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Recognizing illitization progress from diagenesis to very low-grade metamorphism in rocks of the Cantabrian Zone (Spain)

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Abstract

The various sub-populations of illitic assemblages have been investigated, using X-ray diffraction methods, in a suite of Paleozoic pelitic rocks from the Cantabrian Zone, the external part of the Iberian Variscan belt (NW Spain) ranging in age from Cambrian to Carboniferous. Cambrian to Westphalian A-B strata represent a pre-tectonic succession, deposited prior to the Variscan deformation, whereas the Westphalian C-D and Stephanian strata are syn-tectonic and were deposited during the Variscan deformation.

Diffraction data were treated using decomposition methods and the peaks identified were rationalized in terms of specific discrete or mixed-layer illite/smectite phases. Excluding the 14-Å chlorite or chlorite/vermiculite (C/V) peak, three and sometimes four, elementary peaks are required for a good fit to the complex area of the diffraction pattern between 5 and 11 °2θ CuKα. A peak at high 2θ value (~ 8.79 °2θ) can be attributed to well-crystallized illite (WCI), whereas a wider peak at lower 2θ values (~ 8.43 °2θ) is attributed to a poorly-crystallized illite (PCI). A third elementary peak (~ 7.80 °2θ) is mixed-layer I/S that has varying percentages of the two components. The variation in the width of the WCI peak is smaller than that of the PCI peak. In the pre-tectonic succession, both peaks increase in width, as the rocks become stratigraphically younger. In the syn-tectonic samples peaks obtained by decomposition are considerably narrower than in the older pre-tectonic rocks. We interpret these narrower peaks as the signature of detrital white micas inherited from the rapidly uplifting Variscan mountain chain. These micas apparently avoided any significant chemical weathering throughout their short transport history. The I/S peaks(s) are regarded as recording the new phases formed during the retrograde evolution of these detrital micas. The pre-tectonic sequence is interpreted similarly. The narrower peak (WCI) in the older rock samples of the pre-tectonic succession would correspond to detrital white micas. The increase in width with decreasing age reflects their retrograde evolution. The other peaks (PCI and I/S) would record the evolution of newly-formed phases. It could be expected that at thermodynamic equilibrium the width of both populations would attain the same value. Such a tendency can be observed in the Cambrian-Silurian samples studied. With increasing metamorphic grade crystallite thickness distributions (CTDs) change from asymptotic to log-normal. This change implies a modification in the mechanism of crystal growth, and may in fact be more important for establishing the transition from diagenesis to metamorphism than the width of either the unresolved or the decomposed peaks.

Keywords: Illite, X-ray diffraction, decomposition, crystallite thickness distributions (CTDs).

1. Introduction

The boundary between diagenesis and metamorphism in pelitic rocks has been defined using omnipresent minerals such as illite or the illite/smectite mixed-layer mineral series (KÜBLER, 1967; KISCH, 1983; BLENKINSOP, 1988; FREY, 1987; FREY and ROBINSON, 1999; JABOYEDOFF et al., 2001; KÜBLER and GOY-EGGENBERGER, 2001 and references therein). However, in such rocks detrital grains may occur alongside phases formed during burial diagenesis (BAILEY, 1966). Although X-ray diffraction (XRD) methods bias results towards smaller crystals (i.e. neoformed), complete separation of the neoformed and detrital components is impossible. Thus, for example, the 10-Å illite peak may be composed of contributions from both types of components, resulting in complex XRD curves (STERN at al., 1991; LANSON and BESSON, 1992; LANSON et al., 1995, 1996; WANG et al., 1995; GHARRABI et al., 1998).

This paper utilizes XRD methods to investigate more closely the evolution of the different components that generate these curves during the

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course of the evolution of illitic phases and describes the characteristics of the various sub-populations forming the illitic assemblages.

2. Materials and methods

In this study we have investigated a suite of Paleozoic pelitic rocks from the Cantabrian Zone, the external part of the Iberian Variscan belt (NW Spain) ranging in age from Cambrian to Carboniferous. Cambrian to Westphalian A-B age represent a pre-tectonic succession, deposited prior to the Variscan deformation, whereas the Westphalian C-D and Stephanian strata are syn-tectonic and were deposited during the Variscan deformation. These rocks have reached upper anchizone grades, with rare epizonal grades, as the result of sedimentary burial. In most of the area peak temperatures predated deformation (e.g. BRIME and PÉREZ-ESTAÚN, 1980; BRIME, 1981, 1985; GARCÍA-LÓPEZ et al., 1997; BASTIDA et al., 1999; BRIME et al., 2001; BASTIDA et al., 2002 and references therein). We have grouped the samples into age intervals, according to previous studies, as follows: Cambrian-Silurian; Lower Devonian; Middle–Upper Devonian; Carboniferous (Westphalian A-B); Carboniferous (Westphalian C-D) and Stephanian.

X-ray diffraction patterns of the <2 µm fraction of the Paleozoic rocks in question were obtained using a Philips 1710 diffractometer equipped with graphite monochromator and using CuKα radiation. Count data were collected at intervals of 0.02 °2θ for at least 2 s. The instrument was controlled through a PC system using the standard Philips automatic powder diffraction software (APD) V3.6.

Profile fitting to the ~10-Å asymmetric illitic multiphase peak in the range 5–11°2θ was undertaken using the Philips APD software which utilizes a Lorentzian approach (SCHREINER and JENKINS, 1983). Profile decomposition was performed on both air-dried and glycolated preparations using decomposition methods outlined by LANSON and CHAMPION (1991), LANSON and BESSON (1992) and ROBINSON and BEVINS (1994), and the peaks identified were rationalized in terms of specific discrete or mixed-layer illite/smectite phases. The quality of the fit was evaluated using the χ^2 parameter and a visual approach. A fit was considered to be good when the fitted profile was located within the noise of the experimental data, the general shape of the experimental pattern was maintained and the residual was uniform and low.

All decompositions were performed with Gaussian-shaped peaks. Despite contrary statements in the literature (HOWARD and PRESTON, 1989; STERN et al., 1991; WANG et al., 1995) some authors (LANSON and BESSON, 1992; LANSON and VELDE, 1992; ROBINSON and BEVINS, 1994) suggested that the peak shapes of poorly crystallized illite and illite/smectite are in fact symmetrical in the range between 5 and 11°2θ. For comparative purposes a few of the raw patterns were treated with DECOMPXR, the decomposition program developed by LANSON (1990, 1992). Details of the calculation algorithms used by DECOMPXR, can be found in LANSON (1990). The agreement of the results using Philips APD software and DECOMPXR was considered an indication of their consistency.

