INTERSTRATIFIED MONTMORILLONITE–MICA CLAYS FROM SUBSOILS OF THE PRAIRIE PROVINCES, WESTERN CANADA*

by

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ABSTRACT

SAMPLES of glacial till and lacustrine soil clays selected over a distance of 900 miles across the Canadian Prairie Provinces were examined in detail by X-ray, chemical, thermal, infrared-absorption and electron-microscope methods. All of the experimental evidence indicated a marked similarity among the samples. The dominant component was an 18 Å phase when glycerol-solvated, which was identified as a randomly inter-stratified montmorillonite–mica. This was associated with minor amounts of a 10 Å phase (randomly interstratified mica–montmorillonite), kaolinite and quartz. Fourier transforms of the basal series of reflections gave contents of expanded material (mont-morillonite) near 55 per cent, whereas calculation of the chemical formula gave near 80 per cent montmorillonite in the interstratified component. The Fourier transforms confirmed the random nature of the interstratification but also showed a tendency to zonal segregation of the mica and montmorillonite layers. On the basis of the chemical formula, the Greene-Kelly test and the infrared spectra, the swelling layer composition was intermediate between that of beidellite and that of Wyoming bentonite.

Although there were no large differences between the samples, the small differences suggested two trends differentiating the till samples and the lacustrine samples: (1) The proportion of the expanded component of the interstratified mineral tended to be less in the lacustrine than in the till samples. (2) The composition of the expanding layers in the lacustrine samples tended to be nearer to the Wyoming bentonite composition than were the till samples. These trends are likely related to K fixation during or after deposi-tion of the lacustrine materials, whereby the expanding layers with a beidellitic com-position would be pre-empted leaving layers more similar to Wyoming bentonite.

INTRODUCTION

STUDIES of the mineralogical composition of soils from the Prairie Provinces (Christiansen, 1959; Ehrlich, Rice and Ellis, 1955; Gravenor and Bayrock, 1961; Pawluk, 1961; Rice, Forman and Patry, 1959; St. Arnaud and Mortland, 1963; Warder and Dion, 1952) have indicated that montmorillonite or montmorillonite and mica were the dominant clay minerals and that mont-morillonite tended to be concentrated in the parent materials. The object of this investigation was to examine in detail these soil montmorillonitic clays

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Table 1.—Description of the Soil Samples

Sample	Great Soil Group	Soil series	Location	Type of Deposit	Underlying formation and its feature (age)		
SMC 41	Brown	Maleb	Coronation, Alta.	Glacial till[1]	Upper Creta	Bearpaw	sh. s.s.
SMH 24	Grey Wooded	Cooking Lake	Edmonton, Alta.	Glacial till[1]	Upper Creta	Edmonton–St. Mary	s.s. sh. coal
18987	Black	Three Hills	Edmonton, Alta.	Lacustrine[1]	Upper Creta	Edmonton–St. Mary	s.s. sh. coal
SME 22	Brown	Haverhill	Moose Jaw, Sask.	Glacial till[2]	Upper Creta	Bearpaw	sh. s.s.
P4608	Dark Brown	Regina	Regina, Sask.	Lacustrine[2]	Upper Creta	Bearpaw	sh. s.s. congl.
SMC 44	Black	Oxbow	Virden, Man.	Glacial till[3]	Upper Creta	Riding Mtn.	sh. s.s. ls. congl.
SMH 18	Black	Red River	Winnipeg, Man.	Lacustrine[4]	Ordovician	Red River	ls. dolomite

[1] Allan, 1943. [2] Mitchell et al., 1947. [3] Ehrlich, Pratt and Poyser, 1956. [4] Ehrlich et al., 1953.

from the C horizons of widely separated sites in the Prairie Provinces and to compare their structures and compositions.

EXPERIMENTAL

Materials

The seven samples were collected from the C horizons of soils of the Brown, Dark Brown, Black, and Grey Wooded Great Soil Groups. The entire area had been glaciated. Detailed information of the nature of the deposit and underlying bedrock is given in Table 1.

Methods

Preparation of the samples studied

The 0.2–0.04 μ clay fraction was separated by combined sedimentation and supercentrifugation (Jackson, 1956) after removal of carbonate with 1 N HCl, destruction of organic matter with H_2O_2, washing and dispersion at approximately pH 8 by adjustment with 0.1 N NaOH. The separated clay fraction was recovered by freeze–drying (Brydon, Rice and Scott, 1963). The particle shape and size of the samples were observed with a Philips 100 electron microscope.

X-ray analysis

X-ray analysis was carried out with Co radiation (Co $K\alpha : \lambda = 1.78890$ Å) using three separate techniques. A Guinier–de Wolff focusing camera (transmission type)* with a flat powder sample was used for obtaining line profiles of (hkl) or (hk) reflections of the clay minerals and for detecting nonclay minerals. The (00l) reflections were obtained with a Philips diffractometer using oriented aggregates prepared by pipetting some of the clay suspension onto glass slides and by examining after solvation, consecutive heat treatments, acid dissolution and different chemical treatments. A 114.6-mm diameter Debye–Scherrer camera, using powder packed in a thin glass tube of 0.2-mm internal diameter, was used for identification of high temperature phases developed when samples were heated to 1250°C.

Infrared absorption analysis

The absorption spectra in the region 250–700 cm^{-1} were obtained with a Beckman IR7 grating spectrometer housing a CsI-prism†. Samples were prepared as a mull in Nujol and mounted between CsI cover slides. A Perkin–Elmer Model 21 spectrophotometer housing a NaCl-prism was employed for recording in the region 700–4000 cm^{-1}. Samples were prepared as a potassium bromide disk containing about 0.025 per cent of a sample.

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† Through the courtesy of Dr. C. H. Amberg, Division of Applied Chemistry National Research Council, Ottawa.

Thermal analysis

Differential thermal analysis curves were obtained with equipment similar to that described by Morita and Rice (1955). The operating conditions were: temperature range, 25°–1250°C; heating rate, 12°C per min; weight of sample used, 116–120 mg.

Thermogravimetric data were recorded with a Stanton Recording Thermobalance having a constant rate of heating of 6.5°C per min.

Chemical analysis

Total chemical analyses were made on samples oven-dried overnight at 110°C by standard procedures. Free silica or alumina was estimated by 4-hr digestion with 0.5 N NaOH on a water bath (Foster, 1953). Free iron oxides were determined by the dithionite-citrate extraction method (Aguilera and Jackson, 1953). Cation exchange capacity was determined by the method of Clark and Hill (1964). Exchangeable K determination was made by analyzing the supernatant solution in the CEC determination.

