

CONVENIENT TECHNIQUE FOR ESTIMATING SMECTITE LAYER PERCENTAGE IN RANDOMLY INTERSTRATIFIED ILLITE/SMECTITE MINERALS

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Abstract—The validity of the saddle/001 method for estimating the percentage of smectite layers in randomly interstratified illite/smectite (I/S) minerals as a routine laboratory technique has been examined with respect to the effects of the crystallite size distribution (N = number of layers) of I/S and the degree of preferred orientation of crystallites in the prepared specimen. X-ray powder diffraction experiments of I/S clays indicated that the crystallite size distribution was $3 < N < 12$; these values were supported satisfactorily by the variation of the $d(002)$ value of the samples. An analysis of the Lorentz factor concerning the degree of preferred orientation of crystallites indicated that a calibration curve calculated using the random powder Lorentz factor and the above crystallite size distribution fit the data better than an assumption of perfect orientation. Consequently, if a calibration curve of the saddle/001 ratio is used to estimate the percentage of smectite layers in I/S, an error of 10–15% should be expected from the variable crystallite size distribution of actual samples, in which I/S dominates over other phyllosilicate phases that give reflection between 10–14 Å. This method is useful, however, in estimating the relative percentage of smectite layers in randomly interstratified I/S for samples examined under identical experimental conditions.

Key Words—Crystallite size, Illite, Illite/smectite, Orientation, Smectite layer percentage, X-ray powder diffraction.

INTRODUCTION

An empirical method was developed by Weir *et al.* (1975) to determine the percentage of smectite layers (% S) in randomly interstratified illite/smectite (I/S). This method has been called the “saddle/001” method and makes use of the intensity ratio of the 001 peak to the background, downangle from the 001 peak (see Figure 1). Since then, this method has been used as a routine and convenient technique to estimate % S in I/S by many workers (Eslinger and Savin, 1976; Hoffman, 1979; Rettke, 1981).

Random I/S in bentonites and hydrothermal alteration products are usually monomineralic or they occur with easily separable minerals, such as quartz, biotite, and feldspar. They may also occur with clay minerals which do not interfere with the determination of the % S in I/S (e.g., kaolinite). The % S in I/S from such rocks can be determined precisely by comparing the X-ray powder diffraction (XRD) patterns with those computer-simulated by Reynolds (1980) or by plotting peak position of XRD data in the $\Delta 2\theta_1 - \Delta 2\theta_2$ diagram of Watanabe (1981, 1988). I/S clays from sandstones and shales, on the other hand, almost always coexist with discrete illite, which cannot be separated me-

chanically. Large quantities of illite (>70%) can interfere in the determinations of % S by the saddle/001 method (Rettke, 1981; Środoń, 1981). Nevertheless the saddle/001 method may be useful to estimate the % S in I/S because it is more accurate than peak-position measurement methods, even in samples containing discrete illite. The accuracy of the saddle/001 method, if illite is present with the I/S, was determined by Rettke (1981), who reported a series of saddle/001 ratio curves as they are affected by the presence of various amounts of discrete illite. According to Rettke (1981), the error of % S determination is 10% for samples containing $\leq 50\%$ discrete illite. Therefore, the saddle/001 method may still be applicable for many samples. On the other hand, I/S samples commonly give only a few discernible XRD peaks, e.g., 001, 002, and 005, after ethylene-glycol (EG) treatment. Loss of diffraction intensity of peaks is probably caused by small coherent diffraction domain. For such samples, it may be impossible to apply Środoń's peak position method (1980, 1981). The saddle/001 peak intensity ratio can also depend on the coherent diffraction domain (Hower, 1981; Środoń, 1981); however, if the crystallite size distribution can be evaluated independently by other methods (e.g., measurement of width at half height of the XRD peak), a working curve can be calculated for the saddle/001 peak intensity ratio.

