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Basis for meaningful illite crystallinity measurements: an example from the Swiss Prealps

by M. Jaboyedoff¹, B. Kübler², M. Sartori³ and Ph. Thélin¹

Abstract

Combination of the illite crystallinity method (IC) with Newmod© simulations permits characterisation of the mean thickness of coherent scattering domains (CSD) and of the amount of illite in mixed-layered illite/smectite. Illites amenable to study by the IC method are in fact mixed-layered illite/smectite (I/S) with low smectite content. In the anchizone, the IC is an indirect measurement of the mean number of consecutive illite layers (or fundamental particles) of I/S. An example from the Swiss Prealps shows a good correlation between IC and the number of consecutive illites layers (N_{FP}). The number of consecutive illite layers is a more precise quantification of metamorphism than the thickness of CSD.

Keywords: illite crystallinity, fundamental particles, mixed-layer, illite/smectite, anchizone, low-grade metamorphism.

1. Introduction

During the past thirty years application of the illite crystallinity (IC) method (KÜBLER, 1967, 1984; FREY, 1987) has proved useful in the study of low grade metamorphism. The IC method involves measuring the Scherrer width (SW) (full width at half-maximum height) of the illite 10 Å X-ray diffraction peak in $^{\circ}2\theta$ CuK α units on an air-dried (AD) pattern. Although illite may contain up to ~ 5% of expandable interlayers (MOORE and REYNOLDS, 1997, p. 151), the current tendency is to characterise illite by the illite layer in I/S and to its end-member chemistry (MEUNIER and VELDE, 1989; SRODON et al., 1992). Moreover, while it is useful to call this method the illite crystallinity technique, the term "illite" covers a range of minerals measured by the IC (MERRIMAN and PEACOR, 1999). Indeed, the 10 Å diffraction peak is often the result of the diffraction of pure illitic layers interstratified with a small amount of expandable interlayers. The IC value can also be influenced by small amounts of interlayered phases such as chlorite. The expandable interlayers are

usually smectite-like, and can therefore be defined as a mixed-layer mineral illite/smectite (I/S). This is the nomenclature used in this paper, thus the layers in the mixed-layer I/S can be viewed as interlayers surrounded by two half tetrahedral-octahedral layers (ALTANER and YLAGAN, 1997).

The lower and higher anchizone limits correspond to about 25 and 60 layers of pure illite, estimated either by high resolution transmission electron microscopy (TEM) or/and by X-ray diffraction (MERRIMAN et al., 1990; KRUMM, 1992; NIETO and SANCHEZ-NAVAS, 1994; WARR and RICE, 1994; ARKAI et al., 1996; DALLA TORRE et al., 1996). In these studies illites did not contain expandable interlayers, or were removed by treatment. Nevertheless it is often observed that the SW is broader for air dried preparations (AD) than for ethylene-glycol treated preparations (EG) (KISCH and FREY, 1987), which indicates the presence of expandable interlayers. EBERL and VELDE (1989) integrated the number of expandable interlayers using a horizontal background, which is not the usual way for measuring the IC (KÜBLER, 1967). DRITS et al. (1997) developed a method which

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takes into account a log-normal distribution thickness of the coherent scattering domains (CSD) by modifying the Scherrer equation. This method does not lead directly to the simultaneous estimations of the total layer number in the CSD and the amount of smectite interlayers (%S). The expandable interlayers are removed either by heating (DRITS *et al.*, 1998) or by Na-saturated I/S with polyvinylpyrrolidone intercalation (EBERL *et al.*, 1998) leading to an estimate of the CSD and the thickness of fundamental particles, respectively.

The aim of this paper is to link traditional IC measurements (KISCH, 1990) to the amount of interstratification %S and the number of layers in the coherent diffracting domain (CSD), using an example from the Préalpes Médiannes Rigides.

2. Materials and methods

2.1. SAMPLE PREPARATION AND SW MEASUREMENT

Samples of < 2 μm clay fractions from HCl-dissolved limestones and marly limestones from the Préalpes Médiannes Rigides were prepared for XRD analysis (Tab. 1). Both new XRD determinations and those reported in JABOYEDOFF and THÉLIN (1996) are used for this study. The geological setting and XRD data are given in JABOYEDOFF and THÉLIN (1996) and references can be found therein. The oriented specimens were prepared by sedimentation on a glass slide with a concentration of more than 2.5 mg/cm². The XRD data were collected with a 18.5 cm radius Rigaku horizontal powder diffractometer (Geigerflex) equipped with a rotating Cu anode (CuK α radiation, nickel filter, 0.5° divergent and scatter slits, 0.15 mm receiving slit and two soller slits of 5°), and operating conditions of 40 kV and 30 mA with the step scanning method using 0.01° and 1 second steps. XRD diffractograms were performed for AD and EG treatments.

