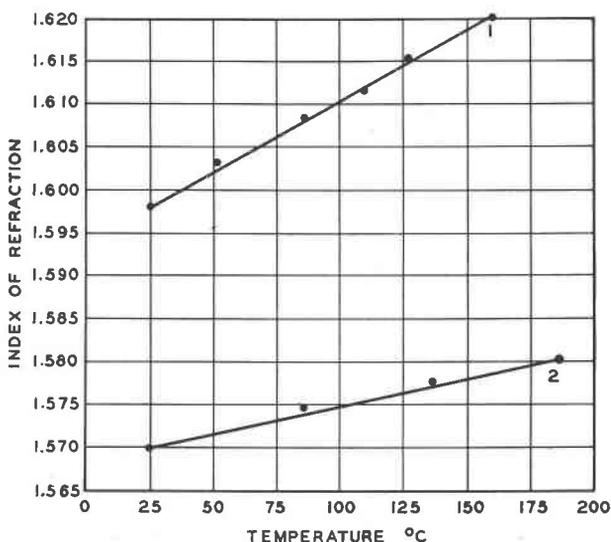


FIG. 1. Relation of index of refraction of illite and muscovite to temperature changes.

No. 1= $\gamma$  values for illite, sample 1 of Table 1.

No. 2=mean values for  $-1\mu$  muscovite.

Data for samples 1 and 2 (Table 1) obtained under the same conditions indicate that the optical properties of illite may show slight variations. Such variations are to be expected in view of the fact that the chemical composition may vary slightly (Table 3). Correns and Mehmel<sup>19</sup> have published results suggesting that for montmorillonite the indices of refraction increase with the Mg content. For illite sample 1 has a higher MgO content and higher indices of refraction than has sample 2. Data are not at hand to evaluate the influence of all components of clay

<sup>19</sup> Correns, C. W., Mehmel, M., *Idem*, 1936.

minerals on their optical properties, so that it must not be concluded that a variation in the MgO content of illite is the only variation in composition which influences optical properties.

The optical properties of illite are similar to those of muscovite except for a smaller value for  $2V$ , and a greater variation of indices of refraction on heating to low temperatures. According to Dana,<sup>20</sup>  $2V$  diminishes in micas, relatively, as the silica increases, from which it would be anticipated (see chemical composition) that  $2V$  for illite would be smaller than for muscovite.

#### *X-ray data.\**

Diffraction data for muscovite, illite (Sample 1 of Table 1), and sericite<sup>21</sup> from the Longfellow mine in California are given in Table 2. Columns one and two give the indices and calculated values of  $d_{hkl}$  for muscovite using the data of Jackson and West.<sup>22</sup> Column three gives values of  $F$  for one molecule calculated from data of Jackson and West and from the present data. Columns four to six give observed values of  $d$  and estimated relative intensities for the indicated samples. The breadth of the diffraction lines increases in the order: muscovite, Longfellow sericite, illite; but no significant variation in  $d$  is found. Powder diffraction diagrams of the illite show only a faint haze between  $d = 4.47\text{\AA}$  and  $d = 3.31\text{\AA}$ , but diffraction from a uniformly oriented flake-shaped aggregate of particles shows perceptible reflections in the  $11l$  and  $02l$  row lines as indicated;  $13l$  and  $20l$  are also more easily recognized in the flake diffractions.

The unit cell of illite seems to be shorter by about one-third of one per cent than the unit cell of muscovite, possibly because of its somewhat lower  $K_2O$  content (see chemical data). Except for this slight difference, the only crystallographic variation between the muscovite, Longfellow sericite, and illite is in particle size, or equivalently, in the perfection of crystallization.

It has been shown<sup>23</sup> that the chemical composition of structures of this type can vary appreciably without pronounced changes in the dif-

<sup>20</sup> Dana, E. S., *Textbook of Mineralogy*, Edited by W. E. Ford. J. Wiley and Sons, New York, 1921.

\* Diffraction patterns were obtained in the laboratory of the University of Illinois through the cooperation of Professor G. L. Clark.

<sup>21</sup> Samples of the sericite were obtained from Prof. P. F. Kerr, Columbia University, New York, N. Y.

