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PATISSIER: software to estimate the smectite content and number of consecutive illite layers in mixed-layer illite-smectite using illite crystallinity data

by *Michel Jaboyedoff* [1, 2, 3] and *Philippe Thélin* [1]

Abstract

Illite crystallinity is an efficient low-grade metamorphic indicator, the value of which depends on several factors. Program NEWMOD allows one to evaluate the dependence of IC for air-dried and ethylene-glycol treated samples on the mean thickness (number of layers) of illite crystallites (N), on swelling interlayer (smectite) content (%S) of illite, and with ordering parameters that describe layer stacking. Using these relationships, software "PATISSIER" allows the estimation in illite-smectite of N, %S, and the number of consecutive illite layers. The latter corresponds to the physical value hidden in IC that best describes metamorphic grade.
Software "PATISSIER" can use old data sets to reinterpret many existing measurements using the IC method, the so-called Kübler index.

Keywords: Illite crystallinity, software, illite-smectite, mixed-layer, XRD.

1. Introduction

Illite crystallinity (IC) was defined as an empirical indicator of incipient metamorphism. It requires the measurement of the illite 10 Å X-ray diffraction (XRD) peak width at half maximum (FWHM is measured from the peak maximum to the background), of an air-dried (AD) oriented sample of the < 2 μm size fraction. In many papers IC has been termed the "Kübler Index". Traditionally, CuKα radiation is used, and the measurement unit is then °Δ2θ CuKα. IC values decrease with increasing metamorphic grade. Use of IC was mainly promoted by two Swiss geologists: firstly by BERNARD KÜBLER (KÜBLER, 1964, 1967, 1968, 1984) and more recently by MARTIN FREY (FREY, 1969, 1987; FREY and ROBINSON, 1999). We dedicate this paper to this two colleagues.

Illite was first defined by GRIM et al. (1937) as a clay-size mica occurring in argillaceous sediments, with a chemical composition slightly different from that of phengite. Illite in diagenetic environments has been re-defined as a true mineral phase (MEUNIER and VELDE, 1989; ŚRODOŃ et al., 1992). The chemical composition corresponds to illite layers within interstratified minerals such as illite-smectite (I-S). For convenience I-S with less than 10% smectite layer content (%S) will be called illite, as albite for feldspars (see MOORE and REYNOLDS, 1997). In fact the term illite used in "Illite Crystallinity" corresponds to this last illite definition and the IC method is valid for samples where %S does not exceed 10%.

IC has been successfully applied to the transition zone between diagenesis and metamorphism. The transition zone was defined by IC values and denoted as the "anchizone", which can be correlated with the sub-greenschist facies. For low-grade diagenesis and high epizone, the IC method is not relevant because IC values have no physical meaning far from the anchizonal IC values (KÜBLER and JABOYEDOFF, 2000). But the IC method is still relevant at high diagenesis conditions and at the beginning of the epizone. These zone limits were originally set to 0.42 °Δ2θ CuKα for diagenesis–anchizone and to 0.25° (KÜBLER, 1967) for anchizone–epizone (greenschist facies) using a Philips diffractometer. IC scale values depend on

