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On the calibration of the illite Kübler index (illite "crystallinity")

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Abstract

The measurement of the Kübler Index KI [i.e., the full width at half maximum (FWHM) value of the X-ray powder diffractometric first basal reflection of illite – dioctahedral K-white mica, formerly called also illite "crystallinity" (IC)], is rather simple and quick; however, the delimitation of the KI zones still remains controversial at present, mostly because of the numerous factors that affect the standardization and inter-laboratory calibration of the KI scale.

The differences reported between the KI values and scales among different laboratories are considered to be due to (1) X-ray diffractometer settings for measurement of FWHM, (2) sample-preparation procedures, and (3) techniques of measuring or obtaining the FWHM values from the line profiles. The effects of (1) are monitored by the use of polished-slate inter-laboratory standards. In order to consider the effects of differences in sample-preparation, Warr and Rice (1993, 1994) distributed inter-laboratory "CIS" standards in the form of (meta)pelitic rock chips and a muscovite flake that require both preparation and measurement in the various laboratories. The "CIS" values given by these authors for these standards were purportedly converted to the Kübler scale using a calibration curve based on measurements on a set of polished-slate slabs and a muscovite flake prepared by Kisch as inter-laboratory KI standards. However, these "CIS" values are much broader than those obtained by virtually all other laboratories, and are considered anomalous; the "raw" data as restored from these "CIS" values are implausibly broad. Comparison with the "raw" values of Warr and Rice suggests that the high- and low-grade boundaries of the anchizone in their "CIS" scale are 0.295 and 0.49–0.50°Δ2θ, much broader than the Kübler-equivalent 0.25 and 0.42°Δ2θ. Similarly, regressions of the "CIS"-equivalent against the slate-slab calibrated Kübler-equivalent values of several laboratories show that the purportedly Kübler-equivalent anchizone-boundary values of 0.25 and 0.42°Δ2θ in the "CIS" scale in fact correspond to much narrower Kübler values.

Consequently, the use of the CIS scale boundaries results in increasing confusion when CIS-calibrated KI values are used for petrogenetic purposes in regional comparisons. This discrepancy is likely to reflect errors in the conversion of the "raw" FWHM values into Kübler equivalents. Data on inter-laboratory chip standards are difficult to evaluate unless the "raw", uncalibrated data are also given: such data should be included in all papers reporting on KI.

Keywords: illite, dioctahedral K-white mica, Kübler index, "crystallinity", inter-laboratory calibration, anchizone boundaries, very low-temperature metamorphism, X-ray powder diffractometry.

Introduction

Metasedimentary rocks of normal marine fine-clastic origin are usually devoid of metamorphic facies-indicating minerals or mineral assemblages in the low temperature realm ranging from diagenesis up to the low temperature part of the greenschist facies. For determining the diagenetic -metamorphic zones (grades) of such rocks that are widespread in sedimentary basins and in the outer fold-and-thrust zones of the orogenic belts, the X-ray powder diffractometric (XRPD) illite Kübler index (KI) method has been successfully applied worldwide. Systematic changes in sharpness of the XRPD 10 Å basal reflection of illite-dioctahedral potassic white mica upon burial (increasing temperature) were recognized first by Weaver (1960), who introduced the so called "sharpness ratio" for petrogenetic purposes. The term illite "crystallinity" (IC) in its current form, the full width at half maximum (FWHM) of the 10 Å XRD peak of illite-muscovite as measured on the <2 μm size fraction, was introduced by Kübler (1967) and has been increasingly used worldwide for determining the grade of diagenesis and very low-grade metamorphism of clay-rich, clastic sedimentary rocks during the subsequent three decades. Because of the complexity and manifold

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interrelations of the mineralogical factors that affect this parameter, the term "crystallinity" has been used by many authors in quotation-marks. Kübler (1984) replaced it by Scherrer Width (largeur de Scherrer). This term is preferentially cited and used as Kübler index (KI) at present (e.g., Merriman and Frey, 1999; Merriman and Peacor, 1999, etc.), and is recommended by the Association Internationale pour l'Étude des Argiles (AIPEA) Nomenclature Committee to be used instead of illite "crystallinity index" or "crystallinity" (Guggenheim et al., 2002). This expression will be used throughout this paper.

It is essential not only for the specialists but also for geologists using the KI results for paleotectonic and geodynamic reconstructions that the boundaries of the Kübler index-based so called diagenetic, anchi- and epizones can be determined and used unequivocally.

