X-RAY POWDER DIFFRACTION IDENTIFICATION
OF ILLITIC MATERIALS

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Abstract—The 10-Å clay components of sedimentary rocks ("illites") are commonly mixtures of 100% nonexpandable illite and an ordered illite/smectite mixed-layer mineral. If the proportion of the illite/ smectite in a mixture is sufficient to produce a measurable reflection between $33–35°2\theta$ (CuKα radiation) that is noncoincident with an illite reflection, the ratio of component layers and type of interstratification for the mixed-layer mineral can be determined. The identification technique developed in this study rests upon the following experimental findings for ordered illite/smectites of diagenetic origin: (1) the thickness of the illite layer in illite/smectites is 9.97 Å; (2) the thickness of smectite-ethylene glycol complex ranges from 16.7 to 16.9 Å; (3) illite/smectites form a continuous sequence of interstratification types—random, random/IS, IS, IS/ISII, ISII—and each type is related to a specific range of expandability.

The new technique broadens the computer simulation method developed by R. C. Reynolds and J. Hower to include those sedimentary materials which are dominated by the presence of discrete illite, are low in illite/smectite, and, as such, have been described previously only by an "illite crystallinity index."

Key Words—Diagenesis, Identification, Illite, Illite/smectite, Interstratification.

INTRODUCTION

The name "illite" was introduced by Grim *et al.* in 1937 "as a general term for the clay mineral constituent of argillaceous sediments belonging to the mica group." Since that time, the term has been widely used in this broad sense to designate material giving a ~10 Å reflection on X-ray powder diffraction patterns of clay-size fractions of sediments and soils. Hence, "illite" is now used as a group name, covering a range of chemical compositions and structural characteristics. The structural variability of illite was quantified by Weaver (1960) in terms of "illite crystallinity" which was based on the "sharpness" of the 10-Å peak. The method has been used extensively, especially in Western Europe, for studying deep diagenesis and low-grade metamorphism (see, e.g., Kisch, 1983). It has never been established precisely what phenomenon gives rise to different illite "crystallinities."

The nature of illite was approached from the structural standpoint by Gaudette *et al.* (1966). Among five well-known illite reference samples, they found one pure and nonexpanding. The other four contained mixed-layer expandable material: illite commonly appeared to be a physical mixture of minerals. This interpretation was confirmed later by Heller-Kallai and Kalman (1972) and Środoń (1979). From an investigation of a range of mixed-layer illite/smectites and illites, Hower and Mowatt (1966) concluded that "illites represent one mineralogical end member of the

montmorillonite-mixed-layer-illite group and remain distinct from true micas until the beginning of metamorphism." This phrase may serve as a definition of the mineral illite, in contrast to the use of the term "illite" in the broad sense, but it does not solve the problem of illite identification. In the following considerations, "illite" refers to the 100% nonexpanding mineral, and "illitic material" is used as a group name implying the possible presence of some expandable layers.

Reynolds and Hower (1970) published a computer method for investigating mixed-layer illite/smectites and defined three types of interstratification of component layers: random, IS-ordered, and ISII-ordered, where I = illite layer, S = smectite layer. Random interstratification was defined by lack of interaction between adjacent layers: the probability of finding an illite layer following a smectite layer in a specified but arbitrary direction simply equals the probability of finding an illite layer in the sequence

$$pS \cdot I = pI.$$

IS-ordering was defined as a nearest-neighbor interaction; the probability of illite following smectite is higher than in the random case. Maximum IS-ordering implies that all smectite layers are followed by illite layers:

$$pS \cdot I = 1 \quad \text{(providing } pI \geq 0.5\text{)}.$$

It should be stressed that, according to the definition of Reynolds and Hower (1970), maximum ordering does not require a 1:1 composition. A mineral with such a composition (pI = pS = 0.5) is only a special

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Table 1. X-ray powder diffraction data for ordered illite/smectites and other illitic materials.

Sample[1]	Reflections from glycolated preparations (°2θ, CuKα radiation)							$\Delta_{002-001}$ (°2θ)	BB1[2] (°2θ)	BB2[2] (°2θ)	Reflections from air-dry preparations (°2θ)		Ir[2]	%S[3]	Type of illitic material[3]
1	5.99	9.84	16.39	26.56	32.91	43.81	47.38	6.55						49	R/IS
2	6.00	9.79	16.42	26.52	32.87	43.80	47.27	6.63						48	R/IS
3	6.64	9.98	16.72	26.88		44.41	48.01	6.74						48	IS
4	6.44	9.73	16.51	26.57	33.38	44.00	47.20	6.78						44	R/IS
5	6.58	9.66	16.67	26.63	33.49	44.14	47.07	7.01						39	R/IS
6	6.64	9.63	16.69	26.62	33.49	44.09	47.16	7.06					7.93	37	IS
7	6.58	9.59	16.66	26.58	33.42	44.07	47.08	7.07					8.48	38	R/IS
8	6.64	9.51	16.69	26.58	33.39	44.02	46.99	7.18					8.50	36	IS
9	6.08	9.50	16.74	26.73	33.33	44.36	sh	7.24						38	R/IS
10	6.89	9.57	16.82	26.67	33.62	44.26	46.93	7.25						32	IS
11	6.71	9.46	16.80	26.65	33.59	44.36	46.90	7.34						32	IS
12	6.75	9.46	16.92	26.75	33.86	44.57	sh	7.46	5.0	4.8				29	IS
13	6.69	9.40	16.86	26.62	33.58	44.29	sh	7.46					8.18	28	IS
14	6.91	9.34	17.00	26.75	33.88	44.65	sh	7.66	5.0	4.0				25	IS
15	7.14	9.28	17.07	26.68	33.98	44.64	46.24	7.79	4.6	4.8	8.16	18.05	3.78	20	IS/ISII
16	7.13	9.26	17.09	26.73	34.09	44.73	sh	7.83	4.8	5.0				20	IS/ISII
17	7.61	9.26	17.17	26.69	34.56	44.66	46.29	7.91	3.8	3.8	8.20	17.60	4.48	17	ISII
18	7.28	8.94	17.27	26.77	34.30	45.02	—	8.33	4.6	5.0				15	IS/ISII
19	7.51	8.98	17.32	26.78	34.74	45.08	—	8.34	4.6	4.6				13	IS/ISII
20	sh	9.06	17.44	26.79	34.93	45.18	—	8.38	3.6	3.8	8.55	17.86	2.38	9	ISII
21	sh	9.05	17.47	26.81	35.08	45.11	—	8.42	3.3	2.8	8.49	17.87	2.32	9	ISII
22	7.30	8.96	17.52	26.79	33.85	45.47	—	8.56	4.9	5.2	8.28		2.61	26	IS + I
23	—	8.94	17.52	26.81	35.1	45.41	—	8.58	3.6	4.0	8.47		2.02	7	ISII
24	—	8.96	17.56	26.78	34.9	45.26	—	8.60	3.2	2.6	8.63	17.86	1.52	6	ISII
25	sh	8.86	17.56	26.78	34.9	45.26	—	8.68	3.6	4.0	8.47		2.46	9	ISII + I
26	—	8.84	17.74	26.87	35.0	45.59	—	8.90	3.6	2.8	8.59	17.94	1.83	8	I + ISII
27	—	8.85	17.76	26.85	—	45.53	—	8.91	3.3	3.0	8.72	17.83	1.58	<15	I + ISII
28	—	8.79	17.69	26.66	c	45.20	—	8.90	3.2	c	8.79	17.71	1.56	<15	I + ISII
29	sh	8.83	17.76	26.68	34.84	45.44	—	8.93	4.7	3.5	8.79	17.77	1.52	9	I + ISII
30	sh	8.81	17.67	26.76	34.0	45.47	—	8.86	4.5	5.0	8.50	17.71	1.50	23	I + IS
31	—	8.84	17.71	26.74	—	45.41	—	8.87	3.2		8.61	17.81	1.47	<15	I + ISII
32	—	8.76	17.68	26.69	—	45.34	—	8.92	3.3	2.8	8.67	17.78	1.40	<15	I + ISII
33	—	8.84	17.75	26.83	—	45.50	—	8.91	2.6	2.6	8.73	17.74	1.37	<15	I + ISII
34	—	8.83	17.72	26.74	—	45.32	—	8.89	3.4	3.0	8.63	17.79	1.30	<15	I + ISII
35	—	8.80	17.76	26.76	diff	45.49	—	8.96	5.0	5.0	8.75	17.86	1.29	>15	I + IS
36	—	8.85	17.77	26.75	c	45.47	—	8.92	3.0	c	8.81	17.79	1.25	<15	I + ISII
37	—	8.82	17.67	26.75	—	45.35	—	8.85	3.0	3.0	8.64	17.81	1.23	<15	I + ISII
38	—	8.84	17.79	26.71	c	45.50	—	9.00	2.3	c	8.80	17.80	1.20	<15	I + ISII
39	—	8.83	17.73	26.79	c	45.45	—	8.90	3.7	c	8.67	17.85	1.18	<15	I + ISII
40	—	8.85	17.76	26.77	c	45.40	—	8.91	2.2	c	8.84	17.75	1.00	0	I
41	—	8.74	17.67	26.66	—	45.27	—	8.93	3.1	1.5	8.74	17.67	0.96	0	I

