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The effects of tectonic strain on crystallinity, apparent mean crystallite size and lattice strain of phyllosilicates in low-temperature metamorphic rocks. A case study from the Glarus overthrust, Switzerland

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Abstract

The relationship between tectonic shearing strain and crystal structural features of illite-muscovite and chlorite has been investigated in a series of metasedimentary rocks (slates, mylonitized conglomerate and calc-mylonite) from two profiles traversing the classic Glarus overthrust, Swiss Helvetic Alps. In addition to the conventional X-ray diffractometric crystallinity measurements of illite-muscovite (IC) and chlorite (ChC), the apparent mean crystallite size and lattice strain were also calculated by the Voigt method, using single-line Fourier analysis.

Small-scale variations in IC and ChC occur as a function of the distance from the Glarus overthrust. In contrast, the apparent mean crystallite size of illite-muscovite decreases while the lattice strain increases towards the overthrust in the Permian Verrucano within an interval of ca. 100–130 m. The reduction in crystallite size caused by increased shearing strain was not followed by recrystallization of illite-muscovite. Opposite trends for chlorite may be explained by syn- and post-overthrusting crystallization. Results from illite-muscovite and chlorite in the calc-mylonite are similar to those of the underlying Tertiary Flysch, in which no systematic changes were found as a function of the distance from the overthrust plane.

Correlation between the crystallinity indices and the apparent mean crystallite size are stronger than those between the crystallinity indices and lattice strain. As crystallite size increases, lattice strain – with large fluctuations – tends to decrease with enhancing crystallinity. In general, lattice strain decreases with increasing crystallite size, although there are considerable differences between the various sample groups.

Recrystallization of illite-muscovite caused by the overthrusting could be observed only within a several dm-wide zone above and below the gliding plane, as indicated by K–Ar dates of the < 2 μm grain-size fraction of illite-muscovite.

Keywords: illite crystallinity, chlorite crystallinity, crystallite size, lattice strain, tectonic strain, Glarus overthrust, Switzerland.

Introduction

There are a number of empirical parameters which are employed to express the continuous structural and chemical changes from illite to muscovite during incipient metamorphism of fine-grained metaclastic rocks. These are, for example, the Kübler index (KÜBLER, 1967a, 1968) or illite crystallinity (IC), the domain size calculated from the IC values by the Scherrer equation (WEBER et al., 1976) and the Flehmig index (FLEHMIG, 1973), the latter of which is based on infrared absorption spectroscopic measurements. Of these indices, the Kübler index (IC) has been used most frequently for metamorphic petrogenetic purposes in the last three decades. On the basis of field observations and comparison with vitrinite reflectance and fluid inclusion data, temperature has been generally regarded as the most important factor influencing IC, although very few laboratory experiments have been carried out to support this assumption (SMYKATZ-KLOSS and ALTHAUS,

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1974; KRUMM, 1984). The additional effects of various lithological, chemical and physical factors have been reviewed by FREY (1987).

Relatively few and rather controversial data exist in the literature on the possible effects of tectonic strain on IC, although this relationship has been investigated on varying scales, ranging from regional, through outcrop, to hand specimen scales. KÜBLER (1967b) did not find any correlation between the first appearance of schistosity and IC. However, reviewing a large number of incipient metamorphic terraines, KISCH (1989) found that slaty cleavage appears mostly within the anchizone, but the corresponding IC values were strongly scattered. This scatter was explained by the "preservation of pre-existing illite 'crystallinities' during subsequent deformation, and the varying time relations of deformation and metamorphism" (KISCH, 1989), in addition to differences in lithology. KÜBLER (1967a), FREY et al. (1973), ALDAHAN and MORAD (1986) and FRANCESCHELLI et al. (1994) described enhanced illite crystallinity in tectonic shear zones. GUTIÉRREZ-ALONSO and NIETO (1996) demonstrated regional-scale inverse correlation between finite strain and white mica crystallinity indices. FLEHMIG and LANGHEINRICH (1974) and NYK (1985) found increasing Flehmig indices (increasing crystallinity) from the tails towards the hinge of outcrop-scale folds. Surprisingly, Kübler indices did not correlate with the increasing tectonic strain in the latter two cases. GRUNER (1976, cited in FREY, 1987) and TEICHMÜLLER et al. (1979) did not find a correlation between deformation and IC in single folds.

According to TEICHMÜLLER et al. (1979), cleavage-forming processes increase lattice distortion and may lower the activation temperature of recrystallization, thus producing non-linear increase of crystallinity within a relatively narrow temperature range (syn-kinematic recrystallization). OFFLER and PRENDERGAST (1985) provided a possible explanation for the enhanced crystallinity in cleavage planes as compared to the bulk rock. In slates and phyllites the phyllosilicate-rich (P) domains are characterized by higher permeability than the quartz-rich (Q) domains, thus enabling substantial changes in chemistry of P domains and their white micas (STEPHENS et al., 1979; WHITE and JOHNSTON, 1981). In this way, illite-muscovite with enhanced crystallinity may form in the P domains due to the increasing removal of interlayer H_2O, and incorporation of K^+ ions (see also AWAN and WOODCOCK, 1991).

Based on a symmetrical pattern of IC values measured across a tight anticline (Cwm Pennant anticline, Northern Wales), ROBERTS and MERRIMAN (1985) presented a direct, regional-scale relationship between IC and tectonic strain, where kinetic effects attending cleavage formation were considered as the main factors enhancing illite crystallinity. In contrast, ROBINSON and BEVINS (1986) did not find a general correlation between the intensity of cleavage formation and IC for the same area. Re-investigating the Cwm Pennant anticline, ROBINSON et al. (1990) did not reproduce the symmetrical "isocryst" pattern given by ROBERTS and MERRIMAN (1985), thus questioning the concept of strain-induced metamorphism. According to MERRIMAN et al. (1990) and ROBERTS et al. (1991), an increase of white mica crystallite size is accelerated by cleavage formation, as cleavage contributes to the simultaneous dissolution and growth of larger mica crystals at the expense of the smaller ones. MERRIMAN et al. (1995) demonstrated differences in strain-induced crystal growth of illite-muscovite and chlorite by integrated TEM and XRD observations. In low-T metamorphic environment, the crystallites of white mica grew more rapidly than those of chlorite, because of their better ability to store strain energy and to recover from subgrain structure. In chlorite, subgrain boundaries developed at high strain rates were only partially recovered at low strain rates.

It is obvious from the review above, that our present knowledge about the possible effect of tectonic strain on the structural and chemical properties of illite-muscovite and other phyllosilicates is incomplete and rather controversial. It seems likely that numerous factors may strongly modify and even eliminate the action of shearing strain on phyllosilicate properties. Such factors may include:

- the modal ratio and the physical and chemical differences between the phyllosilicate in question and the matrix-forming (embedding) portion of the rock during the deformation (can stress be inactivated by the deformation/recrystallization of the matrix or is the phyllosilicate the main phase responding to the strain?);
- the rates of strain;
- the temperature of the rock mass during deformation;
- the fluid/rock ratio and its variations in the deforming medium;
- the time relation of deformation and (re)crystallization, etc.