The IC values were determined from the raw data by measuring the peak width at half distance of the peak maximum and a linear background joining the near peak background. No K α_2 stripping, no background removal and no curve modelling is applied. The noisy XRD data were smoothed with the Savitsky-Golay method. For this study SW measurements were made on both AD preparations (SWAD = IC) and EG preparations (SWEG); the SWEG was not measured for all samples. A Reichweite order R1 is assumed for the data because in the Préalpes Médiannes Rigides we do not have evidence for longer-range orders (JABOYEDOFF and THÉLIN, 1996).

Tab. 1 SW data, SW_c corrected for instrumental broadening and interpretation of results as in figure 2. IC limits for the first column are 0.18° and 0.36° $\Delta 2\theta$ CuK α .

SWAD (IC) ° $\Delta 2\theta$ CuK α	SWEG ° $\Delta 2\theta$ CuK α	SWcAD ° $\Delta 2\theta$ CuK α	SWcEG ° $\Delta 2\theta$ CuK α	%S	N	N _{EP}
0.17 °	0.17 °	0.10 °	0.10 °	0.0%	84	83
0.18 °	0.17 °	0.11 °	0.10 °	0.8%	93	53
0.19 °	0.19 °	0.12 °	0.12 °	0.0%	67	66
0.20 °	0.20 °	0.13 °	0.13 °	0.0%	61	60
0.21 °	0.20 °	0.14 °	0.13 °	0.9%	69	42
0.22 °	0.20 °	0.15 °	0.13 °	1.5%	78	36
0.23 °	0.21 °	0.16 °	0.15 °	1.1%	62	37
0.23 °	0.20 °	0.16 °	0.13 °	1.9%	90	33
0.24 °	0.22 °	0.17 °	0.15 °	1.5%	64	32
0.25 °	0.22 °	0.18 °	0.15 °	2.0%	73	30
0.25 °	0.22 °	0.18 °	0.15 °	2.0%	73	30
0.26 °	0.26 °	0.19 °	0.19 °	0.0%	37	37
0.26 °	0.22 °	0.19 °	0.15 °	2.3%	79	28
0.26 °	0.24 °	0.19 °	0.17 °	1.6%	54	29
0.27 °	0.23 °	0.21 °	0.16 °	2.3%	72	27
0.28 °	0.26 °	0.22 °	0.19 °	1.6%	47	26
0.29 °	0.27 °	0.23 °	0.21 °	1.7%	44	25
0.29 °	0.29 °	0.23 °	0.22 °	0.4%	32	29
0.30 °	0.27 °	0.24 °	0.21 °	2.1%	46	23
0.31 °	0.27 °	0.25 °	0.21 °	2.5%	51	22
0.32 °	0.29 °	0.26 °	0.23 °	2.1%	41	21
0.33 °	0.28 °	0.27 °	0.22 °	2.8%	51	21
0.33 °	0.27 °	0.27 °	0.21 °	3.1%	58	21
0.34 °	0.31 °	0.28 °	0.25 °	2.2%	36	20
0.34 °	0.30 °	0.28 °	0.24 °	2.7%	42	20
0.35 °	0.33 °	0.29 °	0.27 °	1.8%	31	20
0.36 °	0.33 °	0.30 °	0.28 °	2.0%	30	19
0.36 °	0.34 °	0.30 °	0.28 °	1.8%	29	19
0.37 °	0.32 °	0.31 °	0.26 °	2.8%	36	18
0.37 °	0.33 °	0.31 °	0.28 °	2.5%	32	18
0.38 °	0.31 °	0.32 °	0.25 °	3.5%	45	17
0.38 °	0.34 °	0.32 °	0.29 °	2.5%	31	17
0.38 °	0.34 °	0.32 °	0.28 °	2.7%	32	17
0.39 °	0.36 °	0.34 °	0.30 °	2.2%	27	17
0.39 °	0.35 °	0.34 °	0.29 °	2.7%	30	17
0.39 °	0.39 °	0.34 °	0.34 °	0.0%	18	18
0.41 °	0.36 °	0.35 °	0.30 °	3.0%	31	16
0.41 °	0.36 °	0.36 °	0.30 °	3.2%	31	15
0.41 °	0.37 °	0.36 °	0.31 °	3.0%	29	15
0.42 °	0.36 °	0.37 °	0.30 °	3.3%	31	15
0.42 °	0.34 °	0.37 °	0.28 °	3.8%	39	15
0.42 °	0.37 °	0.37 °	0.31 °	3.2%	30	15
0.44 °	0.38 °	0.39 °	0.32 °	3.3%	28	14
0.45 °	0.34 °	0.40 °	0.29 °	4.3%	41	15
0.48 °	0.40 °	0.43 °	0.35 °	3.9%	27	13
0.49 °	0.36 °	0.44 °	0.30 °	4.8%	40	13
0.49 °	0.42 °	0.44 °	0.37 °	3.6%	24	13
0.51 °	0.45 °	0.46 °	0.40 °	3.3%	20	12
0.52 °	0.42 °	0.48 °	0.37 °	4.3%	26	12
0.53 °	0.41 °	0.49 °	0.36 °	4.7%	30	12
0.55 °	0.52 °	0.51 °	0.47 °	2.1%	14	10
0.58 °	0.49 °	0.54 °	0.44 °	4.0%	18	10
0.62 °	0.47 °	0.58 °	0.42 °	5.2%	23	10
0.67 °	0.47 °	0.63 °	0.42 °	5.9%	26	10

