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A RECONNAISSANCE STUDY OF VERY LOW-GRADE METAMORPHISM IN SOUTH DEVON

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The first results of a clay mineral crystallinity survey of south Devon are presented using samples from the Devonian and Carboniferous slates lying north of the Start Peninsula and around the margins of Dartmoor. An overall regional increase in grade from north to south correlates both with stratigraphic and structural depth. Higher grade metamorphic rocks are located within the older and more intensely deformed southern structural zone of the Dartmouth area, whereas less metamorphosed rocks occupy the more flat-lying and commonly less strained northern zone of the Plymouth-Ashburton-Torbay area. The Start-Perranporth Line is confirmed as juxtaposing two areas of different metamorphic grade, and a zone of regional D2, retrogressive growth of kaolin is recognised within the vicinity of the fault zone.

Correlations between illite and chlorite crystallite sizes confirm the metamorphic nature of the phyllosilicate minerals in the majority of samples, and near-equilibrium conditions are suggested. The poor correspondence of crystallite sizes within certain sample groups, such as those recorded from diagenetic slates in the vicinity of Lydford, may be used as a criteria for identifying non-equilibrium assemblages.

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INTRODUCTION

Clay mineral crystallinity (Frey, 1987), as determined by X-ray diffraction (XRD), is an easy and rapid technique for determining the degree of very low-grade metamorphism, utilising illite, chlorite or kaolin-bearing assemblages found in mudstones, slates and tuffs. During the last 15 years, day mineral crystallinity methods, particularly using illite, have formed the principal approach in determining the conditions of very low-grade metamorphism in south-west England (Warr *et al.,* 1991), which reflects the predominance of pelitic lithologies and the rarity of diagnostic mineral facies assemblages in metabasites (Robinson *et al.,* 1994).

Although the clay mineral crystallinity approach forms an empirical and general measure of metamorphic grade, recent advances have been made in both the physical understanding and methodology of clay mineral crystallinity techniques. Combined clay mineral crystallinity and high resolution transmission electron microscopy studies (Eberl *et al.,* 1990; Merriman *et a1.,* 1990; Nieto and Sanchez-Navas, 1994; Jiang and Peacor, 1994), have confirmed the notion that the method provides an empirical measure of the thickness of crystallites (the X-ray scattering domains) in the crystallographic c*-direction (e.g. Weber *et al.,* 1976). Available procedures for calculating crystallite size information directly from the XRD profiles have been tested, such as the Scherrer method (Merriman *et al.,* 1990), the Wilson method (Árkai and Tóth, 1983; Nieto and Sanchez-Navas, 1994), the Warren-Averbach method (Eberl and Środoń, 1988; Eberl and Blum, 1994; Warr and Rice, 1994; Wan, in press) and a Siemens Single-Line method (Warr, in press). Additional procedures have also been developed, such as the computer generated grid of Eberl and Velde (1989), which predicts crystallite size thicknesses from illite crystallinity (IC) and the Intensity ratio (Ir) data of Środoń (1984).

This paper presents clay mineral crystallinity results and crystallite size data, calculated by the Siemen's Single-Line method, for 67 samples collected from south Devon, in order to assess the growth history of the various phyllosilicate minerals and further to produce the first metamorphic map for the region at a reconnaissance level. Although the results presented are not directly comparable to previous IC data which originated largely from studies undertaken at the University of Bristol (see Warr *et al.,*1991 for review), the database has been calibrated to a new internationally recognised Crystallinity Index Standard scale (Warr and Rice, 1994), which enables direct comparisons to be drawn against other laboratories following this method of standardisation.

REGIONAL GEOLOGY

An overview of the regional geology is given by Selwood and Durrance (1982) and Selwood *et al.,* (1982), with detailed accounts of the structure found in Coward and McClay (1983) and Chapman *et al.,* (1984). The main geological units which are presented in Figure 2a are summarised as follows.