Diffraction profiles were calculated using NEWMOD package (REYNOLDS, 1985; REYNOLDS and REYNOLDS, 1996). Comparison of experimental and calculated patterns provides an additional corroboration of the phases identified.

A few samples were selected for analysis using the MudMaster computer program (EBERL et al., 1996) to evaluate the evolution of the thickness, i.e., dimension along the *z* direction of the coherent diffracting domain. This program determines crystallite thickness distributions (CTDs) by the Bertaut-Warren-Aberbach method (DRITS et al., 1998). There are three basic CTD shapes (EBERL et al., 1998b; KILE et al., 2000); *(1) asymptotic,* with greatest frequencies in the smallest size classes, decreasing exponentially as size increases; *(2) log-normal* with crystal sizes normally distributed with a positive skew towards larger sizes; and (3) *universal steady-state shape* with a negative skew. These shapes can be related to different growth mechanisms (EBERL et al., 1998b; KILE et al., 2000; KILE and EBERL, 2000; BRIME and EBERL, 2002). Prior to the X ray study, samples were saturated with Na, and the <2 µm size fraction was separated by centrifugation. Polyvinylpyrrolidone (PVP)-intercalated samples were prepared according to EBERL et al. (1998a) and X-rayed on single crystal Si-wafers. Samples were scanned from 4 to 10 °2θ. The step size was 0.02 °2θ with a count time of 10 s per step.

3. Results

Excluding the 14-Å chlorite, or C/V peaks, three and sometimes four, elementary peaks are required for a good fit to the complex area of the diffraction pattern between 5 and 11 °2θ CuKα (Fig. 1). A first narrow peak, at higher 2θ values (~8.79 °2θ), is unaffected by glycolation, whereas a second wider peak occurs at lower 2θ values (~8.43 °2θ). Positions of these peaks indicate that

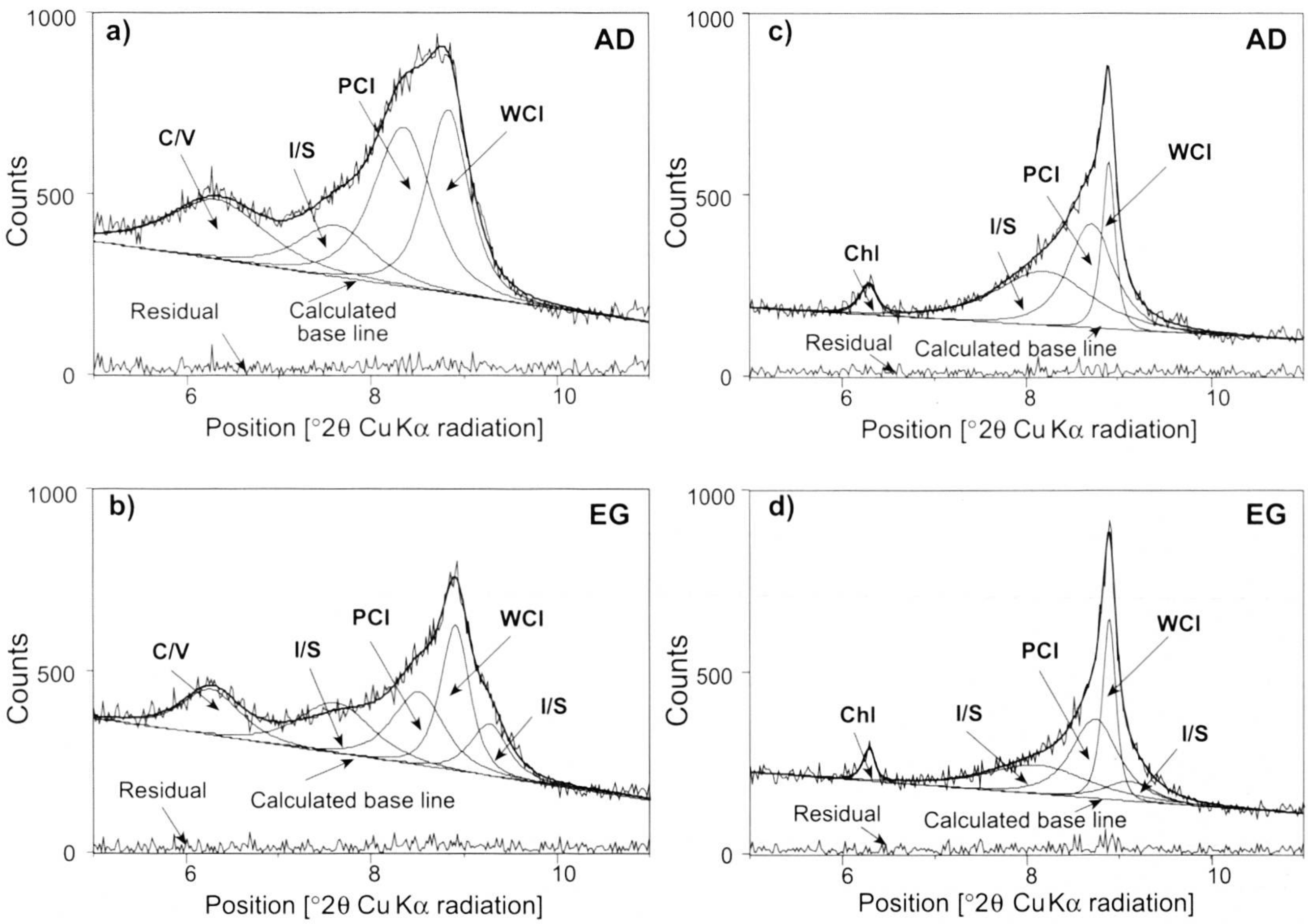

Fig. 1 Illustration of the component peaks of the complex XRD bands of Lower Devonian (a, b) and syn-tectonic Carboniferous (c, d) samples (B299 and CC4 respectively). AD — pattern from air-dried sample; EG — pattern from ethylene glycol-solvated sample; C/V — chlorite/vermiculite mixed-layers; Chl — chlorite; I/S — illite/smectite mixed layer; PCI — poorly-crystallized illite; WCI — well-crystallized illite; thick line — fitted profile (sum of all elementary peaks).

this phase may have up to 12% smectite, although this small amount does not significantly modify the position of the peaks in patterns of the glycol-solvated samples. Following LANSON et al. (1996), we adopted the terms "well-crystallized illite" (WCI) and "poorly-crystallized illite" (PCI), respectively, to describe these two peaks. A third elementary peak (~7.80 °2θ) is a mixed-layer I/S that has varying amounts of smectite but never more than 25%. An extra peak may be required to obtain a satisfactory fit for the glycolated sample, since a new peak appears on the high angle side of the 10-Å peak (Fig. 1b).