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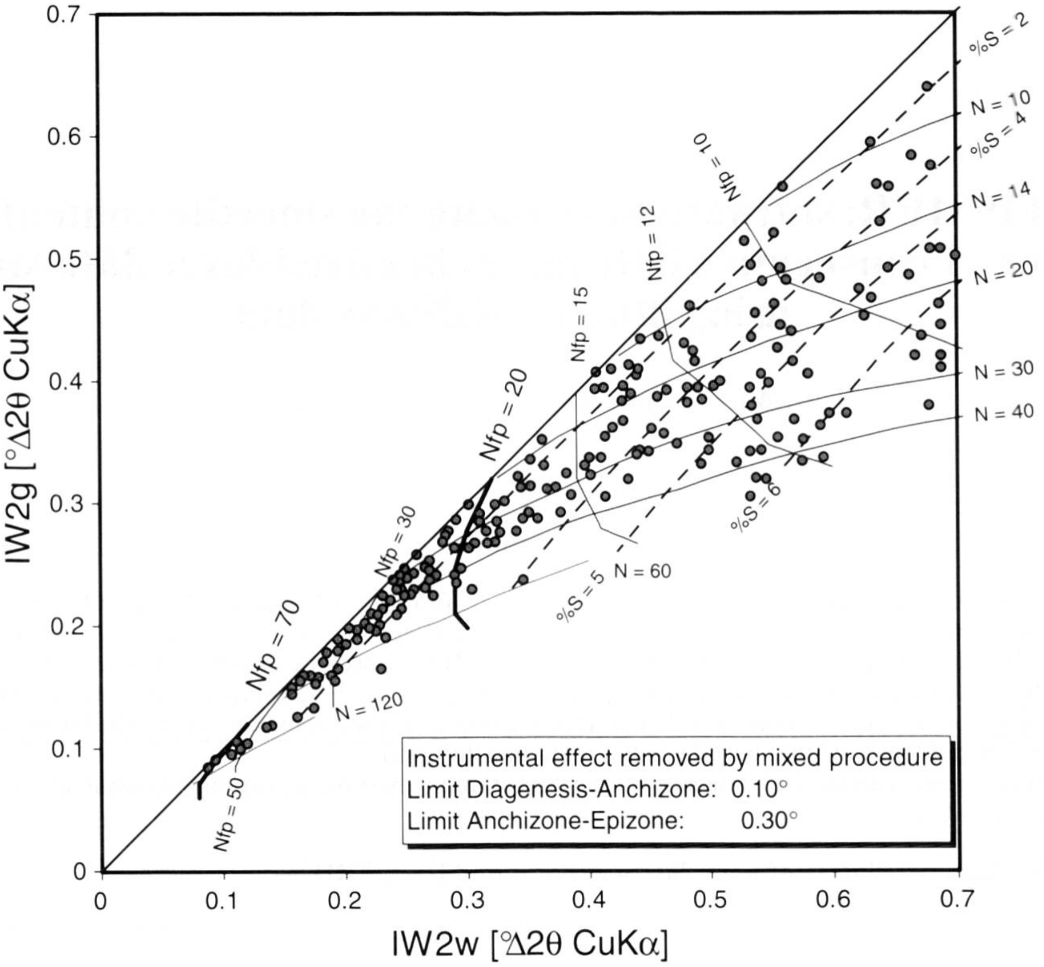


Fig. 1 Plot of IWAD-IWEG data of KISCH and FREY (1987) after instrumental effects were removed from ICAD and ICEG by geometric mean of Lorentzian and Gaussian corrections (= mixed procedure) in a calculated IWAD-IWEG graph (=IW2w-IW2g), 2 water layers (2w) and 2 ethylene-glycol layers (2g) are assumed. The grid lines are output from program "PATISSIER".

several technical factors: geometry of the diffractometer, electronics and XRD profile treatment (KISCH, 1987, 1991). Furthermore, newer diffractometers and associated software may change the way IC is measured (KÜBLER, 1990). Thus, the present tendency is to obtain the anchizone limits corrected for instrumental broadening, which leads to values around 0.10° and 0.30 °Δ2θ CuKα (JABOYEDOFF, 1999; JABOYEDOFF et al., 2001a), independent of diffractometer characteristics.

The difference between the IC (AD), traditionally measured on air-dried samples, and the IC values of ethylene-glycol (EG) treated samples was used to give a rough estimate of the swelling layer content (KÜBLER, 1984; KRUMM, 1984). For convenience and in keeping with terminology used in JABOYEDOFF et al. (2001a) we use the term ICAD for the standard IC value and ICEG for the IC measured on EG specimens (see Table 1 for abbreviations used). Using software NEWMOD (REYNOLDS, 1985; REYNOLDS and REYNOLDS, 1996), x-y graphs ICAD versus ICEG are used (Fig. 1). These graphs permit the determination of the mean number of layers (N) in the coherent scattering domain thickness of I-S, %S (in % of N), and the mean number of consecutive illite layers in I-S layer stacking (N_{fp}). The IC value is an indirect measure of N_{fp} (JABOYEDOFF et al., 2001a). The latter is considered to be a good metamorphic estimator.