To determine the nature of relationships between tectonic strain and illite crystallinity, a large series of detailed studies is required, in which most of the above-listed factors are well constrained. The present paper provides results of such a case study, which was carried out along two profiles crossing the classic Glarus overthrust, Helvetic Alps, Switzerland. Illite and chlorite crystallinity, apparent mean crystallite size and lattice strain data are presented as a function of the distance from the overthrust plane, in order to evaluate the possible effect of quasi-horizontal shearing strain that acted during large-scale nappe transport.

Conditions of metamorphism and thrusting

The samples for the present study were collected from two localities indicated by A and B in figure 1. The sample series from locality A represents the critical part of the profile investigated by FREY (1988), while the samples at locality B were taken from the immediate vicinity of the thrust plane (Tab. 1). These profiles cross the Glarus overthrust along which the Helvetic Nappes, consisting here mainly of Permian Verrucano slates, were thrust over the Infrahelvetic Complex. The latter is built up mainly of carbonate-bearing slates forming the Upper Eocene to Lower Oligocene parautochthonous North Helvetic Flysch and the Middle to Upper Eocene allochthonous South-Helvetic Units. The overthrust plane itself is marked by a thin (ca. 1 m) calc-mylonite (Lochseite limestone of probable Mesozoic age). Using statistically evaluated IC and fluid inclusion data, FREY (1988) demonstrated that the overlying Permian Verrucano suffered epizonal conditions (ca. 300–350 °C), and the Tertiary Flysch medium-grade anchizonal (ca. 250–300 °C) regional metamorphism. The latter proved to be of medium

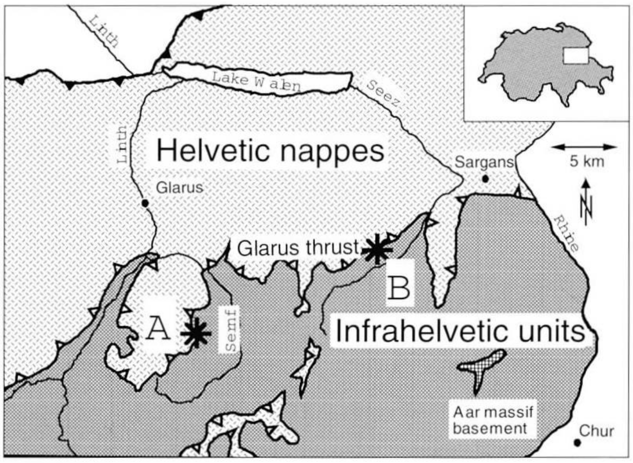

Fig. 1 Geological map of the region between the Linth and Sernf valleys, Glarus Alps, eastern Switzerland (after TRÜMPY, 1967, modified by FREY, 1988). The asterisks denote the localities of the investigated profiles A and B.

pressure type, as evidenced by the distribution of K-white mica *b* lattice parameters.

The main penetrative ductile deformation, including development of a slaty cleavage, folding and thrusting occurred during the Calanda phase (TRÜMPY, 1969; SCHMID, 1975; MILNES and PFIFFNER, 1977; PFIFFNER, 1986). This was followed by the Ruchi phase, producing less ductile, complex heterogeneous deformation during the continued nappe movement along the Glarus overthrust, affecting the pre-existing structures in a 200–300 m wide zone above and below the thrust plane. The main metamorphism was related to the Calanda phase, reaching its temperature climax shortly after this deformation (GROSHONG et al., 1984; PFIFFNER, 1986).

FREY (1988) summarized the radiometric dates obtained on the 0.1–2 µm grain-size fraction of illite-rich samples from the vicinity of the Glarus overthrust, presented by HUNZIKER et al. (1986), HUNZIKER (1987) and J.C. HUNZIKER (1988, unpublished data). 30.5 ± 1.9 Ma Rb–Sr and 24.2 ± 1.3 Ma K–Ar dates were calculated for an epizonal Verrucano sample from 300 m above the Glarus overthrust. According to the authors mentioned above, the ca. 30 Ma may date the main (Calanda) phase of metamorphism, or may be a partially resetted age value, while the values of 23–25 Ma indicate that the translation along the Glarus overthrust lasted until at least the fourth (Ruchi) phase. The discontinuous inverse metamorphic zonation evidenced by FREY (1988) is the result of post-metamorphic thrusting along the Glarus overthrust, providing an example of "transported metamorphism". RAHN et al. (1995) estimated a post-metamorphic displacement of approximately 10 km.

BURKHARD et al. (1992) explained the stable and Sr isotopic compositions of the calc-mylonite by exchange with ^{18}O-depleted and ^{87}Sr-enriched fluids at very high (3 to > 10^3) water/rock ratio, implying fluid advection to the north. These fluids might derive from the underlying flysch by metamorphic dewatering and/or might represent metamorphic fluids or formation brines expelled along the overthrust plane from greater depth (BURKHARD et al., 1992). In mylonitized slates of the overlying Verrucano, concordant oxygen isotopic temperatures of 300–350 °C and 250–300 °C were obtained for assemblages of quartz-chlorite-calcite and quartz-albite-calcite, respectively. These calculated T ranges are in agreement with the earlier estimates of FREY (1988). The calc-mylonite was (re)crystallized at 355 ± 30 °C (calcite-dolomite thermometric data of BURKHARD and KERRICH, 1988). Microstructural features indicate an alternation of ductile and brittle deformations

Tab. 1 List of samples investigated.

sample	elevation (m) a.s.l.*	distance from the overthrust (m)*	tectonic unit, formation	rock type
locality A				
MF - 994	3000	+170	HN/PV	slate
MF - 995	2280	+150	HN/PV	slate
MF - 996	2260	+130	HN/PV	slate
MF - 997	2240	+110	HN/PV	slate
MF - 998	2220	+90	HN/PV	slate
MF - 999	2190	+60	HN/PV	slate
MF - 1000	2170	+40	HN/PV	slate
MF - 1001	2130	0	HN/PV	slate
MF - 1002	2130	0	SHU/TF	slate
MF - 1003	2110	–20	SHU/TF	slate
MF - 1004	2090	–40	SHU/TF	slate
MF - 1005	2070	–60	SHU/TF	slate
MF - 1006	2050	–80	SHU/TF	slate
MF - 1007	2030	–100	SHU/TF	slate
MF - 1008	2010	–120	SHU/TF	slate
MF - 1009	1990	–140	SHU/TF	slate
locality B				
MF - 1995	1210	+10.00	HN/PV	mylonitized, fine-grained metaconglomerate
MF - 1996	1200	+ 0.40	HN/PV	mylonitized, fine-grained metaconglomerate
MF - 1997	1200	–0.05–0.12	CM/GO	calc-mylonite
MF - 1998	1200	–0.50–0.60	SHU/TF	slate

* approximate values for samples MF-994 -1009, and precise values with respect to the overthrust plane for samples MF-1995–1998.