2.2. IC CALIBRATION

The IC values were determined using the Neuchâtel IC scale calibrated with the Lausanne diffractometer using 27 samples (JABOYEDOFF, 1999). The lower and upper limits of the anchizone were determined at 0.22° and $0.39^\circ 2\theta$ $\text{CuK}\alpha$ (Lausanne diffractometer: Rigaku; JABOYEDOFF and THÉLIN, 1996). After a change of filament and of diffractometer geometry in 1996, the limits of our diffractometers shifted slightly leading to the current anchizone limits of 0.18° and $0.36^\circ 2\theta$ $\text{CuK}\alpha$. For reasons of comparison a value of $0.03^\circ 2\theta$ $\text{CuK}\alpha$ has been subtracted from the SW values determined prior to the filament change (Tab. 1). This subtraction is justified because KISCH (1990) showed that differences in instrumental broadening between various configurations of a diffractometer shifts the IC scale by a constant amount.

The IC values from the Préalpes Médiannes Rigides indicate well-defined variations in metamorphism that increase towards the south-east from deep diagenesis to anchizonal values (BAUD, 1987; MOSAR, 1988; JABOYEDOFF and THÉLIN, 1996) (Fig. 1).

2.3. ELIMINATING INSTRUMENTAL BROADENING

Several methods exist to remove instrumental effects from an X-ray diffraction peak to obtain a pure profile. In almost all cases it is necessary to know a diffraction profile from a "perfect crystal powder". In order to obtain such a profile, we used a mica powder imbedded in resin with glass powder used for disorientation and polished in order to obtain a planar surface (Standard Reference Material 675 Mica Powder; U.S. Dept. of Commerce; National Bureau of Standards Washington, DC 20234). Such a powder is assumed to contain crystals with few defects and of sufficiently large sizes to avoid broadening produced by small crystals. This leads to an estimate of instrumental peak equal to $\text{SW } 0.09^\circ 2\theta$ $\text{CuK}\alpha$.

The best method consists in unfolding directly the measured diffraction peak with the instrumental profile leading to the pure profile, and the latter is used to measure the corrected SW for instrumental broadening. In practice it is time consuming to obtain reliable results by unfolding method (STOKES, 1948; ERGUN, 1968;