A method to determine N and %S was proposed by EBERL and VELDE (1989), but their way of measuring IC is not an internationally applied standard technique because the background was traced horizontally. The estimation of N, N_{fp} or the distribution of coherent scattering domain size has been studied recently by the Berthaut-Warren-Averbach (BWA) method (ARKAI et al., 1996; WARR and RICE, 1994; DRITS et al., 1998). In order to properly apply the BWA method, the expandable interlayer must be eliminated by sample expansion to about 50 Å by dispersion in a polymer such as PVP-10 following saturation of clays with Na (LANSON and KÜBLER, 1994; DRITS et al., 1998; EBERL et al., 1998). This method does not take much more time than measuring IC, and only one measurement is needed. But IC remains a widespread method and reinterpretation of ancient data justifies the present approach. Therefore, we developed software "PATISSIER", written in Visual Basic 6.0 (Microsoft©), using IC data (ICAD and ICEG), it permits rapid estimation of N and %S, with no additional treatment (Fig. 1).

2. Theoretical basis

NEWMOD simulations do not take into account instrumental broadening. To use the chart defined by NEWMOD, instrumental effects have to be removed from the measured IC value. Basically, the pure XRD profile [$f(2\theta)$] (without instrumental effects) is broadened by effects of the X-ray source and the geometry of the diffractometer. The shape of instrumental profile [$g(2\theta)$] is estimated using a powder of very well crystallized mica. As shown by KLUG and ALEXANDER (1974), an experimental peak profile corresponds to the convolution of the pure and instrumental profile [$h(2\theta)=f(2\theta)*g(2\theta)$]. Depending on the profile type, a simple expression of the instrumental width can be found (see below). The true illite peak width (IW) corresponds to the IC corrected for instrumental broadening. The IW abbreviation was introduced by JABOYEDOFF et al. (2001a), who describe in detail the method behind the software. IWAD and IWEG correspond, respectively, to ICAD and ICEG corrected for instrumental broadening. IW is mainly controlled by the following factors:

- Coherent scattering domain
  (i) **Mean thickness (N) or mean number of layers**
  (ii) Shape of the histogram of thickness or of the number of layers
- **%Smectite (%S)**
- **Stacking order (R)**
- Chemical composition
- Lattice strain

In the program "PATISSIER", only mean thickness, smectite layer percentage, and stacking order are considered. Thickly stacked layers produce narrower XRD profiles than thinner stacks due to basic principles of wave interference. The presence of expandable layers (%S) is a source of peak broadening because the periodicity of the lattice is broken, whereas order furnishes an increase in lattice periodicity, leading to peak narrowing. Other variables like lattice strain are either fixed or neglected. The distribution of particle thickness plays an important role for the diffraction peak shape, but for most of the distributions, the program gives satisfactory results for the mean thickness. Furthermore, for identical mean and variance of thickness, it can be shown that a different distribution type gives very similar IW values (JABOYEDOFF, 1999; JABOYEDOFF et al., 2001a).

The consecutive illite stacks in I-S may be viewed as a fundamental element of I-S, they are called fundamental particles by NADEAU et al. (1984). I-S may be viewed as stacked packets of fundamental particles separated by smectite type interlayers. The mean thickness N_{fp} of fundamental particles is the number of 10 Å layers. N is defined as the average of the total number of layers including illitic and smectitic layers of an I-S, otherwise known as MacEwan particles, which are therefore stacks of fundamental particles. MacEwan particles consider an I-S crystallite as a single mineral, and this view is compatible with fundamental particle concepts (MOORE and REYNOLDS, 1997; p. 180; ALTANER and YLAGAN, 1997; MEUNIER et al., 2000). Note that $N \geq N_{fp}$ (ŚRODOŃ et al., 1990, 1992) because $N_{fp}=N/(1+(N-1)\times(\%S/100))$. HRTEM results often furnish information on N because expandable layers are collapsed

Table 1 Abbreviations used in text and figures.