The three phases described above are clearly discriminated by their position within each of the groups of samples considered (Fig. 2). However the fields overlap when the groups are compared because both PCI and I/S show a higher *d* value as the rocks become stratigraphically younger. The degree of overlap, even within the same group, is greater when width is considered (Fig. 3). Peak widths decrease from I/S to PCI and to WCI populations, with the I/S populations showing the greater variability. The widths of PCI and I/S increase as rocks become stratigraphically younger, although this trend terminates at the top of the Westphalian A-B rocks, that is, at onset of the syn-tectonic Carboniferous sequences (Fig. 3). WCI shows very little variation in both position and width throughout the succession and this is markedly so with the syn-tectonic sequence (Figs. 2 and 3).

These three sub-populations (WCI, PCI, I/S) form separate domains in the full width at half maximum (FWHM) intensity *versus* peak position diagrams (Fig. 4). A higher position in the diagram indicates a smaller diffracting domain, whereas displacement to the right indicates more smectitic layers. The degree of overlap of WCI , PCI, and I/S domains is greater in width than in position. The pre-tectonic series shows a higher I/S and PCI content in the stratigraphically younger samples. As age increases, the proportion of I/S decreases and the coarse-grained WCI increases relative to the PCI. However, no attempt has been

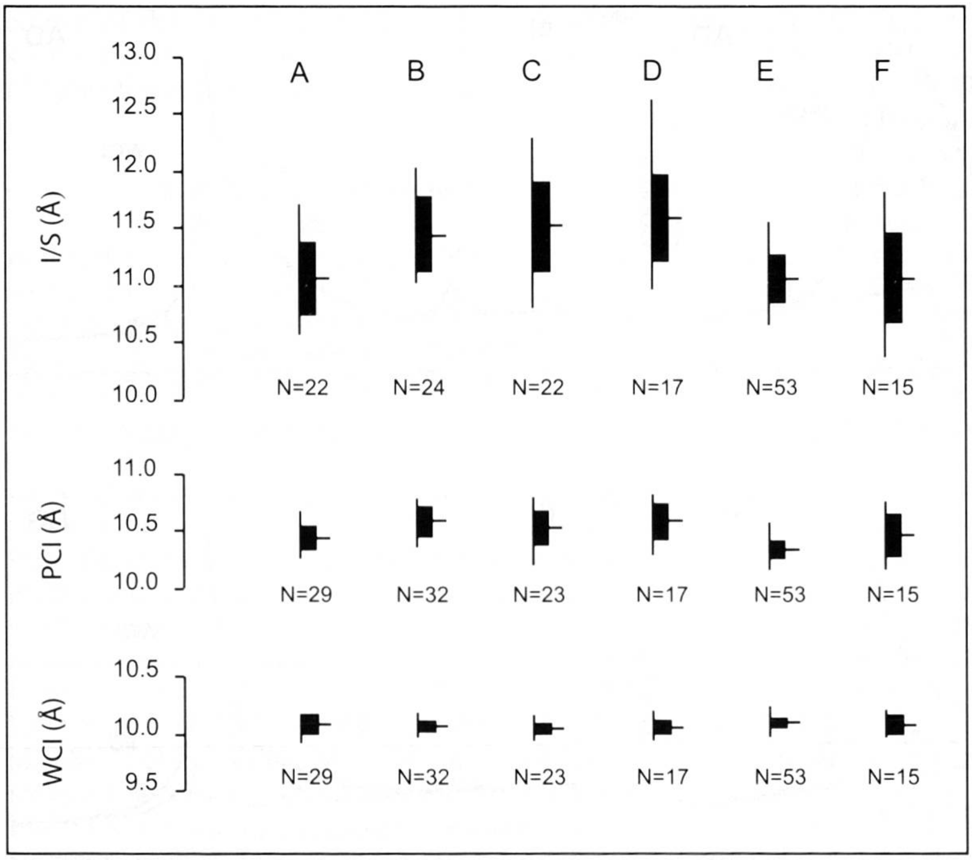

Fig. 2 Position variability for the three elementary peaks fitted on experimental XRD curves. All samples in air dried state. A—Cambrian-Silurian; B—Lower Devonian; C—Middle-Upper Devonian; D—Westphalian A-B; E—Westphalian C-D; F—Stephanian. Vertical lines—range of values; horizontal line—mean value; black box—2 standard deviation.