sh = shoulder; diff = diffuse; c = coincidence with prism refection (resulting from poor orientation).

[1] Samples 1–25 are arranged according to increasing $\Delta_{002-001}$; samples 26–41 according to decreasing Ir. 1, 4, 5, 10, 11 = Carboniferous bentonites, Upper Namurian A, Poland (collected by the author); 2, 6, 7, 8, 13, 15 = Carboniferous bentonites, Westphalian A/B, Poland (collected by the author); 3 = Arkansas rectorite, hydrothermal (collected by D. D. Eberl); 9, 12, 14, 16, 18, 19, 23, 24 = Silurian bentonites, Welsh Borderlands (collected by D. Morgan); 17 = Zempleni clay, Hungary; 20 = Ms-1-75 sample, Fredrickstown, Missouri (R. E. Grim collection); 21 = Silurian bentonite, Kalkberg Fm., New York State (R. E. Grim collection); 22 = H-36 sample, Morris, Illinois (Ward's Natural Science Establishment); 25 = H-41 bentonite, Tazewell, Virginia (Ward's Natural Science Establishment); 26 = Shale break in dolomite, Silurian, Marblehead, Wisconsin (R. E. Grim collection); 27 = Shale break in Madison limestone, Mississippian, Wyoming (R. E. Grim collection); 28 = Illitized tonstein, Upper Carboniferous, Czechoslovakia (collected by J. Králik); 29 = "Fithian illite," Pennsylvanian under-clay, Illinois (Ward's Natural Science Establishment); 30 = G-477 clay from cavity in dolomite, Oregon, Illinois (R. E. Grim collection); 31, 32, 34, 37 = Authigenic sandstone cements, Triassic, Poland (collected by A. Rochiewicz); 33 = Strasbourg bentonite, Virginia (Ward's Natural Science Establishment); 35 = Shale break in carbonates, Missouri (collected by the author); 36 = Shale streak in limestone, Triassic, Poland (collected by M. Muszyński); 38 = Beavers Bend shale, Silurian, Oklahoma (collected by W. D. Keller); 39 = A-1 glauconite (collected by J. Hower); 40 = Ground muscovite, India; 41 = Sericite clay, hydrothermal, Kaube mine, Nara prefecture, Japan (R. E. Grim collection).

[2] These indices are explained in the text.

[3] The last two columns identify the illitic materials by methods developed in this study.

case of maximum IS ordering, a case in which there are no excess illite layers. This exceptional type of ordering should be called regular, according to the definitions of AIPEA Nomenclature Committee (Bailey *et al.,* 1982). Such a mineral has, of course, been found in nature, and is called rectorite (Bailey *et al.,* 1982).

Maximum ISII type of ordering was defined by Reynolds and Hower (1970) as the probability of a smectite layer being followed by three illite layers equaling 1:

$$pS \cdot III = 1.$$

For this arrangement to be possible, $pI \geq 0.75$. If $pI = 0.75$, no excess illite layers exist in the mineral, and the mineral is a regular interstratification. Such a mineral with nearly perfect ISII interstratification has been named tarasovite (Lazarenko and Korolev, 1970; Brindley and Suzuki, 1983).

A complete statistical treatment of mixed-layering can be found in Reynolds (1980). In an alternative and also widely used mathematical approach to mixed-layering (Kakinoki and Komura, 1965), the above-listed types of interstratification are also recognized and called Reichweite 0, 1, and 3, respectively.

In 1980, Šrodoń published an improvement of the Reynolds and Hower method, taking into account the variable thickness of ethylene glycol-smectite complex. The smectitic end of the mixed-layered series was well-documented in the paper, but the illitic end was not because of a lack of sample material. The assumption of a 10-Å-thick illite layer was made according to Reynolds and Hower (1970), without verification. The objectives of the present paper are to investigate the illitic end of the mixed-layered series using a broad range of sample material, and then to apply these data for identification of other illitic materials.

X-RAY DIFFRACTION DATA FOR ILLITIC MATERIALS

The samples analyzed in this study come from several localities in Europe and the United States and represent different ages and sedimentary formations (Table 1). Almost all are of diagenetic origin. The analytical techniques used are identical to those described by Šrodoń (1980). All samples were Na-exchanged and examined by X-ray powder diffraction (XRD) at room humidity (40–50% RH). All peak positions were obtained by step-scanning at $0.01°2\theta$ intervals using CuKα radiation.

Computer-simulated, one-dimensional XRD patterns of highly illitic mixed-layer illite/smectites were inspected in the range of $50–3°2\theta$ in search for the parameters most sensitive to the component layer ratio. In addition to XRD peak positions for ethylene glycol-solvated preparations, the following measurements also were found to be useful:

(1) $\Delta_{002-001}$ = the angular distance between the 002 and 001 reflections from glycolated preparations (the 1 *M* mica indices are used here and hereafter to designate the reflections of illitic material approximately coinciding with the reflections of mica; the noncoincident reflections are designated by their angular values);

(2) Positions of the 001 and 002 reflections from air-dry preparations;

(3) Ir = the intensity ratios (peak heights ratios) of the 001 and 003 reflections from the air-dry and the glycolated samples:

$$Ir = \frac{001/003 \text{ air-dry}}{001/003 \text{ ethylene glycol}}.$$

(4) BB1 = the joint breadth of 001 illite and adjacent illite/smectite reflections, measured in $°2\theta$ from where the tails of the peak join the X-ray background;

(5) BB2 = the joint breadth of 004 illite and adjacent illite/smectite reflections, measured like BB1.

The last two parameters were discussed by Šrodoń (1979). Table 1 presents the above data for most of the samples used in this study. The last two columns identify the illitic materials by methods developed in this study.

THICKNESS OF THE ILLITE LAYER AND THE REFINED TECHNIQUE FOR IDENTIFYING ORDERED ILLITE/SMECTITES

The samples in Table 1 are arranged into two groups: illite/smectites or illitic materials dominated by illite/smectite (1–25), and illites or illitic materials dominated by illite (26–41). From ethylene glycol-treated preparations, the first group is characterized by a non-integral series of basal reflections and a $\Delta_{002-001}$ value of $8.85–9.00°2\theta$; the second group is characterized by an integral series of basal reflections and a $\Delta_{002-001}$ value smaller than $8.7°2\theta$.