The Start Complex of south Devon comprises an interlayered series of highly deformed schistose pelites, and basic igneous rocks with normal-type mid-oceanic ridge basalt (N-MORB) chemistry (Floyd *et* al., 1993). These are separated from the Lower Devonian rocks to the north by the east-west trending Start-Perranporth Line (SPL), a prominent fault zone which is considered to be a line of intense ductile dextral transpression (Holdsworth, 1989). The Lower Devonian rocks of south Devon are divided into the Dartmouth Group (Smith and Humphreys, 1989), located in the core of the regional scale Dartmouth Antiform, and the Meadfoot Group (Seago and Chapman, 1988). The Dartmouth Group is comprised of monotonous slates, phyllites and sandstones which are interpreted as originating largely in a lacustrine environment (Smith and Humphreys, 1989), whereas the Meadfoot Group consists of predominantly grey slates and thin sandstones, interpreted as shallow water estuarine-marine sediments with storm deposits (Richter, 1967). Between Plymouth and Torbay there occurs an area of Middle Devonian rocks consisting of mostly slates and massive limestone fades. Northwards, around the eastern and western sides of Dartmoor, the rocks comprise complex successions of Middle to Upper Devonian slates and limestones, and Lower to Upper Carboniferous clastic successions (Selwood and Durrance, 1982; Isaac, 1981).

The structure of the area is characterised by large scale northward verging thrust nappes (Coward and McClay, 1983; Selwood *et a1.,* 1982), with the position of major thrust boundaries shown in Figure 2a.

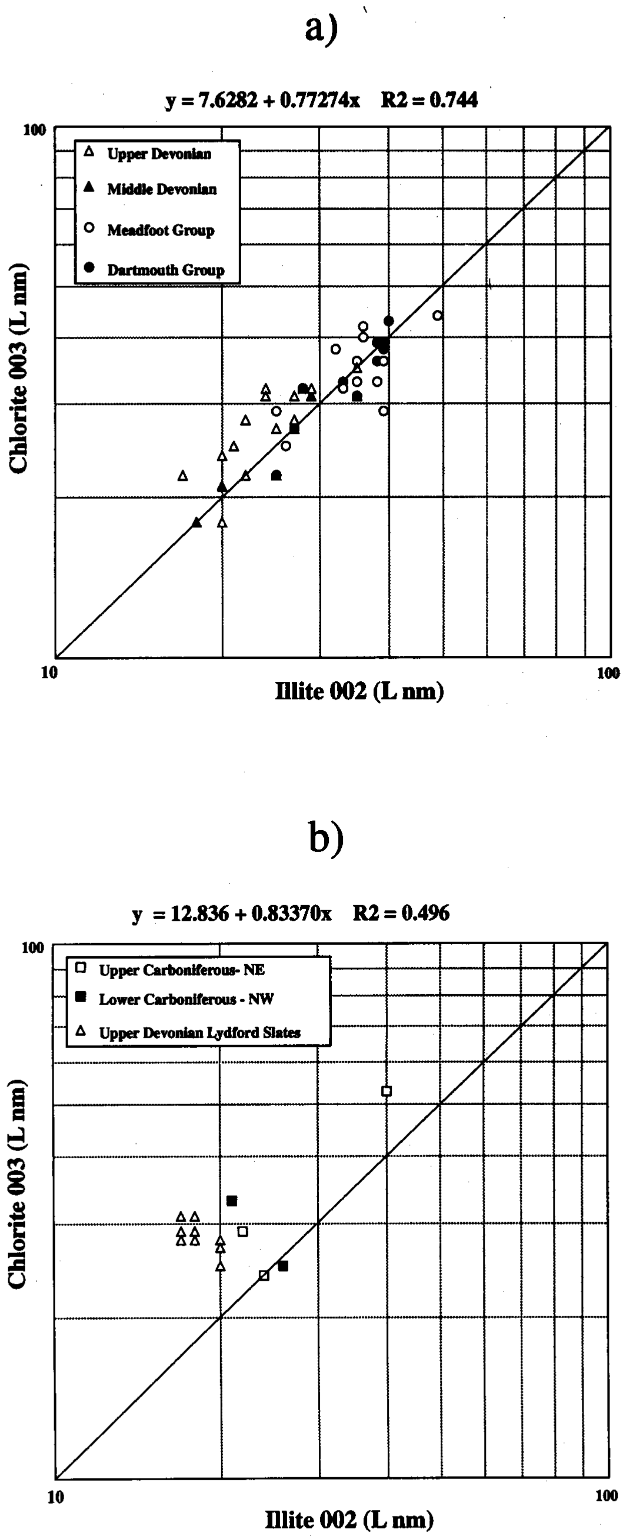

Figure 1. Log-log graph of the crystallite sizes (L nm) of illite (002) and chlorite (003) from Lower to Upper Devonian slates lying north of the Start Complex. A 1:1 line is shown for reference, and the regression equation for the best fit straight line is given above the graph. b) Log-log graph of the crystallite sizes (L nm) of illite (002) and chlorite (003) from Upper Devonian to Lower Carboniferous slates situated around the margins of Dartmoor. A 1:1 line is shown for reference, and the regression equation for the best fit straight line is given above the graph.