However, from the outset there have been differences between the values and scales of KI used by different laboratories. These differences are due to lack of uniformity in three categories of laboratory and measurement procedures, namely:

(1) X-ray diffractometer settings adopted for measurement of the FWHM values (scan rates, time constants, slit widths, use of filters, step scanning vs. diffractometer traces);

(2) sample-preparation procedures (grinding methods, use of acid treatment and cation saturation, grain-size separation methods, clay-layer thickness sedimented or smeared on glass slides, etc.) and

(3) techniques used for measuring or determining the "raw" (uncalibrated) FWHM values.

The effect of factor (1) has been elaborated by Kisch (1990) and can be monitored by the comparison of measurements on polished-slate inter-laboratory standards. In contrast, the effects of the factors (2) are still largely unknown, although an attempt has been made to minimize these effects by the recommendation of uniform preparation procedures by the Working Group for Illite Crystallinity (Kisch, 1991). In theory and also in practice, possible effects of factor (3) can be ruled out, provided the FWHM values of the investigated samples and the standards used for calibration are measured in the same way.

The aims of the present paper are (a) to show some sources of these methodological difficulties, (b) to contribute to the solution of these problems with an attempt at standardization of KI values, based on the distribution of inter-laboratory standards in the form of rock chips that require both preparation (grinding, treatment, separation of the <2 μm size fraction) and measurement by the various laboratories, and (c) to offer suggestions concerning the calibration of the KI scales.

Use of Kübler index (illite "crystallinity") standards

From the early 1970s and onwards, the late Bernard Kübler (Institute of Geology, University of Neuchâtel, Switzerland) and Hanan J. Kisch (Department of Geology and Mineralogy, Ben-Gurion University of the Negev, Beer-Sheva, Israel) distributed polished slate slab standards upon request to different laboratories for the calibration of the KI scales, considering the effects of different instrumental settings (e.g., goniometer speed, etc.) on FWHM. Subsequently, Kübler's and Kisch's scales were correlated against each other, and it was found that Kisch's values were ca. 0.04°Δ2θ narrower. This was due to the use of a

Table 1 Boundary values of the anchizone *sensu* Kübler (1967) as calibrated by Kisch and the equivalent CIS values as obtained using equation (1) of Warr and Rice (1994) and equations (4a) to (7) of the present paper. Values in °Δ2θ, CuK_{α}.

author/laboratory	equation	diagenetic zone/anchizone	anchizone/epizone
Kübler, Neuchâtel		*0.25*	*0.42*
Kisch, Beer-Sheva		*0.21*	*0.38*
Warr and Rice (1994)	(1)	0.29	0.55
Warr, Heidelberg, as measured*	(4a)	0.232	0.422
calculated CIS boundary values	(6)	0.293	0.491
calculated CIS boundary values	(7)	0.289	0.489
Warr, Heidelberg, as measured*	(4b)	0.225	0.424
calculated CIS boundary values	(6)	0.286	0.493
calculated CIS boundary values	(7)	0.282	0.490
Warr, Heidelberg, as measured*	(5)	0.237	0.435
calculated CIS boundary values	(6)	0.298	0.504
calculated CIS boundary values	(7)	0.294	0.502

*from the CIS page of the VLGM web-site, Heidelberg

scanning rate of 0.5°2θ/min by Kisch instead of the 2°2θ/min by Kübler, applying the same time constant (for a discussion of the effects of scanning rates and time constants on FWHM values see Kisch, 1990). As polished slate standards are not ground, size separated, chemically or otherwise treated other than by mechanical polishing, the resulting differences between the half-height peak widths account for effects of diffractometer settings only, and not for any effects introduced by differences in preparation procedures. Kisch's and/or Kübler's polished rock slab standard series have been run by a large number of laboratories worldwide. At present, many slate slab series calibrated against either Kübler's or Kisch's laboratories are in use in various laboratories dealing with KI measurements.