Abbreviations: HN/PV – Helvetic Nappes (Permian Verrucano) above the Glarus overthrust; CM/GO – calc-mylonite, Glarus overthrust; SHU/TF – South-Helvetic Units (Tertiary Flysch).

caused by local and short periods of escape of overpressured fluids into the overlying Verrucano and/or along the thrust plane (BURKHARD et al., 1992). Fluid accumulation led to fluid pressure close to lithostatic pressure in the thrust plane below the less permeable Verrucano nappe, and was responsible for the extreme localization of strain in the calc-mylonite. This enabled large-scale thrusting of lithologically similar nappes over one another without penetrative, thrust-related internal deformation (BURKHARD et al., 1992).

Methods

Illite crystallinity [IC, i.e., the full-width-at-half-maximum (FWHM) of the first, 10 Å basal reflection of illite-muscovite] and chlorite crystallinity indices [i.e., the FWHM values of the first (14 Å) and second (7 Å) basal reflections of chlorite indicated as ChC(001) and ChC(002)] were determined by X-ray diffractometric (XRD) methods. The procedure of preparing < 2 μm grain-size fraction samples was similar to that used by KÜBLER (1968), as described by FREY (1988). The XRD instrumental and measuring conditions in Budapest were: Philips PW-1730 diffractometer with computerized APD system, CuKα radiation, 45 kV / 35 mA, proportional counter, graphite monochromator, divergency and detector slits of 1°, and collection of data with 0.01° 2Θ steps, using measuring time intervals of 1 s. At these conditions the standard deviation was $s = 0.008°\ \Delta 2\Theta$ for a FWHM of $0.215°\ \Delta 2\Theta$ ($n = 10$). Thus, in the case of epizonal samples, the relative standard deviation of the FWHM values is 3.7%. The calibration of IC and ChC values against KÜBLER's IC scale, where the anchizone ranges between 0.25 and 0.42° Δ2Θ, was made using standard rock slab series (Nos 32, 34 and 35) kindly provided by B. KÜBLER. Smaller scale, temporary instrumental changes were corrected by the repeated use of an other, also calibrated standard rock slab series (Nos Á-1, –2 and –3) of the Laboratory for Geochemical Research, Budapest. Applying the least squares' method, the calibration equation is:

IC(KÜBLER) = 1.164 · IC(present work) – 0.038.

The actual boundary ranges of IC, ChC(001) and ChC(002) of the present paper, which correspond to KÜBLER's original anchizone, are 0.247–0.393°, 0.261–0.368° and 0.242–0.296° $\Delta 2\Theta$, respectively (see also ÁRKAI et al., 1995b). All of these boundary values refer to air-dried (AD) mounts.

The apparent mean crystallite size and lattice strain values of illite-muscovite and chlorite were calculated from line-profiles of XRD basal reflections. According to KLUG and ALEXANDER (1974), the line-broadening of an XRD reflection is the resultant of three main factors, namely: instrumental effects, crystallite size (mean size of the domains that coherently scatter X-rays, measured perpendicular to the diffracting planes) and the various kinds of lattice imperfections (local heterogeneity of composition and structure, including also mixed-layering) summarized as lattice strain (also defined relative to the given crystallographic direction).

There are several methods [i.e., single-line methods, such as variance and Voigt method, and multi-line methods, such as the Warren-Averbach (W–A) method] for calculating the effects of crystallite thickness and lattice strain by analyzing XRD line-profiles. In these methods the line-broadening caused by instrumental effects is subtracted by comparison with data for synthetic or natural "standards" with no inherent broadening, i.e., crystal size is relatively large. Finite crystallite size and lattice strain each result in unique line-broadening functions which may be approximated by specific mathematical functions, each with characteristic effects on reflections of different order from the same crystallographic plane, hkl.

Although most of these methods were elaborated and used originally for cold-worked metals with relatively simple (mostly cubic) structures, these methods have also been applied to phyllosilicates (for an extensive review see ÁRKAI et al., 1996), in spite of the fact that the justification of such extended applications has not been checked systematically so far. EBERL and SRODON (1988) and EBERL et al. (1990) used the W-A method (WARREN and AVERBACH, 1950) to determine the mean values and also the distributions of crystallite thicknesses for illite and other phyllosilicates. The assumptions for the latter applications were that XRD line-broadening is caused by crystallite size related to the thicknesses of diffracting units, the broadening is not influenced by stacking faults within the crystallites, and that the interiors of the crystallites are strain-free. Line-broadening by lattice strain for basal reflections of illite was entirely attributed to inter-crystallite swelling which was kept constant at 17 Å by saturation and ethylene glycol-solvation (see also EBERL and BLUM, 1993). As swelling mixed-layers are subordinate or absent and other factors such as stacking faults may considerably affect the strain, these preconditions are not valid for metamorphic K-white micas (LANSON and KÜBLER, 1994). Consequently, the W-A method as suggested by EBERL and SRODON (1988) may not be applicable to metamorphic phyllosilicates. Thus the application of the W-A technique to metamorphic rocks by WARR and RICE (1994) gave different crystallite thickness values from various reflection-pairs, and subsequently WARR (1996) recommended use of the single-line Fourier transform method rather than the multi-line W-A method.

The imperfectness of natural "standards" causes explicitely stronger disturbing effects in case of the multi-line W-A method than in the case of single-line methods. When studying multiphase samples, the intensities of the higher order reflections are relatively weak. The effects of overlapping or interfering reflections of the various phases, especially being frequent at higher order reflections, can be overcome by deconvolution, using certain mathematical functions, thus producing "artificial" intensity distributions. These non-ideal conditions, which are common in rocks, may make the results of the W-A analysis rather arbitrary.

Therefore, the apparent mean crystallite sizes and lattice strains of illite-muscovite and chlorite were calculated from line-profiles of the first order (10 and 14 Å) XRD basal reflections, based on the Voigt method of LANGFORD (1978), as modified by the Philips APD-1700 software package. The so called structural profiles of the basal reflections were calculated using STOKES' (1948) deconvolution method based on Fourier coefficients, which utilizes Fourier transformations of the line-profiles obtained from samples being invesitigated and from the "standard" samples. Final calculations were performed on the profiles as corrected for standard-determined instrumental effects, and therefore theoretically free of such effects. As compared to the original application of the Voigt match (LANGFORD, 1978), this procedure called also single-line Fourier analysis, has the advantage that no errors in integral breadths caused by the finite crystallite sizes and lattice strains can arise from the deconvolution itself, since the Stokes' deconvolution uses no assumptions on the shape of the profiles of the sample and the "standard", whereas LANGFORD's (1978) method assumes that both profiles can be described by Voigt functions. The line-broadening caused by small crystallites is described by a Cauchy (or Lorentzian) type function, whereas that caused by lattice strain is described by a Gaussian function. The integral breadth values resulting from these two components can be obtained from a parabolic fit to the natural logarithms of the Fourier coefficients of the structural profile. The linear coefficient is proportional to the Cauchy function, whereas the parabolic coefficient is related to the Gaussian contribution to line broadening. Having obtained these two integral breadth values, one can use the appropriate Scherrer equation for calculating crystallite size and strain (for further details see the manual of the Philips APD-1700 software package). Lattice strain (here given in %) is the measure of displacement of the basal planes about their nominal positions: $e = \Delta d/d$, where d is the basal spacing and Δd is the local displacement. According to the Scherrer formula, $e = \beta^f_G/4tg\Theta$, where β^f_G is is the integral breadth of the Gaussian component of the structural profile. As the displacements may be positive as well as negative values, the square root of the quadratic lattice strain is calculated (SQRT<e²>). This single-line method was applied using the IBM-PC program written by ATTILA NAGY for the Laboratory for Geochemical Research, Budapest.