<sup>22</sup> Jackson, W. W., and West, J., The crystal structure of muscovite: *Zeits. Krist.*, vol. 76, pp. 211-227, 1930.

<sup>23</sup> Marshall, C. E., Layer lattices and the base-exchange clays: *Zeits. Krist.*, vol. 91, pp. 433-449, 1935.

TABLE 2

Muscovite calculated from Jackson and West with some additions			Muscovite observed (powder)		Sericite Longfellow mine, observed (powder)		Illite (Sample 1 of Table 1) observed (flake and powder)	
Indices	Calc. $d_{hkl}$	$F$ for 1 molecule. Calc. for $MoK\alpha$						
			$\bar{d}$	$I$	$\bar{d}$	$I$	$\bar{d}$	$I$
002	9.96	15.5	9.99	s	9.96	ss	9.98	s
004	4.98	25.6	4.98	m	4.97	s	4.97	w
020	4.51	4						
110	4.48	19	4.47	ss	4.47	ss	4.47	s
11 $\bar{1}$	4.45	20						
021	4.40	11						
111	4.295	9	4.29	w	4.3	ww		
11 $\bar{2}$	4.215	4						
022	4.11	8	4.11	w	4.11	w	4.11	ww
112	3.965	9	3.95	ww	3.95	ww		
11 $\bar{3}$	3.865	19	3.87	db m	3.87	db m		
023	3.73	21	3.72	m	3.75	s	3.7	ww
113	3.58	6	3.55	ww				
11 $\bar{4}$	3.48	38	3.475	m	3.44	s	3.4	ww
024	3.34	24						
006	3.32	40	3.32	ss	3.32	ss	3.31	m
114	3.20	38	3.20	m-s	3.22	s	3.2	ww
11 $\bar{5}$	3.11	12	3.1	ww				
025	2.987	35	2.98	s	2.99	s	2.98	w
115	2.862	29	2.86	m	2.85	m	2.84	ww
11 $\bar{6}$	2.785	33	2.78	m	2.77	m		
13 $\bar{1}$	2.592	35						
200	2.580	37	2.585	w	2.58	w		
116	2.570	29						
131	2.560	43						
20 $\bar{2}$	2.555	46	2.560	ss	2.56	ss	2.56	s
13 $\bar{2}$	2.545	38						
008	2.49	18	2.49	w	2.50	ww		
132	2.485	56						
13 $\bar{3}$	2.458	25	2.46	w	2.45	w	2.44	w
202	2.441	14		ww				
20 $\bar{4}$	2.387	46	2.390	db m				
133	2.380	51	2.376	db m	2.38	m	2.38	m
134	2.26	51						
040	2.255	18						
22 $\bar{1}$	2.245	20	2.245	bd w	2.235	w	2.24	m
041	2.24	18						
220	2.238	4						
13 $\bar{5}$	2.228	10						
221	2.205	21						
22 $\bar{3}$	2.179	34	2.185	w	2.185	w	2.18	w
222	2.142	19						

TABLE 2. (Continued)

Muscovite calculated from Jackson and West with some additions			Muscovite observed (powder)		Sericite Longfellow mine, observed (powder)		Illite (Sample 1 of Table 1) observed (flake and powder)	
Indices	Calc. $d_{hkl}$	$F$ for 1 molecule. Calc. for $\text{MoK}\alpha$						
			$d$	$I$	$d$	$I$	$d$	$I$
20 $\bar{6}$	2.142	33	2.14					
043	2.136	16	2.13	db m	2.13	m	2.11	w
135	2.13	36						
13 $\bar{6}$	2.098	29						
223	2.07	14	2.05	ww	2.05	ww		
044	2.054	14						
136	2.002	7						
0010	1.991	57	1.991	s	1.991	m	1.98	m
206	1.948	24	1.95	w	1.95	w		
04 $\bar{6}$	1.865	23	1.83	ww				
138	1.756	31	1.76	w				
208	1.715	18						
0012	1.660	9						
131 $\bar{0}$	1.660	2	1.654	w	1.66	w	1.65	w
201 $\bar{0}$	1.657	56						
312	1.640	18	1.64	m	1.64	m	1.64	m
313	1.602	20	1.60	w	1.60	ww		
314	1.555	18	1.55	w	1.55			
1310	1.514	50	1.52	w	1.52			
2010	1.509	41						
060	1.503	65	1.504	s	1.50	s	1.50	s
33 $\bar{1}$	1.498	65						
062	1.486	16						
33 $\bar{3}$	1.483	15						
331	1.480	21						
0014	1.423	24						
33 $\bar{7}$	1.371	6						
335	1.345	6	1.344	m	1.344	w	1.34	ww
			1.335	w	1.335	w		
260	1.30	39						
40 $\bar{2}$	1.296	45						
400	1.290	51	1.295	bd w	1.295	w	1.29	m
068	1.288	26						
33 $\bar{9}$	1.289	28						
337	1.287	30						
402	1.263	31						
40 $\bar{6}$	1.245	26						
0016	1.245	30	1.245	bd w	1.245	w	1.24	w
264	1.242	32						