Samples in the second group (when the 2θ values in Table 1 are converted into d-spacings) indicate that the layer spacing of discrete illite is not strictly 10 Å, but varies from 9.96 to 10.03 Å. The most precise value can be obtained from the 003 and 005 reflections because the 001 and, to a lesser extent the 002, are displaced slightly toward low angles due to the effect of small crystallite size and the Lorentz polarization factor (Reynolds, 1968). The question then arises, is the illite layer thickness in the mixed-layer minerals also as variable?

This question can be answered by plotting the data from Table 1 into Figure 8 of Šrodoń (1980). Figure 1 shows the results. Most samples from the illite-dominated group plot within theoretical lines representing illite, computed assuming both infinite thickness for the crystallites, and 7–14 layers. The range of layer thickness for discrete illite is clearly shown. A few points
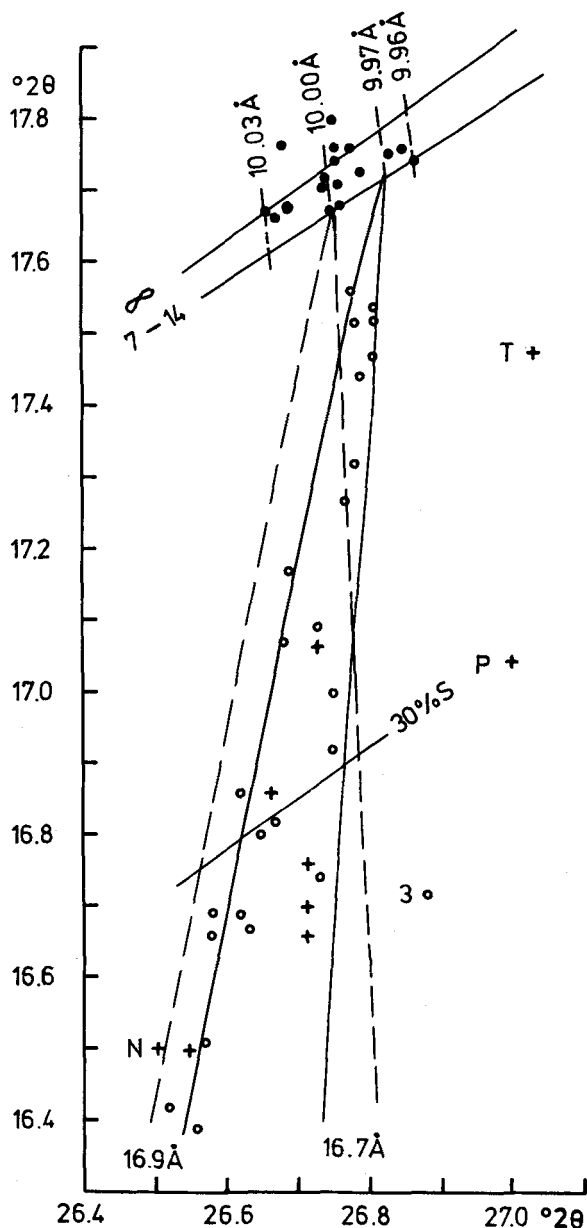
Figure 1. Plot of 002 vs. 003 reflections of illitic materials. Solid circles = samples dominated by illite; open circles and crosses = samples dominated by illite/smectite (crosses represent data from the literature and are not listed in Table 1). Pairs of solid and dashed lines = theoretical plots of ordered illite/smectites with various combinations of component layer thickness. Solid lines marked with ∞ and 7–14 = theoretical plots of illite with an infinite crystallite thickness and 7–14 layers in crystallites. Hydrothermal samples: T is from Lazarenko and Korolev (1970); P is from Gallego and Pérez (1965); 3 is Arkansas rectorite (Table 1, no. 3); N is from Shimoda (1972).

Figure 2. Plot for determining smectite:illite ratio from 002 and 003 reflections in ordered illite/smectites of diagenetic origin, based on the computer data obtained with the following assumptions: 7–14 layers/crystallite, illite layer = 9.97 Å thick, smectite-ethylene glycol layer = 16.7–16.9 Å thick. Solid lines = IS type of ordering; dashed lines = ISII type ordering.

plotting randomly above the infinite thick illite line were found to represent samples contaminated with quartz, in which the quartz 101 reflection interfered

with the 003 of illite and displaced the projection point towards the left side of the figure.

With the exception of samples 22 and 25, all samples from the illite/smectite-dominated group are pure illite/smectites. The criteria for making this distinction are discussed below. In Figure 1, all of these samples plot as a sequence of decreasing expandability that extrapolates to layer thickness smaller than 10 Å for the
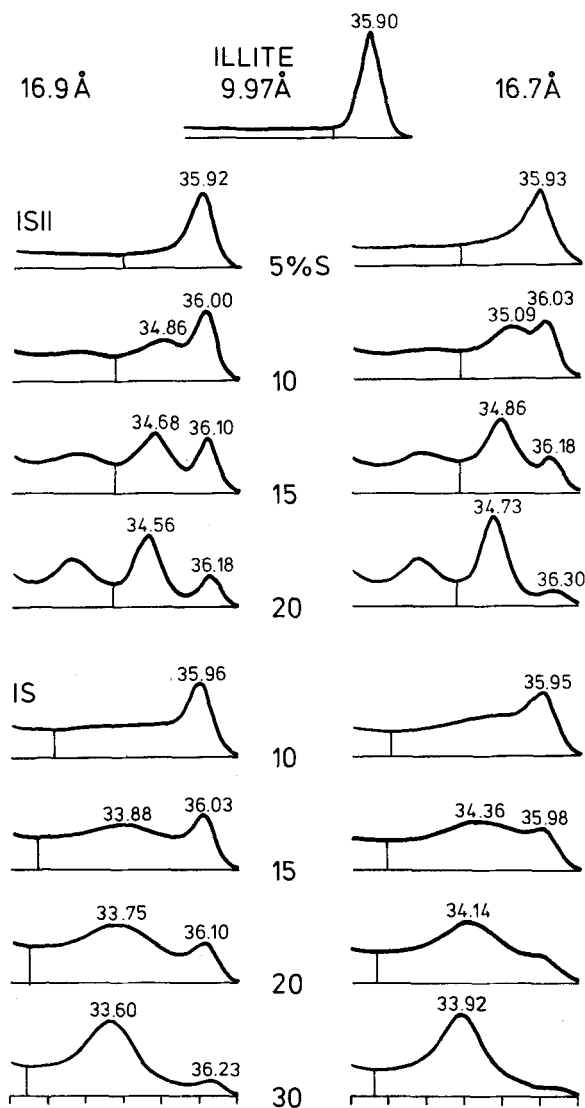
Figure 3. Diffraction profile, 31–37°2θ, highly illitic illite/smectites calculated for 9.97-Å illite layer and various combinations of ordering and thickness for the smectite-ethylene glycol layer. BB2 parameter corresponds to the angular distance between ~37°2θ and the vertical line. The dependence of BB2 on the type of ordering is clearly shown. 10% S IS-ordered minerals and 20% S ISII-ordered minerals are theoretical examples; in nature the type of ordering changes around 15% S (cf. Figures 4 and 9).

Figure 4. Examples of X-ray powder diffraction patterns of glycolated, ordered illite/smectites. Sample numbers refer to Table 1, where the peak positions are listed. Type of ordering, percentage of smectite, and manner of measuring BB1 and BB2 parameters are shown. Samples 15 and 16 have identical expandabilities, but different thicknesses of the smectite-ethylene glycol complex. The difference manifests itself most strongly in the relative intensities of peaks in the 42–48°2θ region.

illite layers. It was found by trial and error that all data points for diagenetic, ordered illite/smectites with 70% or more illite layers can be matched by a combination of the layer thicknesses of 9.97 Å for the illite layer and from 16.7 to 16.9 Å for the ethylene glycol-smectite layer (Figure 1).