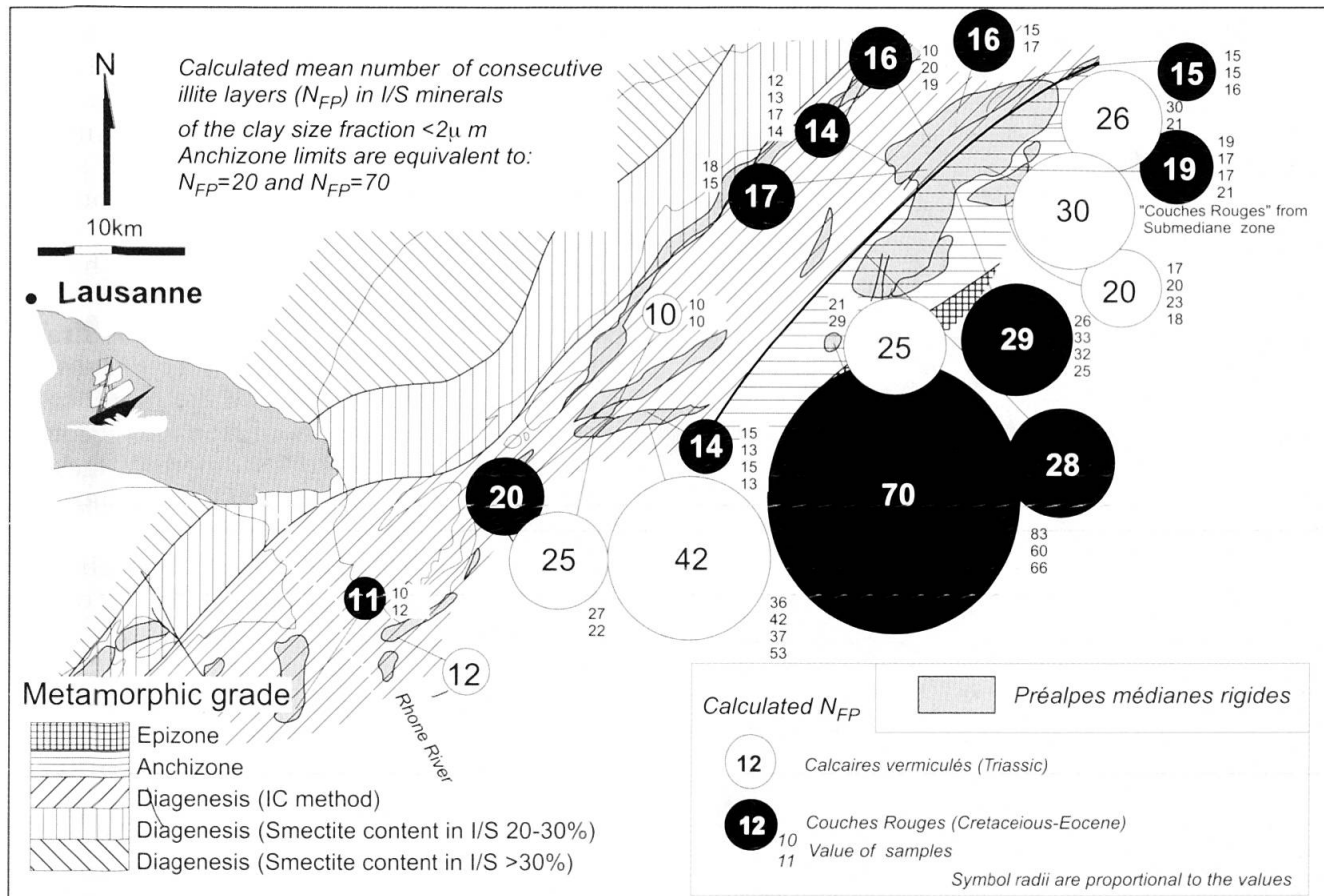


Fig. 1 Estimated fundamental particles thickness N_{FP} within the Préalpes Médiannes Rigides. Modified after JABOYEDOFF and THÉLIN (1996) and JABOYEDOFF and COSCA (1999).

EBERL et al., 1996, 1998; DRITS et al., 1998; JABOYEDOFF, 1999).

In the present study, we used a quicker method. A table of conversion is established between SW and the corrected SW for instrumental broadening (SW_c). This table is calculated by measuring the SW on theoretical peak profile obtained by the convolution of a Pearson VII function of known SW_c with the function corresponding to the instrumental profile (Fig. 2; Tab. 1) (JABOYEDOFF, 1999). The Pearson VII functions of power 2 (HOWARD and PRESTON, 1989) is used and assumed to be realistic for clay diffraction peaks. This method leads to similar results obtained by the simple sum of the SW of the instrumental profile and pure profile (Lorentzian function) for low SW and reaches Gaussian behaviour (square root of the sum of the squared SW) for higher SW.

This procedure, applied to three different diffractometers (Rigaku© in Lausanne, Philips© and Scintag© in Neuchâtel), led to estimates of higher and lower anchizone limits without instrumental effect of 0.10° and $0.30^\circ \Delta 2\theta$ CuK α respectively (JABOYEDOFF, 1999). Using figure 2 and limits defined in this paper (0.18° and $0.36^\circ \Delta 2\theta$ CuK α) the estimated anchizone limits are 0.11° and $0.30^\circ \Delta 2\theta$ CuK α . This small discrepancy with the previous values can be assigned to intrinsic errors caused by the calibration and the rounding of

limits. Therefore, we used the limits determined on the three diffractometers.

3. Interpretation of the IC scale

A physical interpretation of the IC traditional measurement seems possible, despite the probable polyphase "origin" (LANSON, 1997) of the first illite diffraction peak. By using the program Newmod© (REYNOLDS, 1985), nets can be created and used to define the mean number of layers (N) by CSD and the average amount of smectitic layers (%S) (Fig. 3). Nets are generated by measuring the SW of the 10 Å peak on Ca-saturated simulated diffractograms for different values of N and %S. This is performed on AD pattern, assuming two water layers (SW2w), and EG pattern assuming two ethylene-glycol layers (SW2g). The nets were created using the default NEWMOD© parameters (d -value of dioctahedral mica 9.98 Å, dioctahedral smectite-2g 16.9 Å, dioctahedral smectite-2w 15 Å, CuK α radiation, Ca-exchange cation) for the simulations, and a divergent slit of 0.5° , goniometer radius 18.5 cm, sample length 2.8 cm, two soller slits of 5° and $K_{0.9}$ for mica and $Fe_{0.2}$ for mica and smectite. The distribution of the CSD layer number used here for simulations has a constant value between two limits chosen as $[N-d; N+d]$ where d is equal to 10 if $N > 12$ and $[2; (2N-2)]$ for $N \leq 12$. It can be shown that the type of the CSD size distribution for Newmod© simulations has very little effect on peak shape when the mean and the variance are kept constant (JABOYEDOFF, 1999).

Use of the constructed nets requires that instrumental broadening of SWAD and SWEG is removed to obtain SW_{cAD} and SW_{cEG} . Strain-related distortions of the illite lattice (ARKAI et al., 1996; DRITS et al., 1998) and the presence of any exotic interlayers are not taken into account. These nets are established for different stacking orders of Reichweite R0, R1 and R3 (MOORE and REYNOLDS, 1997).

Such nets can be used to explore the anchizone limits assuming that fundamental particles on average consist of N_{FP} consecutive illite layers (Fig. 4) and the MacEwan particles consist of I/S minerals of N illite layers (MOORE and REYNOLDS, 1997). In this case the mean of N, and %S of MacEwan particles are related to N_{FP} (SRONDON et al., 1990, 1992).

