Clays and Clay Minerals, Vol. 33, No. 5, 379–390, 1985.

CURVES FOR THE QUANTIFICATION OF MICA/SMECTITE AND CHLORITE/SMECTITE INTERSTRATIFICATIONS BY X-RAY POWDER DIFFRACTION

KATSUTOSHI TOMITA

Institute of Earth Sciences, Faculty of Science, Kagoshima University
Kagoshima, 890, Japan

HIDEWO TAKAHASHI

Department of Geology, Faculty of Education, Kagoshima University
Kagoshima, 890, Japan

Abstract—X-ray powder diffraction intensities for many interstratified structures of mica/smectite and chlorite/smectite were calculated by changing combinations of probabilities and transition probabilities of two component layers, respectively. The calculated d-values were plotted with P_{MS} and P_{SM} as the axes of coordinates for mica/smectites (where M is a mica layer and S is a smectite layer). These d-values were then linked into equal d-value curves on a graph. Three equal d-value diagrams ranging from 32.5 to 24.5 Å, from 15.4 to 10.25 Å, and from 3.365 to 3.08 Å were constructed for mica/smectites. Several diagrams were also constructed for mica/glycolated-smectites and chlorite/smectites using the same techniques. P_{MS} and P_{SM} values of mica/smectite producing 26.8- and 12.6-Å reflections in its X-ray powder diffraction pattern were obtained from the coordinates of the intersection of the 26.8-Å line of the first diagram and the 12.6-Å line of the second diagram. The components and stacking parameters of mica/smectites and chlorite/smectites were estimated easily using these diagrams. Interstratified mica/smectites were quantified in the air-dry and glycolated states, and chlorite/smectites in the glycolated state. Stacking parameters obtained by this method agreed well with those obtained by MacEwan's method. Stacking parameters for Reichweite (R=0) and (R=1) structures were obtained.

Key Words—Chlorite/smectite, Ethylene glycol, Interstratification, Mica/smectite, Mixed layer quantification, X-ray powder diffraction.

要旨 — ウンモ／スメクタイトと緑泥石／スメクタイト混合層鉱物の多くの混合層構造について，そのX線粉末回折強度を，構成層の存在確率および継続確率をそれぞれ変えて計算した。ウンモ／スメクタイト混合層鉱物については，計算で得たd-値を，P_{MS}とP_{SM}を座標軸にとりプロットした（ここでMはウンモ層，Sはスメクタイト層である）。これらのd-値のうち等しいd-値を線で結んで等d-値曲線を得た。ウンモ／スメクタイト混合層鉱物については，32.5-24.5Å(Fig.2)，15.4-10.25Å(Fig.3)，3.365-3.08Å(Fig.4)の3つの等d-値曲線を作成した。ウンモ／グリコール－スメクタイトと緑泥石／スメクタイト混合層鉱物についても同じ方法でいくつかの等d-値曲線を作成した。そのX線粉末回折パターン中に26.8Åと12.6Åの反射を持つウンモ／スメクタイト混合層鉱物のP_{MS}とP_{SM}の値は，Fig.2 の26.8Å線とFig.3 の12.6Å線との交点から得られる。ウンモ／スメクタイトと緑泥石／スメクタイト混合層鉱物の構成層の積層パラメータはこれらの図を使って容易に推定できる。本論文の図を使って，ウンモ／スメクタイト混合層鉱物については，室温で乾燥した試料とエチレングリコール処理した試料について，また、緑泥石／スメクタイト混合層鉱物については，エチレングリコール処理した試料について積層パラメータを知ることができる。この方法で得た混合層鉱物の積層パラメータは，MacEwan の方法で得たものとよく一致した。本論文の図を使うことにより，Reichweite(R)=0 とR=1 の構造の積層パラメータが得られ，これらの図は，(R)=0 とR=1 についてのみ適用される。

INTRODUCTION

Over the years numerous publications have addressed the problem of the calculation of X-ray powder diffraction patterns of interstratified laminar systems. One of the first significant papers on this subject was by Hendricks and Teller (1942) which dealt with infinite crystallites. For small particle size assemblages, the matrix method of Kakinoki and Komura (1952, 1954a, 1954b, 1965) appears to be the most complete, in that structure factors for all of the layers are introduced and varying degrees of non-random interstratification may be handled. The program of Reynolds and Hower (1970) has these capabilities also. Allegra (1964) showed that the order of the matrices of the intensity equation can be reduced if the complexions of layers are represented by the corresponding complexions of displacement vectors. Allegra's hypothesis is essentially similar to that of Kakinoki and Komura (1965). The direct Fourier transform method of MacEwan (1956, 1958) may be applicable for some different types of layers. MacEwan's methods in somewhat modified form have been used by Reynolds (1967, 1980), Reynolds and Hower (1970), and Tettenhorst and Grim (1975a, 1975b) to calculate diffraction patterns that correspond closely to actual diffraction runs. Sato (1969, 1973), Sato *et al.* (1965), Cradwick (1975),

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

Drits and Sakharov (1976), and Watanabe (1981) applied the hypothesis of Kakinoki and Komura (1952, 1954a, 1954b, 1965) for interstratified minerals.

Środoń (1980) prepared several graphs for the precise identification of illite/smectite interstratifications and investigated the thickness of the ethylene glycol complex in expanding clays. His techniques successfully minimized the error in quantifying the degree of layer ordering, but his method is somewhat complicated. We have calculated X-ray powder diffraction patterns for many interstratified mica/smectites and chlorite/smectites using Kakinoki and Komura's hypothesis (1965) and prepared diagrams to determine the degree of interstratification in these mixed-layer minerals. Reynolds (1980) and Środoń (1980) provided techniques for measuring layer ratio and for estimating the degree of ordering, but junction probabilities cannot be measured using their approach; such measurements are the goal of this paper.

EXPERIMENTAL

X-ray powder diffraction profiles of interstratified structures were calculated using the equation of Kakinoki and Komura (1965). The integrated intensity and the intensity maximum position were calculated by an electronic computer using a slightly modified program of Takahashi (1982). An N of 20 was used in the calculations.

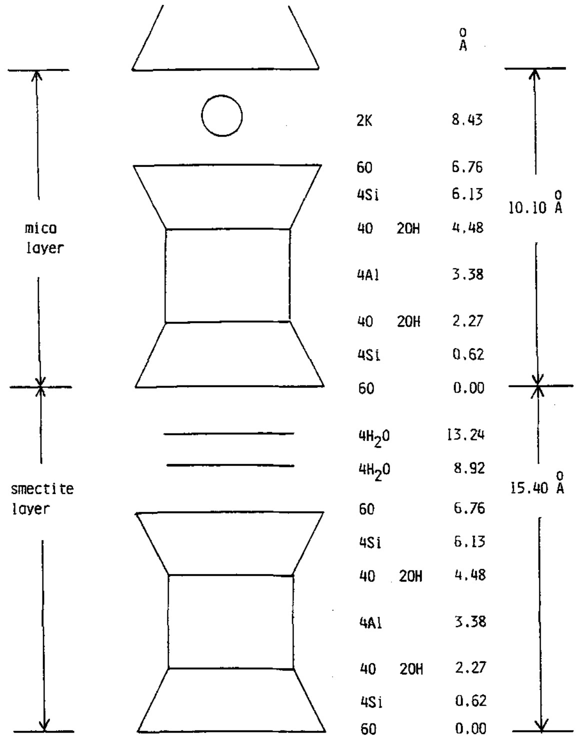

Figure 1. Mica and smectite models used in calculation of X-ray powder diffraction intensity.