Recently a personal computer program to calculate XRD patterns of I/S, NEWMOD program, was de-

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Table 1. Illite/smectite from Shinzan and Kinnekulle.

| Sample | %S | Reichweite | K/O ₁₀ (OH) ₂ ² | Fe/O ₁₀ (OH) ₂ ³ | Impurity |
|-------------------|--|-------------|--|---|-----------------------------|
| Shinzan | | | | | |
| WS-2-183 | 100 | | 0.05 | 0.08 | |
| WS-1-336 | 95 ± 5 | g = 0 | 0.08 | 0.22 | |
| WS-4-329 | 85 ± 5 | g = 0 | 0.11 | 0.13 | |
| WS-4-383 | 73 ± 5 | g = 0 | 0.25 | 0.14 | |
| WS-2-377 | 70 ± 5 | g = 0 | 0.19 | 0.14 | |
| WS-2-407 | 68 ± 5 | g = 0 | 0.27 | 0.13 | |
| WS-2-383 | 55 ± 5 | g = 0 | 0.41 | 0.13 | illite, quartz |
| WS-4-392 | 50 ± 10 | g = 0 | 0.31 | 0.17 | illite |
| Kinnekulle | | | | | |
| B31 | 45 ± 5 (35) ¹ [48] ² | g = 0/g = 1 | 0.38 ⁴ | 0.13 ⁴ | kaolinite, quartz |
| B33 | 50 ± 5 (41) ¹ [56] ² | g = 0/g = 1 | 0.32 ⁴ | 0.15 ⁴ | kaolinite, chlorite, quartz |
| B38 | 55 ± 5 (51) ¹ [64] ² | g = 0 | 0.26 ⁴ | 0.18 ⁴ | kaolinite, chlorite, quartz |
| STMX6 | 60 ± 5 (52) ¹ [66] ² | g = 0 | 0.25 ⁴ | 0.16 ⁴ | kaolinite, quartz |

¹ Percentage of smectite layers from X-ray powder diffraction data by Brusewitz (1986, 1988).

² Percentage of smectite layers obtained from $K_{\text{fix}}/K_{\text{fix,max}}$ by Brusewitz (1986, 1988).

³ Data from Inoue *et al.* (1978, 1987).

⁴ Data from Brusewitz (1986, 1988).

veloped by R. C. Reynolds (Dartmouth College, Hanover, New Hampshire, published by the author). In this paper we re-examine the validity of the saddle/001 method for estimating % S in randomly interstratified I/S by comparing theoretically calculated saddle/001 peak ratios given by the NEWMOD (version 1.0) calculation and experimentally observed peak position and saddle/peak ratios of I/S samples.

MATERIALS AND METHODS

Materials

The randomly interstratified I/S used in the present study were from Shinzan, Japan (8 samples) and Kinnekulle, Sweden (4 samples) (supplied by A. M. Brusewitz). The interstratified structure, chemical composition, and other mineralogical properties of the former and latter samples were described in detail by Inoue *et al.* (1978, 1987, 1988), Inoue and Utada (1983), Inoue (1986), Brusewitz (1986, 1988), and Velde and Brusewitz (1986).

X-ray powder diffraction

The <1- μm fraction of each clay was isolated by centrifugation without any pretreatment. Oriented specimens were prepared by dropping the suspensions on a glass slide with a pipet and drying them in an oven at 60°C. Two different analytical procedures were used: (1) Oriented specimens were saturated with EG liquid using an atomizer and then examined imme-

diately by XRD, using a Philips PW 1730 diffractometer (40 kV, 40 mA). Each sample was scanned at 0.5°/min with CoK α radiation. One-degree divergence and scattering slits were employed throughout this measurement. (2) Oriented specimens were saturated with EG vapor at 60°C overnight and then examined by XRD with a Rigaku RAD I-B diffractometer (40 kV, 20 mA) equipped with a Cu tube and a graphite monochromator and 0.5° divergence and scattering slits at 0.5°/min scanning speed.

The % S in the samples was determined by the $\Delta 2\theta_1 - \Delta 2\theta_2$ method of Watanabe (1981, 1988). The error in this procedure is generally $\pm 5\%$, but contamination by discrete illite probably reduces the precision and accuracy (Inoue *et al.*, 1987). For the Kinnekulle samples, as shown in Table 1, the determined % S values were slightly larger than those previously reported by Brusewitz (1986, 1988) using an XRD method, but smaller than the % S evaluated from the chemical $K_{\text{fix}}/K_{\text{fix,max}}$ data (Brusewitz, 1986).