Variables	
R	Reichweite, number of neighbours affected by the layer type
%S	Percentage of smectite interlayers in mixed-layer illite-smectite
IC	Illite crystallinity index
ICAD	FWHM of the 10 Å illite peak for air-dried sample (= standard IC)
ICEG	FWHM of the 10 Å illite peak for ethylene-glycol treated sample
IW	FWHM of the 10 Å illite peak corrected for instrumental effects
IW2g	Calculated IW assuming two ethylene-glycol layers in the interlayer
IW2w	Calculated IW assuming two water layers in the interlayer
IWAD	IW from air dried sample
IWEG	IW from ethylene-glycol treated sample
k	Constant proportional to %S×N
N	Mean number of layers in a coherent scattering domain
N_{fp}	Mean number of layers in illite fundamental particles

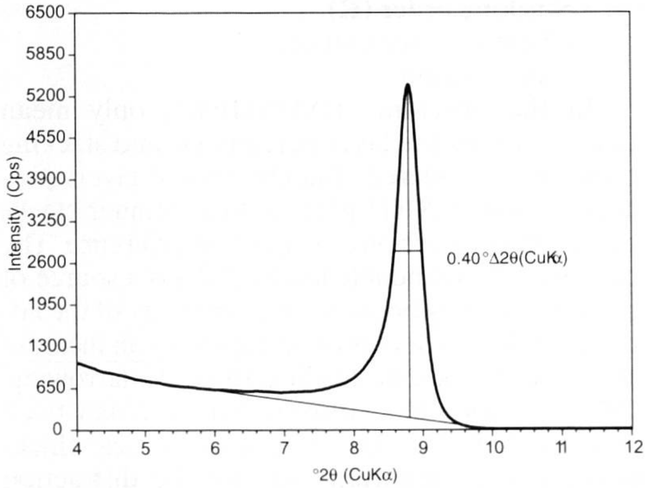


Fig. 2 Illustration of the method used for background and IC or IW measurements.

(Jaboyedoff et al., 2001b). The increase of N_{fp} has been found to be a good indicator of increasing metamorphism (Eberl and Środoń, 1988; Eberl et al., 1987, 1990; Jaboyedoff et al., 2001a). As N_{fp} increases, the stacking order of the I-S increases, with the type of layer depending on its first, second, etc. neighboring layers. R (Reichweite in the sense used for NEWMOD, see Moore and Reynolds, 1997) equals the number of neighbors of one layer, which are affected by its type. For example, an I-S with order R = 1, containing less than 50 %S indicates that every smectite layer possesses at least one illite layer as first neighbor on both sides such that all S layers are linked with one illite layer in a IS sequence, while the remaining illite layers are freely distributed. In other words, no consecutive smectite layers can be found. R = 3 indicates that all smectite layers are involved in an ISII sequence, but this order type cannot occur if %S > 25%. I-S with R = 0 means that a layer has no influence on the type of the neighbor layers.

3. IC and IW measurements

The IW measurements and NEWMOD simulations used for creating the charts were performed in a very simple way. A linear background is traced under the 10 Å illite peak (Fig. 2), this choice makes the method systematic, and usually peaks are sufficiently sharp to use this method, even if some noise is present. Background tracing is realized in such a way as not to cross the base of the peak outside the limits of the peak, and it is tangent to the mean background. The width of the peak is measured at half the distance between the background and the maximum peak intensity.

The NEWMOD simulations are not affected by any instrumental broadening effects. Thus, to compare the simulations with experimental IC data, the XRD data have to be corrected. This is implemented in the program in several ways, but the program also can use data that are already corrected.

The use of the program has some restrictions in the way IC data are measured. If data are smoothed in order to reduce noise, diminishing of the peak maximum must be avoided. If automatic background tracing is used, its subtraction from the signal must use a background calculation not influenced by the peak intensities. This means that the background must not vary directly under a peak.