MICA/SMECTITE INTERSTRATIFICATIONS

Preparation of determinative curves

The calculation of X-ray powder diffraction (XRD) patterns for interstratified structures was based on the model of Sato *et al.* (1965) as shown in Figure 1. A value of 15.4 Å was used for the smectite layer, but the spacing of smectite is not uniform from sample to sample and depends on relative humidity, kinds of cations in interlayers, and differences in layer charges. It is difficult to take all these facts into account; hence, 15.4 Å was used for smectite layer as was used by previous workers (Hendricks and Teller, 1942; MacEwan *et al.*, 1961). Because many clay-size micas (illites) give a spacing near 10.1-Å rather than 10-Å value (Kodama, 1962; Sudo and Shimoda, 1978), the mica model of Sato *et al.* (1965) was used. Diffraction intensities were calculated for many combinations of probabilities and transition probabilities for mica and smectite layers. Equal d-value line diagrams from 32.5 to 24.5 Å, from 15.4 to 10.25 Å, and from 3.365 to 3.08 Å were constructed from the calculated data and are shown in Figures 2, 3, and 4, respectively. In these illustrations P_{MS} is the probability a smectite layer succeeds a mica layer, assuming that the first layer is mica layer. P_{SM} is the probability that a mica layer succeeds a smectite layer assuming that the first layer is smectite.

Interstratification quantification

Samples having high d-spacings. As an example of the practical application of this method, a specimen investigated by Tomita *et al.* (1969) was examined. The d-values of the 001 and 002 reflections are 26.8 and 12.6 Å, respectively. The 26.8-Å contour in Figure 2 was traced along a 26.8-Å line. The tracing paper was placed on the diagram shown in Figure 3, and the 12.6-Å contour was traced in the same way. From the intersection of the two lines shown in Figure 5 at point A, P_{MS} and P_{SM} values of .665 and .875, respectively, were obtained. All remaining probabilities and junction probabilities for nearest-neighbor ordering can be obtained from these data and the statistical relations.

For a two-component interstratification of layers A and B, assuming P_A to be the frequency of occurrence of A, P_B is that of B, and $P_A + P_B = 1$. If P_{AB} is the probability that B succeeds A, given that the first layer is A, P_{AA}, P_{BB}, and P_{BA} are similarly defined. Thus:

$$P_{AA} + P_{AB} = 1,$$

$$P_{BA} + P_{BB} = 1,$$

$$P_AP_{AA} + P_BP_{BA} = P_A,$$

$$P_AP_{AB} + P_BP_{BB} = P_B,$$

$$P_BP_{BA} = P_AP_{AB}, \quad \text{and}$$

$$\frac{P_B}{P_A} = \frac{P_{AB}}{P_{BA}}.$$

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

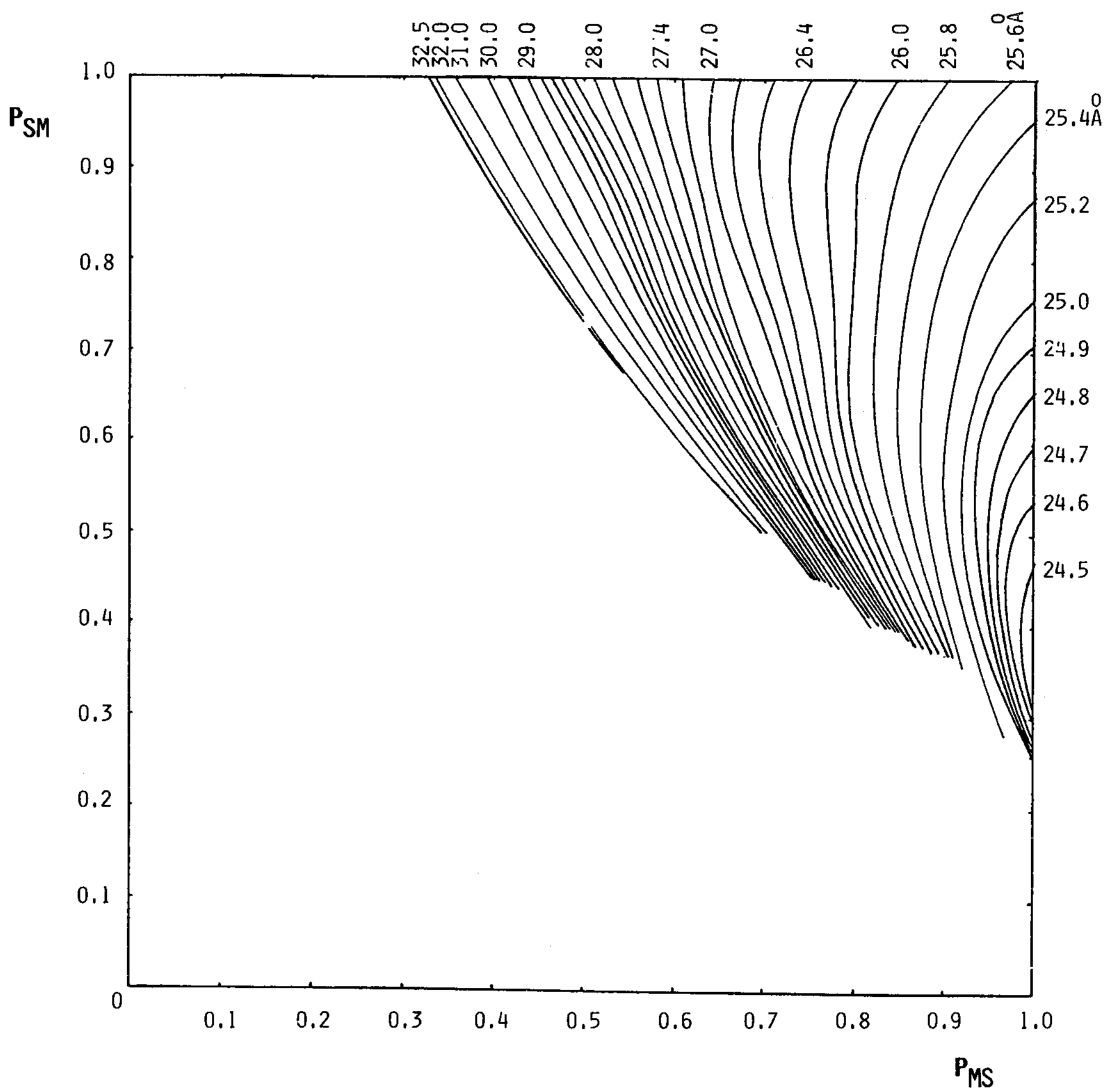

Figure 2. Diagrams of equal d-value lines for 32.5–24.5-Å reflection for quantification of mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

Using these diagrams in Figures 2 to 3, one can obtain P_M (probability of existence of mica layer) = .57, P_S (probability of existence of smectite layer) = .43, P_{MM} = .335, P_{MS} = .665, P_{SM} = .875, and P_{SS} = .125 for this specimen, where M is a mica layer and S is a smectite layer. These results agree well with those obtained using the MacEwan Fourier transform method by Tomita *et al.* (1969). The results obtained by the MacEwan Fourier transform method and the present method are listed in Table 1.

In practice, it is convenient to have the chart photographed on a transparent film or plate. The film then is placed over a sheet of graph paper and viewed against an illuminated opal glass background. The coordinates P_{MS} and P_{SM} may then be easily read for each d-value.