Observed diffractions were obtained with  $\text{CuK}\alpha$  radiation.

Key: ss=very strong, s=strong, m=medium, w=weak, ww=very weak, db=discernible doublet, bd=broad.

fraction pattern. Although illite differs somewhat in chemical composition from muscovite, it is not surprising, therefore, that it should possess about the same structure.

The illite sample (Sample 1 of Table 1) does not contain diffraction lines of quartz.

#### *Chemical data.*

Computed on the basis of the analyses of the purest samples (Nos. 1 and 2 of Table 3), the composition of illite may be approximately ex-

TABLE 3. CHEMICAL ANALYSES OF ILLITE\*

	1	2	3	4	5
SiO <sub>2</sub>	50.10	51.22	47.21	52.23	44.01
Al <sub>2</sub> O <sub>3</sub>	25.12	25.91	21.47	25.85	26.81
Fe <sub>2</sub> O <sub>3</sub>	5.12	4.59	10.73	4.04	11.99
FeO	1.52	1.70			
MgO	3.93	2.84	3.62	2.69	2.43
CaO	0.35	0.16	0.21	0.60	0.11
Na <sub>2</sub> O	0.05	0.17		0.33	0.07
K <sub>2</sub> O	6.93	6.09	5.78	6.56	4.78
TiO <sub>2</sub>	0.50	0.53		0.37	0.64
Ign. Loss†	6.82	7.49	10.99	7.88	9.19
Total	100.44	100.70	100.01	100.55	100.03
H <sub>2</sub> O+	7.18	7.14	6.17	7.88	8.03
H <sub>2</sub> O-	1.90	1.45	3.80	1.13	2.33
SiO <sub>2</sub> /R <sub>2</sub> O <sub>3</sub>	3.00	3.02	2.84	3.13	2.17
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	3.39	3.36	3.51	3.43	2.79

1. Fine colloid fraction, Maquoketa (Ordovician) shale, near Gilead, Calhoun County, Ill.

2. Fine colloid fraction, Pennsylvanian underclay, near Fithian, Vermilion County, Ill.

3. Fine colloid fraction, slightly weathered till, IVb horizon of Clarence soil, Ford County, Ill.

4. Fine colloid fraction, Cretaceous shale, near Thebes, Alexander County, Ill.

5. Fine colloid fraction, Pennsylvanian shale, near Petersburg, Menard County, Ill.

\* Analyses 1, 2, 4 and 5 were made under the supervision of O. W. Rees, Associate Chemist, Illinois State Geological Survey; analysis 3 was made under the supervision of R. H. Bray.

† The loss on ignition values for samples 1, 2, 4 and 5 are based on material dried to 110°C.; for sample 3 it is based on air dried material.

pressed as  $2K_2O \cdot 3MO \cdot 8R_2O_3 \cdot 24SiO_2 \cdot 12H_2O$ . Considerable analytical work shows that the composition of the mineral is subject to some slight variation. Additional analyses of purified material must be gathered before the exact limits of the variations are known.