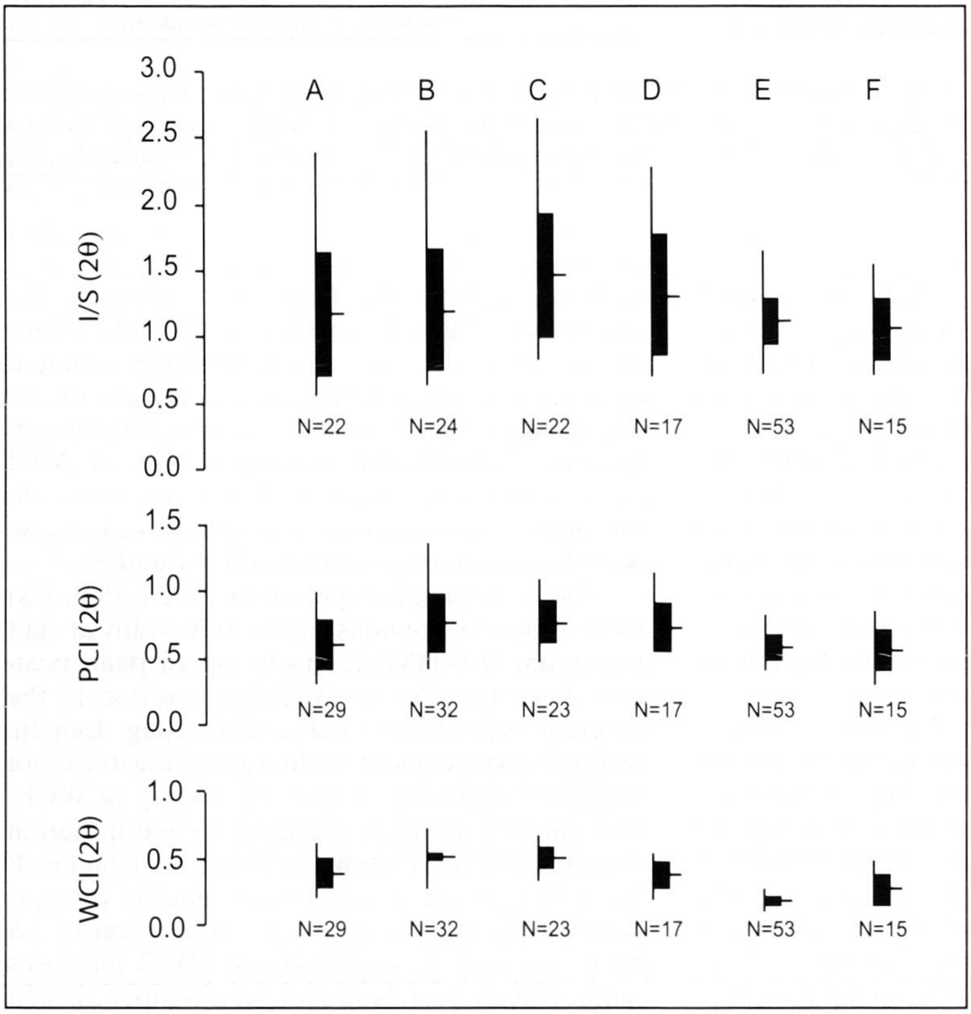

Fig. 3 Width variability for the three elementary peaks fitted on experimental XRD curves. All samples in air dried state. A—Cambrian-Silurian; B—Lower Devonian; C—Middle-Upper Devonian; D—Westphalian A-B; E—Westphalian C-D; F—Stephanian. Vertical lines—range of values; horizontal line—mean value; black box—2 standard deviation.

made to quantify the amount of the different phases on either the oriented preparations or the random powder.

Using NEWMOD modelling, the peaks have been matched most closely using two different illite/smectite mixed layers, one with a long-range order (R = 3) and the other with R = 1. As an example we have selected the modelling prepared to fit sample B299 illustrated in Figs. 1a, b. NEWMOD calculated patterns of both mixed layer phases are shown in Fig. 5. The modelled composite peak in the region between 5 and 11 °2θ for a mechanical mixture of chlorite/vermiculite, illite/smectite (80:20, R = 1) illite/smectite (90:10, R = 3) and muscovite, in proportions 5:20:35:40 (Fig. 6) respectively, provided a good match to the observed pattern.

Study of random powders for polytype determinations, using the method described by MOORE and REYNOLDS (1997), has shown the presence of two different polytypes ($2M_1$ and 1M) on most of the <2 μm fractions studied and these could be related to the WCI and PCI phases, respectively.

Results of the XRD thickness measurements showed two main types of crystal thickness distributions (Fig. 7): asymptotic (Devonian and younger rocks) and log-normal (most pre-Devonian rocks). The log-normal CTDs show a tendency to polymodality with increasing grade. CTDs have been characterized using parameters α and $β^2$ (EBERL et al., 1998b) that describe the mean and variance of the natural logarithms of the crystal thickness, respectively (Fig. 8). The regression for the correlation of α with the mean size of samples is quite high (r = 0.965) and all log-normal CTDs have values of α >1.6. Correlation is not so significant (r = 0.327) when $β^2$ is considered mainly for samples with log-normal CTDs. It is noteworthy that whereas syn-tectonic samples show asymptotic CTDs, they have large size classes, absent in older rocks with asymptotic distributions (Figs. 7 a–b).

4. Discussion

As shown previously, in the case of the pre-tectonic sequence, the characteristics of the three sub-populations obtained by decompostion of XRD peaks evolve in response to increasing