Samples not having high d-spacings. As an example of material not having high d-spacings, the following d-values published by Walker (1951) were used: 11.4, 4.56, 3.41, 3.34, and 2.62 Å. This specimen produced no XRD reflections with high d-spacings. For interstratified mica/smectites, the reflection near 3.30 Å is influenced by discrete illite. Treatment with ethylene glycol is necessary to check the reflections of interstratified minerals. Here, the 11.4-Å line was traced from the diagram in Figure 3, and the 3.34-Å line was traced

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

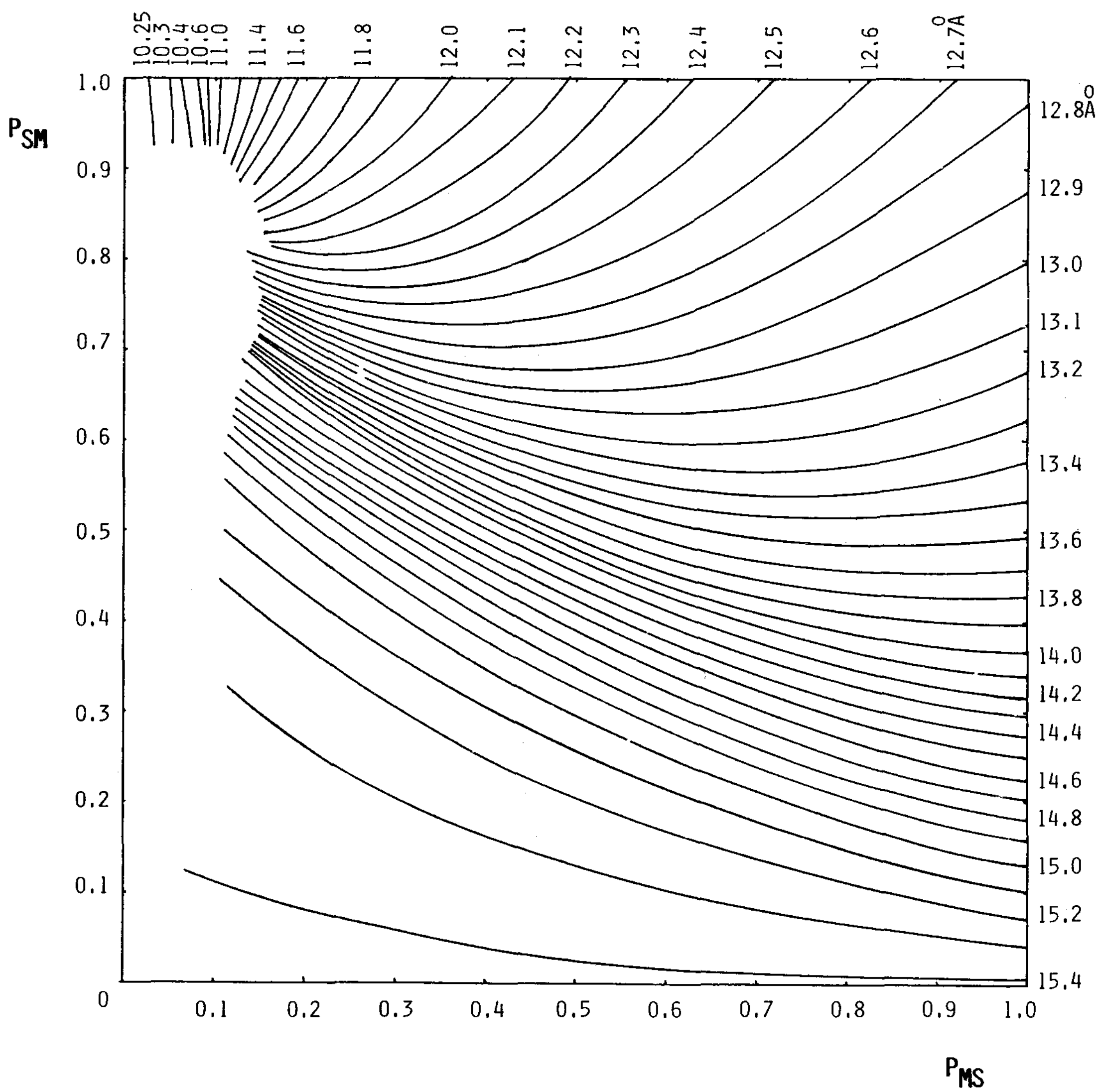

Figure 3. Diagram of equal d-value lines for 15.4–10.25-Å reflection for quantification of mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

Table 1. Comparison of interstratifications of sample described by Tomita *et al.* (1969) obtained by MacEwan's method and the present method.

	Present method	MacEwan method
P_M	.57	.56
P_S	.43	.44
P_{MM}	.335	.34
P_{MS}	.665	.66
P_{SM}	.875	.84
P_{SS}	.125	.16

P_M: probability of existence of mica layer. P_S: probability of existence of smectite layer. P_{MM}: probability that a mica layer succeeds a mica layer given that the first layer is mica layer. P_{MS}, P_{SM}, and P_{SS} are similarly defined.

from the diagram in Figure 4. From the intersection of the two lines, P_{MS} and P_{SM} values of .13 and .945 were obtained, respectively. The ratio of P_M:P_S is .945: .13, i.e., .88:.12. We obtained the following results for the specimen: P_M = .88, P_S = .12, P_{MM} = .87, P_{MS} = .13, P_{SM} = .945, and P_{SS} = .055.

Samples where two contour traces do not intersect

As an example of a sample where the tracings of two d-value contour lines do not intersect, data from Sato *et al.* (1965) were examined. An XRD pattern of the specimen showed reflections at 29.4, 11.9, 5.07, 4.51, and 3.28 Å. When the 29.4- and 11.9-Å curves were drawn using the diagrams in Figures 2 and 3, the two

