A CLAY MINERAL CRYSTALLINITY INVESTIGATION OF THE UPPER CARBONIFEROUS CULM BASIN OF SOUTH-WEST ENGLAND

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Illite, chlorite and kaolinite crystallinity data from two inland traverses across the Upper Carboniferous Culm Basin have revealed important changes in the pattern of regional metamorphism both across and along strike. Metamorphism appears to increase both with stratigraphic depth and along strike from the eastern to the western parts of the basin. Both the range and the distribution of metamorphic grade is therefore comparable with that established for the South Wales Coalfield, and similar tectono-thermal processes are considered likely.

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INTRODUCTION

The preserved Upper Carboniferous Culm Basin of south-west England comprises a 3-km-thick sequence of turbiditic (Crackington Formation), lacustrine shelf or subsea fan (Bude Formation; Melvin, 1986; Higgs, 1991), and deltaic (Bideford Formation; de Raaf et al., 1965) basin-fill sediments which formed in a foreland basin setting (Hartley and Warr, 1990; Warr, 1993; Hecht, 1992). Most of the data on the grade of metamorphism has come from illite crystallinity (IC) studies (see Warr et al., 1991 for summary), which has proved a useful method for establishing the degree of very low grade metamorphism of mudstones and shales (Frey, 1987). However, the crystallinity of other common clay minerals, such as chlorite (ChC) and kaolinite (KaC) have, received little attention in the Variscides of south-west England.

A large proportion of the IC data for the Culm Basin was obtained from the well-exposed coastal areas (Kelm, 1986; Primmer, 1985; Warr and Robinson, 1990; Hecht, 1992), with only a reconnaissance level sampling of the inland region (Kelm, 1986). The metamorphic grade of the inland tract, which represents over one third of the area of the Variscides...
of south-west England, is still poorly known. The absence of economic coal reserves has resulted in a lack of borehole information and associated coalification data, unlike the South Wales Coalfield (White, 1991). Some preliminary vitrinite reflectance results were presented by Cornford et al. (1987), which showed a range of maturities between 1.5 to 5.67% Rm, but this data was geographically restricted to the Atlantic coastline and the area around the northern margin of the Dartmoor granite.

The main objective of this study was to establish a more detailed database from across the basin as a whole, in order to determine the patterns of metamorphic grade in relation to the well-studied coastal section. To achieve this, sampling was concentrated along two inland north-to-south traverses through the basin. The first (A-B, Figure 1), passes from the Bideford area towards the northwest margin of the Dartmoor granite (around Okehampton), and the second (C-D) traverses the area north-west, west and south-west of Exeter. In addition to IC, the ChC and KaC were determined and assessed in terms of metamorphic grade.

ANALYTICAL PROCEDURE
A total of 258 mudstone and shale samples were collected from the Upper Carboniferous rocks of the Culm Basin. The analytical method followed as closely as possible the recommendations of Kisch (1991), except in the usage of a disc mill for disaggregation purposes, and the absence of cation saturation. A preliminary test of the precaution, however, milling times were limited to <30 seconds. The prepared by the pipetteHonHglass method, keeping the specimen thicknesses relatively thin (approx. 1mg/cm²). Measurements were made using a Siemens D5000 diffractometer based at the Geologisch-Paläontologisches Institut in Heidelberg. Full details of the measuring parameters and operating procedures used are listed in Warr and Rice (In press). Crystallinity determinations for the 10Å illite and 7Å chlorite/kaolinite reflections were made using the full-width-half-maximum (FWHM) parameter of the Siemens Diffrac-A1 (version 3) analytical software programme. The experimental results were converted to the scale of HJ. Kisch (1980; 1990) by calibration against a set of five polished slate standards and a muscovite crystal supplied directly by Kisch. The calibration equation used was:

\[ IC \text{ this work} = 1.511558 \times IC \text{ Kisch} - 0.029329 \quad R^2 = 0.945. \]

All results presented in this study have been calibrated to the scale of Kisch (1980) using the above equation, and are comparable with previous IC data determined at the University of Bristol (Warr et al., 1991). The upper and lower boundaries of the anchizone were therefore taken at IC values of 0.21 Δ°2θ and 0.38 Δ°2θ respectively.