It is noteworthy that illite has a  $\text{SiO}_2$  to  $\text{R}_2\text{O}_3$  ratio of about 3. This ratio higher than 2, which is generally characteristic of most mica, is not the result of admixed quartz or other minerals, such as pyrophyllite, with high  $\text{SiO}_2$  to  $\text{Al}_2\text{O}_3$  ratios, as the *x*-ray and optical analyses of samples 1 and 2 show that these samples consist of a single mineral. The MgO and FeO (MO) content are of sufficient magnitude and uniformity to require a place in the formula. The  $\text{K}_2\text{O}$  and  $\text{H}_2\text{O}$  contents, which are relatively low and high, respectively, for micas, are also noteworthy.

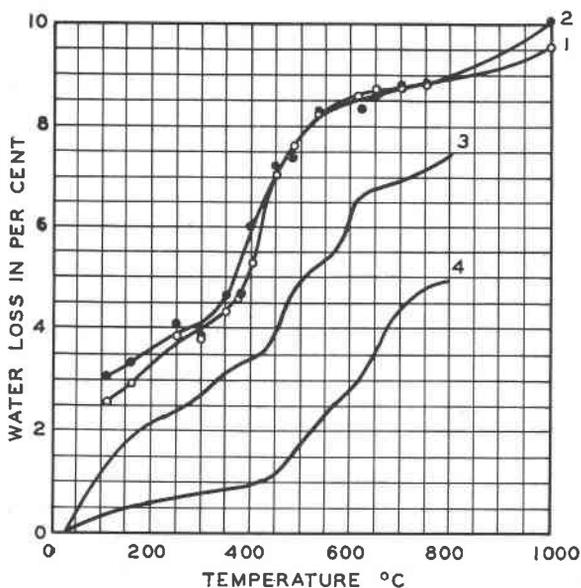


FIG. 2. Dehydration curves of illite and muscovite.

No. 1—Sample 1 of Table 1.

No. 2—Sample 2 of Table 1.

No. 3—Very finely ground muscovite.

No. 4— $-100$  mesh muscovite (coarser than 3).

Curves 3 and 4 after Kelley, Jenny and Brown; *Soil Sci.*, 1936.

Samples 3, 4 and 5 are less pure than 1 and 2. Samples 3 and 5 contain admixed limonitic material, and sample 4 contains quartz. The presence of these constituents must be recognized in considering the significance of the analyses of the samples.

#### *Dehydration data.*

Dehydration curves (Fig. 2) obtained from illite resemble more closely the curve for very finely ground muscovite than that of  $-100$  mesh

muscovite or those of other clay minerals.<sup>24</sup> Dehydration data for other micas are not available for comparative study.

Kelley, Jenny, and Brown<sup>25</sup> have recently shown that very fine grinding of various hydrous minerals tends to shift the inflection point to a lower temperature and to apparently reduce the crystal lattice water with the difference appearing as adsorbed water. This effect is shown by their curves for -100 mesh muscovite and for the same mineral very finely ground (Fig. 2). They have concluded that "differences in the inflection points do not necessarily signify different lattices but may be

TABLE 4. WATER LOSS DETERMINATIONS OF ILLITE\*

Temperature °C.	Water loss in per cent	
	Sample 1	Sample 2
110	2.60	3.03
160	2.92	3.41
250	3.82	4.12
300	3.63	3.84
350	4.45	4.62
375	4.62	4.62
400	5.37	6.01
450	7.03	7.24
485	7.56	7.47
530	8.37	8.31
615	8.54	8.41
650	8.74	8.62
700	8.74	8.79
750	8.77	8.79
1000	9.60	10.14

\* Determinations made under the supervision of O. W. Rees, Associate Chemist, Illinois State Geological Survey. Location of samples given in Tables 1 and 3.

the result merely of variations in particle size." It follows that dehydration curves should not be used as diagnostic characteristics of minerals unless particle size is considered. Until dehydration data can be obtained for illite of varying particle size, the dehydration characteristics of the mineral and their value in identifying it cannot be determined fully.