Samples T, P, 3, and N are of hydrothermal origin and are plotted in Figure 1 to show that the above findings about the layer thicknesses are valid only for
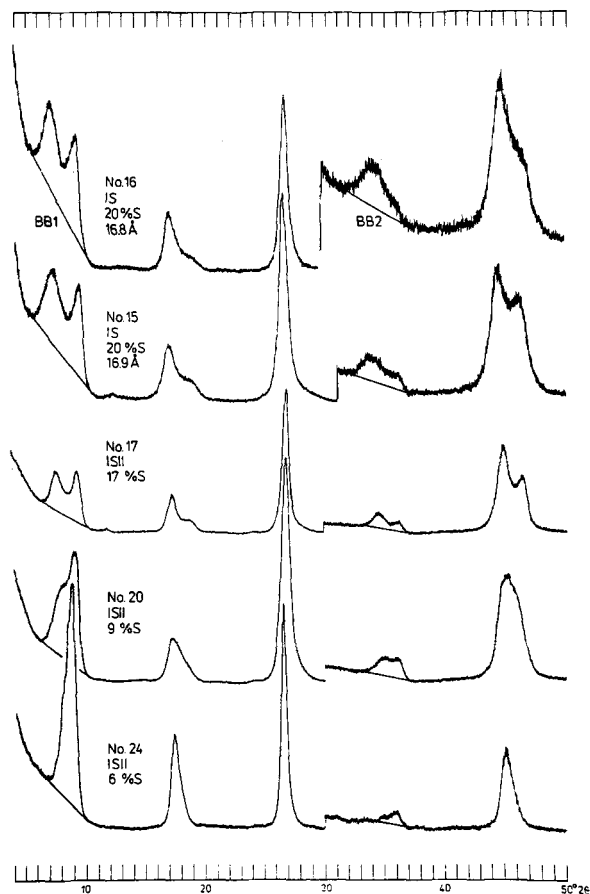
the materials of diagenetic origin. Among the hydrothermal samples the variation is much broader, and it remains to be shown whether the variation is due to changes in illite or ethylene glycol-smectite layer thickness or both.

It is fortunate for identification that the range of thicknesses of smectite-ethylene glycol complexes in diagenetic, ordered illite/smectites is so narrow and that the illite layer thickness is stable. By contrast, in smectite-dominated, diagenetic, mixed-layer minerals, the thickness of the smectite-ethylene glycol complex ranges from 17.2 to 16.6 Å (Środoń, 1980).

A plot based on Figure 1 and calibrated for the percentage of smectite layers with the computer-calculated data (Figure 2) serves for precise measurement of the illite/smectite ratio in highly illitic mixed-layer min-
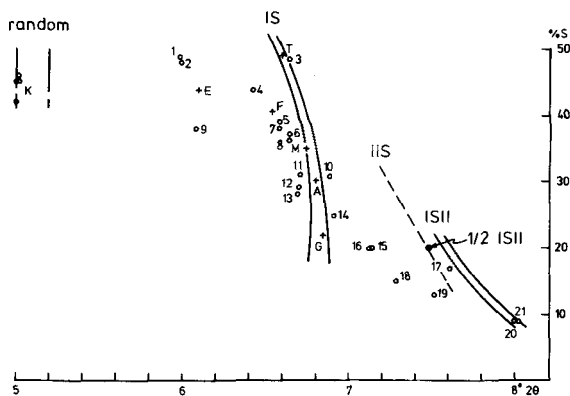
Figure 5. Plot for identifying the type of ordering in illite/smectites. Solid lines = theoretical plots of random, IS, and ISII-ordered types of interstratification, calculated assuming a 9.97-Å illite layer and a 16.9-Å (left line) and 16.7-Å (right line) smectite-ethylene glycol layer. Dashed line = IIS type of ordering (9.97 Å/16.7 Å); solid circle is an example of half-ordered ISII mineral (9.97 Å/16.9 Å). Open circles represent samples listed in Table 1; crosses represent additional samples: T = Tulameen bentonite from Pevear *et al.* (1980); E, F, G, A (A2), M (Two Medicine) = samples from Reynolds and Hower (1970); K = Ordovician bentonites from Kinnekulle, Sweden (author's data).
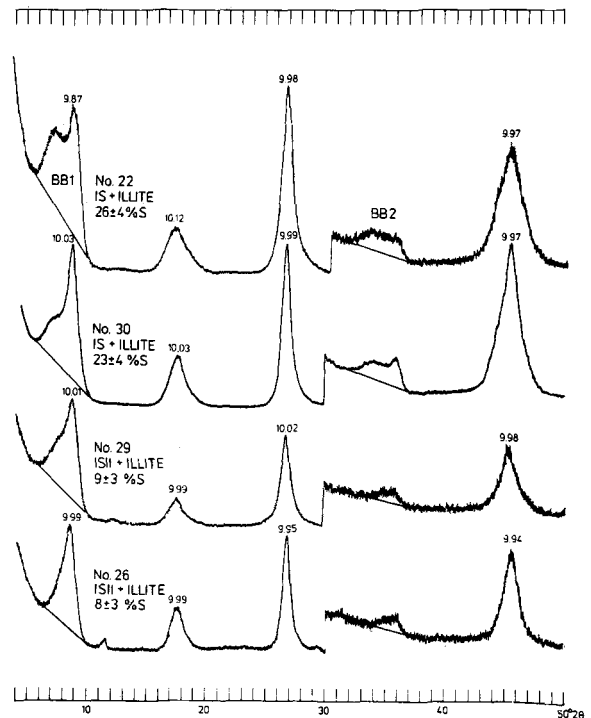

Figure 6. Examples of X-ray powder diffraction patterns of glycolated illitic materials with measurable noncoincident illite/smectite reflections. Sample numbers refer to Table 1. d(001) values are in Ångstrom units. The identification based on Figure 7 with BB1 parameters and d-spacings from Table 1 is given for each sample.

erals. Figure 2 has separate sets of lines for IS and ISII types of ordering. The IIS type of ordering suggested by Drits and Sakharov (1976) and Nadeau and Reynolds (1981) is not introduced for reasons discussed below. To make a determination from Figure 2, one must plot the experimental values of the 002 and 003 reflections and read the percentage of smectite by interpolation between the appropriate lines.

ISII interstratification is characteristic of the extremely illitic minerals (Reynolds and Hower, 1970). At 20% smectite, the lines for the IS and ISII types are very close (Figure 2). At lower values, it must be decided which line is to be used. The distinction between the types of interstratification can be made by measuring the BB1 or BB2 parameter (Środoń, 1979) or the position of the low-angle reflection between 6 and $8°2\theta$. Minerals with prevailing IS characteristics have BB1 and BB2 values larger than $4°2\theta$, and the angle of the reflection is smaller than $7.5°2\theta$. The opposite is true for the ISII type of interstratification. These distinctions were made on the basis of both experimental data (Table 1) and, for BB2, computer-simulated XRD patterns (Figure 3). XRD patterns of the ordered illite/smectites from bentonites, covering the whole range of variation, are given in Figure 4. Figures 3 and 4 also show the method of measuring BB1 and BB2.

More precise identification of the type of interstratification can be obtained from Figure 5 which exploits the reflection at $6-8°2\theta$. This reflection is much more sensitive to the type of interstratification than are the reflections used for measuring the percentage of smec-

tite layers (cf. Figures 2 and 5), and depends little on the thickness of the smectite-ethylene glycol complex (Figure 5). Data from Table 1, unpublished data on random interstratifications, and data from other authors plotted as points in Figure 5 show that in nature distinct populations of different kinds of interstratification do not exist, but rather a continuous series from randomly interstratified to maximum ISII-ordered minerals. The range of possible types of interstratification varies with the component layer ratio. The question arises as to whether in this series there exists an IIS structure between the IS and ISII structures as postulated by Drits and Sakharov (1976).