Because the size distribution of crystallites may considerably influence the shape of line profiles (LANGFORD, 1978; KRUMM, 1995), the mean crystallite size and lattice strain values obtained by the Voigt method from various samples can only be interpreted assuming that there are no significant differences in the crystallite size distributions between the samples compared. At present, no TEM data are available on these size distributions of the studied samples. The approach using the W-A method for size distribution determinations (EBERL and BLUM, 1993) is rather sophisticated and unrealistic for metamorphic phyllosilicates (LANSON and KÜBLER, 1994), and consequently, was not applied in the present study. Taking into consideration all of these uncertainties listed above, only the results within a data set obtained from XRD patterns of a given basal reflection by a given method using a selected (rather arbitrary) "standard" can be compared, evaluating only the relative differences and trends and not the absolute values. Therefore, the obtained parameters are called "apparent" in the present study.

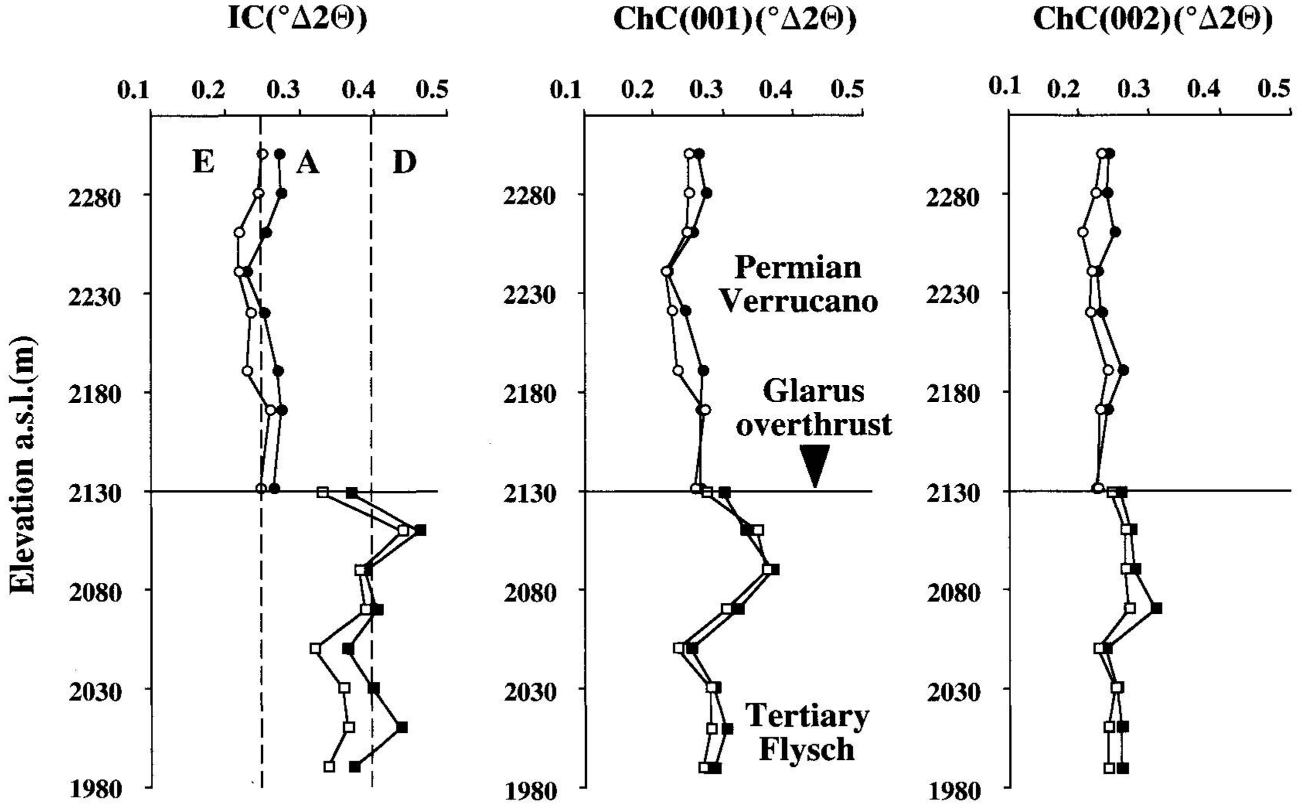

Fig. 2 Changes of illite and chlorite crystallinity indices in a profile crossing the Glarus overthrust at locality A. Legend: circle – Permian Verrucano, square – Tertiary Flysch. Full symbols indicate values measured on air-dried (AD) mounts, open symbols indicate values measured on ethylene glycol-solvated EG) mounts. E – epizone, A – anchizone, D – diagenetic zone.

Tab. 2 Crystallinity, apparent mean crystallite size and lattice strain of illite-muscovite.

sample MF-	air-dried (AD) samples			ethylene glycol-solvated (EG) samples		
	IC, AD Δ°2Θ	crystallite size (Å)	lattice strain (%)	IC, EG Δ°2Θ	crystallite size (Å)	lattice strain (%)
994	0.272	855	0.26	0.250	818	0.41
995	0.275	780	0.31	0.245	741	0.52
996	0.254	831	0.46	0.219	857	0.58
997	0.229	946	0.48	0.218	932	0.55
998	0.252	920	0.39	0.235	1117	0.35
999	0.270	733	0.46	0.230	708	0.69
1000	0.275	556	0.75	0.259	584	0.77
1001	0.264	524	0.89	0.248	484	1.10
1002	0.369	423	0.50	0.330	478	0.52
1003	0.461	281	0.89	0.437	247	1.10
1004	0.386	371	0.73	0.379	342	0.84
1005	0.402	387	0.57	0.387	397	0.56
1006	0.363	404	0.65	0.319	468	0.56
1007	0.396	379	0.57	0.359	372	0.75
1008	0.434	290	0.80	0.362	338	0.83
1009	0.370	421	0.49	0.337	437	0.60
1995	0.256	676	0.66	0.249	709	0.68
1996	0.266	665	0.75	0.234	859	0.55
1997	0.391	451	0.29	0.367	425	0.56
1998	0.392	402	0.47	0.446	366	0.45

For evaluating the significance of the relative differences, the standard deviations of repeated calculations made on XRD line profiles obtained from 5–5 runs of 3 mounts prepared from a given sample are the followings:

- mean crystallite size: 686 Å, s = 33 Å, n = 15
- mean lattice strain: 0.71%, s = 0.06%, n = 15 for illite-muscovite, and
- mean crystallite size: 542 Å, s = 67 Å, n = 15
- mean lattice strain: 1.07%, s = 0.18%, n = 15 for chlorite.

Note that the calculations from the chlorite first basal reflection are less precise than those from the illite-muscovite first basal reflection, mainly due to the increasing uncertainties of determination of the background towards the lower 2Θ angles.

A detailed description of K–Ar dating of the illite-muscovite-rich, < 2 μm grain-size fractions is presented in Árkai et al. (1995a).