The saddle/001 peak intensity ratio, as shown in Figure 1, is the same as that defined by Weir *et al.* (1975). The slope of the base line was determined using a glass slide without clay materials.

Calculations

The saddle/001 peak intensity ratio was calculated by the NEWMOD program as a function of % S. The actual ratio is influenced by many factors, such as in-

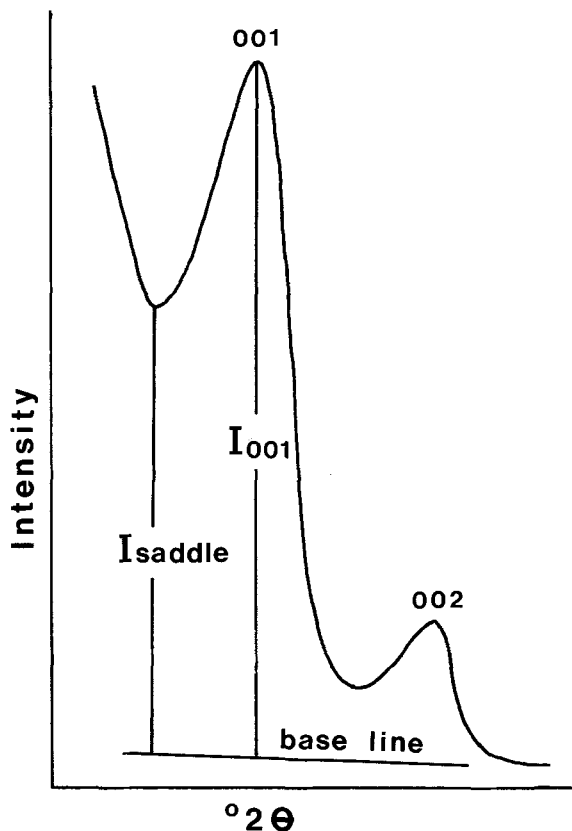


Figure 1. Definition of the saddle/001 peak intensity ratio. The intensity was approximated by the peak height from the base line.

terstratified structure, chemical composition, crystallite size distribution, thickness of the EG-smectite complex layer, morphology of the clay particles, and degree of preferred orientation of the crystallites on a glass slide. The octahedral Fe contents of the samples studied range from 0.1 to 0.2 per half unit cell as shown in Table 1. These values did not affect the saddle/001 peak intensity ratio, as estimated by preliminary calculations. The interlayer K content of the mica layer was assumed to be 0.8 per half unit cell, in accord with the chemical analyses (Inoue *et al.*, 1987; Bruswitz, 1986). The morphology of the clays ranged from flaky at 100% S to lath shape at 50% S in the Shinzan samples (Inoue, 1986; Inoue *et al.*, 1987, 1988) and mostly flaky in the Kinnekulle samples (unpublished data). The crystal morphology certainly can influence the peak height and peak broadening due to effects on preferred orientation and the shape factor in the Scherrer equation (Klug and Alexander, 1974). A quantitative evaluation cannot be easily performed, and, hence, it was neglected in the calculations reported here. The increase in background due to the fluorescence effect also was neglected because the monochromatized and non-

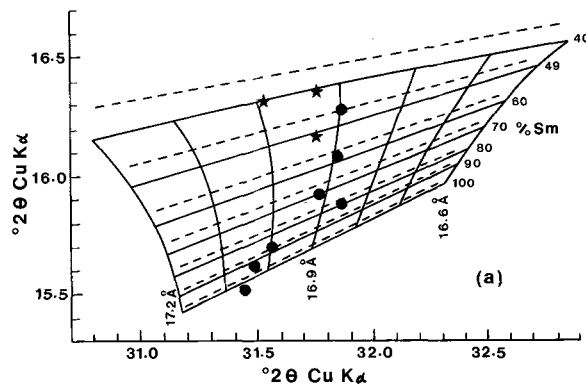


Figure 2. Plots in Šrodoň's (1981) diagram for measuring thickness of ethylene glycol-smectite complex layer in illite/smectite. Solid circles and stars are samples from Shinzan and Kinnekulle, respectively. Solid and dashed lines correspond to $1 < N < 8$ and $1 < N < 14$ of crystallite size distribution.

monochromatized XRD data differed only slightly, as shown in Figure 4.