A modification of the glycerol adsorption method of Kinter and Diamond (1958) was used in which samples were weighed while enclosed in a 110°C oven. The sample holders were suspended consecutively on a Pt wire suspended from the left-hand pan of an analytical balance. One milliliter of 10 per cent glycerol was added to each sample which was then allowed to dry in the oven. Weighings were made after 2 hr, 6 hr, and every 24 hr for several days. A constant final weight was never achieved and the change from fast to slow loss in weight did not occur at the same time with all samples. Consequently, repeated weighings were made, and the slow constant rate of weight-loss was extrapolated back as shown later in Fig. 6.

RESULTS AND DISCUSSION

Color and morphology

The fractionated clays varied in color from gray to brownish yellow. Electron micrographs of these samples showed irregularly shaped and platy particles approximately in the size limits (0.2–0.04 μ) aimed for in the separation. Small opaque particles were observed in all samples and were interpreted as amorphous iron oxides.

X-ray analysis

The X-ray data indicated that the mineral assemblage of all seven samples was practically identical. All of them consisted of a dominant 15 Å phase and small amounts of 10 Å and 7 Å phases and quartz. The till samples contained a trace of feldspar. The presence of a 1.49–1.50 Å reflection of medium intensity on the Guinier films suggested that the dominant clay mineral was dioctahedral.

An example of the diffractometer patterns of the oriented aggregates after various treatments are shown in Fig. 1. Glycerol caused the 15 Å basal reflec-
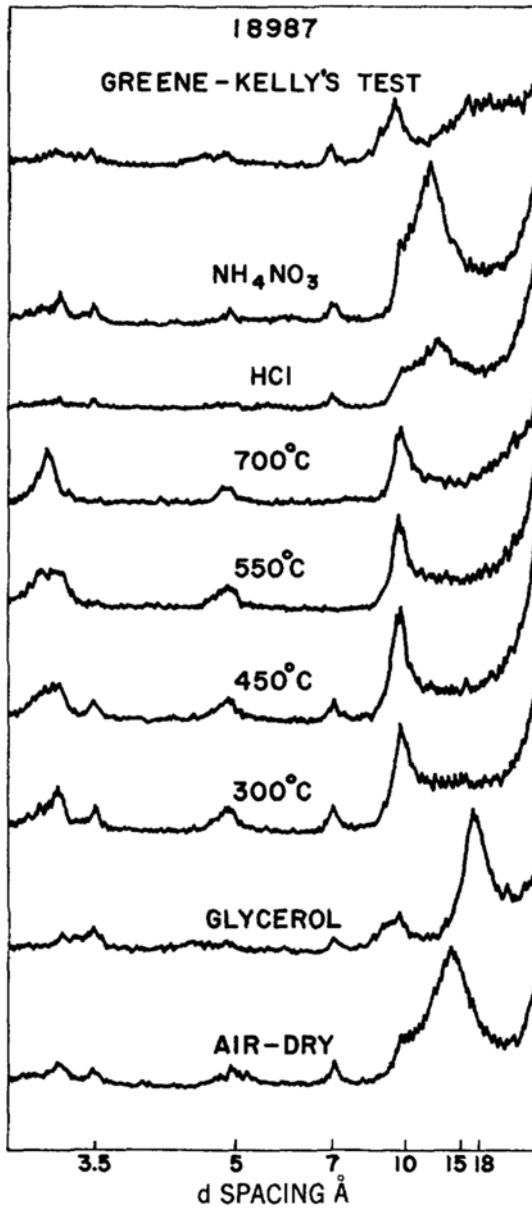
FIG. 1. X-ray diffraction patterns of oriented specimens of the 0.2–0.04 μ fraction from sample 18987 after various treatments.

tion to shift to 18 Å, but higher orders were not a rational series. Upon heating to 550°C, the 15 Å phase had collapsed giving a 9.6–10 Å reflection. The hydrochloric acid treatment (Brydon and Heystek, 1958) caused the 15 Å basal reflection to be replaced by a broad peak having a maximum at 12–13 Å. Similarly, treatment with 1 N NH_4NO_3 (Walker, 1949) caused a shift to 12 Å. After the combined test devised by Greene-Kelly (1953), only sample SMH24 showed a distinct peak at 17.2 Å; the others showed either no peak around 17.5 Å or a very broad diffraction effect. At the same time all samples showed a definite peak at 9.5–9.8 Å with no rational higher orders. These results strongly suggest that the 15 Å phase consists of randomly interstratified layers of montmorillonite and mica and that the expandable layers are probably intermediate between montmorillonite and beidellite in composition.

The small 10 Å peak in the untreated specimen was shifted by glycerolation to 9.69–9.8 Å and was partially merged with the second-order reflection of the 18 Å phase. The 7 Å phase was unaffected by glycerolation, acid treatment and heating to 450°C. At 550°C it had completely disappeared. Judging from these results, the 10 Å phase is randomly interstratified mica–montmorillonite and the 7 Å phase is kaolinite.

A rapid method for quantitative estimation of the clay minerals developed by Oinuma and Kobayashi (1961), based on the idea of Johns, Grim and Bradley (1954), was applied by assuming that the intensity ratios of the basal reflections of the above three clay components are 1.7 : 1.0 : 1.7 respectively. Sudo, Oinuma and Kobayashi (1961) obtained larger values for the 15 Å component ratio and found the best value to be 2.6. On the other hand, our calculations indicate that the intensity of the basal reflection of 70 : 30 interstratified montmorillonite–mica was 3/5 of that of pure montmorillonite. Therefore, the original ratio of 1.7 was used for the estimation of 15 Å material in this case. In addition, since the 7 Å and 10 Å components were weak, any error in applying the above ratios would not greatly change their proportions. These assumptions, although not rigorously correct, should give a reasonable quantititative estimate and the results are given in Table 2.

Crystallite-size determination

The crystallite size, L_{001} was calculated from the half-maximum breadth value of the (001) reflection from the glycerol-solvated oriented specimen. The measured value was corrected for instrumental line broadening using the (001) reflection at 15 Å of p-iodobenzoic acid, which consists of sufficiently large crystals to serve as a standard. The L_{001} values were computed from Jones' equation (1938) and are given in Table 3. All of the L_{001} values range from 45 Å (2.5 × 18 Å) to 90 Å (5 × 18 Å) corresponding to two- to five-layer crystallites.