<sup>24</sup> For dehydration curves of other clay minerals see Ross, C. S., and Kerr, P. F., P.P. 165E, *U.S.G.S.*, 1930; Endell, K., Hofmann, [U., and Maegdefrau, E., *Zement*, vol. 24, 1935; Kelley, W. P., Jenny, H., and Brown, S. M., *Soil Sci.*, vol. 41, 1936.

<sup>25</sup> Kelley, W. P., Jenny, H., and Brown, S. M., Hydration of minerals and soil colloids in relation to crystal structure: *Soil Sci.*, vol. 41, pp. 259-274, 1936.

*Base-exchange data.*

Base-exchange capacity values (Table 5) obtained on samples of illite range from about 20 to 40 milliequivalents per 100 grams. These values are lower than the value (about 50 milliequivalents per 100 grams) assigned by Hofmann, Endell<sup>26</sup> and their coworkers to "glimmerton" which is similar to illite. Base-exchange capacity varies with particle size so that this property should be expressed as ranging between limits. Magnitude rather than specific values are important because of variations in particle size, and also because of variations due to slight differences in composition.

TABLE 5. BASE-EXCHANGE CAPACITY

Sample No. (location Table 1)	1	2	3	4	5
Inorganic base exchange capacity in milliequivalents/100 grams	20.5	35.5	33.0	30.0	21.6

In previous reports<sup>27</sup> the question has been raised as to whether illite (the "sericite-like" mineral) has low base-exchange capacity or no base-exchange capacity. If no base-exchange capacity is assigned to illite in the samples studied, the base-exchange capacity shown by the samples (Table 5) can only be explained by the presence in each sample of a second mineral with high exchange capacity. Assuming for an admixed mineral a base-exchange capacity even of 100 (a high value for montmorillonite which possesses the highest exchange capacity of the clay minerals), the quantity necessary to yield the capacities would be appreciable. As already noted, *x*-ray and optical data reveal no constituent in the samples other than illite, except quartz and limonite in Nos. 3, 4, and 5, and these minerals would tend to decrease rather than increase base-exchange capacity values. Theoretical reasons<sup>28</sup> have been presented showing that the micas should possess base-exchange capacity. In the present research, exchange capacity values of the same order of magnitude shown for illite have been obtained from pure crystalline muscovite after grinding it to a diameter of about 1 micron, the ground material retaining the *x*-ray diffraction pattern characteristic of muscovite.

<sup>26</sup> Endell, K., Hofmann, U., and Maegdefrau, E., Über die Natur des Tonanteils in Rohstoffen der Deutschen Zement-Industrie: *Zement*, vol. 24, pp. 625-632, 1935.

<sup>27</sup> Grim, R. E., and Bray, R. H., *Op. cit.*, 1936.

Grim, R. E., Bray, R. H., and Bradley, W. F., *Op. cit.*, 1936.

<sup>28</sup> Van der Meulen, J. B., Relation between the phenomena of cation exchange with silica-alumina complexes and their crystal structure: *Rec. trav. chim.*, vol. 54, pp. 107-113, 1935.

## RELATION OF ILLITE TO OTHER MICAS

The analytical data provide conclusive evidence that illite is a member of the mica group of minerals and somewhat similar to muscovite. Its optical values agree with those for muscovite, except for a smaller value for 2V, and a greater variation of indices of refraction on heating to low temperatures. Illite and muscovite yield diffraction patterns with lines having the same spacings. The former mineral may be distinguished, however, by the greater width of the lines. The mineral can be differentiated from muscovite on the basis of its chemical composition because of a higher SiO<sub>2</sub> to R<sub>2</sub>O<sub>3</sub> ratio, a higher SiO<sub>2</sub> to K<sub>2</sub>O ratio, a higher R<sub>2</sub>O<sub>3</sub> to K<sub>2</sub>O ratio, an appreciable content of MO (mainly magnesia), and a higher H<sub>2</sub>O content.