The anchizone limits appear very similar to the curves which follow MacEwan particles for different N constructed of fundamental particles of constant N_{FP} (Fig. 4) (MOORE and REYNOLDS, 1997; NADEAU et al., 1984). The increase of fun-

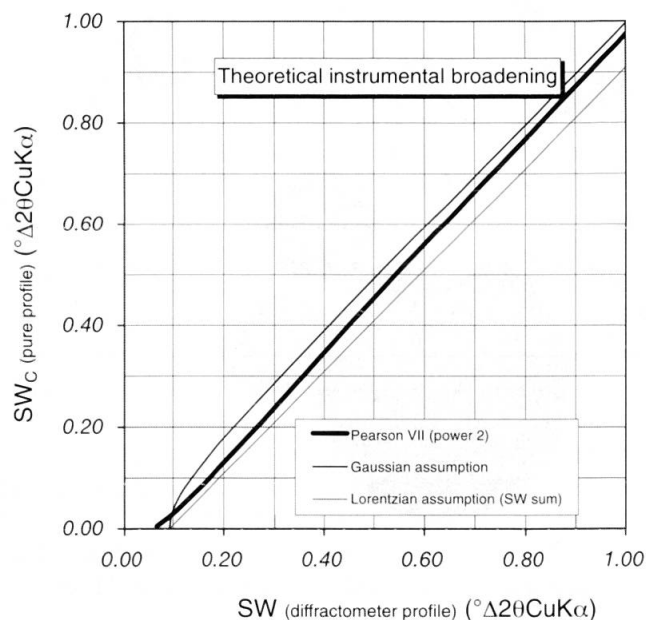


Fig. 2 Relation used to remove the instrumental broadening effect from the SW to obtain the Scherrer width of the pure profile (SW_c). This net is created by the convolution of the instrumental profile with a Pearson VII (power 2) function used as pure profile. It is compared to the relationship obtained assuming Gaussian or Lorentzian profile for both instrumental and pure profiles.

damental particle thickness with IC value was demonstrated by EBERL and BLUM (1993), DRITS et al. (1997, 1998) and EBERL et al. (1998). However, when expandable interlayers are present, IC is not necessarily a measurement of fundamental particles thickness. Note that the composition of fundamental particles are considered as pure illite as defined by MEUNIER and VELDE (1989).

In a net of SW2w and SW2g, MacEwan particles of constant N_{FP} generate curves of almost constant SW2w (IC corrected for instrumental broadening) in the anchizone range. This relationship is less clear the further one moves to shal-

lower diagenesis or to deeper epizone. Making the assumption that the IC scale is equivalent to N_{FP} , the grid area within the anchizone appears as a unique zone where the IC scale is accurate (Fig. 3) (KÜBLER, 1984). This explains partly the observed scattering of the IC measurements in the diagenetic zone and the lack of accuracy of the method in the epizone, because in those zones IC is not correlated to N_{FP} size, which is assumed to be the parameter which quantifies the metamorphism (KÜBLER, 1984).

The diagenesis-anchizone limit corresponds to $N_{FP} \approx 20$ and that of the anchizone-epizone limit