The half-maximum breadth values of the (02,11) and (13,20) reflections were measured from the tracings of the Guinier films by a microphotometer and corrected for instrumental broadening by using quartz reflections. These

TABLE 2.—QUANTITATIVE ESTIMATION OF THE CLAY MINERALS

Sample	Phase	Peak area	Corrected area*	%
SMC 41	15Å	999.5	587.9	91
	10Å	33.5	33.5	5
	7Å	46.5	27.3	4
SMH 24	15Å	1078.0	634.1	93
	10Å	28.5	28.5	4
	7Å	30.5	17.9	3
18987	15Å	1246.0	1732.9	92
	10Å	34.5	34.5	4
	7Å	56.5	33.2	4
SME 22	15Å	1172.5	689.7	94
	10Å	24.5	24.5	3
	7Å	33.5	19.7	3
P 4608	15Å	1187.0	698.2	89
	10Å	53.5	53.5	7
	7Å	53.0	31.1	4
SMC44	15Å	1247.5	1733.8	97
	10Å	7.0	7.0	1
	7Å	26.5	15.5	2
SMH 18	15Å	1341.0	788.8	90
	10Å	56.0	56.0	7
	7Å	49.5	29.1	3

* Factors used: (15Å); (10Å); (7Å) = (1/1.7); (1/1.0); (1/1.7) from Oinuma and Kobayashi (1961).

TABLE 3.—CRYSTALLITE SIZE OF THE INTERSTRATIFIED MONTMORILLONITE–MICA.

Sample	L_{001}(Å)	$L_{02,11}$(Å)	$L_{13,20}$(Å)
SMC 41	85	150	197
SMH 24	80	139	140
18987	81	118	150
SME 22	80	139	186
P 4608	47	102	151
SMC 44	80	139	157
SMH 18	63	104	140

values are affected by the kaolinite and discrete 10 Å minerals so that the absolute values for the interstratified mineral would be in error. However, since the amounts of the former minerals are small and relatively constant, the relative values of $L_{02,11}$ and $L_{13,20}$ as computed from Warren's equation
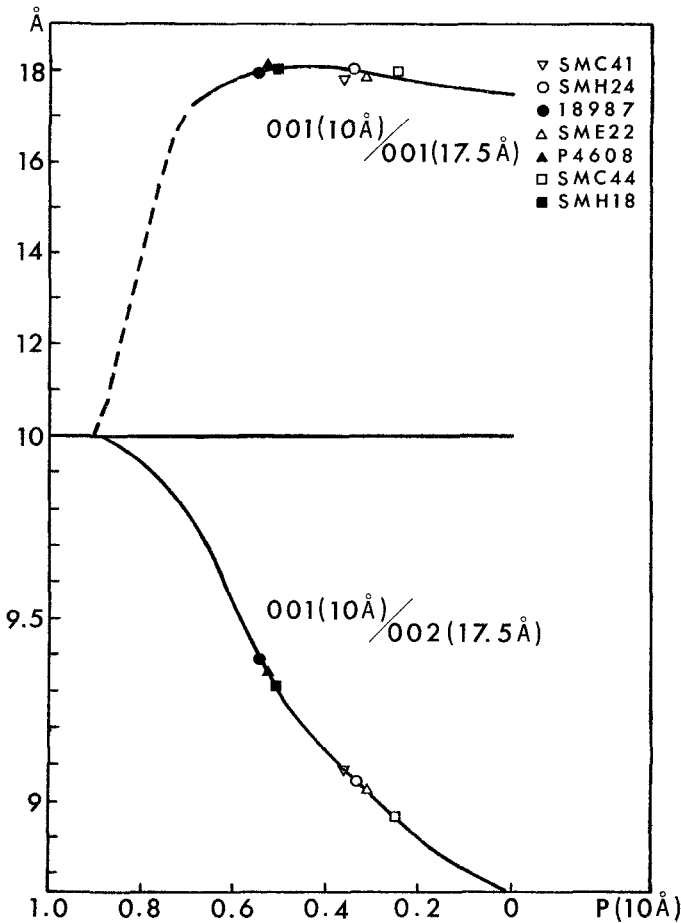
FIG. 2. Peak positions of the (001) reflections of the interstratified mont-
morillonite–mica (Brown, 1961, p. 417).

(1941) should be reliable. There were no remarkable differences, but the
lacustrine samples tended to be smaller than the till samples (Table 3).

Mixing ratio of the interstratified montmorillonite–mica

In order to check the nature of the major interstratified component, the
position of the basal series from the glycerol-solvated oriented specimens were
estimated carefully from X-ray data with a low scanning speed. The positions
of the 001(10 Å)/001(17.5 Å) maxima for all samples were almost identical,
whereas those of the 001(10 Å)/002(17.5 Å) were different. Because the

crystallite size was two to five layers in thickness, these results corresponded to the curve calculated by MacEwan rather than that of Weaver (1956), which was calculated from Hendricks and Teller's (1942) method for infinite crystallites (for both curves, see Brown, 1961, p. 417). Consequently, the positions of the 001(10 Å)/002(17.5 Å) maxima were plotted on the MacEwan curve (Fig. 2) and the corresponding 001(10 Å)/001(17.5 Å) values were plotted using the estimated mica-layer proportions. Fig. 2 shows that the lacustrine samples had a higher proportion of mica layers than the tills and that there was little difference among the tills or among the lacustrine samples.