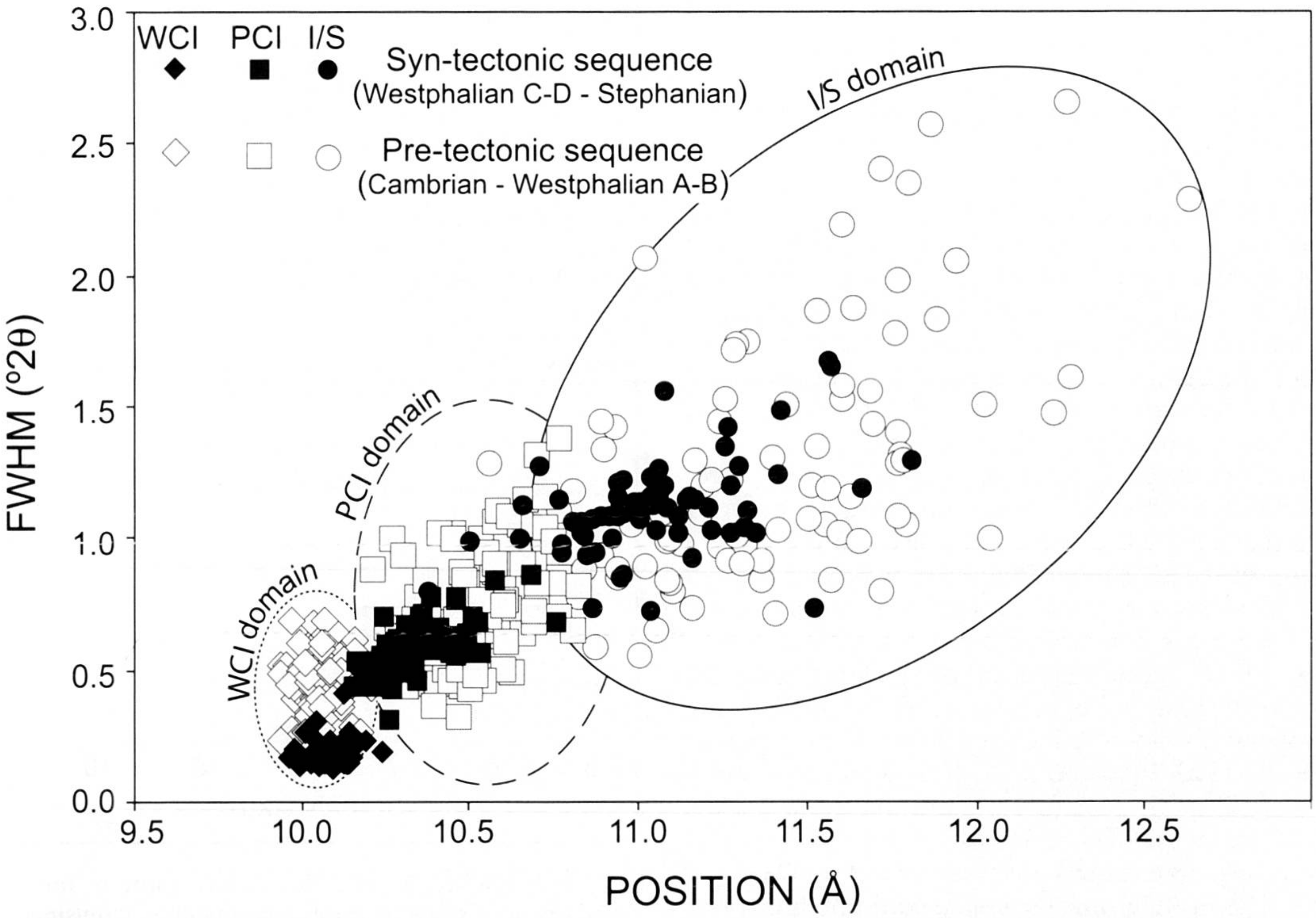

Fig. 4 Full width at half maximum (FWHM, °2θ CuKα) plotted as a function of position (Å) for the various peaks fitted to experimental XRD profiles. All samples in air-dried state.

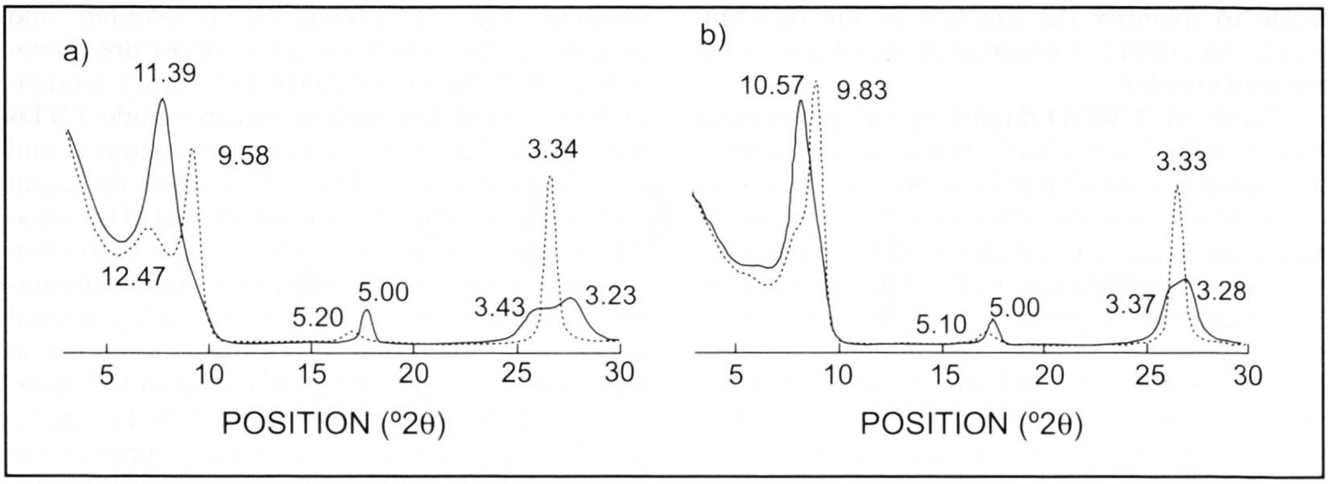

Fig. 5 NEWMOD calculated XRD patterns. (a) Illite/smectite (80:20, R = 1). (b) Illite/smectite (90:10, R = 3). Solid lines—patterns for air-dried sample; dashed lines—patterns from glycolated samples.

depth of burial. Not only does I/S become progressively more illitic (domains extend towards lower d(001)), but in addition, illite crystals grow at the expense of I/S (domains extend towards low FWHM), in agreement with POLLASTRO (1985) and ŚRODOŃ et al. (2000). These trends are mirrored by a change in CTDs from asymptotic to log-normal. A similar change from asymptotic to log-normal CTDs with increasing metamorphic grade has also been described by EBERL et al. (1990).

In the syn-tectonic samples (Westphalian C-D and Stephanian), decomposed peaks are considerably narrower than in the older rocks (Figs. 2 and 3). We have interpreted these narrower peaks as the signature of detrital white micas inherited from the rapidly uplifting Variscan mountain chain. These micas probably avoided any significant chemical weathering during their short transport history. It can be argued that these peaks could correspond to metamorphic phases formed during a previous event. This could be the case were not the peaks too narrow for the average temperatures achieved in the Cantabrian Zone prior to that event (BASTIDA et al., 2002). The I/S peaks are regarded as recording the new phases formed during the retrograde evolution of these detrital white micas. Textural evidence for the formation of I/S by replacement of pre-existing illitic or chloritic material of metamorphic origin has been repeatedly observed during TEM studies (JIANG et al., 1990; PEACOR, 1992; NIETO and SÁNCHEZ-NAVAS, 1994; MERRIMAN and ROBERTS, 2001; among others). These detrital grains could explain the large size classes observed in the CTDs.

The pre-tectonic sequence could be interpreted similarly. The narrower peaks (WCI) in the older rock samples could correspond to detrital white micas. The increase in width with decreasing age reflects their retrograde evolution. The other peaks (PCI and I/S) could record the evolution of newly-formed phases. It could be expected that the width of both populations would attain the same value at thermodynamic equilibrium. Such a tendency can be observed in the Cambrian-Silurian samples studied. The higher grade samples are also characterized by a log-normal CTD.