On the basis of composition, illite is more nearly similar to the phengite variety of mica than to muscovite. The term phengite has been used for white micas whose analyses suggest a higher SiO<sub>2</sub> to R<sub>2</sub>O<sub>3</sub> ratio than 2. Also the presence of MgO in phengite has been noted. Formulae assigned to phengite (K<sub>2</sub>O · (Mg, Fe)O · 2Al<sub>2</sub>O<sub>3</sub> · 7SiO<sub>2</sub> · 2H<sub>2</sub>O by Winchell;<sup>29</sup> K<sub>2</sub>O · 2Al<sub>2</sub>O<sub>3</sub> · 6SiO<sub>2</sub> · 2H<sub>2</sub>O by Tschermak;<sup>30</sup> K<sub>2</sub>O · RO · 2Al<sub>2</sub>O<sub>3</sub> · 6SiO<sub>2</sub> · 2H<sub>2</sub>O by Hallimond)<sup>31</sup> do not agree, particularly in their SiO<sub>2</sub> to K<sub>2</sub>O ratios, with the analyses of illite (Table 3). A search of the literature revealed analyses 1 and 2 (Table 4) of phengite most nearly like the compositions given in Table 3. It is evident that even these specially selected analyses are not closely similar to those of illite, and, therefore, there is no justification, on the basis of composition, for designating the mineral studied as phengite.

The deep red mica from the manganese mines of St. Marcel in Italy named alurgite by Breithaupt and analyzed by Pennfield,<sup>32</sup> and mariposite from the Josephine mine, Bear Valley, California, analyzed by Hillebrand,<sup>33</sup> which Schaller<sup>34</sup> has shown to be the same mineral, is similar in some respects to illite. The analyses (Table 6) show important differences, however, in the H<sub>2</sub>O and K<sub>2</sub>O values. There is also an essential difference in the mode of occurrence.

Schwartz and Leonard<sup>35</sup> described a micaceous alteration product of

<sup>29</sup> Winchell, A. N., *Idem*, 1929.

<sup>30</sup> Tschermak, G., Die Glimmergruppe, *Zeits. Krist.*, vol. 3, pp. 122-167, 1879.

<sup>31</sup> Hallimond, A. F., On the chemical classification of the mica group: *Mineral Mag.*, vol. 20, pp. 305-318, 1925.

<sup>32</sup> Pennfield, S. L., On some minerals from the manganese mines of St. Marcel in Piedmont, Italy: *Am. Jour. Sci.*, 3rd Ser., vol. 46, p. 238, 1893.

<sup>33</sup> Hillebrand, W. F., Mineralogic Notes: *Bull.* 167, *U.S.G.S.*, p. 74, 1900.

<sup>34</sup> Schaller, W. T., Mineralogic Notes, Series 3; *Bull.* 610, *U.S.G.S.*, pp. 139-140, 1916.

<sup>35</sup> Schwartz, G. M., and Leonard, R. J., Alteration of spodumene in Etta Mine, Black Hills, South Dakota: *Am. Jour. Sci.*, 5th Ser., vol. 11, pp. 257-264, 1926.

TABLE 6. CHEMICAL ANALYSES OF MICAS RESEMBLING ILLITE

	1	2	3	4	5	6	7	8	9	10	11	12
SiO <sub>2</sub>	52.28	58.90	46.54	45.72	46.58	40.79	53.22	56.79	47.29	53.47	48.96	47.05
Al <sub>2</sub> O <sub>3</sub>	23.56	25.28	30.39	37.17	37.46	29.98	21.19	25.11	31.31	32.36	30.96	34.90
Fe <sub>2</sub> O <sub>3</sub>		2.30	4.42	2.18	0.80	8.07	1.22	0.63	1.19	0.79		1.50
FeO	5.76		2.98			2.48		0.92	0.37	0.42	0.24	
MgO	2.43	1.49	0.94	2.00	1.16	2.71	6.02	3.29	2.38		1.97	1.95
CaO	0.65	0.65	0.35	0.05	tr	0.45		0.07	0.69	0.17	0.26	
Ni <sub>2</sub> O		1.37	1.44	1.44	0.64	0.38	0.34	0.17	0.88	0.44	1.65	7.96
K <sub>2</sub> O	9.52	5.73	5.57	6.69	6.38	3.47	11.20	8.92	4.07	7.68	8.47	
TiO <sub>2</sub>						1.28		0.18				
CrO <sub>3</sub>							0.87					
MnO							0.18			0.12		
F			0.58								1.04	1.45
Ign. loss	5.94	4.14	5.31	6.79			5.75	4.72		4.07	3.83	4.07
Total	100.44	99.86	98.52	102.04			99.99	100.80			99.38	98.88
H <sub>2</sub> O+					6.06	9.34			9.92			
H <sub>2</sub> O-					0.30	1.20			1.88			
SiO <sub>2</sub> /R <sub>2</sub> O <sub>3</sub>		3.75	2.38	2.02	2.09	1.97	4.10	3.78	2.50	2.76		
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	3.77	3.96	2.61	2.09	2.12	2.31	4.24	3.84	2.57	2.81		