Warr and Rice (1993, 1994) introduced standards referred to as "Crystallinity Index Standard" for KI (hence: CIS standards) in the form of chips of four rock samples (SW1, 2, 4 and 6) and a muscovite flake (MF1). Contemporaneously, Stefan Krumm (Institute for Geology and Mineralogy, University of Erlangen-Nürnberg, Germany) distributed six slate chip standards (ILC1-6) for similar purposes. These standards require preparation involving grinding, separation of the <2 μm size fractions, their sedimentation on glass slides, and measurement of the FWHM values of the 10 Å mica and 7 Å chlorite or kaolinite reflections. Warr and Rice (1993, 1994) established a calibration curve based on their 10 Å FWHM values against those obtained by Kisch on five polished slate slabs and one muscovite crystal distributed by Kisch, deriving a regression equation

$$KI_{<Heidelberg>} = 1.511558 \times KI_{<Kisch>} - 0.029329$$
(Warr and Rice, 1994, p. 144). [1]

(Note that illite Kübler index was abbreviated as IC that time). Warr and Rice (1994) stated that "IC values quoted in the following parts of this (their) study have been converted to the Kübler scale by using the above calibration equation, and then adding a constant of +0.04°Δ2θ, representing the difference between the IC scales employed by H.J. Kisch and B. Kübler (Kisch, 1980, 1990)." Because of the differences in instrumental settings, the boundary values of the anchizone are 0.25 and 0.42°Δ2θ in Kübler's scale which correspond to 0.21 and 0.38°Δ2θ in Kisch's scale (Kisch, 1990).

Rearranged, the conversion formula of Warr and Rice (1993, 1994) to the Kübler-equivalent scale (hence : $KI_{<Kübler-equiv.>}$) then becomes

$$KI_{<Heidelberg>} = 1.511558 \times (KI_{<Kübler-equiv.>} - 0.04) - 0.029329, \quad [2]$$

or

$$KI_{<Kübler-equiv.>} = KI_{<Heidelberg>}/1.511558 + 0.059403 \quad [3]$$

Note that Warr and Rice (1993, 1994) published only equation [1]. Equations [2] and [3] are derived from equation [1] by the present authors. As these linear regression equations used for calibration were obtained on samples unaffected by grinding, size separation, or chemical treatment other than mechanical polishing, they only account for effects of diffractometer settings or techniques used for measuring the FWHM, and *not* for any effects introduced by differences in preparation procedures. FWHM values for the 10 Å and 7 Å peaks of the <2 μm fractions of these standards (hence: "CIS values"), were given by Warr and Rice (1993, 1994). These values are stated to have been "calibrated", that is transformed into Kübler equivalents using equation [1]. Insofar as these CIS values have been thus transformed into Kübler equivalents, the low-grade and high-grade limits of the anchizone in this "CIS scale" should be identical to those established by Kübler, i.e. 0.42°Δ2θ and 0.25°Δ2θ, respectively.

Anomalously broad "calibrated" peak widths of Warr and Rice (1994)

Over the years, Warr distributed the CIS standard sets for many laboratories all over the world; so did also Krumm with the ILC standards. These CIS and ILC standards have been prepared and the FWHM values of the 10-Å and 7-Å reflections of the <2 μm fractions measured by a number of laboratories, including those of the authors. The KI (=IC) values measured on these CIS (SW1-6) and on Krumm's ILC1-6 slate rock chip standards and on the MF1 muscovite flake standard by the various laboratories have been compiled and are available on the web-site of "Very Low-Grade Metamorphism" established and administrated by Krumm and Warr (http://www.rzuser.uni-heidelberg.de/~jr7/vlgm/cis.html).

The values measured and reported by virtually all of these laboratories, presumably the "raw" FWHM values, as measured, are appreciably narrower than the purportedly "calibrated" FWHM data ("CIS values") given by Warr and Rice (1993, 1994). This divergence suggests, but by itself does not prove, that the narrower values measured by these other laboratories are closer to the "true" values, but it warrants some closer inspection of the procedures followed by Warr and

Rice for possible error. The anomalously broad FWHM values measured on the XRPD profiles given by Warr and Rice (3/1993 unpublished report, Fig. 5; 1994 paper, Fig. 3), and the identical CIS values listed in their Tables 2 and 3, are claimed by Warr (written personal communication to H.J. Kisch) to be recalculated "calibrated" Kübler-equivalent traces/values rather than the original diffractometer traces/values "as run". If, on the other hand, these "CIS values" were to be taken to be the "raw", as measured, values before calibration, their Kübler/Kisch equivalents after calibration would be much narrower, and much more in conformity with the values measured by most of the other laboratories that have prepared and measured Warr and Rice's (1994) CIS standards.