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

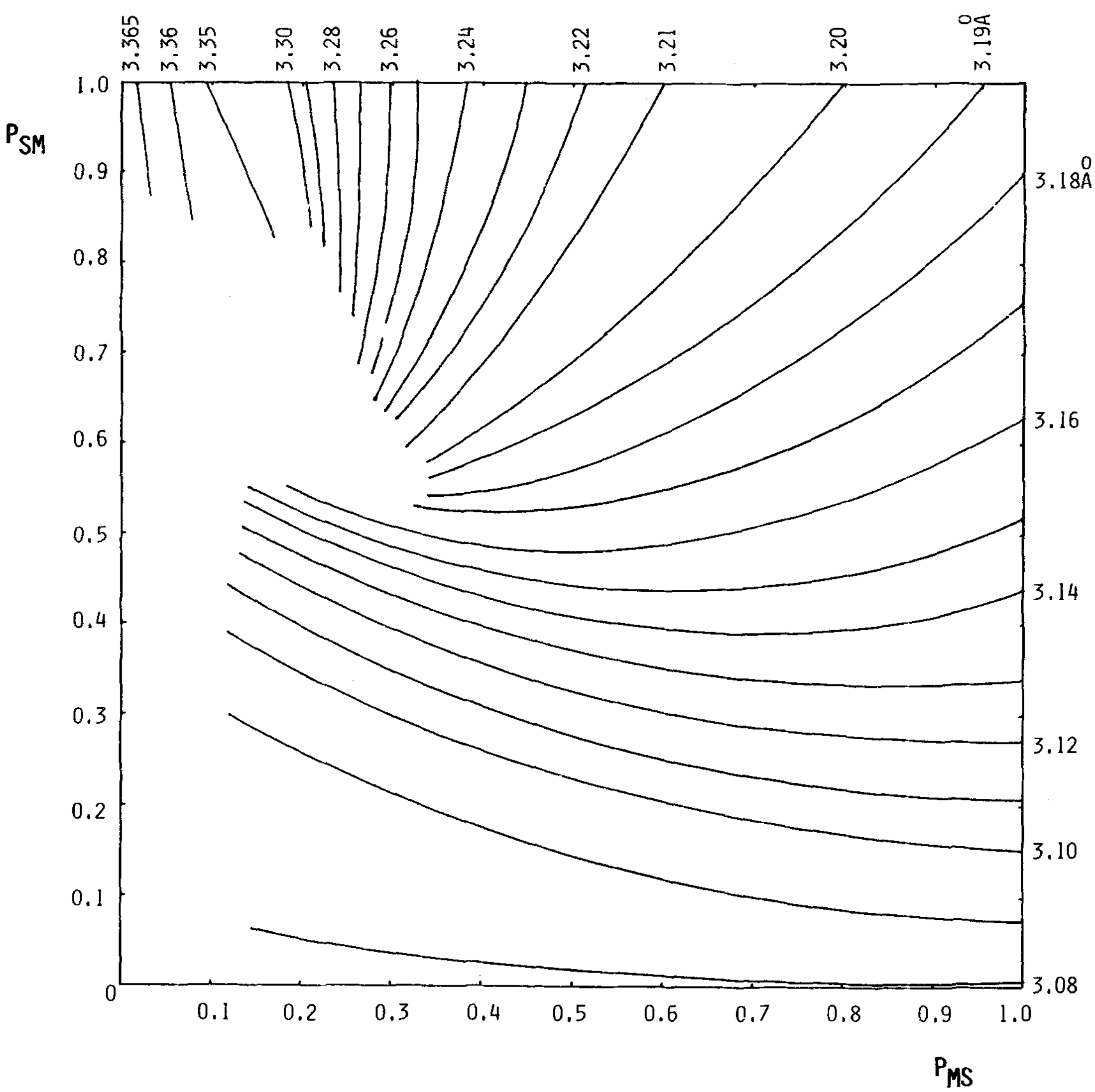

Figure 4. Diagram of equal d-value lines for 3.365–3.08-Å reflection for quantification of mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

curves did not intersect, as shown in Figure 6. Under these circumstances reading errors of peak positions were considered because a point of intersection should exist on the curve between B and C. XRD patterns were calculated (Figure 7) for several intersections on the curve between B and C, and interstratifications for their structures are listed in Table 2. As can be seen, these patterns are not markedly different from one another in the narrow range like the above case. Here, the intersection was estimated by considering the degree of reading errors of d-spacings of the two reflections. The d-values of the 001 and 002 reflections have certain widths due to reading errors of peak positions; hence, the d-value lines were broadened until the two lines intersected as shown in Figure 6. The two lines intersected at point A, and values of .37 for P_{MS} and 1 for P_{SM} were obtained. The quantification of the interstratification of the specimen (Table 3) agree well with the values reported by Sato *et al.* (1965).

Interstratifications can be determined for samples not having high d-spacings in the same manner. A specimen from Kamikita, Japan, investigated by Shimoda *et al.* (1969), gave XRD reflections at 11.2 and 3.30 Å. Values of .14 for P_{MS} and 1 for P_{SM} were obtained using the diagrams in Figures 3 and 4, as shown in Figure 8. Thus, the interstratification of the specimen is as follows: $P_M = .88$, $P_S = .12$, $P_{MM} = .86$, $P_{MS} = .14$, $P_{SM} = 1$, and $P_{SS} = 0$.

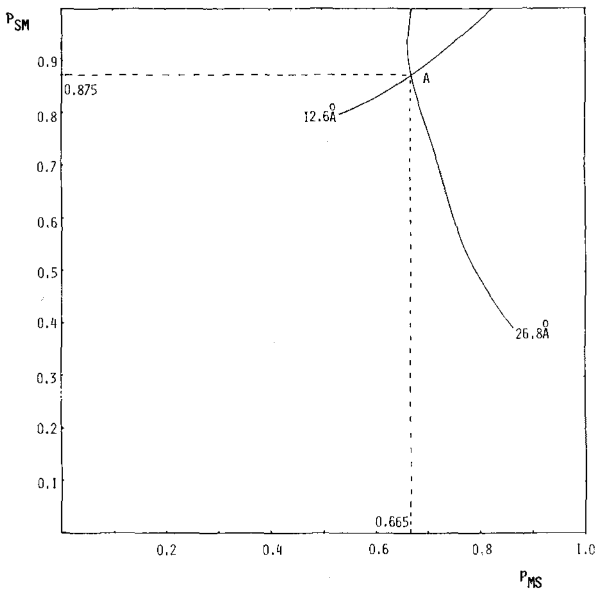

Figure 5. Plot of 26.8- and 12.6-Å lines using the diagrams of Figures 2 and 3 for quantification of the mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

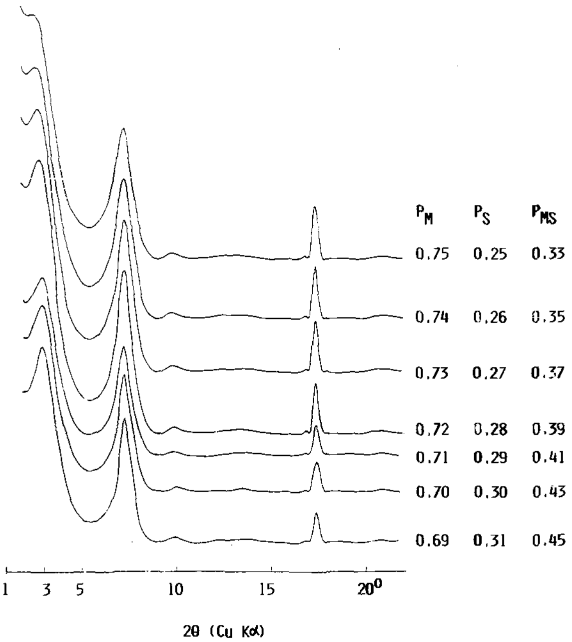

Figure 7. Caluclated X-ray powder diffraction patterns of some interstratified structures having values of P_{MS} and P_{SM} on the line between B and C in the Figure 6. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

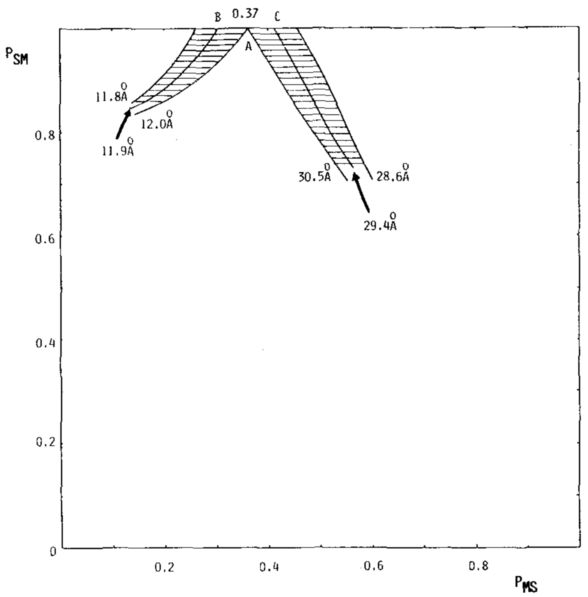

Figure 6. Plot of 29.4- and 11.9-Å lines having certain widths using the diagrams of Figures 2 and 3 for quantification of the mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

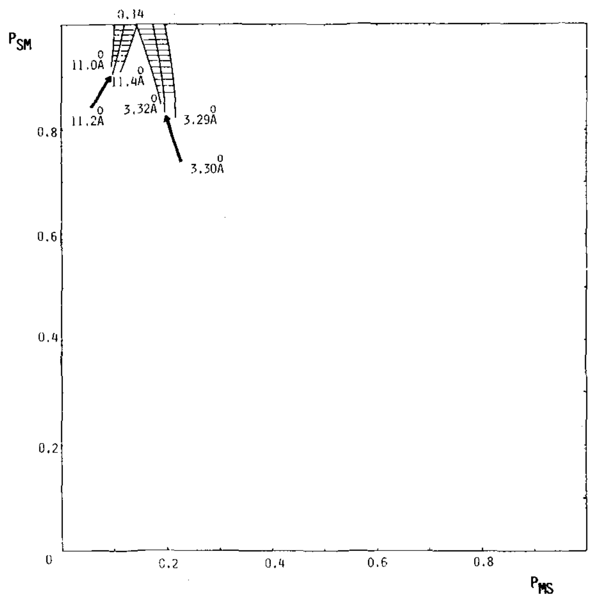

Figure 8. Plot of broad 11.2- and 3.30-Å lines using the diagrams of Figures 3 and 4 for quantification of the mica/smectite interstratifications. P_{MS} is probability that a smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