The criteria for distinguishing between these three structures cannot be based on 001, 002, 003, or 005 reflections because these reflections are insensitive to the type of interstratification. The two sensitive reflections are those at 6–8° and $33-35°2\theta$. To solve the problem, samples 17 and 19, which plot close to the IIS line (Figure 5), were studied in detail by the computer simulation.

It is known from Figure 1 that the smectite-ethylene glycol complex thickness of sample 17 is slightly larger than 16.9 Å, which agrees with the position of this sample in Figure 5 relative to the ISII lines, and dis-

Table 2. Examples of the complete computer fit of X-ray powder diffraction patterns of glycolated illite/smectites.[1]

	Sample parameters			Peak positions (°2θ, CuKα radiation)						
Sample 15	20% S, IS/ISII	16.9 Å	7.14	9.28	17.07	26.68	33.98	44.64	46.24
T	20% S, ISII	16.9/9.97 Å	7.58	9.20	17.10	26.69	34.56	44.64	46.40
T	20% S, IS	16.9/9.97Å	6.76	9.17	17.07	26.68	33.75	44.67	46.21
Sample 17	17% S, ISII	16.9 Å	7.61	9.26	17.17	26.69	34.56	44.66	46.29
T	20% S, ISII	16.9/9.97 Å	7.58	9.20	17.10	26.69	34.56	44.64	46.40
T	15% S, ISII	16.9/9.97 Å	7.71	9.09	17.22	26.72	34.68	44.81	46.21
Sample 20	9% S, ISII	16.8 Å	sh	9.06	17.44	26.79	34.93	45.18	—
T	10% S, ISII	16.9/9.97 Å	7.91	8.94	17.39	26.75	34.86	45.10	—
T	10% S, ISII	16.7/9.97 Å	7.97	8.95	17.44	26.83	35.09	45.19	—

sh = shoulder.

[1] Sample numbers and identification (the percentage of smectite, the type of ordering, the thickness of smectite-ethylene glycol complex) are from Table 1 and Figure 1. The experimental values, except of the reflection at about 9°2θ (see text), fit within the range given by the theoretical data (T).

agrees with the position relative to the IIS line. Thus, sample 17 is an almost perfectly ordered ISII structure (see also Table 2).

The thickness of smectite-ethylene glycol complex of sample 19 is 16.75 Å (Figure 1). If the sample was an IIS structure, it should plot in Figure 5 just to the left of the IIS line, but that is not the case. The alternative explanation is that sample 19 is an intermediate IS/ISII structure (incompletely ordered ISII structure). Figure 5 shows that a one-half-ordered ISII structure plots exactly on the IIS line. Sample 19 is therefore an approximately one-half-ordered ISII structure.

Samples A-2, F, and G of Reynolds and Hower (1970), interpreted as IIS structures by Drits and Sakharov (1976), plot in Figure 5 in the field of perfect or almost perfect IS ordering, as identified by the original authors. The value of the 33–35°2θ reflection was given by Reynolds and Hower (1970) only for sample A-2 and confirms the interpretation of perfect IS ordering. Thus, from the above data the IIS type of ordering does not seem to exist in diagenetic illite/smectites. A similar conclusion was reached by Schultz (1982) in his study of clays from the Montana Disturbed Belt. In the compositional range 50–40%, all varieties of interstratification were encountered from completely random to maximum IS-ordered (Figure 5). Partially ordered IS structures were found down to about 30% S, and maximum-ordered down to 20% S. Between 20 and 12% S partially ordered ISII structures were encountered. Among the maximum-ordered ISII samples known to the author, the most expandable is 17% S and the most illitic is 6% S.

Figure 5 can be used to check the identification of the type of interstratification based on BB1 or BB2. For the intermediate IS/ISII structures, interpolation between the appropriate lines in Figure 2 can be applied to increase the precision of the determination.

Table 2 presents a verification of the described identification technique by a complete computer fit with

the XRD patterns of three samples for the 3–50°2θ range. It appears that all the reflections can be matched satisfactorily, except for 001, which is always displaced in the simulated patterns toward lower angles. This discrepancy is interpreted as the effect of small particle size, which is most strongly manifested in 7–10°2θ region (Reynolds and Hower, 1970). The simulations were made assuming an even distribution of crystallites from 7 to 14 layers thick. The peak position is controlled by the number of layers in the thickest crystallites (Środoń, unpublished data). It is concluded the highly illitic mixed-layer minerals of diagenetic origin have their thickest crystallites larger than 14 layers. Nevertheless, the 7–14-layers model is satisfactory from the identification standpoint (coarser crystallites are very costly to simulate) because for such large crystallites the displacements of higher angle peaks are negligible. The 1–8-layer model, useful when dealing with the smectite-dominated mixed-layer minerals (Środoń, 1980, 1981), is not valid for the illitic ones. Among the data presented in Table 2, the most convincing is the good fit of the reflection at 33–35°2θ because this reflection is exeptionally sensitive to the type of interstratification and to the thickness of the smectite-ethylene glycol complex (Figure 3).

Peak intensity data may also be used to verify the identification procedure. Computer simulation showed that the relative intensities of the two peaks in the 42–48°2θ range depend strongly on the smectite-ethylene glycol complex thickness. The experimental materials, identified according to Figure 2, fit the theoretical model (Figure 4, samples 15 and 16).

The improved identification technique presented in this section allows for the precise measurement of the component layer ratio, the type of interstratification, and the thickness of smectite-ethylene glycol complex in ordered illite/smectites. In terms of the component layers ratio alone, the differences in results between the older (Środoń, 1980) and the new technique are neg-
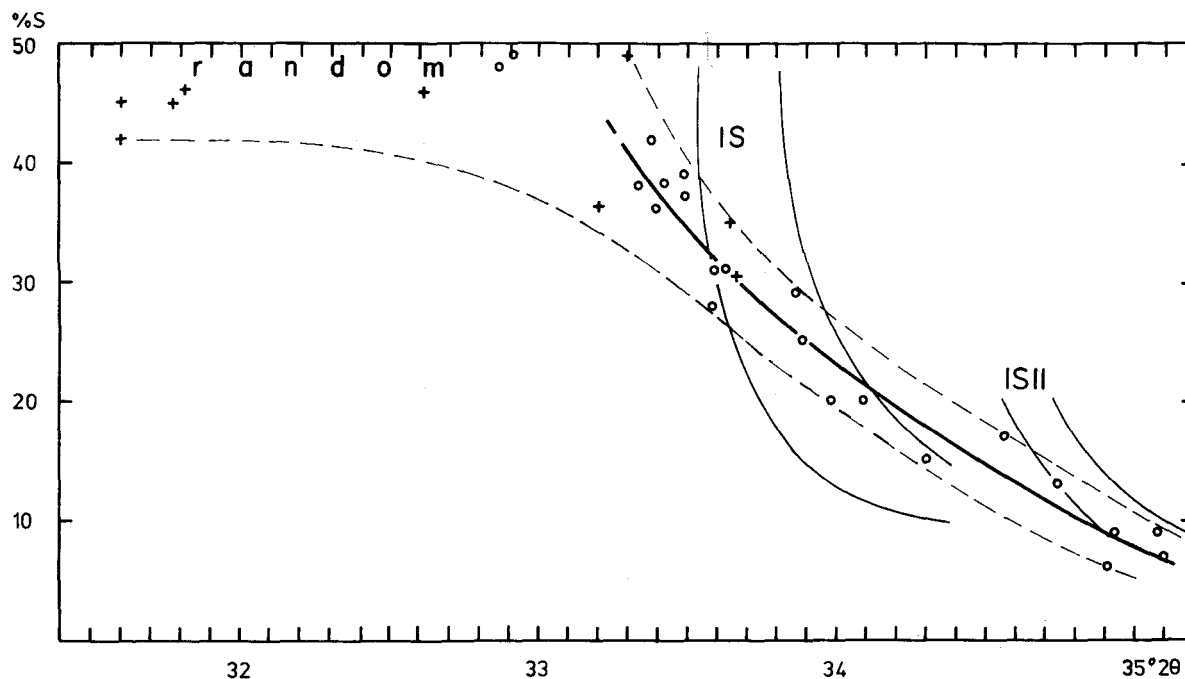
Figure 7. Empirical plot for measuring the smectite:illite ratio from 33–35°2θ reflection in ordered illite/smectites mixed with discrete illite, based on the sample material listed in Table 1 (circles) and on additional unpublished data of the author and from the literature (crosses). The error involved, indicated by the spread of sample points is shown by the dashed lines. Thin solid lines represent theoretical plots of IS and ISII types of ordering, calculated for 9.97 Å/16.9 Å and 9.97 Å/16.7 Å pairs of layer thickness.