Results

ILLITE AND CHLORITE CRYSTALLINITY

Figure 2 shows changes in IC and ChC as a function of the distance from the Glarus overthrust, the numerical values of which are given in tables 2 and 3. The IC and ChC values of the Permian Verrucano correspond to high-grade anchizonal and epizonal conditions, while those of the Tertiary Flysch indicate mostly low-grade anchizonal, and partly transitional into the diagenetic zone conditions. The low-grade anchizonal crystallinity indices of the calc-mylonite correspond fairly well with those of the Tertiary Flysch.

As the Glarus overthrust is approached in the Permian Verrucano at locality A (Fig. 2), a slight decrease in IC and ChC(001) between 2300 and 2240 m, followed by a moderate increase of these indices between 2240 and 2170 m, are seen. The IC and ChC(001) values of the sample taken from the close vicinity of the overthrust plane (2130 m) are practically identical to those of the 2170 m sample. In contrast, ChC(002) shows only slight fluctuations, without notable trends. No systematic changes were found in IC, ChC(001) and ChC(002) values of the Tertiary Flysch either. However, the fluctuations of the FWHM values are rather strong. Based on the ChC indices, perhaps two distinct slices between 2130–2070 m and 2050–1990 m can be distinguished, where the lower portion displays slightly higher metamorphic grade. Ethylene glycol-solvation caused sharpening of the reflections: the FWHM values measured on glycolated mounts are always smaller than those measured on air-dried samples, however, the differences are rather small. This suggests that the proportions in interlayered smectite in both

Tab. 3 Crystallinity, apparent mean crystallite size and lattice strain of chlorite.

sample MF-	air-dried (AD) samples ChC(001) AD Δ°2Θ	ChC(002) AD Δ°2Θ	crystallite size (Å)	lattice strain (%)	ethylene glycol-solvated (EG)samples ChC(001) EG Δ°2Θ	ChC(002) EG Δ°2Θ	crystallite size (Å)	lattice strain (%)
994	0.265	0.244	339	1.23	0.251	0.232	344	1.51
995	0.276	0.241	286	1.65	0.251	0.224	256	1.92
996	0.258	0.252	191	2.43	0.250	0.206	265	1.96
997	0.221	0.226	390	1.39	0.220	0.218	658	1.08
998	0.245	0.231	387	1.20	0.226	0.215	641	0.85
999	0.271	0.261	395	0.87	0.235	0.239	523	0.92
1000	0.269	0.239	305	0.41	0.275	0.229	460	0.31
1001	0.268	0.224	534	0.27	0.261	0.226	614	0.06
1002	0.302	0.258	252	1.68	0.276	0.245	310	1.55
1003	0.332	0.272	294	1.06	0.349	0.265	263	1.62
1004	0.370	0.277	188	2.20	0.362	0.263	199	2.33
1005	0.321	0.308	278	1.24	0.304	0.269	250	1.70
1006	0.254	0.238	402	1.20	0.235	0.226	289	1.84
1007	0.288	0.253	266	1.63	0.283	0.251	259	1.56
1008	0.303	0.258	252	1.60	0.281	0.240	497	0.73
1009	0.288	0.260	243	1.75	0.270	0.239	241	1.87
1995	–	–	–	–	–	–	–	–
1996	0.267	0.218	571	0.12	0.251	0.212	665	0.17
1997	0.323	0.283	285	0.88	0.309	0.259	289	1.29
1998	0.313	0.251	246	1.85	0.347	0.310	338	0.66

the illite-muscovite and chlorite are subordinate. The crystallinity indices of the calc-mylonite from locality B (Tab. 2) agree fairly well with those of the Tertiary Flysch.

APPARENT MEAN CRYSTALLITE SIZE AND LATTICE STRAIN

Illite-muscovite

In contrast to the illite crystallinity distribution (Fig. 2), considerable variations are seen both in the mean crystallite sizes and lattice strains of illite-muscovite (Tab. 2 and Fig. 3). In the Permian Verrucano Fm., a significant and continuous decrease in crystallite size is shown from 2240–2220 m towards the Glarus overthrust. Further from the overthrust plane, between 2240 and 2300 m, only fluctuations (perhaps an insignificant downward increase) are found. Except for one sample (taken from 2220 m), the mean crystallite size values, calculated on XRD profiles obtained from air-dried and glycolated mounts, are practically identical. In comparison with the crystallite size data, the mean lattice strain values show an opposite, symmetrical trend, with the lattice strains increasing toward the main overthrust plane. In the majority of the Verrucano samples, the glycolated mounts exhibit slightly higher lattice strain averages than the air-dried ones. This difference may be the result of interlayered smectitic component, being present in subordinate proportions in the host 10 Å phase.

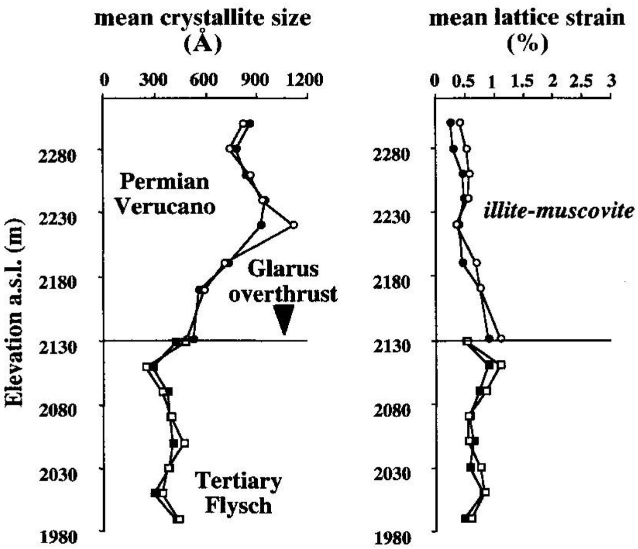

Fig. 3 Variations of apparent mean crystallite size and lattice strain of illite-muscovite across the Glarus overthrust. Values calculated by the Voigt method from the 10 Å basal reflection. For the legend see the caption of figure 2.

In the close vicinity of the main overthrust plane at locality A, the mean crystallite size values of the Permian Verrucano and the Tertiary Flysch are the same for EG, but differ by 101 Å for the AD values, while the mean lattice strain of the latter is significantly lower than that of the Verrucano. Excluding these two samples from the neighbourhood of the Glarus overthrust, the mean crystallite size values from the Permian Verrucano are significantly higher than those of the Tertiary Flysch. In contrast, there is a considerable overlap between the lattice strain values of the two units. Considering the fluctuations in the Tertiary Flysch, two smaller minima in crystallite size, and two more pronounced maxima in lattice strain are found at 2110 and 2010 m, which correspond also to the two maxima in IC (see Fig. 2). Again, opposite trends of crystallite size and lattice strain are shown in the Tertiary Flysch.

At locality B, in the Permian Verrucano samples, the crystallite size values are larger while the lattice strain values are smaller than in the close vicinity of the Glarus overthrust at locality A. However, at locality B, there remains a significant difference in crystallite size and lattice strain between the hangingwall and footwall units: the two Verrucano samples exhibiting larger crystallite size and also larger lattice strain values than the underlying Tertiary Flysch sample. The results of the calc-mylonite are similar to those of the Tertiary Flysch.