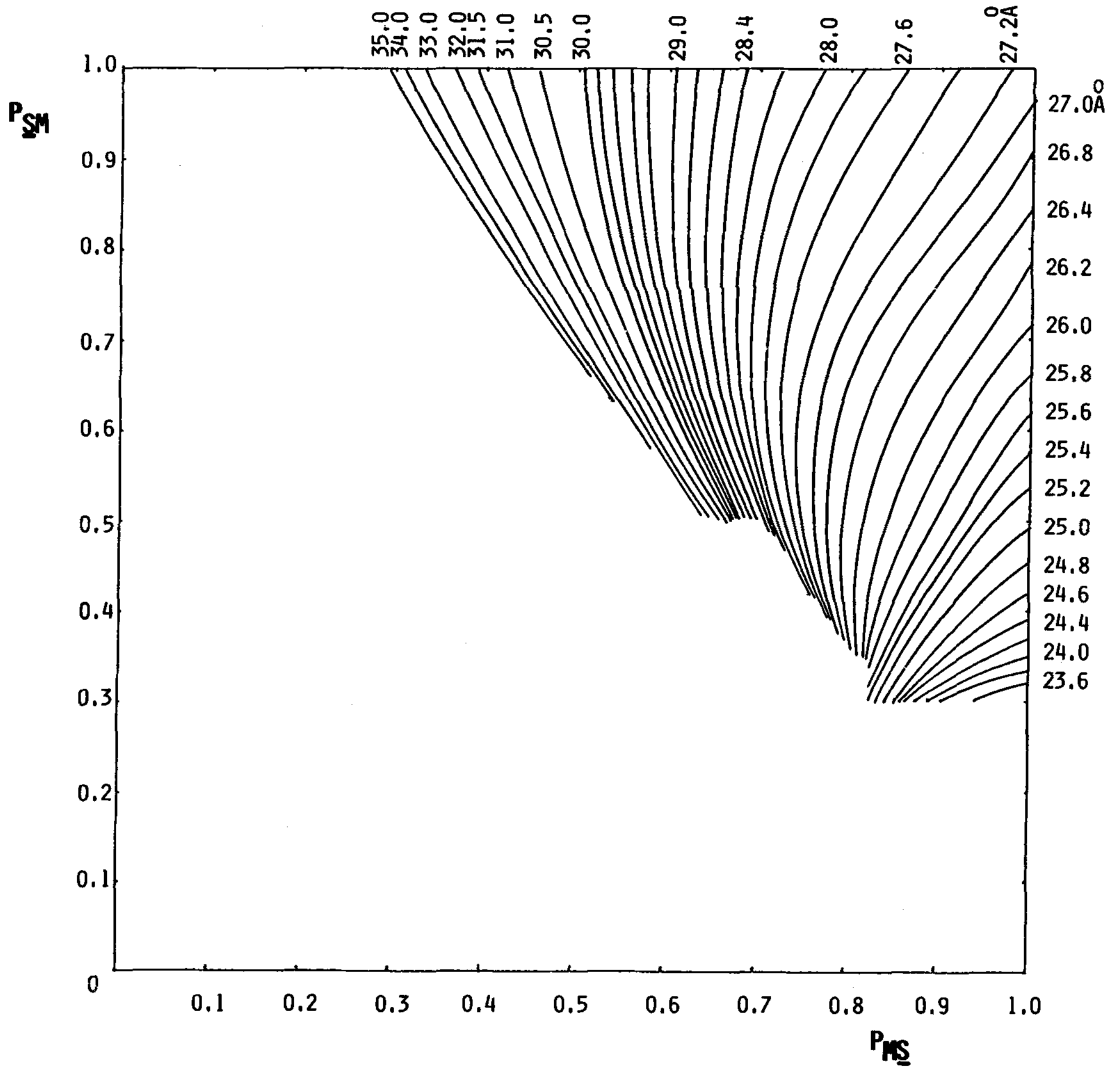

Figure 9. Diagram of equal d-value lines for 35.0–23.6-Å reflection for quantification of mica/glycolated-smectite interstratifications. P_{MS} is probability that a glycolated-smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

Table 2. Calculated interstratifications of some interstratified structures having values of P_{MS} and P_{SM} on the curve between B and C in Figure 6.

P_M	P_S	P_{MM}	P_{MS}	P_{SM}	P_{SS}	d(001) (Å)	d(002) (Å)
.75	.25	.67	.33	1		32.9	11.98
.74	.26	.65	.35	1	0	31.55	12.0
.73	.27	.63	.37	1	0	30.8	12.02
.72	.28	.61	.39	1	0	30.1	12.07
.71	.29	.59	.41	1	0	29.6	12.10
.70	.30	.57	.43	1	0	29.3	12.14
.69	.31	.55	.45	1	0	28.9	12.15

P_M: probability of existence of mica layer. P_S: probability of existence of smectite layer. P_{MS}: probability that a smectite layer succeeds a mica layer given that the first layer is mica layer. P_{MM}, P_{SM}, and P_{SS} are similarly defined.

MICA/GLYCOLATED-SMECTITE INTERSTRATIFICATIONS

Because expandable layers are commonly not uniform from sample to sample, XRD intensities must be calculated for interstratified structures containing a smectite component expanded with ethylene glycol. The disadvantages of using ethylene glycol are: (1) ethylene glycol is not suitable for vermiculite-type complexes; and (2) it is difficult to read accurately the d-value of a reflection (13.0–16.0 Å) of interstratified mica/glycolated-smectites when chlorite is present in the specimen. The advantages of using ethylene glycol complexes are: (1) two-layer complexes are relatively stable under room conditions; and (2) the intensities