Fourier transforms by MacEwan's method

More precise and direct information on the nature of the interstratification was obtained with the Fourier transform method of MacEwan (1956) using the basal reflections from the glycerol-solvated specimens. Since samples SMC44 and 18987 had the highest and lowest contents of expanded layers respectively, and, since the difference between them was small (Fig. 2), only two calculations were made. In the preparation of the intensity data, two errors in the measurement of peak area were minimized as follows: A small hump at 9.8 Å due to interstratification was eliminated by drawing a smooth curve along the trace of the main peak at 9.0 Å. The fifth-order basal reflection of the interstratified component at 3.5 Å was superimposed with the (002) reflection of kaolinite. Since the intensities of the kaolinite (001) and (002) reflections are similar (Brown, 1961), the kaolinite (001) peak area was deducted from the observed peak area at 3.50 Å. The I,obs's were thus corrected and then normalized so that the data for the two samples would be comparable. These data are given as I,corr. in Table 4. Fig. 3 gives the two Fourier transforms of these basal reflections. They showed two fundamental

TABLE 4.—X-RAY DIFFRACTION INTENSITIES USED FOR ONE-DIMENSIONAL FOURIER ANALYSIS

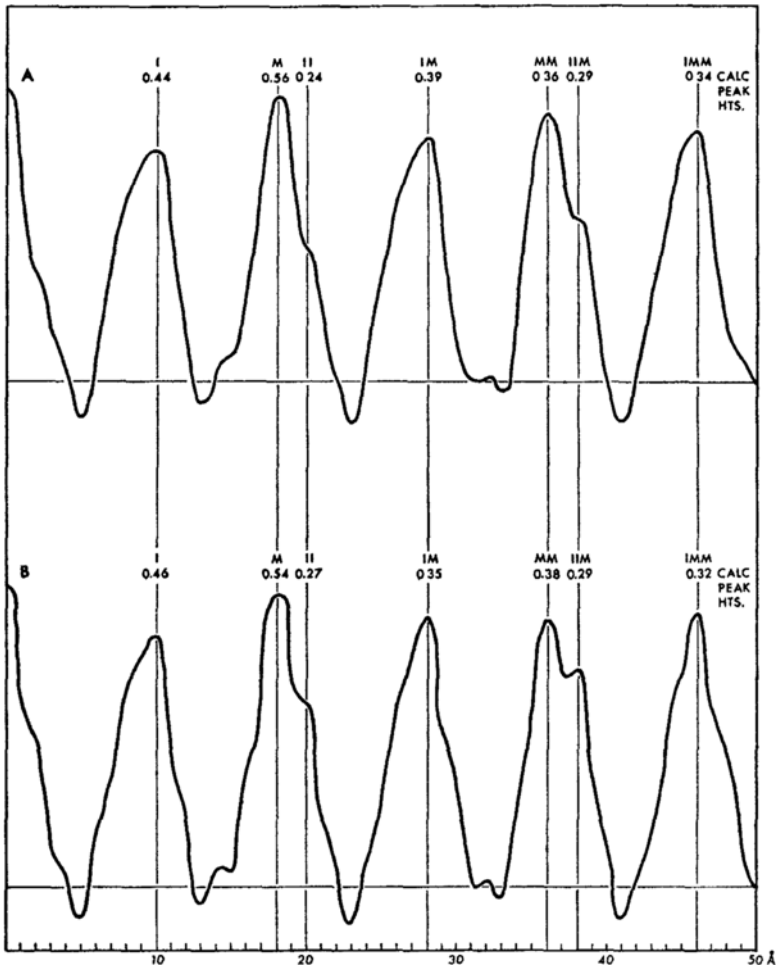| 18987			SMC 44		
Series (Å)	I,obs	I,corr.	Series (Å)	I,obs	I,corr.
1(17.8)	9260	9260	1(17.9)	9260	9260
2(9.16)	2400	2400	2(9.08)	1860	1860
3(5.84)	220	220	3(5.85)	87	87
4(4.48)	610	610	4(4.47)	840	840
5(3.52)	1200	890	5(3.52)	1330	1096
6(2.94)	250	250	6(2.95)	222	222
7(2.53)	86	86	7(2.58)	28	28
789—	—	—	8 —	—	—
9(1.98)	800	800	9(1.98)	188	188
Kaolinite (7Å peak)	310			234	

FIG. 3. Fourier transforms of the (001) reflections of the interstratified montmorillonite–mica in glycerol-solvated specimens of A, Sample SMC44, and B, Sample 18987. (I–10 Å layers, M–18 Å layers).

peaks of 10 Å (I) and 18 Å (M) corresponding to mica and montmorillonite layers respectively.

The probability coefficients were deduced from the observed data of peak heights and are given below.

Sample SMC44		Sample 18987	
$P_I = 0.44$ $P_M = 0.56$		$P_I = 0.46$ $P_M = 0.54$	
$P_{II} = 0.55$ $P_{MM} = 0.65$		$P_{II} = 0.59$ $P_{MM} = 0.65$	
$P_{IM} = 0.45$ $P_{MI} = 0.35$		$P_{IM} = 0.41$ $P_{MI} = 0.35$	

The ratio of the height of the M peak (18 Å) to the I peak (10 Å) is 0.54 : 0.46 for sample 18987 and 0.56 : 0.44 for sample SMC44. These ratios agree roughly with the proportions of mica and montmorillonite layers deduced from the peak positions of the basal spacings.

In both of the samples $P_{II} > P_I$. It could, therefore, be concluded that the montmorillonite and mica layers, which were interstratified in the mixing ratio of 55 : 45, had a partially random structure with a tendency to segregation, i.e. zonal structure.
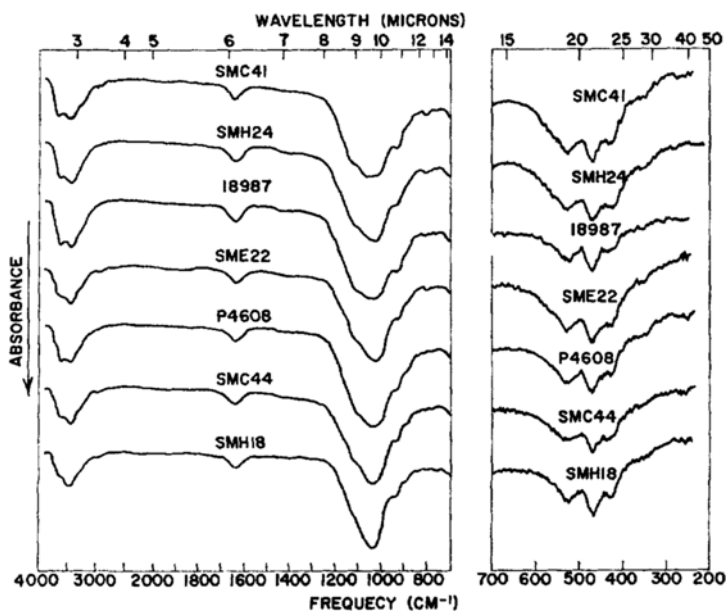

FIG. 4. Infrared absorption curves of the seven clays.