Studies of polytype variations within various size fractions (>4, 2–4, 1–2, 1–0.5, and 0.5–0.1 μm) support this hypothesis. Results have shown an increase in the amount of 1M polytype as particle size decreases, with the opposite behaviour of the $2M_1$ polytype, absent in the finer fraction. If we assume that the 1M polytype is diagenetic and that all the $2M_1$ is detrital (see GRATHOFF et al.,

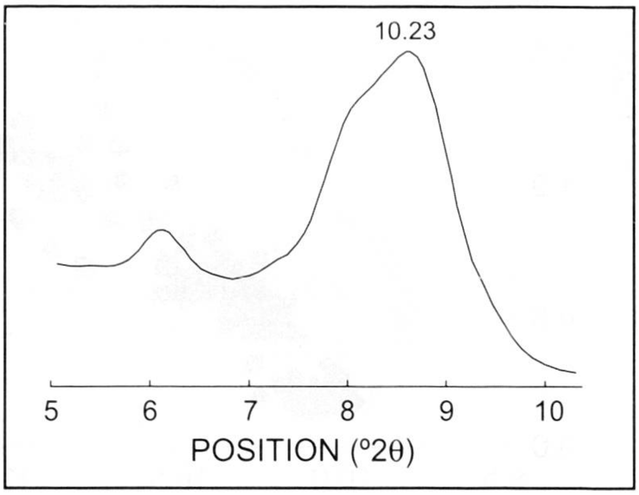

Fig. 6 NEWMOD modelled XRD pattern for a mechanical mixture of chlorite/vermiculite, illite/smectite (80:20, R = 1) illite/smectite (90:10, R = 3) and muscovite, in proportions 5:20:35:40 respectively.

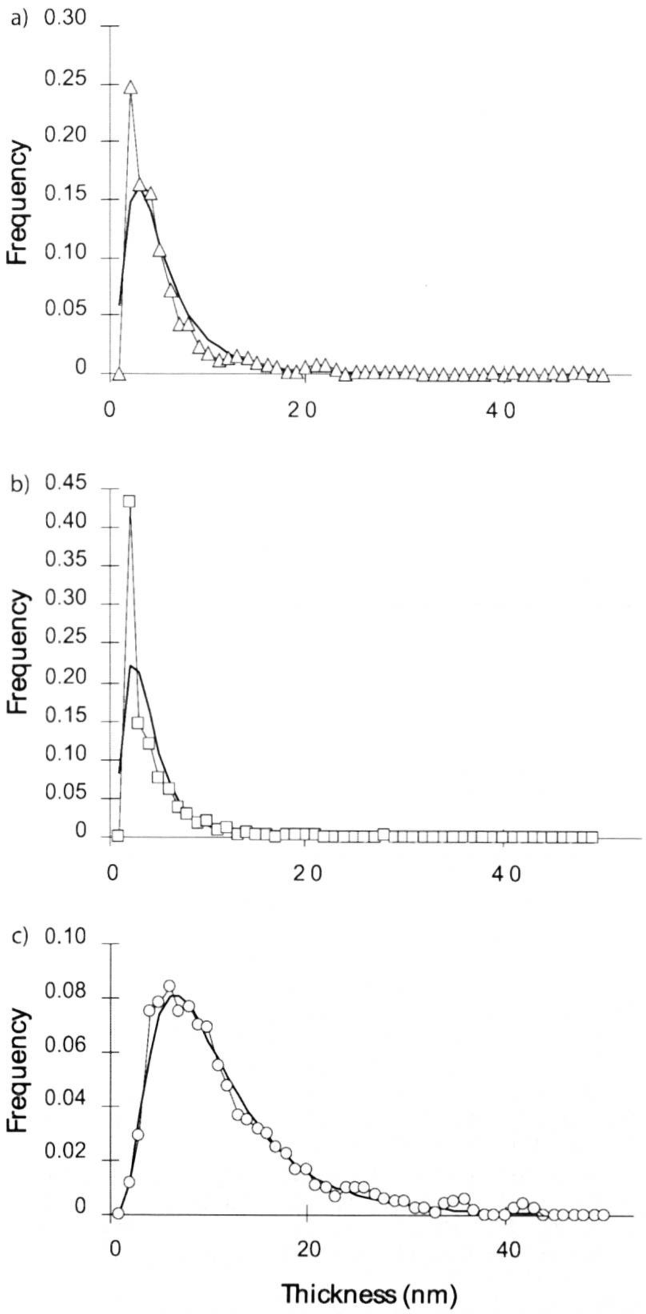

Fig. 7 Illite crystallite thickness distributions. (a–b) Asymptotic shape; (c) Log-normal-like shape. a) Sample B151 from the Westphalian C–D; b) Sample B269 from the Lower Devonian; c) Sample B657 from the Cambrian. The solid lines are log-normal fits to the data.

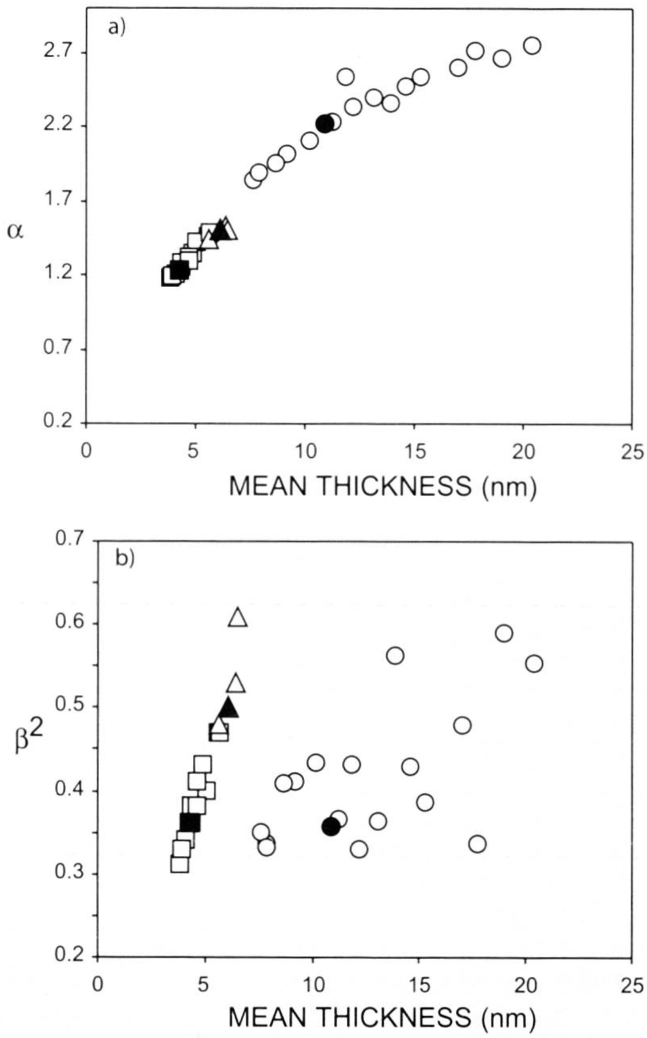

Fig. 8 Correlation between mean thickness (nm) and parameters α (a) and β^2 (b) for area weighted thickness distribution of particles. Triangles—Asymptotic distributions, syn-tectonic sequence (Westphalian C-D and Stephanian); squares—Asymptotic distributions, pre-tectonic sequence (Cambrian–Westphalian A-B); circles—Log-normal distributions, pre-tectonic sequence (Cambrian–Westphalian A-B); Solid symbols—correspond to CTDs plotted on Fig. 7.