A possible yet unproven explanation for the anomalously large FWHM values of the CIS standards is based upon the differences in measuring the "raw" values. Originally, Kübler and Kisch, and most of their followers have measured the "raw" FWHM values manually, using the experimental XRPD peak profiles of the standards and the investigated samples, naturally after subtracting the background. By contrast, Warr and Rice (1994, p. 144) followed an other procedure: "The program FIT [of the Siemens DIFFRAC-AT (version 3) software] was used to determine the crystallinity by first subtracting the background from the raw data, followed by peak fitting using a Split Pearson 7 function. From the fitted data, the crystallinity was measured by the FWHM (full-width-half-maximum) parameter of the program." No information is available on the possible differences between the FWHM values measured manually on the experimental XRPD profiles or calculated with mathematical processing of the natural profiles.

Since Warr and Rice did not publish their measured FWHM values, it is impossible for the outside user to repeat or evaluate their calibration procedures. However, restoration of the original "raw" values from the calibrated CIS values using Warr and Rice's rearranged regression equation [2] yields restored "raw" $KI_{Heidelberg}$ values, for instance 0.86 and 0.62 for the 10-Å peak widths of CIS standard SW1 and SW2 (given CIS values = 0.63 and 0.43, respectively). These values are implausibly broad, even allowing for the fact that Warr and Rice's peak widths for the polished-slate standards are much broader than those obtained by Kisch or by Kübler.

The data base of the VLGM web-site contains the FWHM values of the CIS standards measured by Kisch in 1993 using the same instrumental conditions at which Kübler-equivalent boundary values (0.21 and 0.38°Δ2θ) were determined (Kisch, 1980, 1990). Comparing these data with the "raw" (as measured) FWHM values of the CIS standards measured by Warr in Heidelberg (also given in the VLGM web-site), the following linear regression equations are obtained:

$$KI_{<Warr,\ Heidelberg,\ Siemens>} = 1.12 \times KI_{<Kisch,\ Beer\text{-}Sheva>} - 0.0034\ (r=0.973) \quad [4a]$$

from the FWHM data pairs of four SW slate chip standards, and

$$KI_{<Warr,\ Heidelberg,\ Siemens>} = 1.17 \times KI_{<Kisch,\ Beer\text{-}Sheva>} - 0.0207\ (r = 0.986) \quad [4b],$$

when the data obtained on slate chip standards and a mica flake SW7/MF1 is also included. When data pairs of four SW and four ILC slate chip standards are correlated, the following equation is obtained:

$$KI_{<Warr,\ Heidelberg,\ Siemens>} = 1.164 \times KI_{<Kisch,\ Beer\text{-}Sheva>} - 0.00749\ (r = 0.976). \quad [5]$$

Anchizone boundaries

Substituting the Kübler-equivalent boundary values of the anchizone determined by Kisch (i.e., 0.21 and 0.38°Δ2θ, respectively) in the equations [4a], [4b] and [5], boundary values of 0.232 and 0.422°Δ2θ [equation 4a], 0.225 and 0.424°Δ2θ [equation 4b], and 0.237 and 0.435°Δ2θ [equation 5] are obtained on the "raw" (as measured) scale of Warr given in the VLGM web-site (Table 1). In turn, relating these "raw" FWHM data of Warr with the CIS data (given also in the VLGM web-site), the regression equations are as follows when four SW slate chip standards and one muscovite flake (MF1) are used:

$$KI_{<CIS>} = 1.039613 \times KI_{<Warr,\ Heidelberg>} + 0.051958\ (r = 0.997) \quad [6]$$

and

$$KI_{<CIS>} = 1.047328 \times KI_{<Warr,\ Heidelberg>} + 0.046334\ (r = 0.997) \quad [7]$$

when, in addition to the SW and MF1 standards, the four ILC slate chip standards (ILC-1, 2, 3 and 4) are also included. Consequently, the high- and low-temperature boundaries of the Kübler-equivalent anchizone will be 0.286 and 0.493°Δ2θ (combining equations [4b] and [6]) and 0.295 and 0.500°Δ2θ (combining equations [5] and [7]). These boundary values are considerably (by 0.04–

0.08°Δ2θ) greater than those (0.25 and 0.42°Δ2θ) originally established by Warr and Rice (1993, 1994) on the CIS scale.