Figures 2 and 3 show the data plotted on Šrodoň diagrams (Šrodoň, 1980, 1981). The thickness of the EG-smectite complex layer seems to range from 17.1 Å at 100% S to 16.8 Å at 50% S in the Shinzan samples and range from 17.0 to 16.7 Å in the Kinnekulle samples. Such differences in the EG-smectite complex thickness do not significantly affect the saddle/001 peak intensity ratio within experimental error. Accordingly, in using the NEWMOD program it was assumed that Reichweite = 0, Fe = 0, K = 0.8, and EG-smectite complex thickness = 16.9 Å. Divergence slit, goniometer radius, and soller slit parameters were the appropriate values for the Philips and Rigaku diffractometers used. The parameter, σ^* , used to calculate the Lorentz-polarization factor was taken as a standard value of 12.0°, as given in the NEWMOD program (*vide infra*). Thus, only two factors affecting the saddle/001 peak intensity ratio are critically examined here from a theoretical standpoint: the effect of crystallite size (coherent diffraction domain of c^* direction) and the effect of the degree of preferred orientation of the crystallites that make up the sample.

RESULTS AND DISCUSSION

Measured saddle/001 ratio of the present samples are plotted against the % S in Figure 4, and compared with working curves previously provided. The ratios deviate both from the curve (solid line in Figure 4) of Weir *et al.* (1975) and from the curve (dash-dotted line in Figure 4) calculated using the NEWMOD program. The working curve (dashed line in Figure 4) provided by Rettke (1981) fits the present data comparatively well.

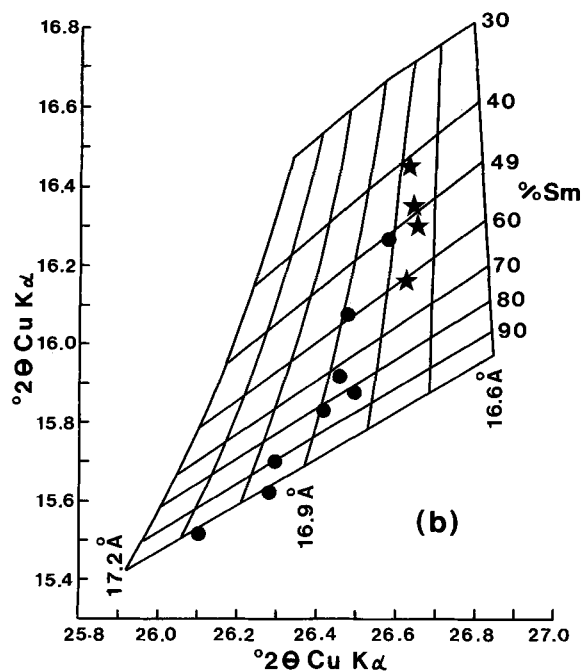


Figure 3. Plots in Šrodoň's (1980) diagram for measuring thickness of ethylene glycol-smectite complex in illite/smectite. Solid circles and stars are samples from Shinzan and Kinnekulle, respectively. Lines were calculated on basis of $1 < N < 8$.

Effect of the crystallite size distribution

Figure 5 shows the variation of the saddle/001 ratio curve as a function of the crystallite size (N) distribution. The calculated curves parallel one another and show an increase of the ratio with decreasing N . In the NEWMOD program, the crystallite size (N) distribution can be varied using the parameters LOW N and HIGH N that refer respectively, to the number of unit cells stacked in the c^* direction that make up the smallest and largest coherent domains. The proportions of each value of N are considered to be the same in the sample, meaning, for example, that if $1 < N < 10$, the mean crystallite size \bar{N} is 5, but equal amounts of $N = 2$ and $N = 9$ sizes are also present.

In Figure 4, the crystallite size distribution used to calculate each working curve was assumed to be 3–7 for Weir *et al.* (1975) and 7–13 for Rettke (1981), as well as for the NEWMOD program. As shown in Figure 5, the experimental data plot in a zone between the $1 < N < 5$ and $1 < N < 10$ curves. Figure 6 shows the calculated variation of $d(002)$ of EG-saturated I/S clays as a function of N . In this illustration, the present data plot in the zone between the $1 < N < 8$ and $1 < N < 14$ –20 curves. The coherent domain size of the c^* direction can then be independently measured by two methods.