1998; MOORE, 2000), these changes support a detrital origin for the WCI peaks in the diagenetic and low anchizonal samples.

5. Conclusions

1. Decomposition of the 10-Å peak band has shown that it is formed by at least two different peaks that are unaffected by glycolation, and a third peak that shifts on glycolation and indicates the presence of mixed-layer illite/smectite with varying amounts of smectite but never more than 25%.

2. The first peak, labelled WCI, is present in all samples and shows very little change. It has been considered as a detrital white mica, altered during retrograde evolution and therefore becoming slightly broader as rocks become stratigraphically younger.

3. The second peak, labelled PCI, decreases in width with age. This decrease indicates an increase in mean size of the diffracting domains in the *z* direction.

4. The third peak, labelled I/S, shows a trend on both position and width with burial. The evolution towards lower *d* values and narrower peaks

could be related to a decrease in the smectitic content and an increase in the mean size of the diffracting domains.

5. The last two peaks could record the evolution of newly-formed phases.

6. CTDs change from asymptotic to log-normal with increasing metamorphic grade. This implies a change in the mechanism of crystal growth (EBERL et al. 1998b), and in fact may be more important for establishing the transition from diagenesis to metamorphism than the width of either the unresolved or the deconvoluted peaks (see BRIME and EBERL, 2002).

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References

BAILEY, S.W. (1966): The status of clay mineral structures. Clay Clay Min. Nat. Conf., 14th, Berkeley, California. Pergamon Press, London and New York, 1–23.

BASTIDA, F., BRIME, C., GARCÍA-LÓPEZ, S. and SARMIENTO, G.N. (1999): Tectono-thermal evolution in a region with thin skinned tectonics: the western nappes in the Cantabrian Zone (Variscan belt of NW Spain). Int. J. Earth Sci. 88, 38–48.

BASTIDA, F., BRIME, C., GARCÍA-LÓPEZ, S., ALLER, J., VALÍN, M.L. and SANZ-LÓPEZ, J. (2002): Tectonothermal evolution of the Cantabrian Zone (NW Spain). In: GARCÍA-LÓPEZ, S. and BASTIDA, F. (eds): Paleozoic conodonts from Northern Spain. Instituto Geológico y Minero de España, serie Cuadernos del Museo Geominero 1, 105–123.

BLENKINSOP, T.G. (1988): Definition of low grade metamorphic zones using illite crystallinity. J. Metamorphic Geol. 6, 623–636.

BRIME, C. (1981): Postdepositional transformation of clays in Palaeozoic rocks of northwest Spain. Clay Min. 16, 421–424.

BRIME, C. (1985): A diagenesis to metamorphism transition in the Hercynian of NW Spain. Mineral. Mag. 49, 481–484.

BRIME, C. and EBERL, D.D. (2002): Growth mechanism of low grade illites based on shape of crystal growth distributions. Schweiz. Mineral. Petrogr. Mitt. 82, 203–209.

BRIME, C., GARCÍA-LÓPEZ, S., BASTIDA, F., VALÍN, M.L., SANZ-LÓPEZ, J. and ALLER, J. (2001): Transition from diagenesis to metamorphism near the front of the Variscan Regional Metamorphism (Cantabrian Zone, Northwestern Spain). J. Geol. 109, 363–379.

BRIME, C. and PÉREZ-ESTAÚN, A. (1980): La transición diagénesis-metamorfismo en la región de Cabo Peñas. Cuadernos Lab. Geol. Laxe 1, 85–97.

DRITS, V.A., EBERL, D.D. and ŚRODOŃ, J. (1998): XRD measurement of mean thickness, thickness distribution and strain for illite and illite-smectite crystallites by the Bertaut-Warren-Averbach technique. Clay Clay Min. 46, 38–50.

EBERL, D.D., ŚRODOŃ, J., KRALIK, M., TAYLOR, B.E. and PETERMAN, P. (1990): Ostwald ripening of clays and metamorphic grade. Science 248, 474–477.

EBERL, D.D., DRITS, V.A. and ŚRODOŃ, J. (1998b): Deducing growth mechanisms for minerals from the shape of crystal size distributions. Am. J. Sci. 298, 499–533.

EBERL, D.D., DRITS, V.A., ŚRODOŃ, J. and NÜESCH, R. (1996): MudMaster: A program for calculating crystallite size distributions and strain from the shapes of X-ray diffraction peaks. U.S. Geol. Surv. Open File Report 96–171, 44 pp.

EBERL, D.D., NÜESCH, R., SUCHA, V. and TSIPURSKY, S. (1998a): Measurement of fundamental illite particle thickness by X ray diffraction using PVP-10 intercalation. Clay Clay Min. 46, 89–97.

FREY, M. (ed.) (1987): Low temperature metamorphism. Blackie , 351 pp.

FREY, M. and ROBINSON, D. (eds) (1999): Low-grade metamorphism. Blackwell Science, 313 pp.

GARCÍA-LÓPEZ, S., BRIME, C., BASTIDA, F. and SARMIENTO, G.N. (1997): Simultaneous use of thermal indicators to analyse the transition from diagenesis to metamorphism: an example from the Variscan Belt of northwest Spain. Geol. Mag. 134, 323–334.

GHARRABI, M., VELDE, B. and SAGON, J.-P. (1998): The transformation of illite to muscovite in pelitic rocks: constrains from X-ray diffraction. Clay Clay Min. 46, 79–88.

GRATHOFF, G.H., MOORE, D.M., HAY, R.L. and WEMMER, K. (1998): Illite polytype quantifications and K/Ar dating of Paleozoic shales: a technique to quantify diagenetic and detrital illite. In: SCHIEBER, J., ZIMMERLE, W. and SETHI, P. (eds): Shales and mudstones. II. Stuttgart, E. Schweizerbart´sche (Nägele u. Obermiller), 161–175.