Krumm et al. (1994) demonstrated that the working definitions of the anchizone used by various laboratories are not in every case equivalent. For example, the definitions used in the laboratories of Kisch (Beer-Sheva) and Frey (Basel) proved to be equivalent (0.21–0.38 and 0.25–0.42°Δ2θ, respectively). By contrast, considerable differences were found between the definitions of anchizone used by Krumm (Erlangen) and the CIS scale proposed by Warr in Heidelberg. Using the FWHM values obtained by Kisch on the CIS and ILC slate chip standards for regression analysis, the 0.25 and 0.42°Δ2θ boundary values on the CIS scale of Warr and Rice (1993, 1994), claimed to be equivalent with Kübler's original boundary values, are ca. 0.28 and 0.50°Δ2θ in the Kübler- or Kisch-equivalent scale (Table 1). Thus, both the equations [4] to [7] of the present paper and the results of Krumm et al. (1994) unequivocally demonstrate that the anchizone boundary values of 0.25 and 0.42°Δ2θ on the CIS scale suggested by Warr and Rice (1993, 1994) are not equivalent to those of the original Kübler's definition of the anchizone. Consequently, the use of the CIS scale boundaries results in increasing confusion when CIS-calibrated KI values are used for petrogenetic purposes in regional comparisons.

Although the CIS (SW and ILC) standards were measured in many laboratories and most of their "raw" (as measured) values are available on the VLGM web-site, only few papers have been published which inform the readers on the relation between the anchizone boundaries as determined by the CIS standardization procedure suggested by Warr and Rice (1993, 1994) and by the polished slate slab standards of Kübler and/or Kisch. Comparing the KI data calibrated to Kübler's and the CIS scales, Brime (1999) showed that there are considerable differences in the boundary values of the anchizone for the two scales. If one adopted the CIS scale to calibrate the KI values, limiting values of 0.33 and 0.59°Δ2θ should be used for the high- and low-temperature boundaries of the anchizone *sensu* Kübler, on the basis of the linear regression equation rearranged from Fig. 1 of Brime (1999, p. 62):

$$KI_{<Brime,\ Kübler\text{-}equiv.>} = 0.664 \times KI_{<Brime,\ CIS\text{-}equiv.>} - 0.031\ (r = 1.000). \quad [8a]$$

Brime et al. (2001) presented two KI data sets standardized by Kisch's polished slate slab series on one hand and by the CIS slate chip standards of Warr and Rice, on the other. Comparing their two sets by linear regression analysis, the following equation is obtained:

$$KI_{<Brime,\ Kübler\text{-}equiv.>} = 0.652 \times KI_{<Brime,\ CIS\text{-}equiv.>} + 0.035\ (r = 1.000). \quad [8b]$$

Leoni (2001) standardized his KI values by using the polished slate slab series of Kübler (Nos. 32, 34 and 35) and by the CIS standards. Using Leoni's (2001) equations (1) and (2), the following relation is found between the Kübler's and CIS scales:

$$KI_{<Leoni,\ Kübler\text{-}equiv.>} = 0.991 \times KI_{<Leoni,\ CIS\text{-}equiv.>} - 0.036\ (r = 1.000) \quad [9]$$

Árkai (1991), Árkai et al. (1995, 1996, 2000) calibrated the "raw" FWHM values also using the polished slate slab series Nos. 32, 34 and 35 provided by Bernard Kübler. In 1998 Árkai (unpublished results, partly submitted to the VLGM web-site) measured simultaneously the polished slate slab series and mica flake standard of Kisch, the CIS slate chip standards SW1-6 and the mica flake MF1 of Warr and Rice (1993, 1994), the ILC1-6 rock chip standards of Krumm, and his own polished slate slabs Nos A-1–3 (the latter allowing a comparison with Kübler's slabs Nos. 32, 34 and 35). Instrumental drifts with time in the measured FWHM values have been taken into consideration by Árkai. Such possible technical causes may be: change of diffractometer or some of its main units, change or aging of the X-ray tube, small-scale shifts in geometric conditions of the goniometer, etc. The following regression equation was valid that time:

$$KI_{<Árkai,\ Kübler\text{-}equiv.>} = 1.00126 \times KI_{<Árkai,\ measured>} + 0.02853 \quad [10].$$

Using this equation, the actual boundary values of the anchizone which correspond to Kübler's 0.25 and 0.42°Δ2θ values were 0.221°Δ2θ and 0.391°Δ2θ on the scale of the "raw" (as measured) data in Budapest in 1998.

Having measured Kisch's polished slate slab and muscovite flake standards at the same time, the relation

$$KI_{<Kisch>} = 0.845 \times KI_{<Árkai,\ measured>} + 0.02307\ r = 0.999) \quad [11]$$

was obtained. According to this relation, the 0.21 and 0.38°Δ2θ boundary values of the Kübler-equivalent anchizone of Kisch correspond to 0.221 and 0.422°Δ2θ on Árkai's measured data scale, providing excellent agreement at the high-

temperature boundary, while differing by ca. 0.03–0.05°Δ2θ at the low-temperature boundary of the anchizone, Kisch's calibration giving a wider range.