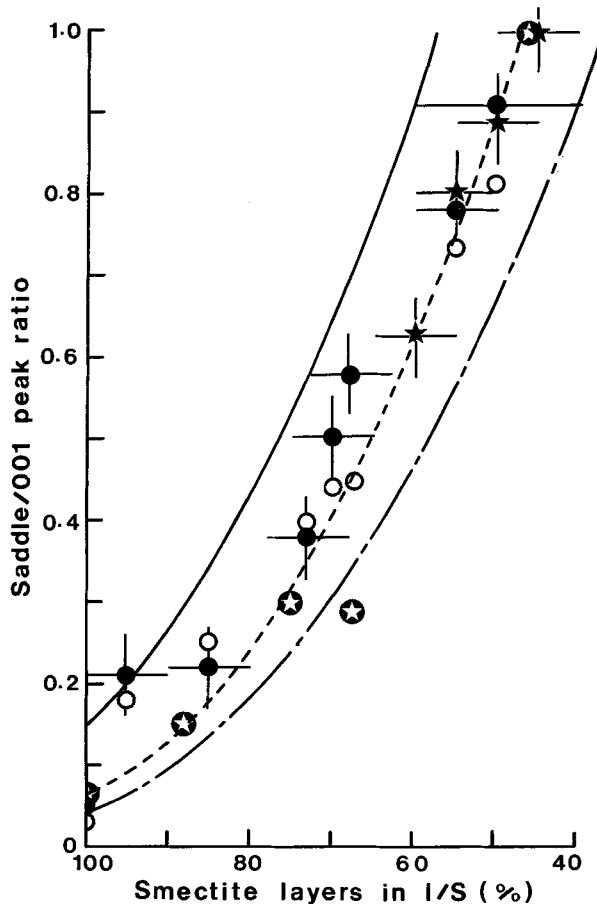


Figure 4. Plots of saddle/001 peak ratio vs. smectite layer percentage in illite/smectite determined by peak position method from Shinzan (O, ●), Kinnekulle (★), and Šrodoň (1981) (⊙). Solid and open circles correspond to data collected using $\text{CoK}\alpha$ and $\text{CuK}\alpha$ radiation, respectively. Solid line: working curve of Weir *et al.* (1975) ($3 < N < 7$), dashed line: working curve of Rettke (1981) ($7 < N < 13$) and dash-dotted line: curve calculated using NEWMOD program, $\sigma^* = 12$, and $7 < N < 13$.

The Scherrer equation (Brindley, 1980), which is generally applied to determine the relation between N and the X-ray diffraction peak breadth B , is:

$$B = 0.91\lambda/Nc^* \cos \theta,$$

where λ is the wavelength of the X-rays, θ is the Bragg angle, and c^* is the basal spacing. In using the width at half height of the 005 peak in the EG-saturated samples, the peak most insensitive with respect to coherent domain size (Ross, 1968; Reynolds, 1968; Tettendorf and Roberson, 1973), the apparent thickness (Nc^*) of the 100% S sample was ~ 200 Å, corresponding to an N value of 11–12 layers. If the Scherrer equation can be applied to interstratified minerals, the apparent thickness of the 50% S sample was ~ 120 Å, corresponding to an N value of about $9(I + S)$ layers.