1. Phengite from Wildschpachtal. *Doelter's Handbuch der Mineralchemie*, 2, Abt. 2, p. 428, 1914.
2. Phengite from Witliches Tal. *Doelter's Handbuch der Mineralchemie*, 2, Abt. 2, p. 428, 1914.
3. Secondary muscovite, from Epprechtstein. *Doelter's Handbuch der Mineralchemie*, 2, Abt. 2, p. 426, 1914.
4. Damourit, from Fen, Norway. *Doelter's Handbuch der Mineralchemie*, 2, Abt. 2, p. 422, 1914.
5. Sericite, Carroll-Driscoll mine, Idaho, from E. V. Shannon, *Bull.* 131, U. S. Nat. Mus., p. 367, 1926.
6. Hydromica, North Carolina, from W. S. Bayley, *Ec. Geol.*, vol. 15, pp. 236-246, 1920.
7. Alurgite, Manganese mines of St. Marcel, in Piedmont, Italy, from S. L. Pennfield, *Am. Jour. Sci.*, vol. 46, pp. 288-295, 1890.
8. White mariposite, Josephine mine, Bear Valley, California, from W. L. Hillebrand, *Bull.* 167, U.S.G.S., p. 74, 1900.
9. Secondary mica, Etta mine, S. Dakota, from G. M. Schwartz and R. J. Seward, *Am. Jour. Sci.*, vol. 11, p. 262, 1926.
10. Secondary mica, Branchville, Conn., from G. J. Brush and E. S. Dana, *Am. Jour. Sci.*, vol. 20, p. 274, 1886.
11. Gilbertite, from Ehrenfriedersdorf, *Dana's System of Mineralogy*, p. 618, 1914.
12. Margarodite, from Zillerthal, *Dana's System of Mineralogy*, p. 618, 1914.

spodumene from Etta mine, South Dakota, which has values for  $K_2O$ ,  $MgO$ , and  $H_2O$  approaching those of illite. It has, however, the  $SiO_2$  to  $Al_2O_3$  ratio of muscovite and is believed to be of hydrothermal origin. Earlier Brush and Dana<sup>36</sup> described an alteration product of spodumene from Branchville, Connecticut, which probably had the same origin as the material from Etta. Except for a slightly higher silica value (Table 6), which was interpreted as indicative of a small amount of free silica, the analysis corresponds to the formula for muscovite.

The names damourite, gilbertite, and margarodite have been given to secondary micas of the muscovite type. Available chemical analyses of these minerals (Table 6) are notably different from those of illite (Table 3). Also, damourite, bilgertite, and margarodite appear mainly to have resulted from hydrothermal alteration.

The mineral described as sericite by Shannon<sup>37</sup> from gouge from the Carroll-Driscoll mine in Idaho (No. 5, Table 6), possesses optical and *x*-ray characteristics similar to those of illite. Chemically, however, there are important differences, particularly in the  $SiO_2$  to  $Al_2O_3$  ratio.

<sup>36</sup> Brush, G. J., and Dana, E. S., Spodumene and the results of its alteration: *Am. Jour. Sci.*, 3rd Ser. vol. 20, pp. 257-285, 1880.

<sup>37</sup> Shannon, E. V., The Minerals of Idaho: *U. S. Nat. Museum, Bull.* 131, 1926. A sample of Shannon's original material was kindly furnished us for *x*-ray analysis. For this courtesy we are indebted to the U. S. Nat. Museum and Dr. W. F. Foshag, Curator of Mineralogy.