ligible for compositions with more than 20% S. The overestimation of the illite layer thickness (10 Å) in the older technique resulted in the underestimation of the smectite-ethylene glycol complex thickness but did not influence significantly the value of the component layers ratio.

IDENTIFYING ILLITE/SMECTITES MIXED WITH DISCRETE ILLITE

Illitic materials with discrete illite/smectite reflections

In soils and epiclastic sedimentary rocks, illite/smectite most commonly occurs mixed with discrete illite, and it is not possible to separate mechanically these two materials. In most instances, the heterogenous nature of the sample is apparent (Figure 6, except sample 22); all coinciding reflections of illite and illite/smectite are dominated by illite, forming a rational series of d(001), but usually two noncoincident mixed-layer reflections can be seen at 6–8° and 33–35°2θ, or at least between 33–35°2θ. Experiments with artificial mixtures have shown that the rational series of illite d(001) persists to at least 30% admixture of an ordered illite/smectite.

In mixtures dominated by illite/smectite, the series of coinciding reflections becomes nonintegral, but an experienced eye can distinguish these reflections from those of pure illite/smectite just from the general appearance of the XRD pattern (compare Figure 4, samples 15 and 16 with Figure 6, sample 22). More objective techniques for making this distinction are discussed at the end of this section.

For both types of samples the noncoincident illite/smectite reflections can be used for identification purposes. The reflection at 6–8°2θ seems less appropriate because it is so close to the intense 001 illite peak that its position is strongly affected. It is commonly reduced to a shoulder on the illite reflection (Figure 6). The reflection at 33–35°2θ avoids these disadvantages, but it is very sensitive to the type of ordering and the thickness of smectite-ethylene glycol complex (Figure 3). Consequently, a theoretical identification plot cannot be constructed. Fortunately, the mixed-layer minerals form a continuous series of types and degrees of ordering (Figure 5) and the variation of the thickness of the expanding layer is relatively small (Figure 1). These factors make possible the identification of diagenetic illite/smectites in mixtures by the use of an empirical working curve based on the data from pure illite/smectites (Table 1). Figure 7 shows the curve and the error involved in this measurement.

The identification can be checked with BB1 and BB2 values (*vide supra*). These values are not affected by the presence of discrete illite because in both, the illite
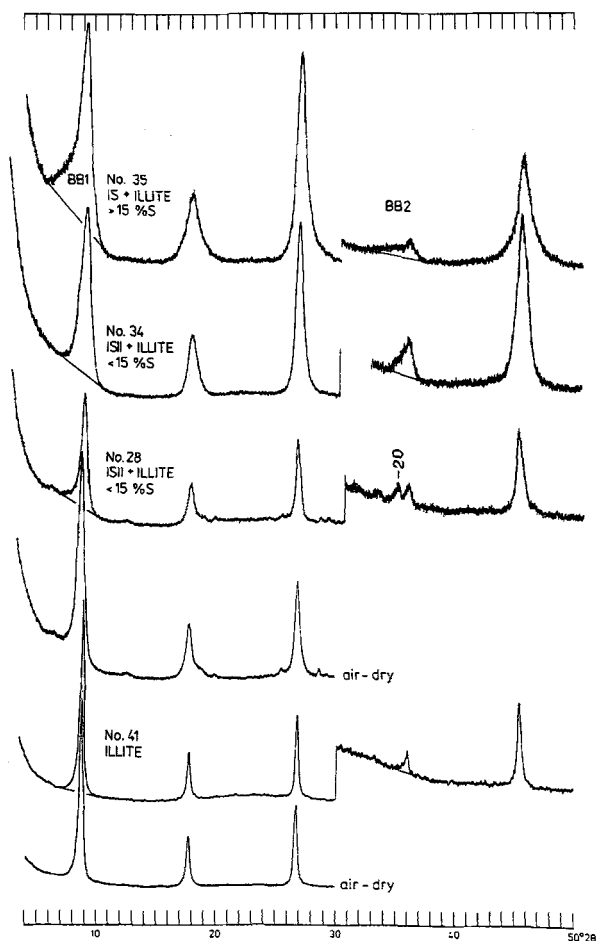
Figure 8. Examples of X-ray powder diffraction patterns of pure illite and illitic materials lacking noncoincident illite/smectite reflections. Sample numbers refer to Table 1. The identification based on BB1, BB2, and Ir parameters is given for each sample. For samples 28 and 41, air-dry patterns are added to the patterns of the glycolated preparations to show 001 and 003 peak intensities, applied in the identification procedure as an Ir parameter. Sample 28 exemplifies a poorly oriented preparation where the $34-36°2\theta$ region cannot be exploited due to the presence of a 20 reflection.

peak lies between the two illite/smectite peaks being measured. If a sample is monomineralic illite/smectite, the precise value of percent smectite measured in Figure 2 should fit within the range obtained from Figure 7. If a sample is a mixture of illite/smectite and illite, the percentage of S from Figure 2 should be lower than the range given by Figure 7. The real value is the one obtained from Figure 7. The other one is biased due to the interference of illite 002 and 003 peaks.

    Example 1: Sample 22 from Table 1 and Figure 7.
    Reflections: 17.52°, 26.79°, 33.85°2θ.
        BB1 = 4.9° and BB = 5.2° suggest prevailing IS type of ordering. From Figure 2: 8% S; from Figure 7: 26 ± 4% S.

Conclusion: a mixture of illite and IS-ordered illite/smectite of about 26% smectite layers.
    Example 2: Sample 12 from Table 1.
    Reflections: 16.92°, 26.75°, 33.86°2θ.
        BB1 = 5.0° and BB2 = 4.8° suggest IS ordering. From Figure 2: 28% S; from Figure 7; 26 ± 4% S.
    Conclusion: pure illite/smectite containing 28% S.

Illite/smectite may be distinguished from a mixture rich in this mineral by comparing the XRD patterns of glycolated different grain-size fractions, e.g., <0.2 µm and 0.2–0.5 µm. Illite is always more coarse-grained than illite/smectite and will tend to accumulate in coarser fractions. Size separation makes the XRD pattern of the coarser fraction look "more illitic" or results in a separation of illitic peaks. On the other hand, in the author's experience, pure illite/smectites do not show significant differences in the component layers ratio in different size fractions.

The sample materials used for the construction of Figure 7 come from bentonites of various ages and locations (Table 1) and is considered to be representative of diagenetic materials. The precision of the determination of the percentage of smectite from Figure 7 grows with increasing illite content. The identification of hydrothermal materials, in particular paragonite/beidellite (Kodama, 1966), remains an open problem. The thickness of the component layers of hydrothermal minerals is more variable (Figure 1), and the variation of the type of ordering vs. expandability has not been investigated systematically.