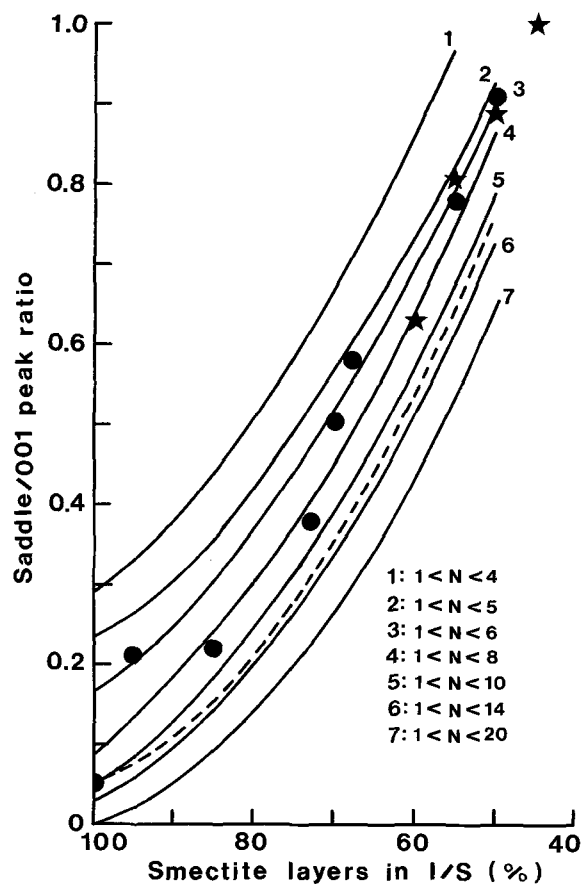


Figure 5. Variation of saddle/001 peak ratio curve as a function of crystallite size distribution. Solid circles and stars represent the Shinzan and Kinnekulle samples, respectively. Dashed curve is a curve calculated on the basis of $3 < N < 12$.

The apparent thickness of the other samples ranged from 90 to 200 Å, i.e., $5 < N < 12$. These values are fairly consistent with the data previously reported for smectite, I/S, and illite (Tettenhorst and Roberson, 1973; Weir *et al.*, 1962; Güven *et al.*, 1980; McHardy *et al.*, 1982). On the other hand, the direct measurement of the particle thickness of the present samples by means of transmission electron microscope (TEM) gave < 50 Å for most clay particles, corresponding to a value of $N < 5$ (Inoue *et al.*, 1987). The discrepancy between the XRD and TEM data may be reconciled by postulating an interparticle diffraction effect, as pointed out by Nadeau *et al.* (1984). Taking into account the plots in Figures 5 and 6 and the measurement of coherent domain using Scherrer equation, the crystallite size distribution may be assumed to range from 3 to 12 in the sedimented aggregates examined here.

Using the calibration curves for 002 peak position as a function of % S for different ranges of N, the best fit with the experimental data is for $3 < N < 12$ (dashed line in Figure 6). If the saddle/001 peak ratio vs. % S

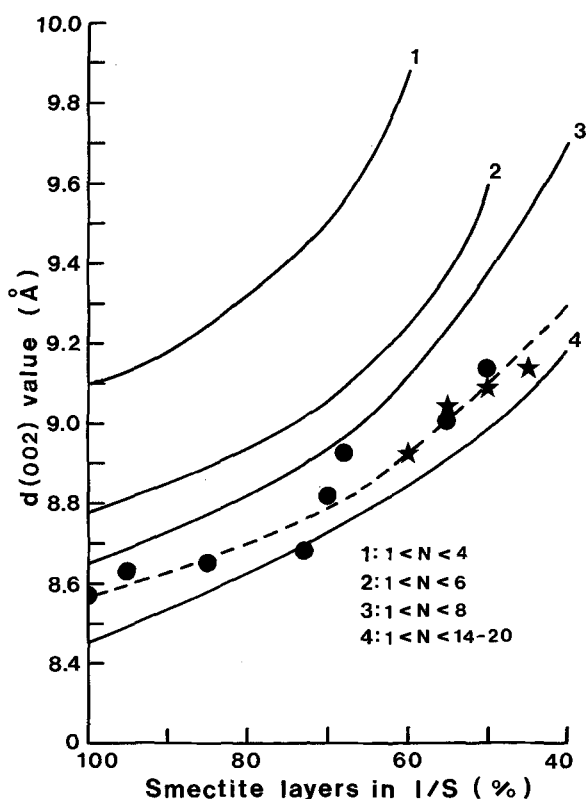


Figure 6. Variation of $d(002)$ value of ethylene-glycol saturated illite/smectite as function of crystallite size distribution. Solid circles and stars represent the Shinzan and Kinnekulle samples, respectively. Dashed curve is calculated on basis of $3 < N < 12$.