Two principal tectonic zones have been long recognised (e.g. Sanderson and Dearman, 1973; Coward and McClay, 1983); a southerly zone of largely inclined, tight to isoclinal, steeply-dipping folds (Zone 8 of Sanderson and Dearman, 1973), and a northerly zone of flat-lying recumbent folds (Zone 9 of Sanderson and Dearman, 1973). The east-west trending division between these two areas runs roughly parallel with the Lower-Middle Devonian boundary.

Although both zones are characterised by polyphase folding and cleavage development, the southern zone is more intensely and complexly deformed. Within this zone, Coward and McClay (1983) attributed the prominent second phase folding as giving rise to the Dartmouth Antiform, and a southerly intensifying S2 crenulation cleavage. They recorded the highest strains just south of Dartmouth (at Blackpool), while the lowest strains were determined in the northern zone, in poorly cleaved limestones around Torbay. These authors suggested this overall strain pattern of apparent northward-decreasing intensity of deformation as possibly reflecting competency differences in the deformed rocks.

ANALYTICAL METHODS AND DATA CALIBRATION

The 64 pelites were prepared and analysed using the equipment and methods described in Warr and Rice (1994). Two XRD slide preparations from each sample were scanned in the air-dried condition from 2 to 50°, at a scan-rate of 0.6°2θ/min^{-1} and a step-width of 0.01°. All data presented are mean values based on the two slide preparations per sample (n=2). When mixtures of chlorite and kaolin (terminology of Ehrenberg *et al.,* 1993) were suspected, further analysis of the clay mineral assemblages was undertaken by using a combination of air dried, glycolated, heated and occasional dimethyl sulfoxide treated specimens (Moore and Reynolds, 1989).

Full details of the analytical procedure for treatment of the raw data files are given in Warr (in press), in which the Siemen's Single-Line method (Siemens Win-Crysize, 1991) has been utilised. Following removal of a linear background level, basal 001 (≈10Å), 002 (≈5Å), illite (white mica) reflections, 002 (≈7Å) chlorite reflections, and 001 (≈7.Å) kaolin reflections were fitted using a Split Pearson VII function by the program FIT (Siemens version 3). Although Kα_2 was retained for peak-breadth (crystallinity) measurements at low 2θ angles (to maintain higher counting statistics), the influence of this reflection was removed by deconvolution prior to crystallite size calculations. Suitably fitted profiles were transferred to the WIN-CRYSIZE program (version 1.0) and calculations made from single peaks by the Single-Line method. This method employs the Fourier Transform of the pure broadening function, and assumes both that small crystallites cause peak broadening in the form of a Lorentzian curve and that strain causes peak broadening in the form of a Gaussian curve. All crystallite size calculations were made using machine broadening profiles determined on a single crystal muscovite standard (MF1c).

The data presented in this paper has been calibrated to the Crystallinity Index Standard (CIS) scale of Warr and Rice (1994). Experimental half-peak-widths were converted to standardised CIS values by the linear regression equation data presented in Warr (in press). Direct comparisons with previous IC studies from south-west England (Warr *et al.,* 1991) should be avoided, until the old databases have been suitably calibrated. Despite these present restrictions, the relative pattern of metamorphism should, however, remain unchanged.

RESULTS

Mite, chlorite and kaolin crystallite sizes

The results of crystallite size determinations made by the Siemens Single-Line method are presented in Figure 1.

A correlation between the crystallite size of illites and chlorites for diagenetic to epizonal Devonian slates (Figure la) shows an overall