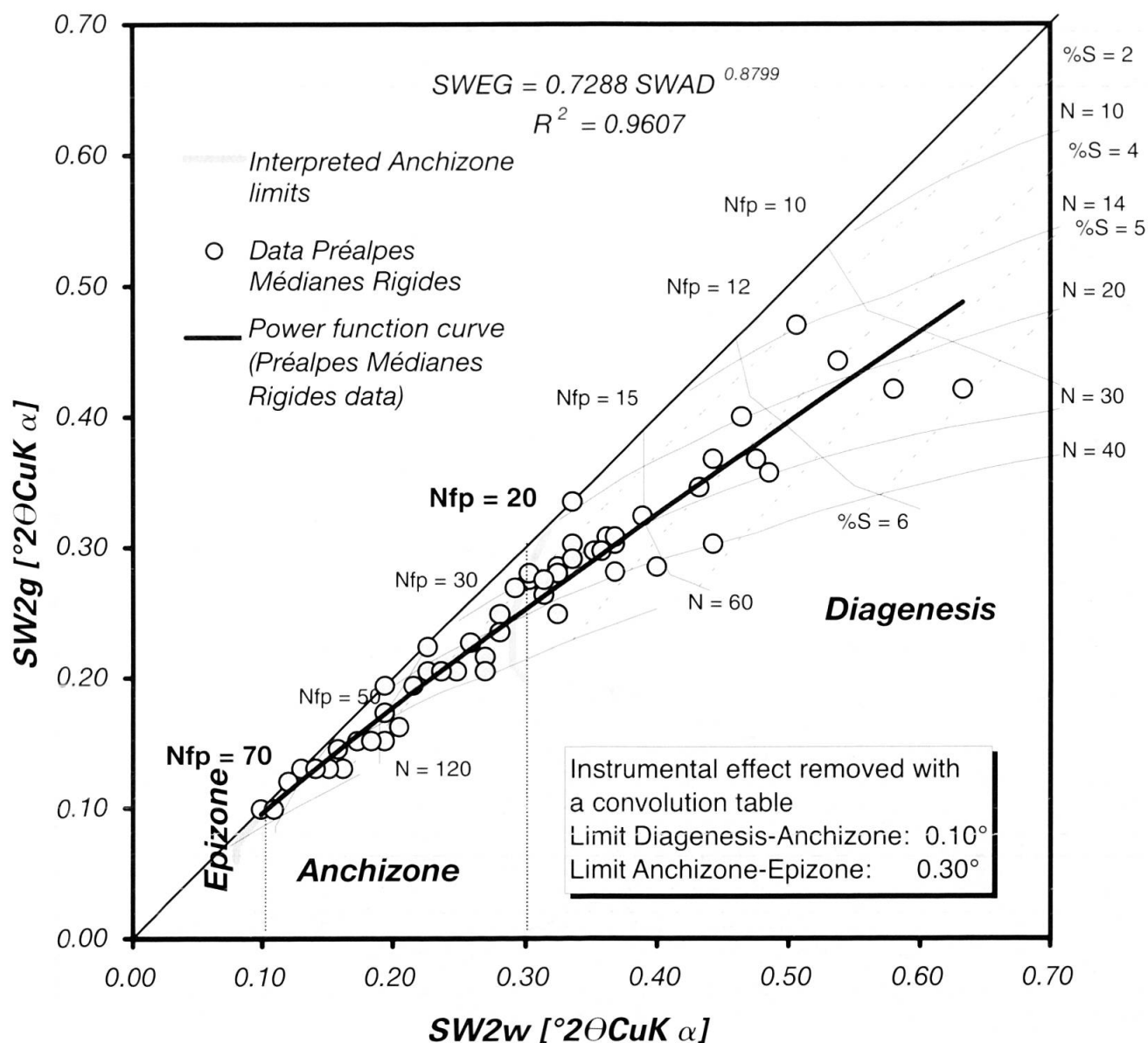


Fig. 3 Net used to find mean number of layers (N), average number of layers of fundamental particles (N_{FP}) and amount of smectite layers ($\%S$) for the order R1. The inputs are the SW_{cAD} (SW2w) and the SW_{cEG} (SW2g) (i.e. SW after removal of the instrumental broadening). The full width at half maximum is measured from a linear background traced on a diffractogram with no background removal and no $K\alpha_2$ stripping. The limits of the anchizone are very similar to the curve, $N_{FP} = 20$ and 70. The fitting of the points by a curve (power function) is also represented. It represents the mean behaviour of I/S responding to metamorphism of the Couches Rouges within the Préalpes Médiannes Rigides.

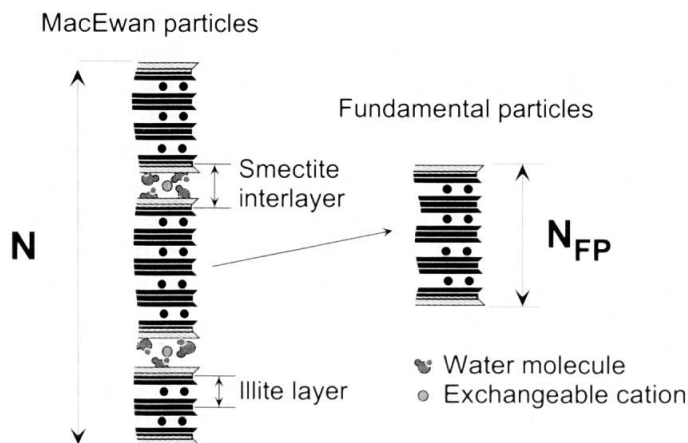


Fig. 4 I/S mixed-layers (= MacEwan particles) with high illite content can be considered as a stack of N illite layers joined by smectite interlayers. These illite stacks are the fundamental particles of NADEAU et al. (1984).

to $N_{FP} \approx 70$. Even if detrital micas can influence the IC, within the anchizone field neo-formed illite dominates the diffraction peak shape (JABOYEDOFF and THÉLIN, 1996). The increase of N_{FP} with weaker metamorphic conditions does not have a direct significance from the crystalline growth viewpoint (ALTANER and YLAGAN, 1997). For example, if some smectite interlayers in an I/S mineral are converted to illite by solid state transformation or through dissolution-precipitation, it will display an increase in N_{FP} .