4. Method

In order to obtain a value of N, %S or N_{fp} from IWAD and IWEG, several measurements of IW using NEWMOD simulations are necessary to create interpretative charts. NEWMOD simulations were performed using the standard parameters (d-value of illite of 9.98 Å, smectite-2g of 16.9 Å, smectite-2w of 15 Å; for Ca exchange cation, generally giving 2 water layers). This implies that IWAD simulations are performed using 2 water layers for smectite interlayers, requiring Ca-saturation of clays, and IWEG 2 ethylene-glycol layers. Theoretical results are abbreviated IW2w and IW2g (Fig. 1). For a given stacking order, simulations yield iso-values of N, %S and N_{fp} in a graph IW2w-IW2g. Plotting in this graph, IWAD and IWEG values lead to estimates of N and %S. Details can be found in Jaboyedoff et al. (2001a).

For simulation, the following instrumental parameters are used:

The divergent slit is set to 0.5°; goniometer radius to 18.5 cm; sample length to 2.8 cm, two soller slits to 5°; and composition to $K_{0.9}$ ($Al_{1.8}$, $Fe^{2+}_{0.2}$) ($Si_{3.3}$, $Al_{0.7}$) O_{10} $(OH)_2$ for illite and $Ca_{0.17}$ ($Al_{1.66}$, $Fe^{2+}_{0.2}$, $Mg_{0.14}$) (Si_4) O_{10} $(OH)_2$ for smectite. The histogram of crystallite number of layer by CSD is assumed uniform. For instance, this means that crystallites containing 8, 9, 10, 11 and 12 layers are in equal number in the diffracting powder. They have a uniform histogram with a mean N equal to 10. Following the same principle, the histogram values are chosen constant for the interval [N – 10; N + 10] and zero outside this interval. For example, if N = 22, the simulation is performed using the same number of crystallites containing 12, 13, … or 32 layers. For $N \leq 12$, the same constant thickness histogram is used, but the interval of the constant histogram value is set to [2; (2N – 2)], be-