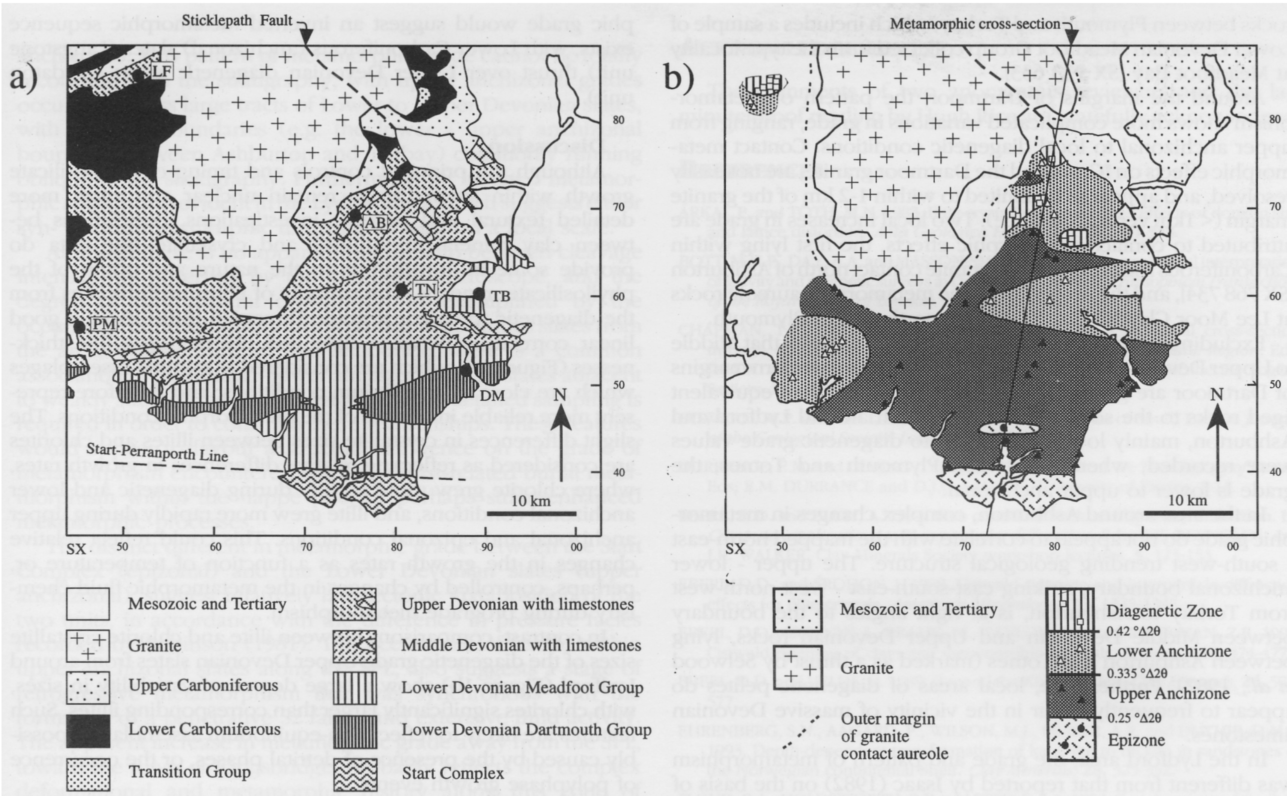

Figure 2. a) Generalised geological map of south Devon (modified after Durrance and Laming, 1982) PM = Plymouth, LF = Lydford, AB = Ashburton, TN = Totnes, TB = Torbay, DM = Dartmouth. b) Metamorphic map of south Devon compiled from IC data measured on the Crystallinity Index standard scale of Warr and Rice (1994).

good straight line regression (R^2 = 0.744), confirming the metamorphic character of the phyllosilicate minerals. Although a straight line regression provides the best line fit, it is noticeable that in pelites of diagenetic to lower anchizonal grade, the chlorites are usually slightly thicker than the illites (points falling above the 1:1 line), whereas in upper anchizonal to epizonal grades, it is the illites that are often the thicker of the two (points falling below the 1:1 line).

In contrast, a number of diagenetic and anchizonal samples which show significantly thicker chlorite sizes than illite, mainly from the Lydford area, are plotted in Figure lb. A rather poor linear correlation exists (R^2 = 0.495), suggesting the illites and chlorite may not have grown simultaneously. Such a distribution probably reflects phyllosilicates that are not in textural equilibrium with each other.

The regional variation in the mean crystallite sizes of illite, chlorite and kaolin, shown in the metamorphic cross-section of Figure 3a, is mirrored by changes in IC, ChC (chlorite crystallinity), and KC (kaolin crystallinity), displayed in Figure 3b. This is in accordance with the suggestion that the principal control on clay mineral crystallinity is that of the effective crystallite size, as expressed by the Scherrer equation (Merriman *et al.,* 1990).