Simultaneously measuring the slate chip standards in Budapest, the following equations were obtained when only the FWHM values of the 10-Å basal reflections of the SW and MF1 standards of Warr and Rice (1993, 1994) were used:

$$KI_{<Árkai, CIS\text{-}equiv.>} = 1.17084 \times KI_{<Árkai, measured>} + 0.02424 \; (r = 0.987, n = 5) \quad [12]$$

and

$$KI_{<Árkai, CIS\text{-}equiv.>} = 1.16365 \times KI_{<Árkai, measured>} + 0.01364 \; (r = 0.975, n = 10) \quad [13]$$

when the 10 Å reflections of the ILC slate chip standards were also included. The KI values that correspond to the 0.25 and 0.42°Δ2θ values of the CIS scale (supposed to be equivalent to the boundary values of Kübler) range between 0.193–0.203 and 0.338–0.349°Δ2θ on the scale of "raw" (as measured) data of Árkai in 1998. These values, especially considering the anchizone/diagenesis boundary, are significantly smaller than those obtained by calibrations via the polished slate slab standards of Kübler and Kisch.

Figure 1 illustrates the linear regressions determined between the Kübler-equivalent KI scale and the CIS scale in the various laboratories discussed previously. In this figure, the Kübler-equivalent KI scale (horizontal axis) represents the data obtained from "raw" measured values by calibration using the polished slate slab standards of Kübler or Kisch, while the CIS scale of KI values correspond to that published by Warr and Rice (1993, 1994) and also given in the VLGM web-site. It is worth mentioning that even the regression equation of Warr's (Heidelberg) results, submitted to the VLGM web-site, differs strongly from the "ideal" 1:1 relation between Kübler's and CIS scales (the regression of Warr's data was calculated by combining equations [5] and [7]). For constructing regression lines of Brime et al. (2001) and Leoni (2001), equations Nos. [8] and [9] were used, respectively. The regression line of Kisch's data was established by relating the SW and ILC values measured in Beer-Sheva, Israel given in the VLGM web-site with the corresponding CIS values, on the basis of the equation

$$KI_{<Kisch, CIS\text{-}equiv.>} = 1.25287 \times KI_{<Kisch, Beer\text{-}Sheva>} + 0.02443 \; (r = 0.989, n = 10) \quad [14],$$

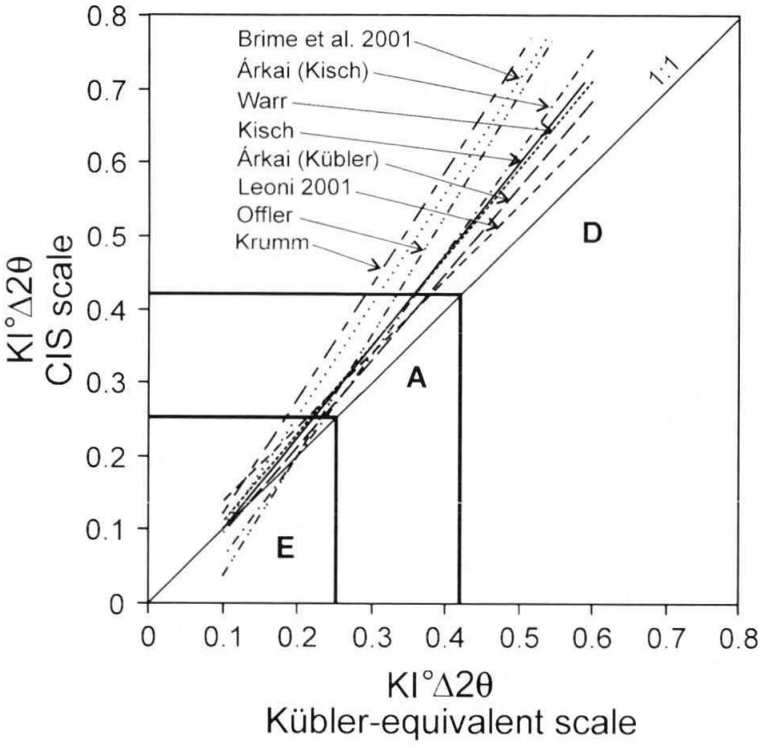

Fig. 1 Linear regressions between the Kübler-equivalent KI (illite "crystallinity") values of various laboratories and the CIS data of Warr and Rice (1994). Abbreviations: E—epizone, A—anchizone, D—diagenetic zone. Detailed explanation in the text.

and adding 0.04°Δ2θ values to the $KI_{<Kisch, Beer Sheva>}$ data in order to obtain the Kübler-equivalent values.