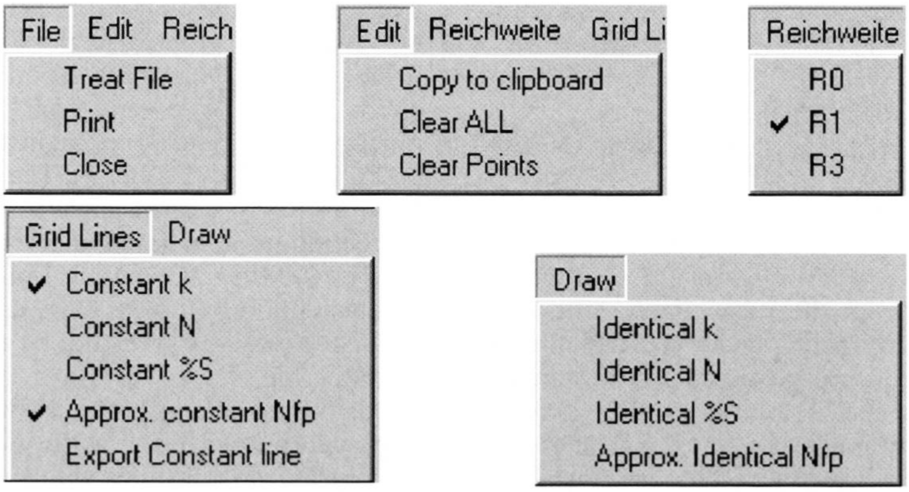
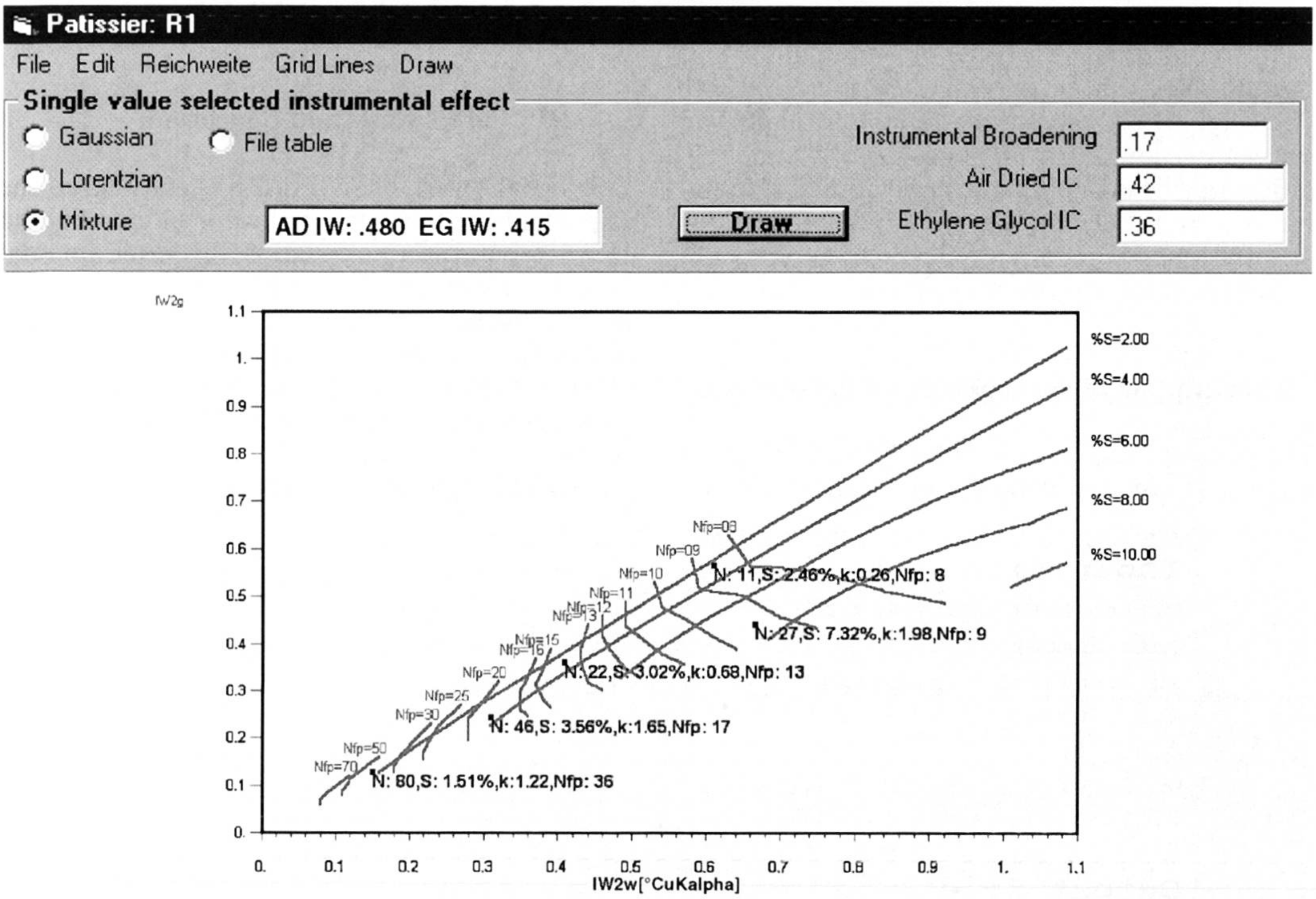


Fig. 3 Menus from the program "PATISSIER", with settings used in the Fig 4.


Patissier: R1

Single value selected instrumental effect

○ Gaussian ○ File table

○ Lorentzian

● Mixture

AD IW: .480 EG IW: .415

Instrumental Broadening .17

Air Dried IC .42

Ethylene Glycol IC .36

Fig. 4 Window of the program "PATISSIER" with an example including the correction for an instrumental broadening of 0.17 °Δ2θ CuKα and other five examples obtain by clicking directly on the window.

cause if 10 is used, then the lower boundary of the interval is negative.