Although too few samples of kaolin-bearing pelites were analysed to establish the relationships between the growth of illites and kaolin (presumably dickite), the limited samples from along the SPL (Figure 3b) show KC to be similar to that of IC.

Regional variations in metamorphic grade

A reconnaissance metamorphic map compiled from the IC data is presented in Figure 2b, which is supplemented by the generalised north-north-east - south-south-west metamorphic cross-section given in Figure 3. Across the region, the grade of metamorphism ranges from local epizonal values in the south, through a large central area of anchizonal slates, to local diagenetic grades around the margins of Dartmoor. All pelitic samples were examined under the binocular microscope, and without exception, comprise slates with closely spaced to penetrative (slaty) fabrics.

A sample of pelitic schist from the Start Complex [SX 738 380] yielded the lowest crystallinity values (IC =0.226° Δ2θ, ChC =0.24° Δ2θ) from the survey, in accordance with lower greenschist facies assemblages seen in metabasites (Tilley, 1923). North of the Start Schists there is a clear decrease in metamorphic grade, with upper anchizonal slates within the Meadfoot Group north of the SPL. A number of kaolin-bearing slates were identified in the vicinity of the fault zone, associated with intense polyphase deformation and mineral veining. Thin section analysis of a pelitic sample collected from 50 m north of the fault contact at Outer Hope [SX 675 403], revealed abundant kaolin packets (presumably dickite) defining a locally strong slaty fabric, probably S2, which is axial planar to subvertical isoclinal folds and overprints both bedding and networks of subparallel thick quartz veins. Microscopic grains of mylonitic ribboned and sutured quartz were also observed, along with abundant pre- and post-slaty cleavage calcite growth.

North of the Start Complex, a trend toward epizonal grades at the northern limit of the Kingsbridge Estuary occurs (Figures 2b and 3a, b), with illite and chlorite crystallite thicknesses ranging from around 35 nm just north of the SPL, to 50 nm thicknesses at Kingsbridge [SX 740 447]. This trend is local in nature and does not appear to correspond with any gravity or magnetic anomalies (Bott *et al*., 1958) that may reflect underlying igneous rocks. Along the coastal section to the east, no corresponding variations in metamorphic grade appear, with upper anchizonal grades recorded as far north as Dartmouth. Overall, there is no equatable variation in IC with the regionally mapped Dartmouth Antiform, or across its supposed thrusted northern limb (Coward and McClay, 1983; Chapman *et al.,* 1984). A notable band of lower anchizonal slates does occur mostly within Middle Devonian rocks between Plymouth and Torbay, which includes a sample of Lower Devonian Meadfoot Group collected from the type locality at Meadfoot Bay [SX 933 633].

Around the margins of Dartmoor, the pattern of metamorphism shows more complicated variations in grade, ranging from upper anchizonal to local diagenetic conditions. Contact metamorphic effects on IC, around the Dartmoor granite, are not easily resolved, and appear to be limited to within 1-2 km of the granite margin (< 1 km vertical distance). Two local increases in grade are attributed to contact metamorphic effects, the first lying within Carboniferous rocks close to the granite contact north of Ashburton [SX 768 734], and the second from the metamorphic aureole rocks at Lee Moor Clay Works [SX 55 60], north-east of Plymouth.

Excluding contact metamorphic effects, it appears that Middle to Upper Devonian slates around the eastern and western margins of Dartmoor are generally less metamorphosed than equivalent aged rocks to the south. In the areas both around Lydford and Ashburton, mainly lower anchizonal to diagenetic grade values were recorded, whereas between Plymouth and Totnes, the grade is lower to upper anchizonal.

In the area around Ashburton, complex changes in metamorphic grade do not appear to correlate with the mapped north-east - south-west trending geological structure. The upper - lower anchizonal boundary, striking east-south-east - west-north-west from Torbay to Ashburton, is at right angles to the boundary between Middle Devonian and Upper Devonian rocks, lying between Ashburton and Totnes (marked as a thrust by Selwood *et al.,* 1982). Furthermore, local areas of diagenetic pelites do appear to frequently occur in the vicinity of massive Devonian limestones.

In the Lydford area, the grade and pattern of metamorphism was different from that reported by Isaac (1982) on the basis of clay mineralogy. Firstly, the Upper Devonian Lydford slates yielded diagenetic grades, with mixed-layered illite/smectite, rather than the supposed anchizonal/epizonal grades (illitechlorite zone with no mixed-layering), and secondly, the grade of metamorphism increases into the Lower Carboniferous slates immediately to the south, rather than decreases. Employing the regional structure of Isaac (1981), this distribution of metamorphic grade would suggest an inverted metamorphic sequence exists, with Lower Carboniferous (anchizonal) slates (Langston unit) thrust over Upper Devonian diagenetic slates (Liddaton unit).