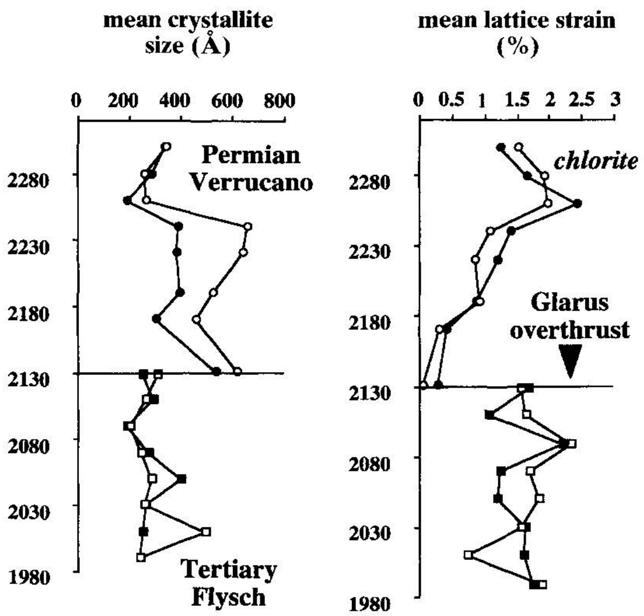

Fig. 4 Variations of apparent mean crystallite size and lattice strain of chlorite across the Glarus overthrust. Values calculated by the Voigt method from the 14 Å basal reflection. For legend see the caption of figure 2.

Chlorite

The chlorite properties show opposite trends in the upper (Permian Verrucano) part of the profile A than that of illite-muscovite. The mean crystallite size of chlorite measured on air-dried samples seems to increase, although with considerable fluctuations, towards the Glarus overthrust (Tab. 3 and Fig. 4). In the case of the glycolated samples, these fluctuations are even larger. Having a maximum at 2250 m, the lattice strain decreases towards the overthrust plane. This latter trend is more well defined than the fluctuating increase in crystallite size. Also in contrast to illite-muscovite, there are relatively large differences between the hangingwall and footwall of the Glarus overthrust at both localities, the crystallite size values being larger and the lattice strain values smaller in the Permian Verrucano than in the Tertiary Flysch. In the underlying Tertiary Flysch itself, only rather strong fluctuations without clear trends are observed both in crystallite size and lattice strain. Considering the whole profile at locality A, there are large overlaps in cyrstallite size and lattice strain values between the Permian Verrucano and the Tertiary Flysch. At locality B, similarly close to the overthrust plane as at locality A, the chlorite from the Verrucano sample immediately above the overthrust plane is characterized by large crystallite size and extremely small lattice strain values, while that of the Flysch by significantly smaller crystallite size and very high lattice strain data. In the calc-mylonite sample, the crystallite size of chlorite is similar (somewhat higher) than that of the underlying Flysch, while the lattice strain is intermediate between those of the Verrucano and Flysch samples.

Tab. 4 K–Ar ages of the < 2 μm illite-muscovite-rich grain-size fractions.

sample MF-	K (%)	^{40}Ar(rad) cc STP/g	^{40}Ar(rad)/^{40}Ar(tot)	age(Ma)* ± 1δ
998	7.93	8.023×10^{-6}	0.548	25.8 ± 1.1
1000	4.03	5.205×10^{-6}	0.385	32.9 ± 1.6
1001	0.859	9.19×10^{-7}	0.082	27.3 ± 4.6
1002	3.70	8.014×10^{-6}	0.450	54.8 ± 2.4
1003	4.802	1.644×10^{-5}	0.713	86.0 ± 3.4
1004	4.35	2.283×10^{-5}	0.679	130.2 ± 5.1
1005	5.31	1.707×10^{-5}	0.763	80.9 ± 3.1
1995	4.56	5.508×10^{-6}	0.194	30.8 ± 2.3
1996	5.35	4.729×10^{-6}	0.354	22.6 ± 1.1
1997	5.76	3.106×10^{-6}	0.267	13.8 ± 0.8
1998	5.15	3.410×10^{-6}	0.382	17.0 ± 0.8

* Calculated using atomic constants of STEIGER and JÄGER (1977).

K–Ar ISOTOPE GEOCHRONOLOGY

Table 4 contains new K–Ar isotopic data on the sample series investigated in the present study. Out of the samples listed in table 4, sample MF-998 has already been analyzed by HUNZIKER et al. (1986). Their result (24.9 ± 1.5 Ma) and the new date (25.8 ± 1.1 Ma) – considering also the standard deviations – agree fairly well. Consequently, the two data sets can be directly compared.

Discussion

Illite and chlorite crystallinity data from two profiles across the Glarus overthrust confirm the earlier observations of FREY (1988) on the metamorphic conditions of the overthrusted Permian Verrucano and the underlying Tertiary Flysch, as well as the post-metamorphic nature of the overthrusting ("transported metamorphism"). The IC data presented here suggest that the grade of the South Helvetic Unit that forms the direct footwall of the overthrust was slightly lower (transitional diagenetic/low-grade anchizonal) than that of the North Helvetic Unit, which was characterized earlier by medium-grade anchizonal IC values.

There are only small-scale fluctuations in IC and ChC as a function of the distance from the Glarus overthrust. In contrast, especially in the hangingwall Permian Verrucano, significant trends are demonstrated in mean crystallite size and lattice strain. On the basis of the trends shown in figures 2, 3 and 4, the tectonic shearing strain, which presumably varied as a function of the distance measured from the main overthrust plane, had no or only very limited effect on the illite and chlorite crystallinity values, but has strongly modified the apparent mean crystallite size and lattice strain relations of these two phyllosilicates. However, strain intensity varies in the different tectonic units of the investigated profiles. Strain is high in the hangingwall Permian Verrucano within a distance of ca. 100–130 m from the main overthrust plane, and in contrast, hardly observable or rather irregularly variable in the underlying Tertiary Flysch. The apparent contradiction found between the trends of crystallinity, crystallite size and lattice strain distributions may be sufficiently explained by the methods used. While the crystallinity reflects only the FWHM values of the basal reflections, the Voigt method uses the whole shape of the structural line-profile. Thus, various size and strain values may be obtained from peaks of equal crystallinity.

Not only the different tectonic units but also the different phyllosilicates responded to the tectonic strain in various ways. Considering illite-muscovite, the decrease of its mean crystallite size and the increase of its lattice strain reflect the growing tectonic shearing strain towards the Glarus overthrust. The increase in tectonic strain might cause advanced reduction of size of the coherent crystallites, and simultaneously, an increase of lattice strain within the crystallites, as the muscovite structure has a great capacity to store strain energy (see also MERRIMAN et al., 1995). In this respect, the crystallization of the (main population) of illite-muscovite in the Permian Verrucano should be considered as pre-tectonic (more precisely, pre-overthrusting). The structural damage (destruction) of illite-muscovite caused by shearing strain connected to the overthrusting was not followed by syn- and post-overthrusting "recovery", i.e., by annealing of sub-grains (= crystallites) and migration of dislocations, all of which might have resulted in an increase of crystallite size and a decrease of lattice strain.