4. Interpretation and discussion

In the Préalpes Médiannes Rigides the distribution of N_{FP} and IC values follow the same trend (Fig. 1; Tab. 1). The N_{FP} values for conditions of diagen-

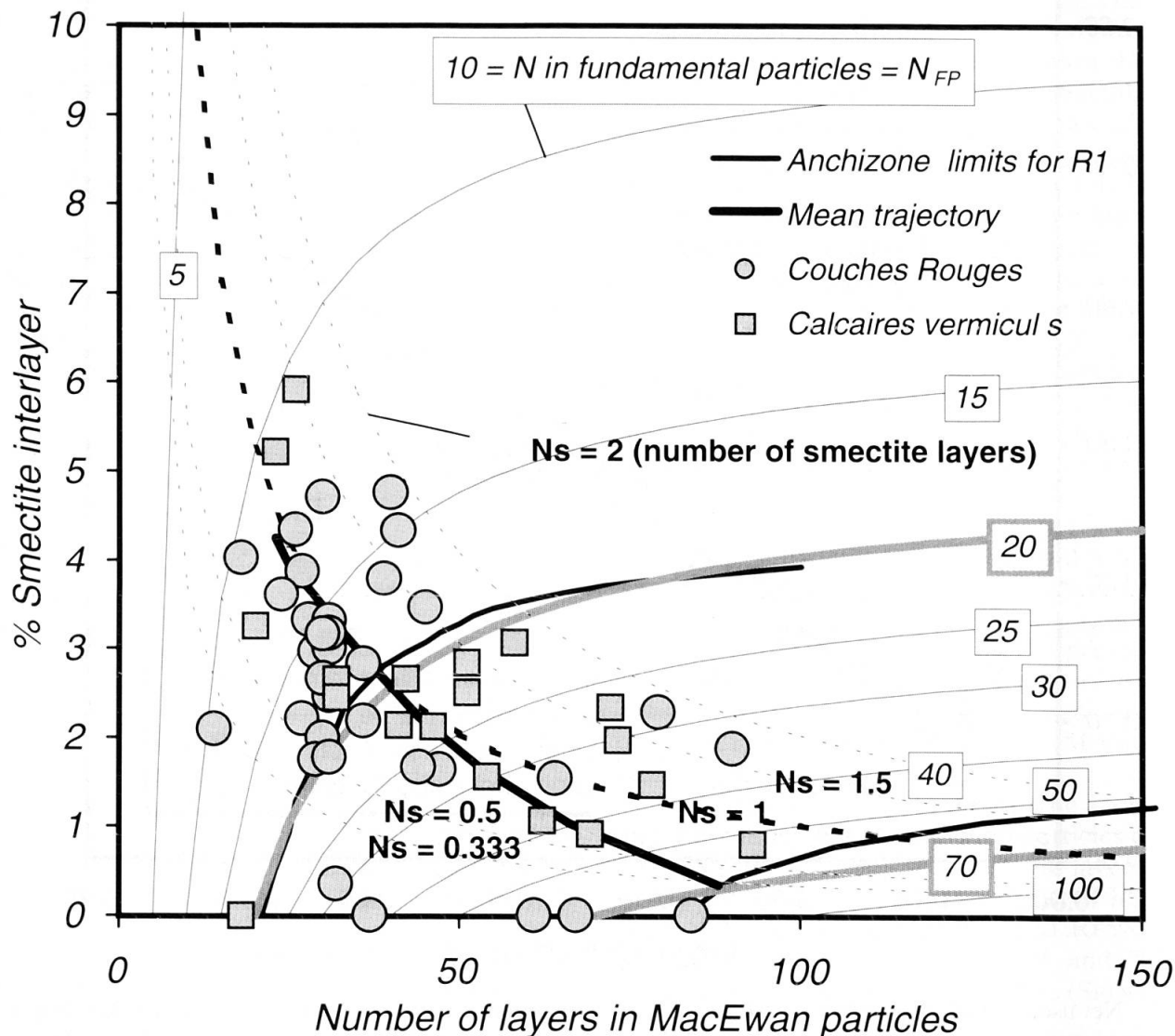


Fig. 5 Plot of the Préalpes Médiannes Rigides data interpreted with the net of figure 3. I/S (MacEwan particles) with constant N_{FP} are plotted and indicated in squares. The dashed curves show the ideal trajectory followed by an I/S crystal thickening along c^* by adding illite layers keeping N_s smectite interlayer trapped by the I/S stack. The I/S curve crosses the curve of constant N_s , indicating a decrease in smectite interlayers. At the beginning of the epizone smectite layers have almost disappeared.

sis are less variable than the classical IC values, but for deep anchizone the variability of N_{FP} is greater. The epizone limit is reached in the more internal part of the Préalpes Médiannes Rigides. The anchizone limits of $N_{FP} = 20$ and 70 agree with the IC boundaries. The N_{FP} values are consistent throughout the entire diagenetic area, whereas they show strong variations in the neighbouring epizone (Fig. 5). This is because IC is approximately inversely proportional to the layer numbers N_{FP} in I/S. As a result, when metamorphic grade increases, N_{FP} increases exponentially and the scale is less sensitive. For conditions of diagenesis N_{FP} seems to be a more reliable quantification of low-grade metamorphism than IC.

The mean path of I/S with increasing metamorphic condition in figures 3 and 5 indicates a simultaneous increase of N and N_{FP} and a decrease in %S. This involves either illitic growth along and parallel to the c^* crystallographic axis or by transformation of the smectite interlayer in illite or both. The growth of the fundamental particles does not imply that I/S does not exist as MacEwan particles. These entities can grow even within the centre of the MacEwan particles, assuming that the expandable interlayers are interfaces of illite growth which have smectitic interfaces.