Figure 7 is useful if the amount of illite/smectite in a sample is sufficient to give measurable reflection at $33-35°2\theta$ and illite/smectite has at least 6–7% smectite layers (at about this value, the reflection at $33-35°2\theta$ disappears; see Figure 3). The remaining illitic materials must be analyzed differently.

Illitic materials lacking discrete illite/smectite reflections

XRD patterns of illitic materials lacking discrete illite/smectite reflections resemble closely the patterns of pure illite (Figure 8). Only detailed analysis of the positions, shapes, and intensities of the basal reflections reveals the presence of the expandable layers. The first step in identification consists of distinguishing between a homogenous, highly illitic mixed-layer mineral (<6% S) and illite or a mixture of illite and a mixed-layer mineral. This distinction can be made using Figure 2. If the experimental point fits into the illite/smectite field, the sample is a highly illitic mixed-layer mineral, and the percentage of smectite can be measured. The computer simulation has shown that even at the level of 2% the positions of the analytical reflections are measurably different from those of pure illite. A monomineralic sample of this kind must have

BB1 and BB2 parameters smaller than 4°2θ (ISII ordering), as established from the analysis of the relation between the type of ordering and the expandability (Figure 5). A sample of such material was not available to the author.

If the projection point in Figure 2 lies in the illite field, the sample is pure illite or a mixture of this mineral with an ordered illite/smectite. From studies of artificial mixtures, the most sensitive parameter for detecting trace amounts of the expandable material is Ir, the ratio of intensity ratios of 001 and 003 reflections from the same preparation, recorded in the air-dry and the glycolated state (Table 1). If Ir = 1.0, the material is 100% nonexpandable (samples 40 and 41, Table 1 and Figure 8). If Ir > 1.0, an admixture of expandable material is evident. When Ir parameter is used, the preparations must be sufficiently thick to absorb X-rays completely.

Another useful criterion for detecting minor amounts of illite/smectite is the shifting of 001 and 002 reflections after glycolation in the opposite directions, identically to the behavior of samples identified in the previous section (Table 1). Using BB1 and BB2 parameters admixtures of IS or ISII-ordered illite/smectite (Figure 8) can be distinguished. It is equivalent, as shown above, to the approximate estimation of whether or not the expandable material has more or less than 15% S. The methods used do not allow for more precise identification of such materials. Although the Ir parameter depends strongly on the layers ratio of illite/smectite (Table 1), it also depends on the proportion of illite and illite/smectite in a mixture. It should be noted that the distinction based on Ir between illite and a mixture containing a small amount of an ordered illite/smectite can be made only if smectite or randomly interstratified illite/smectite are absent from the sample. If one of them is present, it will influence the intensities of 001 and 002 illite in both air-dry and glycolated samples. In surface or shallow buried sediments, the coexistence of 10-Å and 17-Å material is common, e.g., the shallowest sample in Figure 9. In such samples the identification of 10-Å material cannot be carried beyond stating that it is "illitic material lacking discrete illite/smectite reflections."

APPLICATION OF THE OUTLINED METHODS IN DIAGENETIC STUDIES

In applying the developed techniques to a diagenetic sequence, several pertinent points should be emphasized:

1. Appearance of an expanding component: Ir > 1, displacement of 001 reflection after glycolation toward higher angles;

2. At about 6–7% expanding component in illite/smectite: appearance of the reflection at about 35°2θ (CuKα radiation).

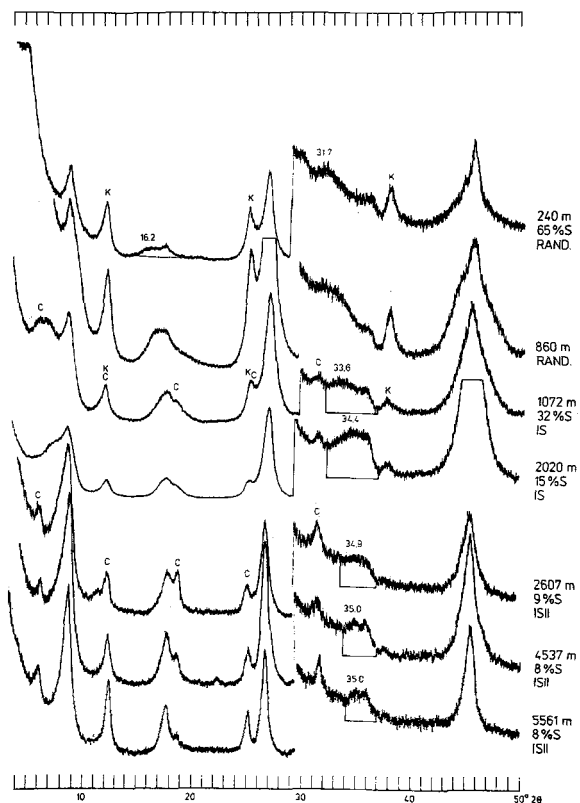
Figure 9. Representative X-ray powder diffraction patterns of glycolated <0.2-μm fractions of shales from the Września IG-1 borehole (central Poland). K = kaolinite, C = chlorite. Peaks used for the identification of illite/smectite are marked with 2θ values. The shallowest sample is identified according to Środoń (1981). The BB2 parameter is shown for IS and ISII minerals. 240 m and 860 m = Upper Jurrasic; 1072 m = Lower Jurrasic; 2020 m = Keuper; 2607 m = Buntsandstein; 4537 m = Rotliegendes; 5561 m = Upper Carboniferous. The 860-m sample illustrates a material that cannot be identified precisely (40–50% S, beginning of IS ordering, but predominant random characteristics).

3. Transition from ISII to IS type of ordering at about 15% S: BB1 and BB2 > 4°2θ.

4. Transition from IS to random interstratification at about 40–50% S: inflection or a small reflection at ~5°2θ and a diffuse band at 32–33°2θ.

Beginning at 6% S, measurements of expandability are possible in mixtures with illite, providing the illite/smectite is sufficiently abundant to give a measurable reflection at 33–35°2θ.

The most complete example of the illite/smectite evolution with depth known to the author is shown in Figure 9. The samples are shales from the Września IG-1 well in central Poland. The deepest samples are from Carboniferous rocks at ~5500 m below the surface, with a temperature of ~200°C and a vitrinite reflectance near 3.0. The XRD patterns show that mixed-layer illite/smectite is present in all samples but
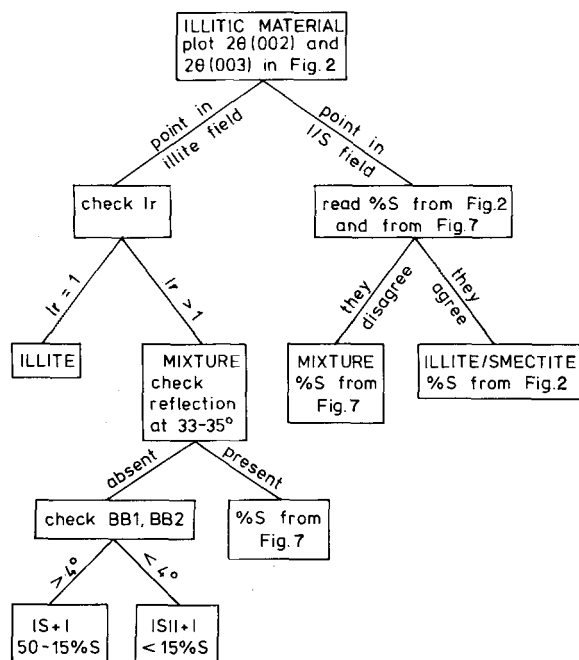
Figure 10. Flow sheet for X-ray powder diffraction identification of illitic materials from their 001 characteristics.

that illite is an abundant component dominating the patterns at about 26.7°2θ and commonly, at about 16–18°2θ. For these reasons, the techniques of Reynolds and Hower (1970), Środoń (1980), and that presented in Figure 2 cannot be used to measure the expandability of the illite/smectite. The sample from 240 m contains randomly interstratified illite/smectite as evidenced by small 17-Å reflection. The percentage of smectite layers was measured following Środoń's (1981) technique. The sample from 860 m also contains random illite/smectite, showing as the broad band between 31 and 34°2θ. This reflection was too diffuse to measure and expandability could not be established. The remaining samples contain ordered illite/smectites as evidenced by a non-coincident reflection between 33 and 35°2θ. This peak position was measured precisely by step-counting technique, and Figure 7 was used to measure expandability and to establish the type of ordering. The identification is consistent with BB2 values shown on the figure.