relations are calculated for various ranges of N, however, the curve for $3 < N < 12$ does not fit the experimental points, which fall to lower apparent N values. A large deviation occurs at 95% S (Figure 5), similar to that reported by Sudo (1985). This large deviation probably results from the variable crystallite size distribution in more expandable I/S. Consequently, the variable crystallite size distribution in actual I/S specimens will lead to the error of 10–15%, if a calibration curve is used in the saddle/001 method. Such a large error may be particularly detrimental to estimating % S of I/S richer in smectite layers. On the other hand, although the variation of the $d(002)$ value supports the assumption of $3 < N < 12$ for the present samples, an inconsistency in the saddle/001 peak ratio vs. % S relation remains. To reconcile the differences between the 002 peak position and saddle/001 ratio in determinations of % S, the effect of preferred orientation of crystallites which make up the samples, should be examined.

Effect of the degree of preferred orientation

The effect of the degree of preferred orientation of crystallites on saddle/001 peak ratio may be estimated

by taking into account the Lorentz factor. Reynolds (1986) estimated quantitatively the dependence of the Lorentz factor on the degree of preferred orientation of crystallites. He introduced a Gaussian distribution function ψ into the Lorentz factor in the NEWMOD program, including the standard deviation σ^* , which describes the frequency of tilt angles about the plane defined by the sample surface. The Lorentz-polarization factor (Lp) can be varied from a random powder to a single crystal form by changing the ψ value.

In randomly interstratified I/S, the 2θ positions of the saddle and the 001 peak are near 4° and 6° for EG-saturated samples. For simplicity, if the crystallite size distribution is the same for each sample, the XRD intensity approximates $A \cdot Lp \cdot F^2$, where F is the structure factor and A is a constant. Furthermore, the F^2 values at 4° and $6^\circ 2\theta$ are constant for either a Lorentz factor of a single crystal or for a random powder form. Consequently, the saddle/001 peak intensity ratio depends on the Lp factor: $I_{\text{saddle}}/I_{001} = A' \cdot [Lp(4^\circ)/Lp(6^\circ)]$, where A' is a constant including F^2 and A . The Lp values are 819.5 at 4° and 363.6 at 6° for a random powder, $(1 + \cos^2 2\theta)/\sin 2\theta \sin \theta$, and 14.3 at 4° and 9.5 at 6° for a single crystal, $(1 + \cos^2 2\theta)/2 \sin 2\theta$. The $Lp(4^\circ)/Lp(6^\circ)$ ratio is 2.25 for a random powder and 1.50 for a single crystal form. The ψ function used in the NEWMOD program describes the deviation of crystallite orientation from a single crystal to a random powder. This is expressed by $Lp^R = \psi \cdot Lp^S$, when Lp^R is the Lp factor for a random powder and Lp^S is the Lp factor for a single crystal. Because $Lp^S(4^\circ)/Lp^S(6^\circ) = 1.50$ and $Lp^R(4^\circ)/Lp^R(6^\circ) = 2.25$, $Lp^R(4^\circ)/Lp^S(6^\circ) = 1.5 \cdot [\psi(4^\circ)/\psi(6^\circ)]$ and $1 \leq \psi(4^\circ)/\psi(6^\circ) \leq 1.5$. If $\psi(4^\circ)/\psi(6^\circ) = 1$, the Lp factor is that of the single crystal form, and if $\psi(4^\circ)/\psi(6^\circ) = 1.5$, the Lp factor is that of the random powder form. This result indicates that the saddle/001 peak intensity ratio varies with the degree of preferred orientation of crystallites from a perfectly oriented specimen, such as a single crystal, to a random powder by a factor of 1.5. This relation is diagrammatically given in Figure 7. If σ^* (the average tilt angle) is assumed to be 12° as in the NEWMOD calculation, which is a standard value recommended by Reynolds (1986), $\psi(4^\circ)/\psi(6^\circ) = 1.136$, and the obtained saddle/001 ratio curve is intermediate between the single crystal curve and the random powder curve (Figure 7). The experimental data fit the curve calculated for the random powder form of the Lp factor better than those which can be calculated for the single crystal or the standard forms of the Lp factor in the NEWMOD program assuming an average 12° tilt angle. Watanabe (see Sudo, 1985) also calculated a working curve of the saddle/001 ratio, based on the random powder form of the Lp factor, the average domain size $\bar{N} = 10$ with a Gaussian distribution of coherent domain from $N = 4$ to 16, and the XRD intensity equation by Kakinoki and Komura (1952) (Watanabe, 1988). The work of