When metamorphic conditions reach the epizone, as expected, fundamental particles N_{FP} in I/S (MacEwan particles) rapidly increase in size. Mineral transformations become important because the thickness and volume reached by fundamental particles (N_{FP}) overtake those in I/S (N) generated at lower metamorphic grade. If there is no external chemical contribution to the system other than rock forming minerals (feldspar, quartz,

etc.), some fundamental particles must grow to become phengites by dissolution of the smaller particles (EBERL et al., 1990; EBERL and BLUM, 1993; JABOYEDOFF, 1999). As a consequence, the number of particles diminish with increasing metamorphism. Because the mineral chemistry changes, a dissolution-precipitation process is preferred to Ostwald Ripening to describe metamorphic evolution (illite \rightarrow phengite) (BARONNET, 1991; PRIMMER, 1994; ALTANER and YLAGAN, 1997). When the greenschist facies is reached, these transformations are characterised by drastic changes in particles size (thickness and dimension along the layers) and micas become visible in hand specimens. This can be observed in the Barrhorn unit, which is equivalent to the Préalpes Médiannes Rigides in the Penninic Alps (SARTORI, 1990).

The relation between N_{FP} and the IC transposed to the Neuchâtel scale is much better than that obtained with the average number of layers N of the I/S (Fig. 6). This shows that IC can be interpreted as an empirical measurement of N_{FP} (MERRIMAN and PEACOR, 1999) in the anchizone. The average trajectory of samples from the Préalpes Médiannes Rigides shows that I/S in the anchizone has 2–3% of S, and that the epizone has approximately 0.5% of S (Fig. 5).

The results obtained by interpreting IC data using a grid from which the N_{FP} are deduced, indicates that it can be considered as a reliable and easy tool for a preliminary interpretation of IC values. This approach can give the general trend of mineral growth in a weakly metamorphosed area. The choice of another Reichweite will slightly shift the N_{FP} value but does not change the trend. The CSD thickness distribution can affect the peak shapes when the variance is very different from the one chosen in our model, but this can be evaluated by Newmod© simulations. Strain has not been considered, and its influence on I/S containing expandable interlayers remains to be studied.

5. Conclusion

The method used here is an improvement of the EBERL and VELDE (1989) method because it requires only two measurements instead of five (SW + four intensity measurements) and the quartz diffraction peak does not disturb the measurements.

Assuming that the variations of IC are essentially caused by the size effect (Scherrer equation) and by a low mixed-layering with smectite, it can be concluded that the IC is an indirect measurement of N_{FP} in the anchizone. This establishes a

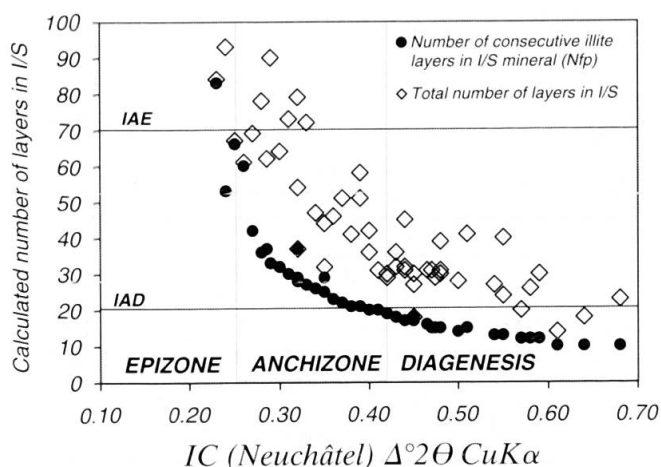


Fig. 6 Interpretation of the Neuchâtel IC scale (anchizone limits of 0.25° and $0.42^\circ \Delta 2\theta \text{ CuK}\alpha$, based on the data of the PMR, interpreted with the method outlined here (see text for explanation). IAE: interpreted anchizone – epizone limit ($N_{FP} = 70$), IAD: interpreted anchizone – diagenesis limit ($N_{FP} = 20$).

link between IC and microscopic parameters. Because IC is not sensitive to expandable layers in the anchizone, it explains why the Scherrer equation has been applied successfully (ARKAI et al., 1996). KÜBLER (1984) indicated that the IC scale is imprecise for conditions of diagenesis and epizone. Considering N_{FP} as the true metamorphic quantification, this imprecision is a consequence of poor quantification of N_{FP} by IC. The Préalpes data set (Figs 1, 5 and 6) shows that IC can be replaced by N_{FP} in order to describe low metamorphic grade conditions (EBERL and BLUM, 1993; DRITS et al., 1997, 1998; EBERL et al., 1998), especially in deep diagenesis where N_{FP} seems more accurate. But such an interpretation needs still some adjustments because of detrital micas which slightly affect the 10 Å peak. The limits of the N_{FP} scale will be slightly lowered when such effect are quantified. The role of defects must also be clarified.

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