Even in the deepest samples (at 200°C), the diagenetic stage of pure nonexpanding illite was not reached, which is consistent with the data of Boles and Franks (1979). The successive stages of illite/smectite diagenesis appear in this profile at much lower depths and temperatures than in the well-known Gulf Coast profiles (Perry and Hower, 1970; Hower et al., 1976; Boles and Franks, 1979). This phenomenon needs further investigation and argues against using illite/smectite as

a paleogeothermometer at the present state of knowledge.

As shown in Figure 9, small but identifiable amounts of illite/smectite are common in fine-grained sediments from all kinds of sedimentary formations. The methods presented in this paper and in Środoń (1981) allow for the identification of this mineral. It makes illite/smectite a universal tool in diagenetic studies, providing that the possible effects of age, temperature, pore water chemistry, and lithology on expandability are recognized. The methods are intended to substitute for measurements of "crystallinity index" ("Kubler index") in the diagenetic zone. The crystallinity index is not a precise measure of diagenetic stage because it is dependent, like the Ir index, on both the quantity and quality of the mixed-layer component of a sample (Środoń, 1979). The concept of crystallinity index is, however, applicable at the illite stage, when this index becomes a function of the growing size of the coherent scattering domain as illite is transformed into muscovite.

CONCLUSIONS

The present study of numerous diagenetic samples of ordered illite/smectites indicates that:

(1) The thickness of the illite layer in illite/smectites is stable and equals 9.97 Å.

(2) The thickness of the smectite-ethylene glycol complex is 16.7–16.9 Å (for Na-saturated samples, run at room temperature and humidity).

(3) Diagenetic illite/smectites form a continuous series from random, through incomplete IS, IS, incomplete ISII, to ISII type of ordering, each type being related to a range of expandability. The IIS type of ordering has not yet been detected in diagenetic materials.

(4) IS and ISII types of ordering can be distinguished by the peak-shape parameters BB1 and BB2.

(5) Even at the level of a few percent smectite layers, homogenous illite/smectite can be distinguished by the 002 and 003 peak positions from a mixture of illite/smectite and discrete illite.

(6) The peak intensity parameter Ir is the most sensitive tool for revealing small amounts of expandable material accompanying illite.

The above information has led to a refined identification technique for illite/smectites and for the identification of illite/smectites in mixtures with discrete illite. Figure 10 presents a complete flow sheet for identification of illitic materials.

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Резюме— 10-Å глинистые компоненты осадочных пород ("иллиты") обычно являются смесями 100% нерасширяемого иллита и упорядоченного минерала типа смешанно-слойного иллита/смектита (ИС). Отношение составляющих слоев и тип переслаивания для смешанно-слойного минерала могут быть определены, если пропорция иллита/смектита в смеси достаточна, чтобы вызвать измеряемое отражение между 33–35°2θ (излучение CuKα), которое не совпадает с отражением иллита. Техника идентификации, разработанная в этой статье, основывается на последовательных экспериментальных данных для упорядоченных иллитов/смектитов диагенетического происхождения: (1) толщина иллитового слоя в иллите/смектите равна 9,97 Å; (2) толщина комплекса смектита с этиленовым гликолом изменяется в диапазоне от 16,7 до 16,9 Å; (3) иллиты/смектиты образовывают непрерывный ряд типов прослоев—беспорядочный, беспорядочный/ИС, ИС, ИС/ИСII, ИСII—и каждый тип связан со специфическим диапазоном расширяемости.

Эта новая техника расширяет метод компьютерного моделирования, развитый Рейнольдсом и Гоуером и включает такие осадочные материалы, в которых находится отдельный иллит, которые имеют малые количества иллита/смектита и которые, как таковые, предварительно описывались только при помощи "индекса кристальности иллита." [E.G.]

Resümee—Die 10-Å Tonkomponenten von sedimentären Gesteinen ("Illite") sind gewöhnlich Mischungen aus 100% nicht expandierbarem Illit und einem regelmäßigen Illit/Smektit-Wechsellagerungsmineral. Wenn das Verhältnis von Illit/Smektit in einer Mischung ausreicht, um einen meßbaren Reflex zwischen 33 und 35°2θ (CuKα-Strahlung) zu erzeugen, der nicht mit einem Illitreflex zusammenfällt, dann kann das Verhältnis der Komponentenschichten und die Art der Wechsellagerung für das Wechsellagerungsmineral bestimmt werden. Die Identifikationstechnik, die in dieser Untersuchung entwickelt wurde, beruht auf den folgenden experimentellen Ergebnissen für geordnete Illit/Smektit-Wechsellagerungen diagenetischen Ursprungs: (1) Die Dicke der Illitlagen in den Illit/Smektit-Wechsellagerungen beträgt 9,97 Å; (2) die Dicke des Smektit-Äthylenglykolkomplexes reicht von 16,7–16,9 Å; (3) Illit-Smektitwechsellagerungen bilden eine kontinuierliche Abfolge von Wechsellagerungstypen—unregelmäßige, unregelmäßige/IS, IS, IS/ISII, ISII—und jeder Typ gehört zu einem bestimmten Bereich von Expandierbarkeit.

Die neue Untersuchungsmethode baut die Computersimulationsmethode aus, die von R. C. Reynolds und J. Hower entwickelt wurde, um solche sedimentären Materialien mit einzuschließen, bei denen diskreter Illit vorherrscht, die wenig Illit/Smektit enthalten, und die, als solche, früher nur durch einen "Illit-Kristallinitätsindex" beschrieben wurden. [U.W.]

Résumé—Les composés argile de 10 Å de roches sédimentaires ("illites") sont communément des mélanges d'illite 100% non expansible et d'un minéral ordonné à couches melangées illite/smectite. Si la proportion d'illite/smectite dans un melange est suffisante pour produire une réflection mesurable entre 33–35°2θ (radiation CuKα) qui ne coïncide pas avec une réflection illite, on peut déterminer la proportion de couches du composé et le genre d'interstratification du minéral à couches mélangées. La technique d'identification développée dans cette étude est basée sur les trouvailles expérimentales suivantes pour des illite/smectites d'origine diagénétique: (1) l'épaisseur de la couche illite dans les illite/smectites est 9,97 Å: (2) l'épaisseur du complexe glycol smectite-éthylène s'étend de 16,7 à 16,9 Å; (3) les illite/smectites forment une séquence continuelle de types d'interstratification—au hasard, au hasard IS, IS, IS/ISII, ISII—et chaque type est apparenté à une étendue spécifique de pouvoir de dilatation.

La nouvelle technique élargit la méthode de simulation à l'ordinateur développée par R. C. Reynolds et J. Hower pour inclure les matériaux sédimentaires qui sont dominées par la présence d'illite discrète, ont un bas contenu en illite/smectite, et, en tant que tels, n'ont jusqu'à présent été décrits que par un "indexe de cristallinité d'illite." [D.J.]