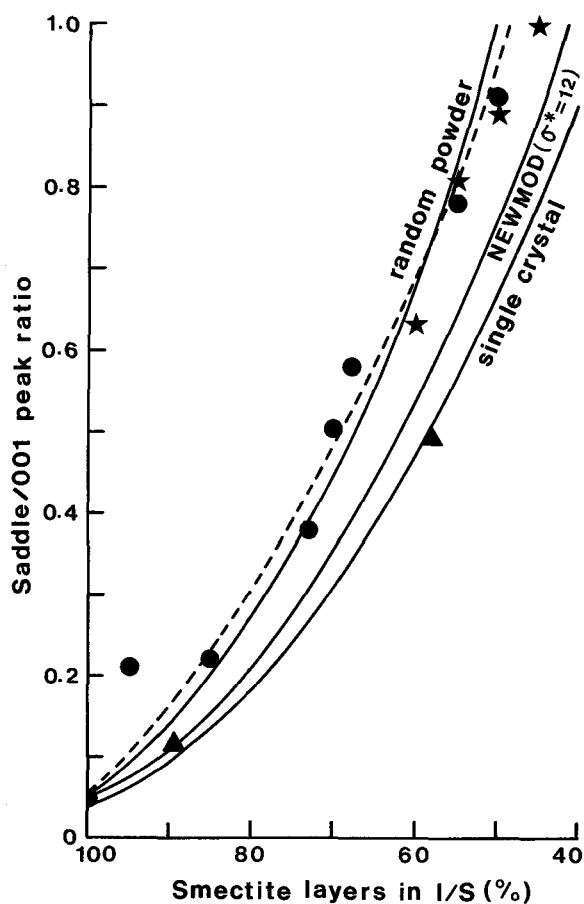


Figure 7. Curves of saddle/001 peak ratio vs. smectite layer percentage calculated using different Lorentz factor forms for $3 < N < 12$. Dashed curve is from work of Watanabe (from Sudo, 1985). Solid circles and stars represent Shinzan and Kinnekulle samples, respectively. Triangles are data from Srodoń *et al.* (1986).

Watanabe also indicates a random powder form of the Lp factor compared with the present data. The degree of preferred orientation of crystallites on a glass slide will vary with the employed preparation procedures (Reynolds, 1986). The experimental curve in Figure 7 indicates that the maximum error of % S by the saddle/001 method is about 10% for I/S having 50% S due to the unknown degree of preferred orientation.

The data from Srodoń *et al.* (1986), as plotted in Figure 7, show a large departure from the present data, probably due to the use of different XRD equipment. Srodoń *et al.* (1986) used a new Siemens D-500 diffractometer, which employs no primary-beam soller slits. The two diffractometers used in the present study utilized two soller slits. As pointed out by Reynolds (1986), the relative intensities of low-angle peaks from oriented specimens cannot be directly compared between the apparatus with one soller slit and a machine

with two soller slits, unless provision is made for difference in the effective Lorentz factor.

SUMMARY AND CONCLUSIONS

The saddle/001 peak intensity ratio changed systematically with the % S in randomly interstratified I/S, as demonstrated previously by Weir *et al.* (1975) and Rettke (1981). The ratio, however, also depended on the crystallite size distribution and the degree of preferred orientation. The XRD experiments indicated that the crystallite size distribution was $3 < N < 12$ for the studied samples. If a calibration curve was calculated on the basis of these data and the Lp factor was standardized in the NEWMOD program, the experimental data showed a significant departure from the curve because of the effect of preferred orientation of crystallites. The analysis of Lorentz factor indicated that the Lorentz factor of a random powder form should be applied to calculation of the calibration curve for specimens prepared by convenient procedure, such as the smear method or by dropping with a pipet. In conclusion, if the saddle/001 method is used routinely to determine % S in randomly interstratified I/S clays, an error of 10–15% should be expected, taking into account the effects of the crystallite size distribution and the unknown degree of preferred orientation in the prepared specimens. The presence of discrete illite will also reduce the accuracy as demonstrated by Rettke (1981) and Srodoń (1981).

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