Strongly differing from white mica, chlorite reacts to high strain rates by polygonization (BONS, 1988, cited by MERRIMAN et al., 1995). Because of the relatively slow migration of dislocations in the structure, the post-deformational growth of chlorite crystallites in a prograde system is retarded, compared with that of white mica. Thus, a more pronounced decrease in crystallite size and an increase in strain was expected in the Permian Verrucano section towards the Glarus overthrust. Surprisingly, the chlorite properties (Fig. 4) show trends that are opposite to those of the illite-muscovite (Fig. 3). The increase in crystallite size and the decrease in lattice strain of chlorite towards the Glarus overthrust can only be explained by a syn- and post-overthrusting crystallization of chlorite. This, perhaps rather speculative interpretation is supported by the microscopic observations of BURKHARD et al. (1992), who found that chlorite preferably forms pressure fringes around pyrite porphyroblasts and also fills foliation-parallel veins.

The crystallite size and strain values of illite-muscovite and the crystallite size of the chlorite from the calc-mylonite are similar to those of the underlying Tertiary Flysch, whereas the lattice strain of chlorite is intermediate between the Permian Verrucano and the Tertiary Flysch.

Plotting mean crystallite size of illite-muscovite as a function of IC, hyperbolic relations are shown in figure 5. The sample groups of the Permian Verrucano and the Tertiary Flysch are clearly separated not only in IC but also in crystallite size. The two sample groups plot also as distinct fields in the IC vs lattice strain diagrams (Fig. 6),

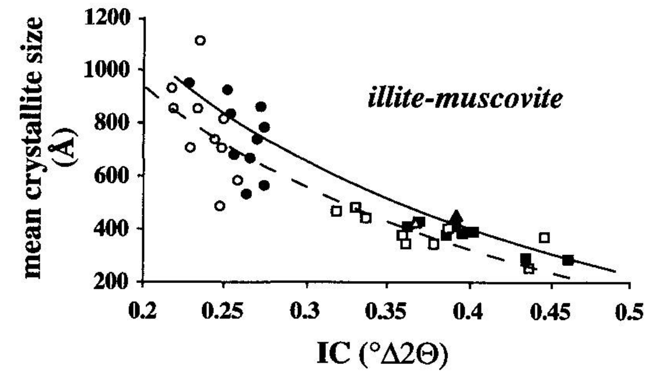

Fig. 5 Apparent mean crystallite size of illite-muscovite as a function of illite crystallinity. Legend: circle – Permian Verrucano, triangle – calc-mylonite, square – Tertiary Flysch. Full symbols – air-dried (AD), open symbols – ethylene glycol-solvated (EG) mounts.

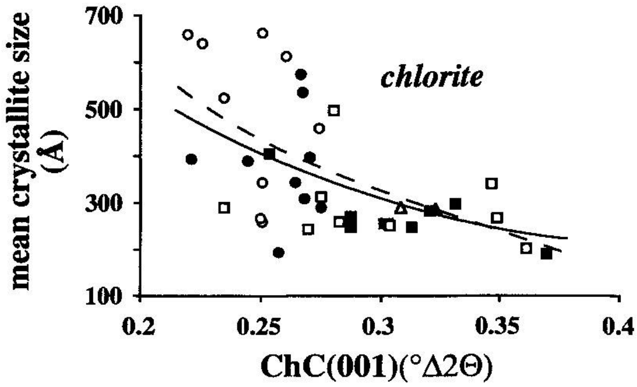

Fig. 8 Apparent mean crystallite size of chlorite as a function of chlorite crystallinity. For legend see the caption of figure 5.

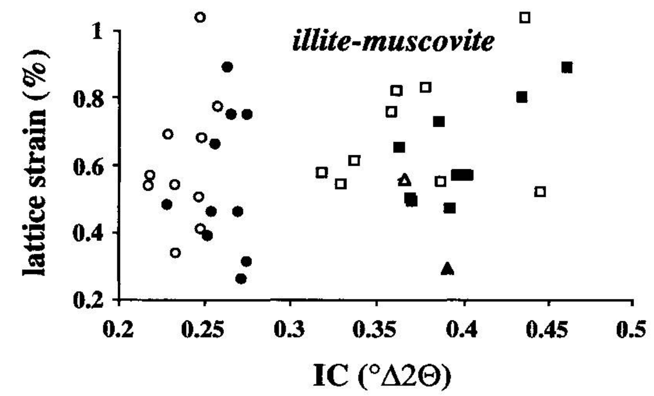

Fig. 6 Apparent mean lattice strain versus illite crystallinity. For legend see the caption of figure. 5.

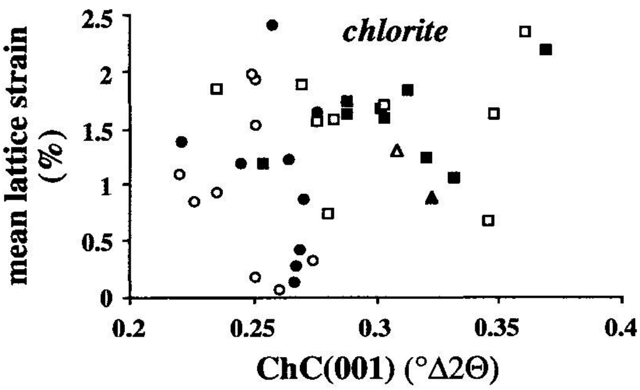

Fig. 9 Apparent mean lattice strain versus chlorite crystallinity. For legend see the caption of figure 5.

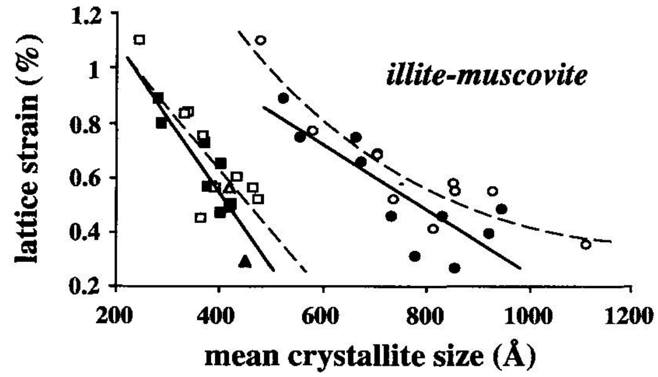

Fig. 7 Apparent mean lattice strain versus mean crystallite size of illite-muscovite. For legend see the caption of figure 5.