Clay mineral identifications of the <2µm fraction were determined on air dried and glycolated preparations, with selected heating and/or acid treatments used to distinguish between kaolinite and chlorite peaks (Moore and Reynolds, 1989). To determine the relationships between IC and crystallite (X-ray scattering domain) size, two mudstone samples (SW1 and SW2), which were considered typical of the Bude and Crackington Formations respectively, were selected from the coastal section (Figure 1). Calculations were performed by the Warren-Averbach method (Warren and Averbach, 1950), using the procedure of Eberl and Srodon (1988). Full description of these analyses are given in Warr and Rice (In press).

RESULTS
Clay mineralogy of the <2µm fraction
The clay mineralogy recorded from the mudstones and shales is much in agreement with the identifications made by Grainger and George (1978) and Grainger and Witte (1981) from the Crackington shales. The <2µm fraction consists mostly of illite/smectite, illite/muscovite, chlorite (both Fe and Mg varieties), and kaolinite. Although all of the four group assemblages described by Grainger and Witte (1981) were recognised, by far the most common of these contained illite and chlorite occurring together, with kaolinite mostly absent. Kaolinite-bearing assemblages, with chlorite absent, were most frequently recorded from the Crackington Formation, with about 18% of samples belonging to this group. This was in contrast to the Bude and Bideford Formations, where only 6 to 7% of the samples were clearly kaolinite-bearing. Significant mixtures of both kaolinite and chlorite were not identified in this study.

Ilite crystallinity
IC results from the shales and mudstones of the Crackington, Bude and Bideford Formations are presented in Figures 1 to 6. Overall the results show the same range of metamorphic grade as presented by Kelm (1986) and Kelm and Robinson (1989), reflecting middle anchizone to upper diagenetic conditions. In Figure 1 the IC data are presented as averaged values (expressed in Δ°2θ) for particular areas of common stratigraphy. Two IC profiles (A-B and C-D) constructed from the traverse data across the middle and eastern parts of the basin are shown in Figures 2 and 3.

![Figure 2: IC profile along the traverse A-B. CK = Crackington Formation, BD = Bude Formation, BF = Bideford Formation. The dotted lines mark the approximate boundary positions between formations.](image2.png)

![Figure 3: IC profile along the traverse C-D. CK = Crackington Formation, BD = Bude Formation and CT = Crediton Trough.](image3.png)
The northern region
The northern region of the Culm Basin (Figure 1) is taken to include the Crackington, Bideford and Bude Formations, which run along strike through the Bideford area. A continuous decrease in the grade of metamorphism can be traced southwards along the River Torridge from lower anchizonal to upper diagenetic grades (Figure 2). This corresponds with a conformable 1200 m-thick stratigraphic succession (de Raaf et al., 1967; Xu Li 1990) passing from the northernmost occurrence of Crackington Formation (Westward Ho! Formation of de Raaf et al., 1965) into the Bideford Formation. The upper diagenetic values of the Bideford Formation ($X = 0.35 \Delta^20$, $N = 30$) are, on average, the highest IC values recorded in the Culm Basin, indicating the least metamorphosed rocks. This is supported by the lowest range of vitrinite reflectance values recorded by Cornford et al. (1987) of between 1.5 to 2.7% Rm.

Continuing further south along traverse A-B (Figure 2), a sharp change in metamorphic conditions from upper diagenetic to lower anchizonal grades occurs around the boundary between the Bideford Formation ($0.6 \Delta^20$, $N = 30$) and the Bude Formation ($X = 0.42 \Delta^20$, $N = 17$). Due to limited sampling of the narrow outcrops of Bude and Crackington rocks which lie immediately south of the Bideford Formation (Figure 1), the exact position of this change is unclear. However, the most likely place for this jump in grade is along the northern boundary of the narrow outcrop of Crackington Formation, which is marked as a fault on the British Geological Survey 1:50,000 Sheet 292.