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

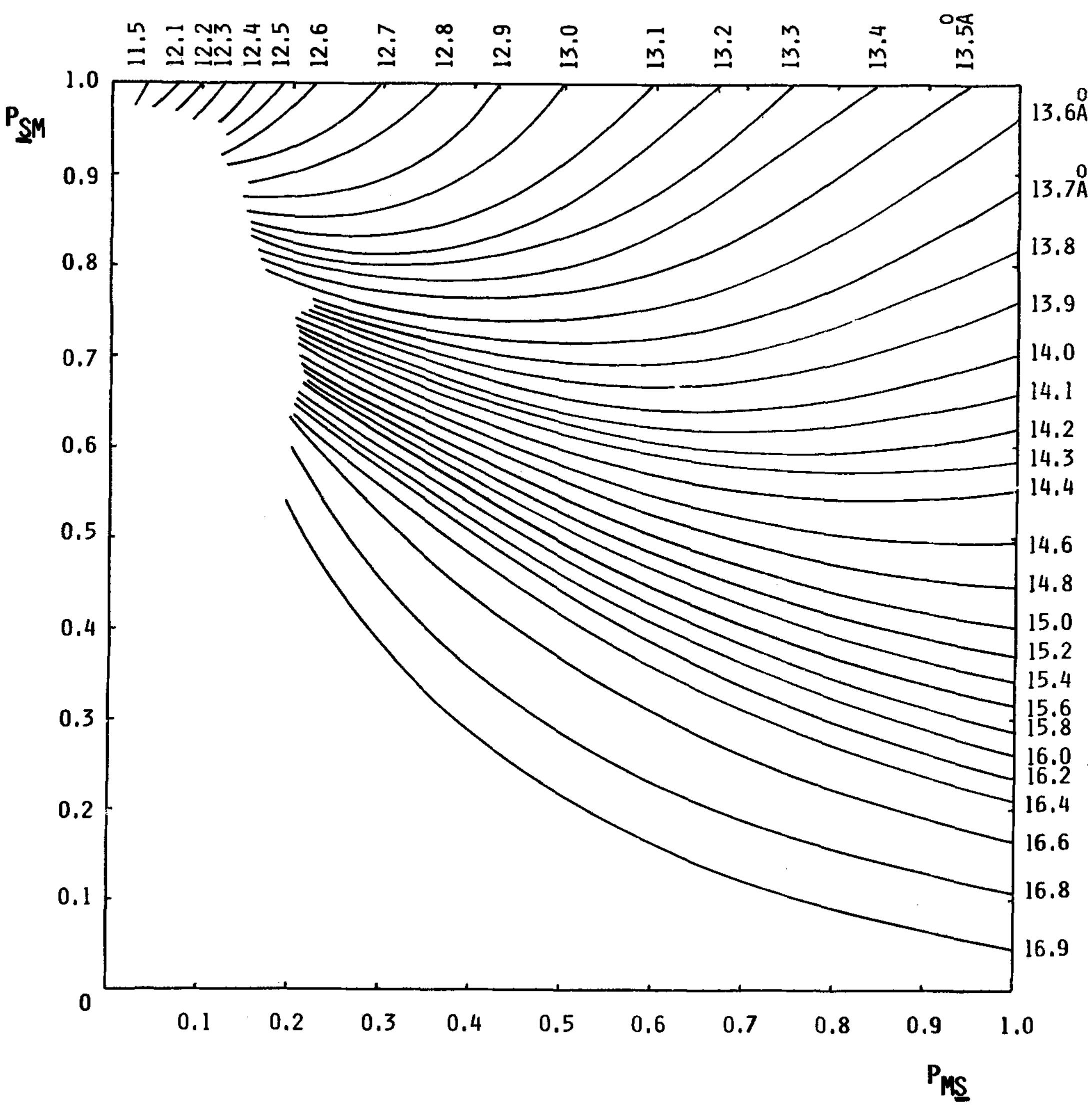

Figure 10. Diagram of equal d-value lines for 16.9–11.5-Å reflection for quantification of mica/glycolated-smectite interstratifications. P_{MS} is probability that a glycolated-smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

Table 3. Interstratification of the 29.4-Å interstratified mineral described by Sato *et al.* (1965).

	Present study	Sato *et al.* (1965)
P_M	.73	.72
P_S	.27	.28
P_{MM}	.63	.611
P_{MS}	.37	.389
P_{SM}	1	1
P_{SS}	0	0

P_M: probability of existence of mica layer. P_S: probability of existence of smectite layer. P_{MS}: probability that a smectite layer succeeds a mica layer given that the first layer is mica layer. P_{MM}, P_{SM}, and P_{SS} are similarly defined.

of second and higher order reflections are intensified. Three diagrams for interstratified mica/glycolated-smectites were constructed from the calculated data and are shown in Figures 9, 10, and 11, respectively. These diagrams were applied to glycolated specimens instead of the diagrams shown in Figures 2, 3, and 4, respectively, in the same manner as mentioned above.

CHLORITE/SMECTITE INTERSTRATIFICATIONS

The calculation of intensities for various interstratified structures of chlorite/smectite was based on the model shown in Figure 12. A smectite structure having

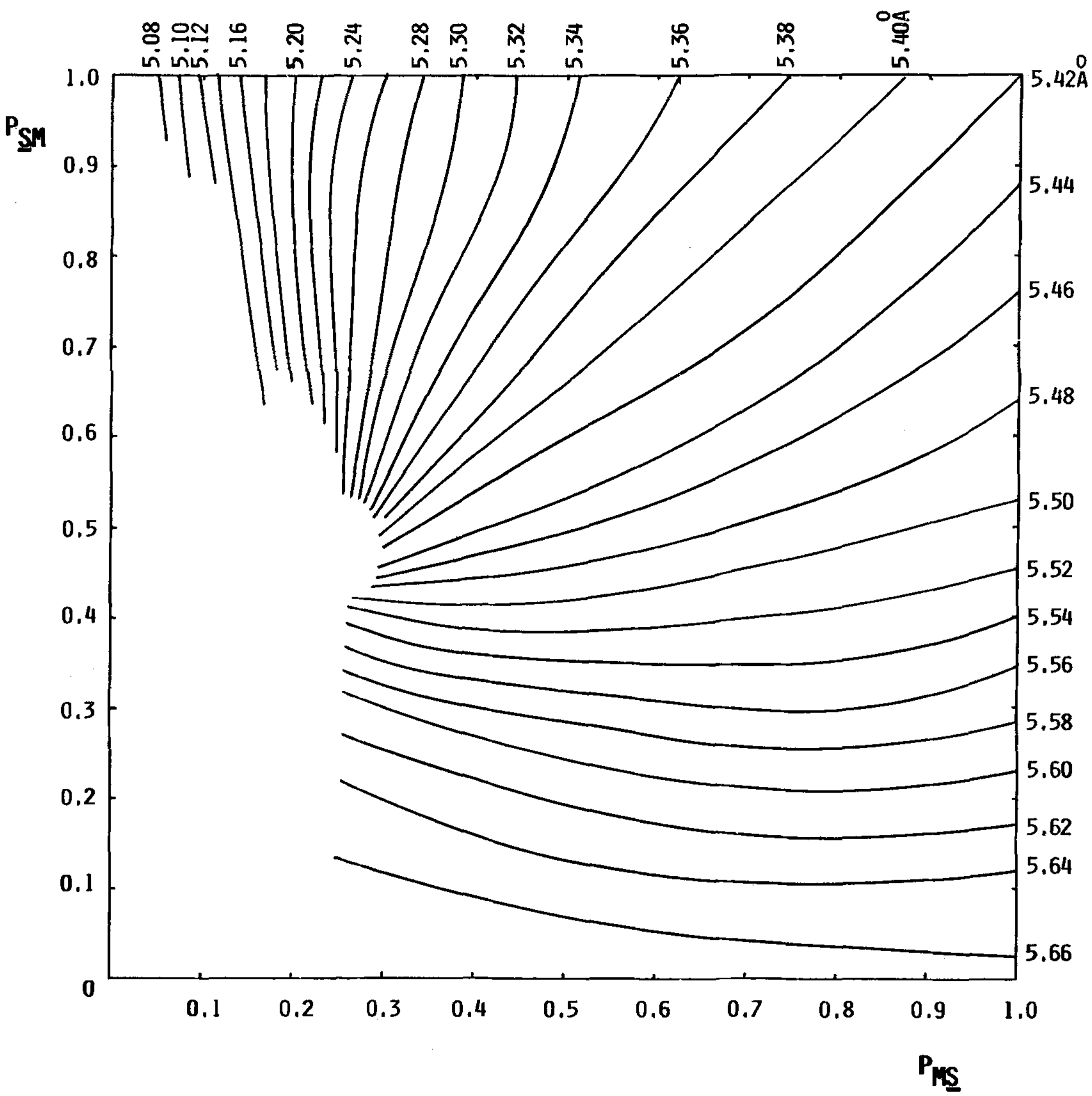

Figure 11. Diagram of equal d-value lines for 5.66–5.08-Å reflection for quantification of mica/glycolated-smectite interstratifications. P_{MS} is probability that a glycolated-smectite layer succeeds a mica layer given that the first layer is mica layer; P_{SM} is similarly defined.

ethylene glycol molecules in interlayers was used in the calculation. A value of 17 Å for an ethylene glycol-smectite layer was used. The 16.6–17.2-Å range found in natural smectites (Środoń, 1980) was not taken into account to simplify the calculations. XRD intensities were calculated for many possibilities by changing existing probabilities and transition probabilities for the chlorite and smectite layers. Three diagrams of equal d-value lines were constructed in the same way as for interstratified mica/smectites and are shown in Figures 13, 14, and 15, respectively. Practical determinations of layer sequences for chlorite/smectites using these diagrams are the same as mentioned above.