DISCUSSION

Although the principal controls and timing of phyllosilicate growth within south Devon remain unclear and await more detailed textural and isotopic investigations, correlations between clay mineral crystallinity and crystallite size data do provide some information as to the nature and origin of the phyllosilicate minerals. The majority of illites and chlorites from the diagenetic to epizonal grade Devonian slates show a good linear correlation between illite and chlorite crystallite thicknesses (Figure la), which are considered as mineral assemblages which are close to equilibrium conditions, and therefore represent more reliable indicators of the metamorphic conditions. The slight differences in crystallite sizes between illites and chlorites are considered as reflecting relative differences in growth rates, where chlorite grew more rapidly during diagenetic and lower anchizonal conditions, and illite grew more rapidly during upper anchizonal and epizonal conditions. This could reflect relative changes in the growth rates as a function of temperature or, perhaps, controlled by changes in the metamorphic fluid chemistry during prograde metamorphism.

In contrast, comparisons between illite and chlorite crystallite sizes of the diagenetic grade Upper Devonian slates from around Lydford (Figure lb) show a large degree of variability in sizes, with chlorites significantly larger than corresponding illites. Such features are likely to reflect non-equilibrium assemblages, possibly caused by the presence of detrital phases, or the occurrence of polyphase growth events.

Despite the uncertainties as to the exact causes of metamorphism, the reconnaissance map of very low-grade metamorphism for the south Devon region does reveal that the older Devonian rocks found to the north of the Start Complex are generally more metamorphosed (upper anchizone - epizone) than the Devonian and Carboniferous successions cropping out around the eastern and western parts of