Along-strike changes in grade also occur within the northern region. This is evident in the Bude Formation, where lower anchizone values ($X = 0.35 \Delta^20$, $N = 32$) in the area to the south of Bideford, pass laterally into upper diagenetic values ($X = 0.42 \Delta^20$, $N = 17$) in the northeastern part of the basin (Figure 1).

The central region
The central region comprises the Crackington and Bude Formations which run along the strike from the coastline between Bude and Hartland Point. The Crackington Formation in the Hartland coastal area is lower anchizone in grade ($X = 0.31 \Delta^20$, $N = 32$), but there are not enough data to make along-strike comparisons. In contrast, the Bude Formation which occupies the central part of the Culm Basin, is of upper diagenetic grade and shows a decrease in metamorphism from the west ($X = 0.44 \Delta^20$, $N = 37$ in the Bude area) to the east ($X = 0.52 \Delta^20$, $N = 16$ in the area to the north of the Crediton Trough).

The southern region
The southern region is comprised of a 10-to-15 km-wide belt of Crackington Formation rocks which extends from the coastal section south of Bude, through the Okehampton area, to the Exeter area south of the Crediton Trough. The metamorphic grade changes from predominantly lower anchizonal conditions in the west, between Dartmoor and the Atlantic coastline ($0.37 \Delta^20$, $N = 37$), to upper diagenetic grades in the east, around the Exeter area ($0.45 \Delta^20$, $N = 38$). Traverse A-B (Figure 2) shows a clear increase in grade towards the south, which is consistent with the pattern established from the coastal area (see Fig. 3 in Primmer, 1985 or Fig. 4 in Warr and Robinson, 1990). In contrast, in traverse C-D, south of the Crediton Trough, the grade decreases towards the south, despite the proximity to the Dartmoor granite.

Chlorite and kaolinite crystallinity
Graphical plots of IC, ChC and KaC are presented in Figure 4 for the Bideford Formation, and in Figures 5 and 6 for the Bude and Crackington Formations lying north and south of the Crediton Trough. Relationships are shown by plotting the FWHM of the 14Å (chlorite) and 7Å (chlorite or kaolinite) peaks against the FWHM of the 10Å (illite) peak. Overall the regional changes in IC described are not so clearly reflected by changes in ChC and KaC. No clear difference can be seen between the FWHM of the 7Å peaks in the Bideford and Bude Formations, but the Crackington Formation does show a slight decrease in the average 7Å peak-widths. It is also evident that the FWHM of the 14Å and 7Å peaks have similar values which are narrower than the 10Å peaks in all samples. Correlative plots for the Bude and Crackington Formations (Figures 5 and 6) show clustered distributions, in contrast to that of the Bideford Formation (Figure 4) which shows a characteristic spread of IC values but with a restricted range of ChC and KaC values. This probably reflects a larger amount of illite/smectite interlayering in the diagenetic rocks of the Bideford Formation which causes more variability in the peak-widths.

Crystallite (domain) size determinations of illite
Preliminary results from the crystallite size calculations determined by the Warren-Averbach method indicate a good correspondence between IC and the size of the diffracting crystallites (Figure 7), with increasing thicknesses of the crystallite size corresponding with decreasing IC values (i.e. narrowing of the 10Å peak). Both samples show positively skewed asymmetrical populations for the diffracting crystallites.

DISCUSSION AND CONCLUSIONS
Clay mineral crystallinity as an indicator of metamorphism
IC is the most widely used of the crystallinity methods for determining metamorphic grade due to both its relative technical simplicity and because changes in peak width occur over the full range of very low grade conditions (upper diagenesis to
lower greenschist facies). However, utilizing more that one method for grade determination is preferred for petrogenic purposes, and yields more information than using IC alone (Arkai, 1991). The main restrictions in an integrated IC, ChC and KaC approach are the more time consuming identification procedures required to distinguish between kaolinite and chlorite minerals, which have very similar basal peaks reflections. Additional difficulties also arise if significant mixtures of chlorite and kaolinite are encountered, but such cases were not identified in this study.