For a glycolated interstratification of chlorite/smectite having the 31.6- and 15.6-Å XRD reflections reported by Sudo (1954), the interstratification of the specimen was determined as follows: point A was determined as the intersection of the 31.6- and 15.6-Å lines using the diagrams of Figures 13 and 14, as shown in Figure 16. From Figure 16 the following values were determined: P_C:P_S = 1:.935, i.e., .517:.483. Accordingly, P_C:P_S = .52:.48, P_{CS} = .935, P_{CC} = .065, P_{SC} = 1, and P_{SS} = 0, where P_C is the probability of the existence of a chlorite layer, P_S is that of a smectite layer, and P_{CS} is the probability that a smectite layer succeeds a chlorite layer, assuming that the first layer is chlorite. P_{CC}, P_{SC}, and P_{SS} are similarly defined.

For chlorite/smectites not showing high d-spacings

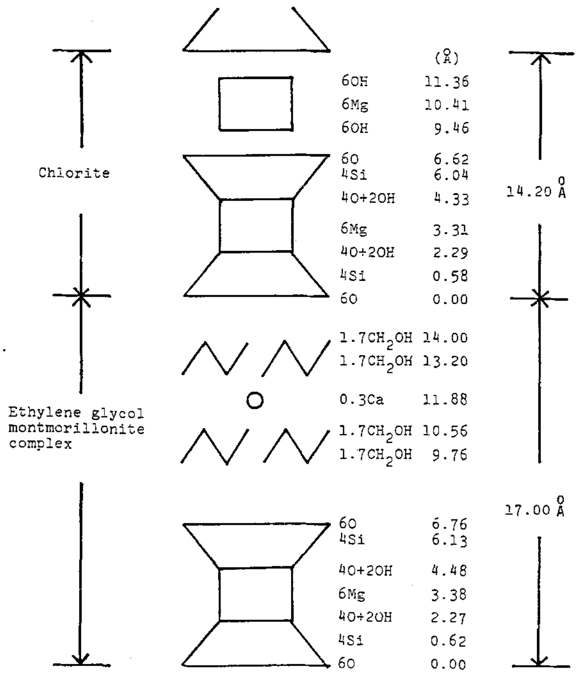

Figure 12. Chlorite and glycolated-smectite models used in calculation of X-ray powder diffraction intensity.

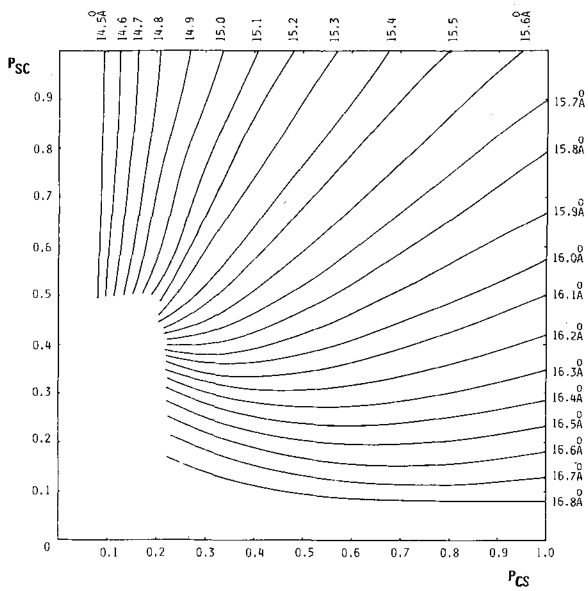

Figure 14. Diagram of equal d-value lines for 16.8–14.5-Å reflection for quantification of chlorite/glycolated-smectite interstratifications. P_{CS} is probability that a glycolated-smectite layer succeeds a chlorite layer given that first layer is chlorite layer; P_{SC} is similarly defined.

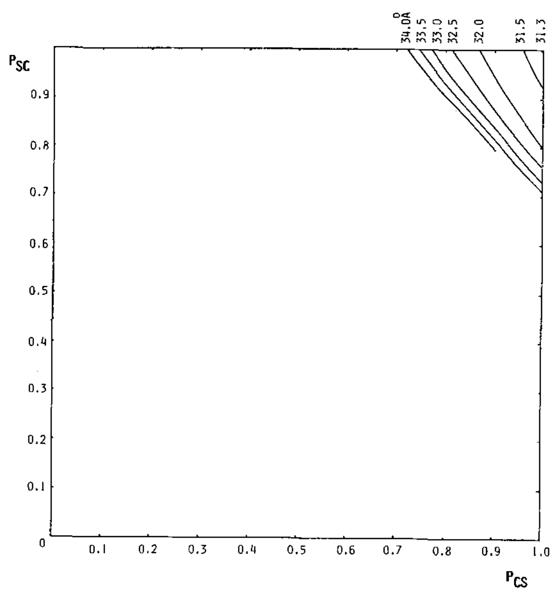

Figure 13. Diagram of equal d-value lines for 34.0–31.3-Å reflection for quantification of chlorite/glycolated-smectite interstratifications. P_{CS} is probability that a glycolated-smectite layer succeeds a chlorite layer given that the first layer is chlorite layer.

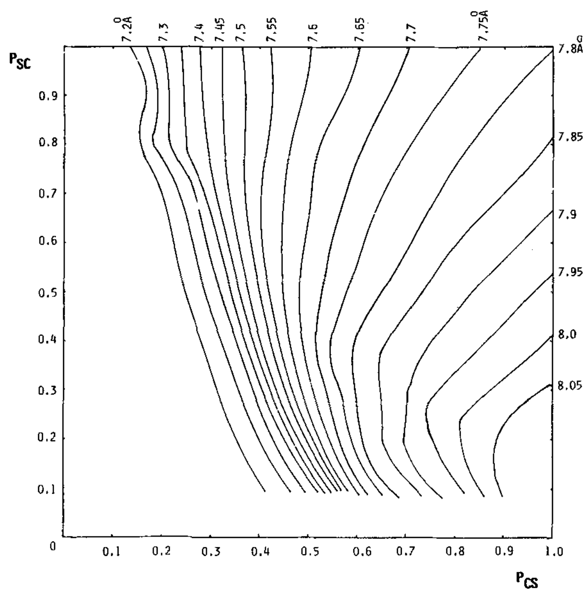

Figure 15. Diagram of equal d-value lines for 8.05–7.2-Å reflection for quantification of chlorite/glycolated-smectite interstratifications. P_{CS} is probability that a glycolated-smectite layer succeeds a chlorite layer given that the first layer is chlorite layer; P_{SC} is similarly defined.

https://doi.org/10.1346/CCMN.1985.0330502 Published online by Cambridge University Press

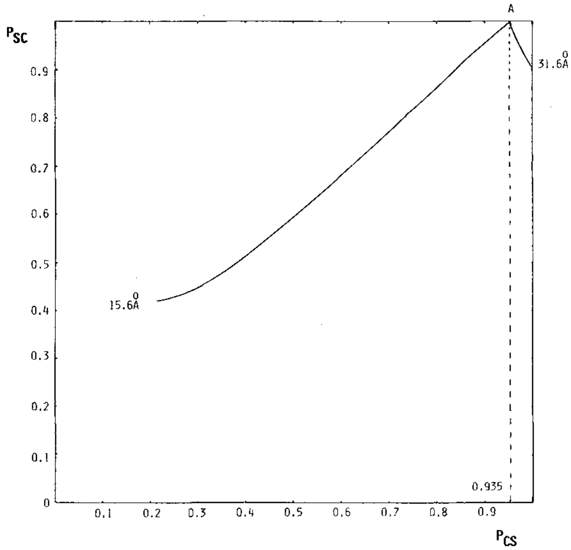

Figure 16. Plot of 31.6- and 15.6-Å lines using the diagrams of Figures 13 and 14 for quantification of the chlorite/glycolated-smectite interstratifications. P_{CS} is probability that a glycolated-smectite layer succeeds a chlorite layer given that the first layer is chlorite layer; P_{SC} is similarly defined.

in their XRD patterns, interstratified structures can be determined by using the diagrams of Figures 14 and 15.