A further example is obtained by using Robin Offler's (Newcastle, Australia) measurements carried out on H.J. Kisch's polished slate slab series (Offler, pers. comm. to Kisch) and on Warr's rock chip standards and mica flake No. MF1 (VLGM web-site and Table 2 of the present paper):

$$KI_{<Offler, Kisch\text{-}equiv.>} = 1.08932 \times KI_{<Offler, measured>} - 0.01318 \ (r = 0.971) \quad [15a].$$

From equation [15a] the $KI_{<Offler, Kübler\text{-}equiv.>}$ values can be obtained by adding 0.04°Δ2θ values to the $KI_{<Offler, Kisch\text{-}equiv.>}$ data. On the other hand, using Offler's KI data obtained from Warr and Rice's standard rock chips, the following equations can be set up:

$$KI_{<Offler, CIS\text{-}equiv.>} = 1.66534 \times KI_{<Offler, Kübler\text{-}equiv.>} - 0.13788 \ (r = 1.000, n = 4: SW1, 2, 3 and 4, without muscovite flake MF1) \quad [15b]$$

and

$$KI_{<Offler, CIS\text{-}equiv.>} = 1.75109 \times KI_{<Offler, Kübler\text{-}equiv.>} - 0.17063 \ (r = 0.999, n = 5: SW1, 2, 4 and 6, and muscovite flake MF1) \quad [16].$$

The $KI_{<Offler, Kübler\text{-}equivalent>}$ values that correspond to the 0.25 and 0.42°Δ2θ values of the CIS scale (supposed to be equivalent with the boundary values of Kübler) are 0.233 and 0.335°Δ2θ, on the basis of equation [15b], and correspond to 0.240 and 0.337°Δ2θ, using equation [16].

Calculating the $KI_{<Kübler\text{-}equiv.>}$ data of Stefan Krumm (Erlangen Germany) from the $KI_{<Kisch\text{-}equiv.>}$ results (Table 2) in a similar way, and combining them with the corresponding CIS values, the following equation is obtained:

$$KI_{<CIS>} = 1.55642 \times KI_{<Krumm, Kübler\text{-}equiv.>} - 0.03374 \ (r = 0.987, n = 9: SW1, 2, 4 and 6, and ILC1-5) \quad [17].$$

Using this equation, 0.182 and 0.292°Δ2θ values on the Kübler-equivalent scale would correspond to the so called CIS boundaries of the anchizone.

Árkai's data are presented in Fig. 1 using the calibrations via Kübler's polished slate slab standards (equation [10]) plotted against the CIS values and via Kisch's standards (equation [11]), also plotted against the CIS values.

The fact that all of the regression lines are lying above the 1:1 line of Fig. 1 suggests that a significant discrepancy exists between Kübler's original anchizone boundaries and those suggested as equivalent CIS boundary values by Warr and Rice (1993, 1994).

Table 2 FWHM values of the 10-Å basal reflections of illite-muscovite on Warr and Rice's (1993, 1994) CIS and Krumm's ILC standards, as measured in various laboratories, and Kisch equivalents calculated using regressions given (values in °Δ2θ, CuK_α).

sample	CIS	Warr	Kisch	Offler		Krumm		Brime		Árkai	
Nr.		as measured*	as measured	as measured	Kisch-equiv.	as measured	Kisch-equiv.	as measured	Kisch-equiv.	as measured	Kisch-equiv.
SW1	0.630	0.57	0.480	0.40	0.422	0.448	0.394	0.3828	0.372	0.534	0.474
SW2	0.470	0.38	0.385	0.31	0.324	0.323	0.293	0.2677	0.275	0.333	0.304
SW4	0.380	0.31	0.269	0.26	0.270	0.247	0.232	0.2219	0.236	0.316	0.290
SW6	0.250	0.20	0.170	0.19	0.194	0.154	0.156	0.1387	0.166	0.206	0.197
MF1	0.110	0.06	0.079	0.13	0.128					0.079	0.090
ILC1	0.424	0.36	0.300			0.260	0.242			0.364	0.331
ILC2	0.282	0.23	0.204			0.150	0.153			0.229	0.217
ILC3	0.533	0.46	0.380			0.330	0.299			0.411	0.370
ILC4	0.293	0.24	0.226			0.175	0.173			0.274	0.255
ILC5	0.453		0.365			0.309	0.282			0.411	0.370
ILC6		0.36	0.921			0.348	0.313			0.659	0.580