The main influence affecting crystallinity is considered to be temperature (Kübler, 1967; 1968), although many other factors have been proposed to exert effects (Frey, 1987). Crystallinity is considered to be mostly a function of the crystallite size distribution at anchizonal and epizonal levels of metamorphism (Merriman et al., 1990), whereas in diagenetic rocks, structural disorders (such as swelling) may be of greater significance (Eberl and Srodon, 1988). The crystallite size results from the two samples of upper diagenetic grade (SW1 and SW2) suggest that the changes in crystallinity largely reflect the crystallite size distribution.

There has been much discussion concerning the role of detrital micas on crystallinity results from the Culm. Some of the apparent discrepancies of grade in relation to the stratigraphy, for example the low IC values of the Hartland area, may be due to the influence of detrital muscovites (Kel'm and Robinson, 1989). Kel'm and Robinson (1989) demonstrated that at lower than mid-anchizonal grades, the IC value varies depending on the size of grain fraction analysed. The lower IC values of the coarser grain sizes reflecting the presence of relict detrital grains. In this case the asymmetrical peak geometries of the illite 10Å peak observed in many of the samples most likely reflect two overlapping illite phases; an illite/smectite phase and an illite-smectite phase (Stern et al., 1991). However, attempted deconvolution of these two overlapping phases proved unsuccessful, with single peak fitting of the 10Å peak profile frequently yielding the most reliable results.

Although it is clear that there is a component of detrital mica within the <2µm fraction analysed and that local variations may result from varying amounts of detrital micas, it is suggested that the effects on the regional metamorphic grade was more limited. Firstly, the regional differences are reflected in the IC data, and partly in the ChC and KaC data, with kaolinite being unlikely to be of detrital origin. Secondly, the range of lithologies sampled from each region was consistent, i.e. a mixture of shales and mudstones, most with visible detrital mica, and thirdly, the regional pattern in crystallinity is consistent with variations in the vitrinite reflectance data of Cornford et al. (1987).

**The regional pattern of metamorphism**

In general there is a broad correlation within the basin between the higher grades of metamorphism and the older stratigraphic units. A sedimentary burial type model for the metamorphism therefore appears the most applicable (Cornford et al., 1987; Warr et al., 1991). In detail, however, regional variations in grade are recognised within the same stratigraphic units both across and along strike. The regional increase in the metamorphic grade towards the western part of the Culm Basin may reflect either an increase in the sedimentary burial depth of the rocks, or an increase in the paleogeothermal gradient. The stratigraphic and palaeogeothermal constraints are, however, not yet sufficient to differentiate between these two possibilities.

The temperature estimates made by Cornford et al. (1987) of around 2200°C for the Bideford Formation relate well to the crystallinity data indicating an upper diagenetic grade. However, the reflectance values from the southern part of the Culm Basin (4.7 to 5.18% Rm) are somewhat high for kaolinite and...
illite/smectite-bearing rocks, and are more characteristic of the upper anchizone and epizone levels of metamorphism (Kisch, 1987). Further work is required to resolve these anomalously high reflectance values.

The regional pattern presented in this study has a number of similarities with that of the South Wales Coalfield, where the conditions of metamorphism have been well constrained from coal rank (White, 1991) and clay mineral data (Gill et al., 1977). Firstly the range of metamorphic grades are very similar, ranging from upper diagenesis to anchizonal conditions, and secondly, increases in grade occur both laterally toward the west, as well as with the stratigraphic depth. Additional similarities in their basin histories are that they have similar subsidence curves and preserved stratigraphic thicknesses (Hecht, 1992; Gaye et al., 1992), which supports the suggestion that both the Culm Basin and the South Wales Coalfield are foreland basins, developed in advance of the deforming Variscan orogen (e.g. Hartley and Warr, 1990; Gaye et al., 1992). In this respect the similarities in the pattern of metamorphism between these basins most likely reflects common palaeo-geothermal processes and tectonic controls within the developing foreland. Further elucidation of these patterns is therefore important to enable these underlying controls to be established.

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REFERENCES