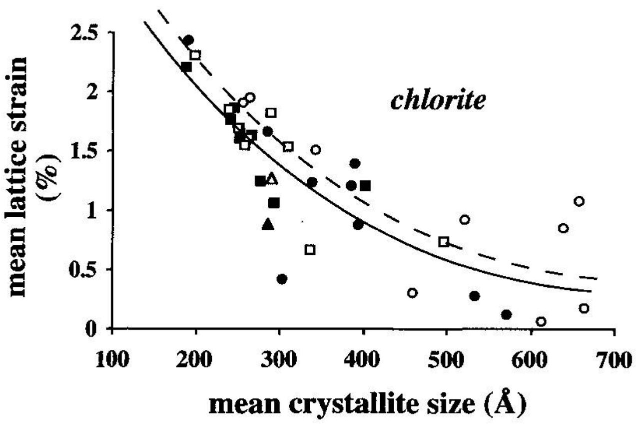

Fig. 10 Apparent mean lattice strain versus mean crystallite size of chlorite. For legend see the caption of figure 5.

however, there are no significant correlations between the IC and lattice strain values. Figures 5 and 6 indicate that the illite crystallinity is influenced mainly by the size of the coherently scattering crystallites, whereas the effect of lattice strain on IC is less important. Similar conclusions were already reached by ÁRKAI and TÓTH (1983), using the results of variance method calculations. Generally, the lattice strain decreases with increasing mean crystallite size (Fig. 7), although the correlation is rather weak. The two groups of the samples are clearly separated also in figure 7, each group exhibiting similar but parallel trends shifted along the crystallite size (x) axis.

Similar inferences can be drawn for the chlorite data: with decreasing ChC(001) the mean crystallite size increases (Fig. 8), whereas no correlation exists between ChC(001) and lattice strain (Fig. 9). A negative correlation is shown between the mean crystallite size and lattice strain values (Fig. 10). In constrast to illite-muscovite, there is considerable overlap between the two sample groups in each type of plot.

Disregarding the sample MF-1996, which is located in the closest vicinity of the Glarus overthrust, i.e., at 0.4 m above the main thrust plane, no systematic changes are found in the K–Ar dates as a function of the distance from the Glarus overthrust; the dates of Verrucano samples scatter between 25.8 and 32.9 Ma, giving an average of 27.0 ± 3.5 Ma (n = 5, the dates of HUNZIKER et al., 1986, are also included).

K–Ar dates older than the stratigraphic age (ca. 35 Ma) were reported by FREY (1988) for the North Helvetic Flysch: 57.7 ± 2.0, 50.7 ± 2.2 and 49.9 ± 1.8 Ma from 450, 900 and 1100 m below the Glarus overthrust. These dates were interpreted as "mixed ages" between the ages of detrital and newly formed (metamorphic) illite-muscovite, as the temperature was not high enough for a complete resetting. In the present study an even wider range was found for the samples of the South Helvetic Unit, where, disregarding sample MF-1998, the apparent K–Ar dates scatter between 54.8 and 130.2 Ma (Tab. 4). Note that the date of sample MF-1002 taken from the direct footwall of the Glarus overthrust plane at locality A is much younger (54.8 Ma) than the ages obtained from more distant samples of the Tertiary Flysch. In addition to this presumed rejuvenating effect of the overthrusting, the great scatter may refer either to stronger variation in the proportions of the inherited and newly formed white micas, and/or to an eventual lower-T anchizonal metamorphism of the South Helvetic Unit, as compared to the underlying, medium-T anchizonal North Helvetic Flysch.

The K–Ar age of the calc-mylonite sample MF-1997 (13.8 ± 0.8 Ma) is considerably younger than the concordant K–Ar and Rb–Sr ages of ca. 23 Ma obtained on a calc-mylonite sample from the Lochseite type locality by HUNZIKER et al. (1986). This new Miocene age may be correlated with the final emplacement of the frontal parts of the Helvetic Säntis thrust sheet which is thought to represent the frontal emergence of the Glarus overthrust (see also BURKHARD et al., 1992). In addition, this very young age indicates that the K–Ar system of illite-muscovite remained open even in the late stages of thrusting. This phenomenon may be the result either of the enhanced temperature caused by the advecting fluids in the closest vicinity of the overthrust plane, and/or by the chemical effects of these fluids, and/or by the tectonic shearing effect on white mica, which might also cause liberation of radiogenic argon from the white mica structure. Younger ages were obtained also on samples located 0.4 m above and 0.5 m below the overthrust plane as compared to the other samples of the units. Thus, the effects of the Glarus overthrust on K–Ar dates of the illite-muscovite-rich, < 2 μm fraction samples can be proved only within a narrow (several dm wide) zone below and above the calc-mylonite. This rejuvenation seems to be stronger in the footwall Flysch (17.0 ± 0.8 Ma) than in the hangingwall Verrucano (22.6 ± 1.1 Ma).

Conclusions

Evaluating the changes of illite-muscovite and chlorite properties as a function of distance from the Glarus overthrust, which has been characterized by high water/rock ratio, advection of hot (ca. 350 °C) fluids, penetrating also the hangingwall Permian Verrucano (BURKHARD et al., 1992) that enabled a large-scale (ca. 10 km) horizontal nappe displacement on this gliding plane (RAHN et al., 1995), the following conclusions can be drawn.

The new crystallinity data confirm the earlier observations of FREY (1988) on the mainly epizonal metamorphism of the overthrusted Permian Verrucano and the mostly anchizonal features of the underlying Tertiary Flysch slates, as well as the post-metamorphic nature ("transported metamorphism") of the classic Glarus overthrust in the Swiss Helvetic Alps.

There are only small-scale variations in IC and ChC as a function of the distance from the Glarus overthrust. In contrast, the mean crystallite size and lattice strain values of phyllosilicates change significantly in the Permian Verrucano, within an interval ranging up to ca. 100–130 m upwards from the overthrust plane. Thus, in the case of illite-muscovite, the apparent mean crystallite size decreases, the average lattice strain increases towards the Glarus overthrust. These variations may be the result of increasing structural damage caused by growing tectonic shearing strain. The destruction of crystallites was not followed by syn- and post-overthrusting (re)crystallization of illite-muscovite. As compared to illite-muscovite, opposite trends, i.e., an increase in crystallite size and a decrease in lattice strain have been proved for chlorite. These trends can only be explained by syn- and/or post-overthrusting crystallization. The crystallinity, mean crystallite size and lattice strain properties of illite-muscovite and chlorite from the calc-mylonite that formed the "gliding plane" for the nappe movement, are similar to those of the footwall Tertiary Flysch, in which no systematic changes could be shown. All the above mentioned changes can be explained by the sequential effects of tectonic shearing strain, and infiltration, migration of hot fluids along the overthrust plane and upwards, into the Permian Verrucano. The intensity of these effects should have decreased, getting farther from the overthrust plane.

In general, with decreasing illite and chlorite crystallinity (i.e., with increasing metamorphic grade), the apparent mean crystallite size increases. This relation may be approximated by a hyperbolic function. In contrast, there are no significant correlations between the crystallinity indices and the apparent mean lattice strain values. Consequently, the illite and chlorite crystallinity indices are influenced mainly by the size of the crystallites or domains that scatter coherently the X-ray (see also ÁRKAI and TÓTH, 1983). In general, the lattice strain of the investigated phyllosilicates decreases with increasing crystallite size, although there are considerable deviations between the various sample groups.

The mechanical, thermal and chemical (fluid advection, infiltration) effects of the overthrusting that caused considerable rejuvenation of K–Ar dates, obtained on < 2 μm illite-muscovite from slates, can be demonstrated only within a rather narrow (several dm wide) zone above and below the gliding plane. The illite-muscovite K–Ar date of the calc-mylonite itself may refer to the end of the nappe movement.

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