DISCUSSION

The layer sequences of interstratified mica/smectite and chlorite/smectites were determined easily using the diagrams developed in this study. These diagrams are useful for the rapid quantification of layer sequences in many interstratified minerals. Differences in chemical compositions of elementary layers of interstratified minerals did not influence the determination of interstratification using these diagrams. Composition only affects the intensities of reflections and has no influence on the d-values of reflections. For interstratified mica/smectite, however, the kind of cation in the interlayers of the expandable component may influence the d-spacing of the expandable layer. If cations such as Na^+ and K^+ are dominant in the interlayers of the expandable component, the d-spacings of the expandable layers are smaller than the d-spacing used in the calculations in this paper. To quantify the interstratification of such minerals, they should be washed with a solution containing Ca^{2+} or Mg^{2+} before obtaining the XRD patterns. Two sets of diagrams for interstratified mica/smectites have been developed: one for mica/smectites and the other for mica/glycolated-smectites. The advantages of using ethylene glycol complexes are: (1) increased intensities of second- and higher-order reflections; and (2) the relative stability of the two-layer complexes under room conditions. Ethylene glycol complexes are less suitable for specimens containing chlorite because it is not easy to read accurately the d-value of a reflection in the 13.0–16.0-Å range for mica/glycolated-smectites. For mica/smectites, the reflection near 3.30 Å is influenced by discrete illite; hence, treatment with ethylene glycol is necessary. XRD intensities of interstratified structures were calculated using an N value of 20. If crystallites are thin, the peaks will shift from their normal positions. If it is still impossible to quantify the interstratification of such materials even after an ethylene glycol treatment, the sample must contain three or more kinds of layers, i.e., have a Reichweite value >1 and layer spacings much different from the model. It is impossible to determine the interstratification of such specimens using the present diagrams. Diagrams for such samples are in preparation. The authors used a spacing of 15.4 Å for smectite; however the spacing of smectite is not uniform from sample to sample and depends on relative humidity, the kinds of cation in interlayers, and differences in layer charges. A value of 17 Å was used for the ethylene glycol-smectite layer as was used by previous workers, although Środoń (1980) reported a range of 16.6–17.2 Å for natural smectites. Some error may be expected because of this simplification as mentioned by Środoń (1980).

ACKNOWLEDGMENTS

This study was supported in part by a Grant-in-Aid for Scientific Research from the Ministry of Education. The authors thank R. C. Reynolds and J. Środoń for many valuable comments and critical review of the manuscript. F. A. Mumpton kindly improved the English of this manuscript.

REFERENCES

Allegra, G. (1964) The calculation of the intensity of X-rays diffracted by monodimensionally disordered structures: *Acta Crystallogr.* **17**, 579–586.

Cradwick, P. D. (1975) On the calculation of one-dimensional X-ray scattering from interstratified material: *Clay Miner.* **10**, 347–356.

Drits, V. A. and Sakharov, B. A. (1976) *X-ray Structural Analysis of Mixed-Layer Minerals:* Acad. Sci. U.S.S.R., Moscow, 256 pp. (in Russian).

Hendricks, S. B. and Teller, E. (1942) X-ray interference in partially ordered lattices: *J. Chem. Phys.* **10**, 147–167.

Kakinoki, J. and Komura, Y. (1952) Intensity of X-ray diffraction by a one-dimensionally disordered crystal. I. General derivation in cases of the "Reichweite" S=0 and 1: *J. Phys. Soc. Japan* **7**, 30–35.

Kakinoki, J. and Komura, Y. (1954a) Intensity of X-ray diffraction by a one-dimensionally disordered crystal. II. General derivation in the case of the correlation range S=2: *J. Phys. Soc. Japan* **9**, 169–176.

Kakinoki, J. and Komura, Y. (1954b) Intensity of X-ray diffraction by a one-dimensionally disordered crystal. III. The close-packed structure: *J. Phys. Soc. Japan* **9**, 177–183.

Kakinoki, J. and Komura, Y. (1965) Diffraction by a one-dimensionally disordered crystal. I. The intensity equation: *Acta Crystallogr.* **19,** 137–147.

Kodama, H. (1962) Interpretation of X-ray powder patterns of some hydromuscovites from Japan with reference to their alkali contents: *Clay Sci.* **1,** 89–99.

MacEwan, D. M. C. (1956) Fourier transform methods. I. A direct method of analysing interstratified mixtures: *Kolloidzeitschr.* **149,** 96–108.

MacEwan, D. M. C. (1958) Fourier transform methods. II. Calculation of diffraction effects for different types of interstratification: *Kolloidzeitschr.* **156,** 61–67.

MacEwan, D. M. C., Ruiz Amil, A., and Brown, G. (1961) Interstratified clay minerals: in *The X-Ray Identification and Crystal Structures of Clay Minerals,* G. Brown, ed., Mineralogical Society, London, 393–445.

Reynolds, R. C. (1967) Interstratified clay systems: calculation of the total one-dimensional diffraction function: *Amer. Mineral.* **52,** 661–672.

Reynolds, R. C. (1980) Interstratified clay minerals: in *Crystal Structures of Clay Minerals and Their X-Ray Identification,* G. W. Brindley and G. Brown, eds., Mineralogical Society, London, 249–303.

Reynolds, R. C. and Hower, J. (1970) The nature of interlayering in mixed-layer illite-montmorillonite: *Clays & Clay Minerals* **18,** 25–36.

Sato, M. (1969) Interstratified structures: *Z. Kristallogr.* **129,** 388–395.

Sato, M. (1973) X-ray analysis of interstratified structure: *Nendo Kagaku (Jour. Clay Soc. Japan)* **13,** 39–47 (in Japanese).

Sato, M., Oinuma, K., and Kobayashi, K. (1965) Interstratified minerals of illite and montmorillonite: *Nature* **208,** 179–180.

Shimoda, S., Sudo, T., and Oinuma, K. (1969) Differential thermal analysis curves of mica clay minerals: in *Proc. Int. Clay Conf., Tokyo, 1969, Vol. 1,* L. Heller, ed., Israel Univ. Press, Jerusalem, 197–206.

Środoń, J. (1980) Precise identification of illite/smectite interstratifications by X-ray powder diffraction: *Clays & Clay Minerals* **28,** 401–411.

Sudo, T. (1954) Long spacing at about 30 Å confirmed from certain clays from Japan: *Clay Miner. Bull.* **2,** 193–203.

Sudo, T. and Shimoda, S. (1978) *Clays and Clay Minerals of Japan:* Elsevier, Amsterdam, p. 55.

Takahashi, H. (1982) Electronic computer's program for calculation of diffracted intensity by close-packed structures with stacking faults: *Bull. Fac. Educ. Kagoshima Univ.* **34,** 1–14 (in Japanese).

Tettenhorst, R. and Grim, R. E. (1975a) Interstratified clays, I. Theoretical: *Amer. Mineral.* **60,** 49–59.

Tettenhorst, R. and Grim, R. E. (1975b) Interstratified clays, II. Some experimental results: *Amer. Mineral.* **60,** 60–65.

Tomita, K., Yamashita, H., and Oba, N. (1969) An interstratified mineral found in altered andesite: *J. Japan. Assoc. Min. Pet. Econ. Geol.* **61,** 25–34.

Walker, G. F. (1951) *The X-ray Identification and Crystal Structures of Clay Minerals:* Mineralogical Society, London, p. 222.

Watanabe, T. (1981) Identification of illite/montmorillonite interstratifications by X-ray powder diffraction: *J. Mineral. Soc. Japan* **15,** 32–41 (in Japanese with English abstract).

(Received 21 September 1984; accepted 23 February 1985; Ms. 1415)