Infrared absorption spectra

The absorption spectra (Fig. 4) of seven samples were virtually identical. The slight difference in the intensities of the two bands at 465 cm^{-1} and 523 cm^{-1} suggested the layer lattice material to be aluminous (Stubičan and Roy, 1961). A shoulderlike small peak around 3710 cm^{-1} supported the previous estimate (5 per cent or less) of kaolinite (Kodama and Oinuma, 1963), and agrees with the estimation by the X-ray method. A characteristic double band from quartz around 800 cm^{-1} was very faint or doubtful. Consequently, the amount was estimated as low as 1 per cent (Hunt and Turner, 1953).

TABLE 5.—DIFFERENTIAL THERMAL DATA FOR THE SEVEN SAMPLES (25°–1250°C)

Sample No.	Temperature of troughs and endotherm peaks (°C)										Products in fired materials
	Endo-thermic	Endo-thermic	Exo-thermic	Endo-thermic	Endo-thermic	Endo-thermic	Exo-thermic	Exo-thermic	Exo-thermic		
SMC 41	120	187 Shoulder	310	540	—	876	920	—	1157		M., α-Fe$_2$O$_3$
SMH 24	120	190 Shoulder	310	535	650 Inflexion	880	920	—	1150		M., α-Fe$_2$O$_3$
18987	125	195 Shoulder	320	540	650 Inflexion	868	918	—	1160		M., α-Fe$_2$O$_3$
SME 22	128	195 Shoulder	320	540	650 Inflexion	858	900	1120	1185		M., α-Fe$_2$O$_3$ Cor., Crist.
P 4608	135	195 Inflexion	328	545	—	850	900	—	1170		M., α-Fe$_2$O$_3$ Hercy.
SMC 44	132	200 Shoulder	322	540	—	860	900	1108	1180		M., α-Fe$_2$O$_3$ Cor., Crist.
SMH 18	128	195 Inflexion	333	548	650 Inflexion	860	898	—	1178		M., α-Fe$_2$O$_3$ Hercy.

M.: Mullite; Cor.: Cordierite; Crist.: Cristobalite; Hercy.: Hercynite.

Thermal analysis

The DTA data summarized in Table 5 indicate the similarity of the samples. An important feature is that the temperature of the third endothermic peak (540°C) is in the range of " normal " illite, " abnormal " montmorillonite or interstratified montmorillonite–illite (Cole, 1955).

The samples fired to 1250°C were examined by X-ray and the phases identified are listed also in Table 5. Mullite and α-Fe_2O_3 were found in all of the samples. In addition, small amounts of cordierite and cristobalite (as the α-form) were detected in samples SME22 and SMC44, and a trace of hercynite was recognized in samples P4608 and SMH18. The appearance of cordierite and cristobalite seems to be related to the exothermic peaks at 1108°–1120°C. The general occurrence of mullite suggests all samples to be aluminous and to be more similar to the Wyoming-type than to the Cheto-type montmorillonites (Grim and Kulbicki, 1961).
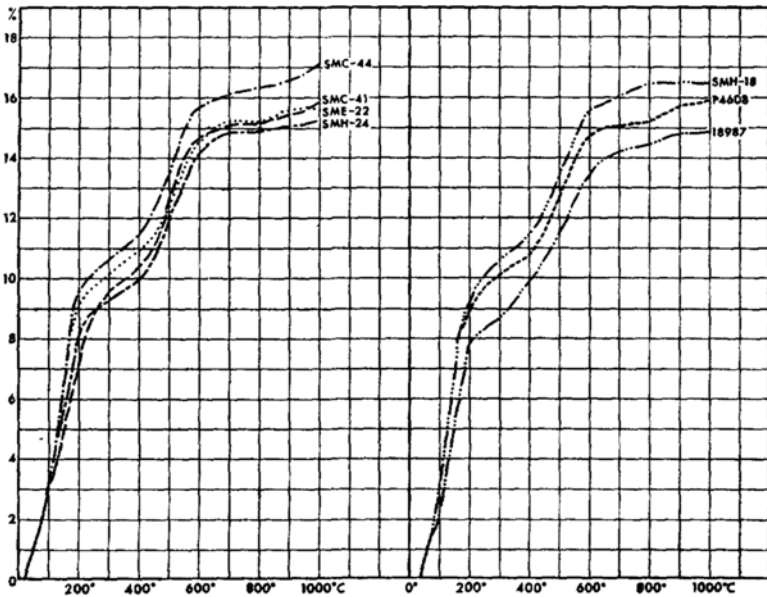

FIG. 5. Thermogravimetric analysis curves of the seven clays.

The thermogravimetric curves in Fig. 5 were obtained by replotting thermogravimetric data as weight loss (per cent) against temperature. By reference to the DTA data, the weight loss in the temperature range 25°–400°C was considered due to loss of absorbed water and combustion of organic matter remaining from the H_2O_2 treatment, and that in the temperature range 400°–1000°C (Table 6) was considered due to loss of structural hydroxyls. Two samples, 18987 and SME22, showed 400°–1000°C losses, which were inter-

TABLE 6.—Chemical Data of the Seven Clays

Sample no.	SMC 41	SMH 24	18987	SME 22	P 4608	SMC 44	SMH 18
Locality	Coronation, Alta.	Edmonton, Alta.	Edmonton, Alta.	Moose Jaw, Sask.	Regina, Sask.	Virden, Man.	Winnipeg, Man.
Origin	Glacial till	Glacial till	Lacustrine	Glacial till	Lacustrine	Glacial till	Lacustrine
Composition, %							
SiO_2	49.54	48.54	52.41	52.27	52.07	52.17	52.27
TiO_2	0.01	tr.	none	none	none	0.01	none
Al_2O_3	26.42	26.22	26.78	24.57	25.02	24.29	25.13
Fe_2O_3	7.73	7.23	5.75	7.40	6.30	7.56	6.08
MnO	tr.	tr.	tr.	tr.	tr.	tr.	tr.
MgO	0.23	2.79	2.18	3.73	1.84	3.79	1.38
CaO	1.45	1.28	1.20	1.16	1.10	1.44	0.93
Na_2O	0.18	0.24	0.59	0.24	0.67	0.19	0.24
K_2O	2.82	2.61	2.91	2.39	2.95	2.11	2.91
H_2O (ig. loss)	10.90	10.49	8.75	9.00	10.22	8.84	10.71
P_2O_5	tr.	tr.	tr.	tr.	tr.	tr.	none
Total	99.28	99.40	100.57	100.70	100.17	100.40	99.65
Free SiO_2	—	1.26	—	0.27	—	0.10	—
Free Al_2O_3	1.49	—	0.24	—	1.11	—	0.13
Free Fe_2O_3	5.18	3.85	2.19	3.40	2.31	3.91	1.95
Exch. K_2O	0.37	0.22	0.19	0.24	0.19	0.24	0.19
Non-exch. K_2O	2.45	2.39	2.72	2.15	2.76	1.87	2.72
Structural Water*	5.57	5.30	4.88	4.69	5.11	5.73	4.99
CEC Meq/100 g	59.4	60.9	59.4	62.7	61.2	65.8	61.8
Glycerol adsorbed mg/gm clay	n.d.	110.0	100.0	125.0	97.0	129.0	109.0