Regressions calculated using the rock slab series and muscovite flake distributed by H.J. Kisch:

$KI_{Kisch\text{-}equivalent} = 1.0888 \times KI_{Offler, measured} - 0.01318$ (r = 0.971). Measurements on 10 slabs and one muscovite flake (Offler's data of 2/92 and 7/92; Kisch's data up to 5/95)

$KI_{Kisch\text{-}equivalent} = 0.8081 \times KI_{Krumm, measured} + 0.0320$ (r = 0.993). Measurements on 9 slabs and one muscovite flake (Krumm's data of 4/94)

$KI_{Kisch\text{-}equivalent} = 0.844 \times KI_{Brime, measured} + 0.0488$ (r = 0.992). Measurements on 8 slabs (Kisch's data up to10/94; Brime's data of 11/01)

$KI_{Kisch\text{-}equivalent} = 0.845 \times KI_{Árkai, measured} + 0.02307$ (r = 0.999). Measurements on 6 slabs and one muscovite flake (Árkai's data of 5/98; Kisch's data up to 9/98)

* as given by Warr in the CIS page of the VLGM (very low-grade metamorphism) web-site in Heidelberg

Possible explanations for the broad CIS values reported by Warr and Rice

There are two possible explanations for the broad values obtained by Warr and Rice on their CIS standards, and consequently, for the discrepancy demonstrated in Fig. 1.

(1) Error in the calibration procedures followed by Warr and Rice (1993, 1994), that is conversion of the "raw" FWHM into Kübler equivalents using a conversion algorithm differing from the regression based on the values measured on Kisch's slab standards.

(2) Preparation procedures producing a size fraction much finer than <2 μm, and consequently resulting in measured KI peak widths much broader than measured on the <2 μm size fractions separated by most other laboratories.

The method used by Warr and Rice (1993, 1994) for obtaining the uncalibrated ("raw") FWHM data, which strongly differ from those used originally by Kübler, Kisch, and later on, by many other laboratories, may explain the unusually broad CIS values, but cannot explain the considerable discrepancies in the boundary values of the anchizone found between those of Warr and Rice (1993, 1994) on one hand and the majority of the other laboratories, on the other.

The present authors tend to favour the first interpretation. If this interpretation is correct, and the CIS scale is not truly Kübler equivalent, then neither are the limits of the anchizone in the CIS scale equivalent to Kübler's 0.42°Δ2θ and 0.25°Δ2θ: they must be at appreciably broader values. However, in order to ascertain the cause of the divergency, it is imperative to know Warr and Rice's "raw", as measured, half-height peak-width values and, preferably, the equation or equations actually used in the calibration. In the absence of such information it is impossible to make a well-considered, unequivocal choice between the above options.

Recommendations: data to be included in studies of KI (illite "crystallinity") calibrated with inter-laboratory slate-chip standards

In order to avoid such doubts about the significance of "calibrated" FWHM values of centrally distributed KI standards, be they Warr and Rice's or other in the future, researchers using such standards should always report the "raw", as measured values on the <2 μm fractions separated by them from such standards. In case they give "calibrated" values on their material, i.e. Kübler- or Kisch-equivalent FWHM values based on the relationship of the peak widths measured on polished-slate standards, they should also give the calibration regressions used. The procedures recommended by us therefore are as follows:

(1) All laboratories using the Warr and Rice standards or any other chip/powder standards should establish their own calibration curve using the polished slate slab standards against Kübler or Kisch. These calibration curves and/or the resulting regression equations should be given in their papers, as well as the low-grade and high-grade limits of the anchizone corresponding to 0.42/0.25°Δ2θ of Kübler and the equivalent 0.38/0.21°Δ2θ of Kisch; preferably, they should also give the FWHM value of the narrowest 10-Å peak as measured on a well-crystallized muscovite flake, preferably a muscovite flake or strip that has not ground.

(2) Laboratories should give the "raw" as measured half-height peak widths as measured in their laboratories as well as the "recalculated" half-height peak widths as obtained using their own calibration curve established in step (1). (In case of page limitations frequently applied by various journals, authors are encouraged to put their "raw" data sets on their own web-sites, thus providing easy access for the readers interested.

(3) Researchers should indicate how the "raw" KI values have been measured or obtained on the diffractograms in order to facilitate assessment of the artifacts that may have been introduced.

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