For convenience of computation, NEWMOD simulations were performed using a relationship-linking %S and N:

$$\frac{\%S}{100} = \frac{k}{N}$$

where k is a constant. For instance k=1 indicates that for N=10 %S =10%. For k=0 and %S=0 IW2w is equal to IW2g which defines a 1/1 straight line in IW2w – IW2g plot. Curves for k > 0 are more distant from the 1/1 line with increasing k.

Convolution of simple peak shape such as Gaussian or Lorentzian leads to a simple relation between peak widths at half maximum measurement. Three simple correction types are implemented in the program:

- $IW = (IC^2 - IW_{inst}^2)^{1/2}$ (Gaussian, KLUG and ALEXANDER, 1974)
- $IW = (IC - IW_{inst})$ (Lorentzian, KLUG and ALEXANDER, 1974)
- $IW = [(IC - IW_{inst}) \times (IC^2 - IW_{inst}^2)^{1/2}]^{1/2}$ (Mixed, BALASINGH et al., 1991)

where IW_{inst} is an estimate of the instrumental broadening width for the 10 Å peak position. Another solution is to read from a file a table linking the values without correction and the corresponding corrected values. This file can be created from convolution method (JABOYEDOFF, 1999; JABOYEDOFF et al., 2000). Unfolding (deconvolution) may also be used to remove instrumental broadening effects.

From the values IWAD and IWEG and for a given R-value, k, N, %S and N_{fp} are estimated in two steps. The first step consists in defining the k value by using a fifth order polynomial fit that passes through origin on simulated points for k=0, 0.5, 1, 2, 3. The k value is found by iteration using linear interpolation of the known polynomial coefficients; the weight of each polynomial is found (w_1 and w_2). Polynomials also are obtained for known k and IW2w versus 1/N. Then, using w_1 and w_2 for the same k values, N is found.

5. Description of the capability of the software

The software displays a graph of IW2w-IW2g for a given R which can be chosen from the menu Reichweite between R = 0, 1, 3 (Fig. 3). By clicking in the window graph it is possible to obtain the value of k, %S, N and corresponding N_{fp}. If needed, the values IWAD and IWEG may be entered numerically in fields, and the instrumental broadening IW_{inst} also can be set. The data IWAD and IWEG and IW_{inst} also can be read from a file. The correction is calculated after choosing among four possibilities: Gaussian, Lorentzian, geometric mean of both (mixed) or by using a file (Fig. 4). This choice can be set either by checkbox or read from the file.

Grid lines of constant k, N, %S or N_{fp} may be displayed and saved in a file. For any point the corresponding grid lines with the same k, N, %S or N_{fp} may be drawn (Fig. 3). Data read from a file are saved in an output file (Fig. 5). A bitmap copy of the results window can be saved or printed in high quality.

6. Example and Conclusion

Because the variable N_{fp} of I-S is considered the important metamorphic indicator hidden behind IC (JABOYEDOFF et al., 2001a), this program enables the transformation of IC to N_{fp}, and that indeed constitutes the main advantage of this technique. For example it is possible to interpret IWAD-IWEG data (Figure A.1 in KISCH and FREY, 1987), using software PATISSIER in terms of the number of consecutive illite layers (Figs. 1 and 6). This method supports the re-evaluation of

```
Input file
Title: FREY AND KISCH DATA
Data number: 210
Instrumental broadening: 0.17
0.81, 0.75
0.83, 0.73
0.78, 0.74
0.73, 0.69
...
Output file
FREY AND KISCH DATA
Reichweite: R1
Instrumental broadening: 0.168
Instrumental broadening type: Lorentzian-Gaussian geometric mean
IWAD        IWEG             N          %S      k        Nfd
0.715       0.652            9          2.58    0.23     7
0.730       0.630            10         3.6     0.35     7
0.678       0.639            9          1.84    0.16     7
0.632       0.594            10         1.9     0.18     8
...         ...              ...        ...     ...      ...
```

Fig. 5 Format of the input and output files.