* Based upon weight-loss data between 400°–1000°C from the thermogravimetric curves.

mediate between the theoretical values for muscovite and montmorillonite (4.52 and 4.93 per cent respectively). The others were somewhat higher than both of the theoretical values. This discrepancy may be attributed to the error in assuming that only lattice hydroxyls were involved in this temperature range. The loss may have been associated with edge hydroxyls, hydroxyls associated with amorphous materials and/or H_3O^+ replacing K^+ in mica (Brown and Norrish, 1952).

Chemical analyses

The total analyses and the data for free oxides of the seven samples are reported in Table 6. Although the analyses of all samples except SMC41 are very similar, the lacustrine samples tend to have a higher content of K_2O and lower content of MgO than the till samples. The analyses suggest a clay with a composition intermediate between montmorillonite and mica.

The CEC values (Table 6) are in the range of 59.4 and 65.8 meq per 100 g. The highest value agrees well with the value of 65 meq per 100 g for Kinnekulle I clay (2.5 per cent K_2O), which was described as 1 : 4 illite : montmorillonite mixed-layer by Byström (1954). The lacustrine clays have higher nonexchangeable potassium contents yet show lower exchangeable potassium values than the tills.
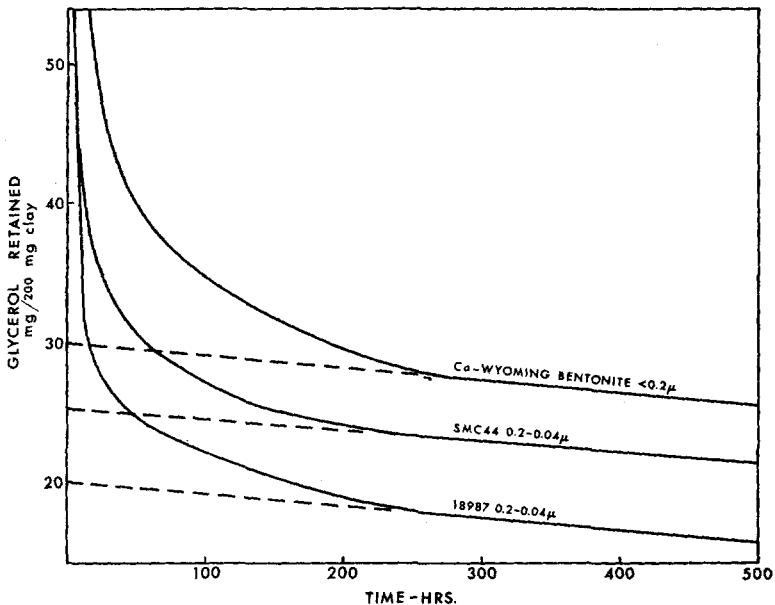

FIG. 6. Drying curves of representative samples of glycerol-saturated clays.

Glycerol adsorption

The drying curves of glycerol-saturated Wyoming bentonite and the soil clays are shown in Fig. 6. All curves show an initial fast loss in weight in approximately 10 hr, a second slower and changing rate of loss over a period of 200–300 hr and finally a very slow loss in weight at a constant rate. These rates were rather similar for the samples used. The straight lines were projected back to zero time and the intercept values reported in Table 6 as the glycerol retention values corresponding to a double layer of glycerol. The retention values range from 62–84 per cent of the value for fractionated Wyoming bentonite. The lacustrine samples had lower values than the till samples.
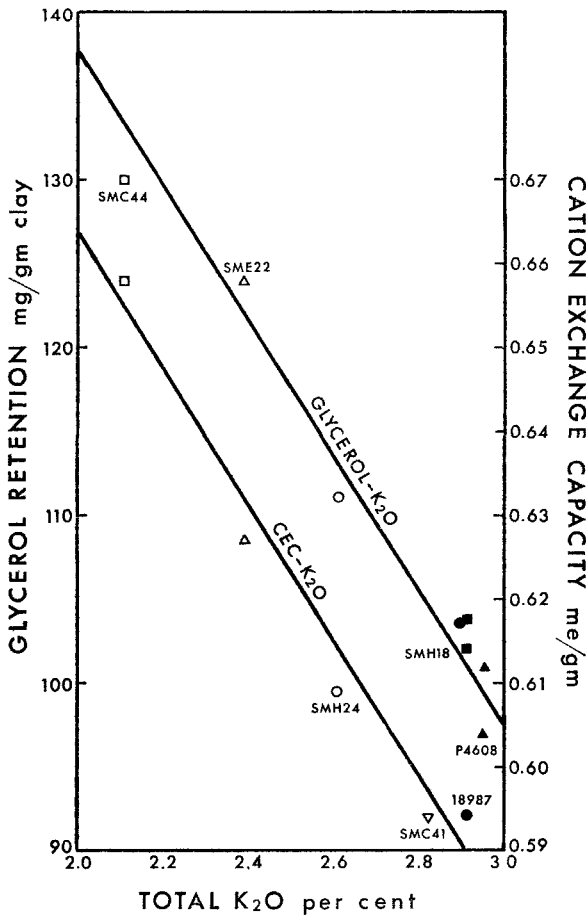
FIG. 7. relationship of glycerol and of cation exchange capacity with the total potassium content of the seven clays.