HOWARD, S.A. and PRESTON, K.D. (1989): Profile fitting of powder difraction patterns. In: BISH, D.L. and POST, J.E. (eds): Modern Powder Diffraction. Rev. in Mineral. 20, 217–275.

JABOYEDOFF, M., BUSSY, F., KÜBLER, B. and THÉLIN, P. (2001): Illite "Crystallinity" Revisited. Clay Clay Mineral. 49, 156–167.

JIANG, W.T., PEACOR, D.R., MERIMAN, R.J. and ROBERTS, B. (1990): Transmission and analytical electron microscopic study of mixed layer illite/smectite formed as a replacement product of diagenetic illite. Clay Clay Min. 38, 449–468.

KILE, D.E. and EBERL, D.D. (2000): Crystal growth mechanisms in miarolitic cavities in the Lake George ring complex and vicinity, Colorado. Am. Mineral. 84, 718–724.

KILE, D.E., EBERL, D.D., HOCH, A.R. and REDDY, M.M. (2000): An assessment of calcite crystal growth mechanisms based on crystal growth distributions. Geochim. Cosmochim. Acta 64, 2973–2950.

KISCH, H.J. (1983): Mineralogy and petrology of burial diagenesis (burial metamorphism) and incipient metamorphism in clastic rocks. In: LARSEN, G. and

CHILINGAR, G. V. (eds): Diagenesis in Sediments and Sedimentary Rocks, 2, Elsevier Amsterdam, p. 289–493 and p. 513–541.

KÜBLER, B. (1967): La cristallinité de l' illite et les zones tout à fait supérieures du métamorphism. Etages tectoniques, Colloque de Neuchâtel, 105–122.

KÜBLER, B. and GOY-EGGENBERGER, D. (2001): La cristallinité de l'illite revisitée: un bilan des connaissances acquises ces trente dernières années. Clay Min. 36, 143–157.

LANSON, B. (1990): Mise en évidence des mécanismes de transformations des interstratifiés illite/smectite au cours de la diagenèse. Ph.D. thesis, Univ. Paris 7-Jussieu, France.

LANSON, B. (1992): Application de la décomposition des diffractogrammes de rayons-X à l'identification des minéraux argileux. Comptes-rendus du Colloque rayons X, Paris 1992, Siemens, ed. Vol 2.

LANSON, B. and BESSON, G. (1992): Characterization of the end of smectite-to-illite transformation: Decomposition of X ray patterns. Clay Clay Min. 40, 40–52.

LANSON, B. and CHAMPION, D. (1991): The I/S-to-illite reaction in the late stage diagenesis. Am. J. Sci. 291, 473–506.

LANSON, B. and VELDE, B. (1992): Decomposition of X-ray diffraction patterns: A convenient way to describe complex I/S diagenetic evolution. Clay Clay Min. 40, 629–642.

LANSON, B., BEAUFORT, D., BERGER, G., BARADAT, J. and LACHARPAGNE, J.C. (1996): Illitization of diagenetic kaolinite-to-illite conversion series: late-stage diagenesis of the Lower Permian Rotliegend sandstone reservoir, offshore of the Netherlands. J. Sed. Res. 66, 501–518.

LANSON, B., BEAUFORT, D., BERGER, G., PETTIT, D. and LACHARPAGNE, J.C. (1995): Evolution de la structure cristallographique des minéraux argileux dans le réservoir gréseux Rotliegend des Pays-Bas. Centre de Recherches Exploration-Production Elf Aquitanie, Bull. 19, 149–165.

MERRIMAN, R.J. and ROBERTS, B. (2001): Low grade metamorphism in the Scottish Southern Uplands terrane: deciphering the patterns of accretionary burial, shearing and cryptic aureoles. Transact. Royal Soc. of Edinburgh Earth Sci. 91, 521–537.

MOORE, D.M. (2000): Diagenesis of the Purington Shale in the Illinois basin and implications for the diagenetic state of sedimentary rocks of shallow Paleozoic basins. J. Geol. 108, 553–567.

MOORE, D.M. and REYNOLDS, R.C. JR (1997): X-ray diffraction and the identification and analysis of clay minerals (2nd ed.). New York, Oxford University Press, 378 pp.

NIETO, F. and SÁNCHEZ-NAVAS, A. (1994): A comparative XRD and TEM study of the physical meaning of the white mica 'crystallinity' index. Eur. J. Mineral. 6, 611–621.

PEACOR, D.R. (1992): Diagenesis and low grade metamorphism of shales and slates. In: BUSECK, P.R. (ed.): Minerals and reactions at the atomic scale: Transmission Electron Microscopy. Rev. Mineral. 27, 335–380.

POLLASTRO, R.M. (1985): Mineralogical and morphological evidence for the formation of illite at the expense of illite/smectite. Clay Clay Min. 33, 265–274

REYNOLDS, R.C. Jr. (1985): NEWMOD©, a computer program for the calculation of one-dimensional diffraction patterns of mixed-layer clays. R.C. Reynolds, Jr., 8 Brook Dr., Hanover NH 03755.

REYNOLDS, R.C. Jr. and REYNOLDS, R.C. III (1996): NEWMOD-FOR-WINDOWS™. The calculation of one-dimensional diffraction patterns of mixed-layered clay minerals. R.C. Reynolds, Jr., 8 Brook Dr., Hanover NH 03755.

ROBINSON, D. and BEVINS, R.E. (1994): Mafic phyllosilicates in low-grade metabasites. Characterization using deconvolution analysis. Clay Min. 29, 223–237.

SCHREINER, M.N. and JENKINS, R. (1983): Profile fitting for quantitative analysis in X ray powder diffraction. Adv. X-ray Analysis 26, 66–69.

ŚRODOŃ, J., EBERL, D.D. and DRITS, V.A. (2000): Evolution of fundamental particle size during illitization of smectite and implications for the illitization mechanism. Clay Clay Min. 48, 446–458.

STERN, W.B., MULLIS, J., RAHN, M. and FREY, M. (1991): Deconvolution of the first 'illite' basal reflection. Schweiz. Mineral. Petrogr. Mitt. 71, 453–462.

WANG, H., STERN, W.B. and FREY, M. (1995): Deconvolution of the X-ray 'Illite' 10-Å complex: a case study of the Helvetic sediments from eastern Switzerland. Schweiz. Mineral. Petrogr. Mitt. 75, 187–199.

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