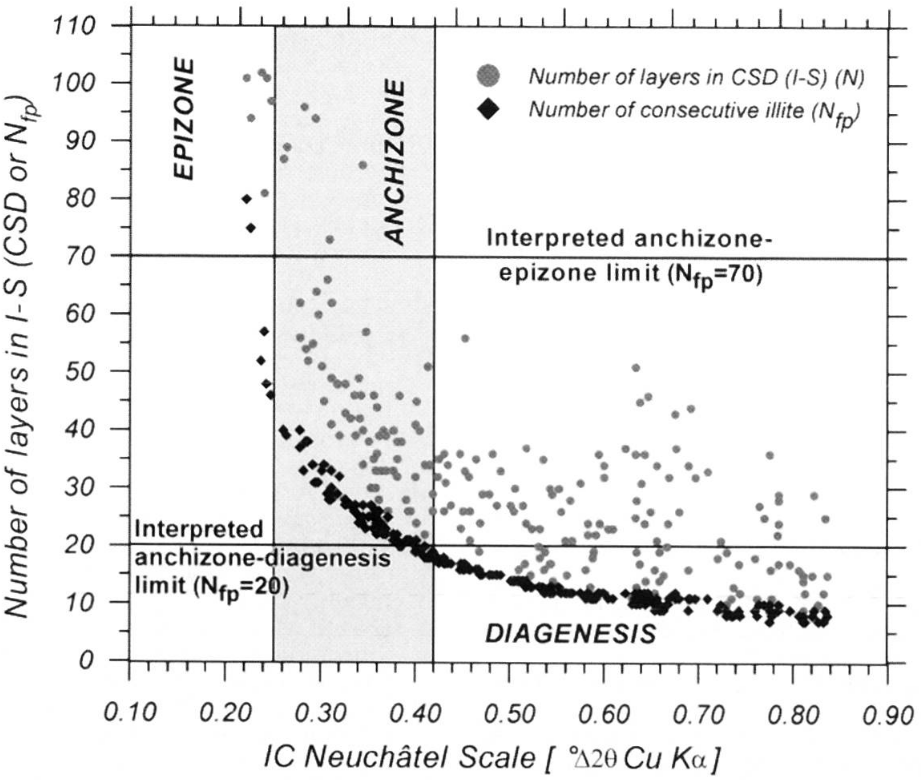


Fig. 6 N and N_{fp} calculated by the program "PATISSIER" for data from Fig. 1 (Kisch and Frey, 1987). The IC scale is the original scale from Neuchâtel. Note the relation existing between IC and number of layers in I-S and N_{fp}.

previously published data sets, such as ICAD-ICEG graphs (Krumm, 1984; Yang and Hesse, 1991). In this case, the instrumental broadening IW_{inst} was the value used from the original IC scale of Neuchâtel (Philips diffractometer), i.e. 0.17 °Δ2θ CuKα. Figure 6 clearly shows that N is not a value relevant to quantify metamorphism. On the other hand, it is clear from the data that IC values are related to N_{fp} values. The method was applied in a case study from the Prealps by Jaboyedoff et al. (2000) and the use of N_{fp} as a low grade metamorphic descriptor is well confirmed.

Grid lines of constant N_{fp}, %S, N values are very useful. They can be used to interpret data, for example in terms of regional metamorphic gradient, because they show which of the variables N_{fp}, %S or N are changing with increasing metamorphism (Jaboyedoff et al., 2000). Such trends can give information on the processes of crystallization. Average values like N or N_{fp} are often sufficient to invoke working hypotheses about crystallization mechanisms.

The advantage of this software is that standard data of IC can be used for a reinterpretation, even from published data sets, provided ICEG exits, as it is often the case. The problem then is to obtain or estimate the instrumental broadening. The results may be used as a start to analyse in detail XRD profiles by NEWMOD simulations (see for example Jaboyedoff et al., 2001a). The program PATISSIER is available on the web at http://www.quanterra.org.

Acknowledgement

Bernard Kübler and Martin Frey are thanked for their enthusiasm over the past 40 years during which they stimulated research on low-grade metamorphism in Switzerland and elsewhere.
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