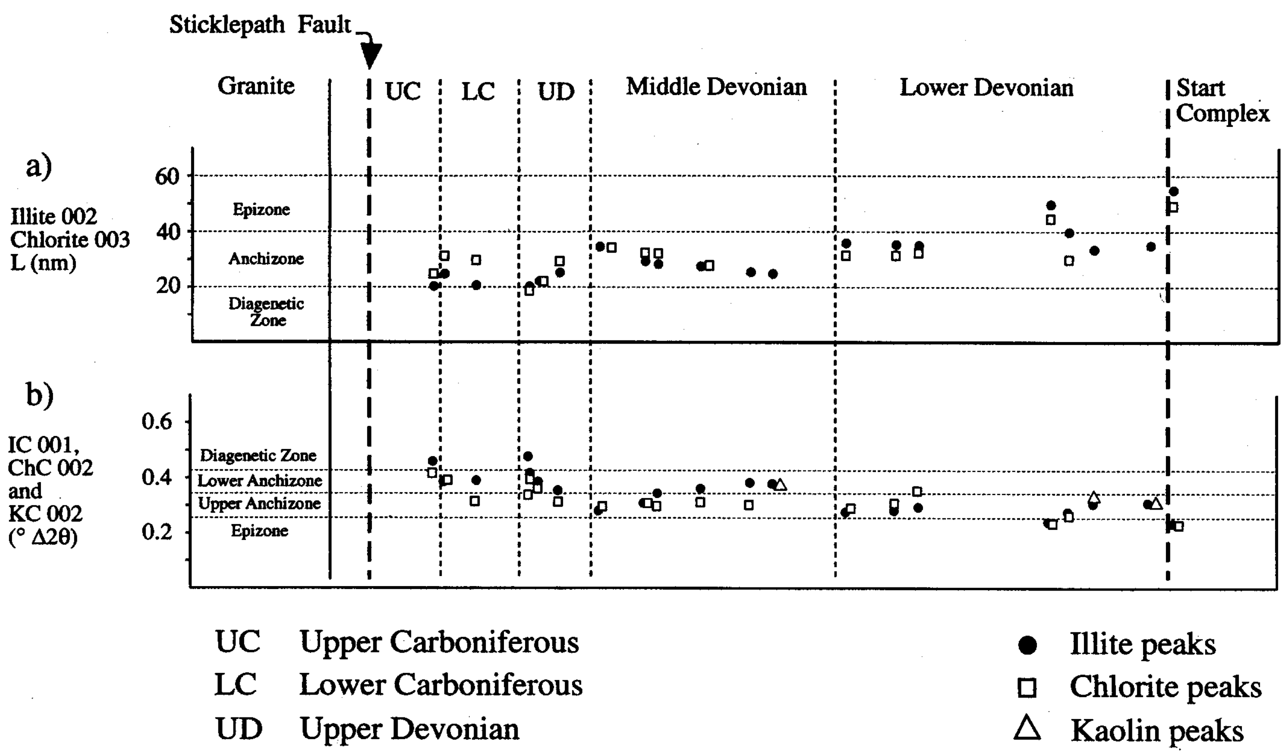

Figure 3. Generalised cross-section across south Devon showing the patterns of clay mineral crystallinity and crystallite size. a). Illite, chlorite and kaolin crystallite size data (L nm). b) Illite, chorite and kaolin crystallinity data (°Δ2θ).

Dartmoor (diagenetic zone - local upper anchizone). This pattern of metamorphic grade cannot be totally reconciled with the stratigraphy, with upper anchizonal grades occurring across large tracts of Lower to Upper Devonian slates, with isocryst boundaries (e.g. the lower - upper anchizonal boundary between Ashburton and Torbay) commonly running oblique to the stratigraphy. Furthermore, an inverted metamorphic sequence is evident in the Lydford area which suggests either syn- or post-metamorphic thrusting, at least on a local scale.

Although there is no apparent relationship between cleavage intensity, as observed by the binocular microscope, and the degree of clay mineral crystallinity, as exemplified by the occurrence of penetratively cleaved diagenetic grades slates from the areas around Lydford and Ashburton, there is a common association of diagenetic - lower anchizonal grade slates adjacent to weakly strained massive limestones. More detailed sampling is required in order to constrain these relationships. These features would appear to favour a structural influence on the grade of metamorphism encountered, such as thrust-related burial metamorphism within the nappe pile, or perhaps strain-enhanced metamorphic processes.

The distinct different in metamorphic grade between the Start Complex (epizonal) and the Lower Devonian slates (upper anchizonal), suggest relatively late tectonic juxtaposition of the two units, in accordance with the difference in pressure facies recorded by Robinson (1981). The occurrence of kaolin within upper anchizonal slates along the SPL, also suggests a stage of retrogressive metamorphism that was synchronous with the formation of a penetrative S2 fabric and extensive fluid activity. The apparent increase in metamorphic grade away from the SPL, toward the town of Kingsbridge, probably reflects the complex deformational and metamorphic history along this zone of intense strike-slip movement. Late tectonic movements along the SPL are also supported by differences in IC across the fault zone in west Cornwall (Primmer, 1983), where higher metamorphic grades were found on the northern side of the fault, rather than to the south.

The principal division of the region into an upright-folded, intensely deformed southern structural zone and a recumbent lying, less deformed northern zone (Coward and McClay, 1983) is partly reflected by the distribution of metamorphic grade. Within the southern zone, the grade is dominantly upper anchizonal with some areas of epizonal values, whereas the northern zone is mainly of lower anchizonal to diagenetic grade. The division between these structural zones cannot, however, be defined on the basis of the metamorphic data, perhaps a reflection of the low sample density across this central region. Overall, however, the pattern is equatable with the structural scenario of deeper crustal levels of higher metamorphic grade lying towards the south.

SUMMARY

1) Devonian and Carboniferous slates of south Devon range from local epizonal grades in the south, through extensive areas of anchizonal grades between Plymouth and Torbay, to local diagenetic grades around the margins of Dartmoor.

2) Generally, the increase in grade from north to south can be related both to stratigraphic and structural depth, with the higher grade metamorphic rocks (upper anchizone - epizone) located within the older and more intensely deformed southern structural zone (Zone 8 of Sanderson and Dearman, 1973), and less metamorphosed rocks (upper anchizone - diagenetic zone) occupying the more flat-lying and commonly less strained northern zone (Zone 9 of Sanderson and Dearman, 1973).

3) The SPL is confirmed as dividing rocks of different metamorphic grade, with epizone values south of the fault within the Start Complex, and upper anchizone values to the north of the fault within Devonian strata. Additionally, a zone of kaolin growth is recognised, synchronous to the formation of the steeply-dipping S2 cleavage, which is suggested to have occurred during retrogressive conditions that post-dated both upper anchizonal conditions and mylonitisation along the fault zone.

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