The inter-relationship of the cation exchange capacity, total K_2O content and the glycerol retention values are shown in Fig. 7. The plot of glycerol retention vs. K_2O gave an excellent negative straight-line relationship. The cation exchange capacity vs. K_2O also gave a negative straight-line relationship except for two values from the lacustrine samples SMH18 and P4608. These relationships are rather unusual, since the effect of edges was not eliminated from either the glycerol retention or the CEC values. Both plots corroborate the hypothesis that a K-bearing phyllosilicate will gain in inter-layer surface and available charges upon losing K (St. Arnaud and Mortland, 1963). The slope of the plot is 3 rather than unity as theory would require and probably reflects the edge effects.

Calculation of structural formula

In the calculation of the structural formula, the quartz and feldspar contamination were neglected because of the small amount, and the muscovite composition, $KAl_2(Si_3Al)O_{10}(OH)_2$, was used for the micaceous-layer material of both the 10 Å-phase and 15 Å-phase material. A combination of chemical analysis, structural water content from weight-loss data and quantitative estimation of clay minerals was used in calculating the formula. Table 7 shows an example of the calculation for sample 18987 done in two ways. In one, corrections were made for the kaolinite and muscovite corresponding to the 7 Å and 10 Å components. In the second, (figures in parentheses in Table 7) the entire K_2O content was calculated as muscovite, and the structural composition of the swelling layer alone was calculated. The analysis in weight % (col. 2) was corrected for free oxides (col. 3) and then normalized to give a total of 100 (col. 4). A correction was made (col. 5) for the muscovite and kaolinite contents from Table 2. The net analysis (col. 6) was converted to relative moles (col. 7) and relative ions (col. 8). Thus, the final figures in column 9 were obtained by scaling the total cationic charges to 22, which here is equal to the total anionic charges: $22 = 2x+y$, where x and y are subscripts as $O_x(OH)_y$.

In a second calculation, since the basic formula of the nonswelling material is $KAl_2(Si_3Al)O_{10}(OH)_2$ by definition, the number of K ions gives the proportion of nonswelling layers. The calculated formulae for all samples are listed in Table 8.

The calculated (OH) content varied from 2.02 to 2.62 atoms per unit cell and was undoubtedly caused by error in evaluating the structural water content from thermogravimetric analysis.

The proportion of montmorillonite in the interstratified material varies from 0.76 to 0.82. These must be maximum values because less than the assumed ideal K content would raise the mica content over the calculated value. In fact, the calculated proportion of montmorillonite is higher in the above calculation than the proportion estimated from peak positions or the Fourier transform. Nevertheless, the order of the different samples is similar.

The aluminous, dioctahedral character of the material is shown by either

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TABLE 7.—CALCULATION OF CHEMICAL FORMULAE FOR SAMPLE 18987

(1)	(2) Wt.%	(3) Free oxides, Wt.%	(4) $\dfrac{100 \times [(2)-(3)]}{\Sigma[(2)-(3)]}$	(5) K*	(5) M†	(6) (4) − (5)	(7) Mole proportion		(8) Atom proportion		(9) Atom ratio	
SiO_2	52.41	—	55.60	1.86	1.81 (11.82)	51.93 (41.92)	Si	0.864 (0.697)	0.864	(0.697)	3.584	(3.759)
Al_2O_3	26.78	0.24	28.15	1.58	1.54 (10.03)	25.03 (16.54)	Al	0.245 (0.162)	0.491	(0.324)	2.036	(1.747)
Fe_2O_3	5.75	2.19	3.78			3.78 (3.78)	$Fe^{3}{}^{+}$ 0.023$_6$		0.047	(0.047)	0.194	(0.253)
MgO	2.18	—	2.31			2.31 (2.31)	Mg	0.057	0.057	(0.057)	0.236	(0.307)
CaO	1.20	—	1.27			1.27 (1.27)	Ca	0.022	0.022	(0.022)	0.091	(0.118)
Na_2O	0.59	—	0.62			0.62 (0.62)	Na	0.010	0.020	(0.020)	0.082	(0.107)
K_2O	2.91	—	3.09		0.47 (3.09)	2.62 —	K	0.027$_5$	0.055		0.228	—
$H_2O(+)$	4.88	—	5.18	0.56	0.18 (1.18)	4.44 (3.44)	OH	0.246 (0.190$_9$)	0.492	(0.382)	2.041	(2.060)

* K = Kaolinite: Using the weight ratio of the oxides from the ideal formula $Al_2Si_2O_5(OH)_4$.

† M = Muscovite: Using the weight ratio of the oxides from the ideal formula $KAl_3Si_3O_{10}(OH)_2$.

TABLE 8.—SUMMARY OF THE CHEMICAL FORMULAE FOR THE SEVEN CLAYS

	Overall layer	Swelling layer	Nonswelling layer
SMC 41	$(K_{0.14}Na_{0.03}Ca_{0.12})(Al_{1.77}Fe_{0.15}Mg_{0.03})$ $-(Si_{4.70}Al_{0.30})O_{9.69}(OH)_{2.62}$	$0.76 \times (Na_{0.03}Ca_{0.16})(Al_{1.69}Fe_{0.20}Mg_{0.03})$ $-(Si_{3.91}Al_{0.09})O_{9.60}(OH)_{2.80}$	$0.24 \times KAl_2(Si_3Al)O_{10}(OH)_2$
SMH 24	$(K_{0.21}Na_{0.03}Ca_{0.11})(Al_{1.63}Fe_{0.19}Mg_{0.32})$ $-(Si_{3.45}Al_{0.55})O_{9.77}(OH)_{2.46}$	$0.78 \times (Na_{0.03}Ca_{0.14})(Al_{1.55}Fe_{0.24}Mg_{0.41})$ $-(Si_{3.57}Al_{0.43})O_{9.71}(OH)_{2.58}$	$0.22 \times KAl_2(Si_3Al)O_{10}(OH)_2$
18987	$(K_{0.35}Na_{0.05}Ca_{0.09})(Al_{1.62}Fe_{0.19}Mg_{0.24})$ $-(Si_{3.65}Al_{0.45})O_{9.68}(OH)_{2.04}$	$0.77 \times (Na_{0.11}Ca_{0.12})(Al_{1.51}Fe_{0.25}Mg_{0.31})$ $-(Si_{3.76}Al_{0.24})O_{9.97}(OH)_{2.06}$	$0.23 \times KAl_2(Si_3Al)O_{10}(OH)_2$
SME 22	$(K_{0.18}Na_{0.03}Ca_{0.09})(Al_{1.52}Fe_{0.22}Mg_{0.41})$ $-(Si_{3.60}Al_{0.40})O_{9.99}(OH)_{2.01}$	$0.81 \times (Na_{0.04}Ca_{0.11})(Al_{1.40}Fe_{0.27}Mg_{0.50})$ $-(Si_{3.74}Al_{0.26})O_{9.965}(OH)_{2.03}$	$0.19 \times KAl_2(Si_3Al)O_{10}(OH)_2$
P 4608	$(K_{0.22}Na_{0.10}Ca_{0.08})(Al_{1.55}Fe_{0.23}Mg_{0.32})$ $-(Si_{3.71}Al_{0.29})O_{9.875}(OH)_{2.25}$	$0.78 \times (Na_{0.13}Ca_{0.12})(Al_{1.43}Fe_{0.30}Mg_{0.38})$ $-(Si_{3.91}Al_{0.09})O_{9.84}(OH)_{2.32}$	$0.22 \times KAl_2(Si_3Al)O_{10}(OH)_2$
SMC 44	$(K_{0.19}Na_{0.03}Ca_{0.11})(Al_{1.56}Fe_{0.16}Mg_{0.40})$ $-(Si_{1.93}Al_{0.36})O_{9.21}(OH)_{2.48}$	$0.82 \times (Na_{0.03}Ca_{0.12})(Al_{1.46}Fe_{0.20}Mg_{0.40})$ $-(Si_{3.75}Al_{0.25})O_{9.145}(OH)_{2.71}$	$0.18 \times KAl_2(Si_3Al)O_{10}(OH)_2$
SMH 18	$(K_{0.31}Na_{0.03}Ca_{0.08})(Al_{1.53}Fe_{0.24}Mg_{0.16})$ $-(Si_{3.87}Al_{0.31})O_{9.395}(OH)_{2.21}$	$0.79 \times (Na_{0.04}Ca_{0.10})(Al_{1.33}Fe_{0.30}Mg_{0.30})$ $-(Si_{3.87}Al_{0.13})O_{9.27}(OH)_{2.26}$	$0.21 \times KAl_2(Si_3Al)O_{10}(OH)_2$

the overall layer formula or the swelling-layer formula. Most of the formulae are intermediate between that of montmorillonite and beidellite. The exception, SMH24, had a tetrahedral Al content of 0.43 atoms per half unit cell and was the only sample that showed the definite presence of a beidellitic lattice by the Li-test (Greene-Kelly, 1953).

Summary of the clay-mineral data

The 0.2–0.04 μ fraction from each of these samples was predominantly a dioctahedral randomly interstratified mixture of montmorillonite- and mica-type layers. Based upon peak positions of the basal reflections, the proportion of montmorillonite was about 50 per cent in the lacustrine samples and 65 per cent in the till samples. Fourier transforms gave a value of 55 per cent montmorillonite for both a till and a lacustrine clay. Assigning the K content to muscovite yielded values of about 80 per cent montmorillonite with the till clays being higher than the lacustrine clays. The crystallite size was calculated to be equivalent to 2- to 5-layers thick in the c-dimension, and the Fourier transform confirmed the essentially random nature of the interstratification with a tendency to zonal segregation of mica-type and montmorillonite-type layers. The calculated formula for the swelling layers and the Greene-Kelly test both led to the suggestion that the swelling-layer material was intermediate in composition between beidellite and montmorillonite.

Most of the reports on the clay mineralogy of the soils of the Prairie Provinces have indicated montmorillonite as the dominant mineral associated with varying amounts of mica. Presumably, these interpretations have been based largely upon a strong 18 Å-basal spacing from a glycerol-saturated specimen. As has been pointed out (MacEwan, 1958) a high proportion of mixed layering gives an 18 Å reflection and, since the higher order reflections are weak, a determination of the degree of mixing on the basis of peak positions is difficult. The high K_2O contents of fine clay fractions showing no 10 Å peak (Warder and Dion, 1952) can be accounted for by interstratification of mica- and montmorillonite-type layers. Interstratification of this kind is therefore a possibility in other materials (Rice, Forman and Patry, 1959; Erhlich, Rice and Ellis, 1955; Pawluk, 1961; and St. Arnaud and Mortland, 1963). Two lacustrine clays, Regina and Three Hills (Rice, Forman and Patry, 1959) and the Cooking Lake (a sample similar to that reported by Pawluk, 1961), which had been previously described as predominantly montmorillonite, are here described on the basis of K_2O content as an interstratified montmorillonite–mica with 20 per cent mica layers.

Origin of the interstratified mineral

It is generally believed (St. Arnaud and Mortland, 1963) that the major source of the soils in question is the thick Upper Cretaceous sediments that cover the west-central two-thirds of the three Prairie Provinces. The Upper Cretaceous is comprised mainly of shales and sandstones with some bentonite beds and lenses. Byrne and Farvolden (1959) indicated that the Bearpaw

shales of southern Alberta were composed of montmorillonite, mica and chlorite, generally in that order of abundance and that there were no marked differences in composition throughout the section. Forman and Rice (1959) reported that the Bearpaw shale of southern Saskatchewan consisted mainly of montmorillonite and mica in the fine clay fraction. Recently, Lerbekmo (1963) showed that mica, montmorillonite and mixed-layer montmorillonite–mica were abundant in the nonmarine shale of the Belly River formation in Alberta. He also showed that the overlying Bearpaw shale had a considerable proportion of mixed-layer montmorillonite–mica. He considered that the mica was detrital and that the montmorillonite and mixed-layer montmorillonite–mica were derived from diagenetic alteration of volcanic ash. There are enough similarities between these analyses of Upper Cretaceous sediments to strengthen the interpretation that the clay minerals in the tills are inherited without much alteration.

Although the seven samples were collected from locations separated by as far as 850 miles, the uniformity of their mineralogical and chemical character was a striking feature. This is further evidence that the interstratified mineral had been formed by diagenesis in the Upper Cretaceous rocks and thence had been inherited by the soils.

In the present investigation, there were no large differences between the lacustrine and glacial-till samples but there were two trends. The proportion of montmorillonite layers in the interstratified mineral tended to be greater in the till samples than in the lacustrine samples. Also, on the basis of the calculated formulae, the expanding layers of the lacustrine samples tended to be nearer to the montmorillonite rather than to the beidellite composition. This is likely related to fixation of K during or after the deposition of the lacustrine materials, whereby the expanding layers with a beidellite-type composition would be pre-empted, leaving montmorillonite-type layers as suggested by Weaver (1959).

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