# Development of slaty cleavage in a mudstone unit from the Cantabrian Mountains, northern Spain

R. J. NORRIS

Department of Geology, University of Otago, P.O. Box 56, Dunedin, New Zealand

and

## N. A. RUPKE

Department of Geology, University of Oxford, England

(Received 24 June 1985; accepted in revised form 17 February 1986)

Abstract—A 200 m thick mudstone unit in the Carboniferous of the Cantabrian Mountains, northern Spain, exhibits an increase in intensity of the slaty cleavage from top to bottom which appears to be correlated with a decrease in the mean grain size, average bed thickness and quartz:mica ratio. Anastomosing cleavage domains, formed by pressure solution and by kinking and rotation of the detrital micas, become closer spaced, wider and more continuous towards the finer-grained base of the unit. Growth of strongly oriented new micas within the cleavage domains also appears to be correlated with the intensity of the domain development and hence with the initial lithology. Clay minerals have been completely replaced throughout the mudstone by muscovite, paragonite and pyrophyllite during low-grade metamorphism. The growth of an oriented mica fabric, however, is restricted to samples with well-developed cleavage domains.

## **INTRODUCTION**

THE rocks described in this paper form part of a small deep-sea fan of Carboniferous age in the Cantabrian Mountains of northern Spain (Fig. 1) (Rupke 1975, 1977). During a sedimentological study, a section was measured and samples collected along a more or less continuous road exposure west of Valdeprado (Rupke 1977). A 200 m thick mudstone unit within this section shows a progressive development of slaty cleavage towards its base. This note presents microstructural and mineralogical data on a suite of samples through the mudstone unit in order to document the development of cleavage across an intensity gradient.



Fig. 1. Geological and lithofacies map of the field area showing locality of section studied. The road between Potes and Cervera de Pisuerga is shown by a double line.

## FIELD DESCRIPTION

The section described forms part of a 5-10 km thick succession of Namurian to Westphalian age (Rupke 1977). Along the road section (Fig. 1) the bedding is subvertical with low-dipping cleavage at a high angle to bedding. The mudstone unit has a total thickness along the line of section of 200 m and sampling was carried out at 20 m intervals. The mudstone coarsens appreciably upwards (Fig. 2) from clay to very coarse siltstone in average grain-size. Over most of the unit, however, sedimentary lamination is present with thin silty or fine sandy graded laminae ranging in thickness from less than 2 mm near the base to over 3 cm in average thickness at the top (Fig. 2). Because of these laminations, it is possible to compare microstructures in rocks of roughly equivalent grain-size despite the change in average grain-size through the section.

Quartz and chlorite decrease whereas mica increases in abundance downwards and pyrophyllite appears in the lower 100 m (Fig. 2). Cleavage structure is prominent in the lower half of the section although only the basal 50 m can be considered good slates. It is clear that in a general way the increase in cleavage intensity coincides with a decrease in average grain-size and bed thickness and an increase in the proportion of phyllosilicates. Thin quartz veins subparallel to cleavage are occasionally present in the middle and lower portions of the section.

## PETROGRAPHY AND MICROSTRUCTURE

Detrital quartz grains are abundant in all samples except no. 1 (for stratigraphic location see Fig. 2). Large



Fig. 2. Vertical section through the mudstone unit showing sample locations and variation in mean grain-size, bed thickness and mineral composition [ $\phi$  scale =  $-\log_2$  (diameter in mm)].

(up to  $150 \,\mu$ m) detrital muscovite grains are conspicuous throughout the sequence and have a fairly pronounced preferred orientation parallel to bedding. Chlorite also occurs as detrital grains parallel to bedding in the upper part but towards the base of the mudstone is more difficult to distinguish optically. Matrix phyllosilicates of less than 10  $\mu$ m grain-size occur throughout the section, increasing in abundance downwards.

Apart from the bedding fabric, the most conspicuous microstructure in samples from the top of the section are spaced discontinuous mica films (Williams 1972, Means 1975) or cleavage domains (Borradaile et al. 1982) (Fig. 7a). These tend to wrap around larger detrital quartz grains and are defined by a concentration of opaque material and very fine-grained matrix phyllosilicates along a very thin domain that is generally beyond resolution under the optical microscope. The domains are anastomosing and commonly appear on the limbs of crenulations in the bedding fabric (Figs. 7a & b). Large detrital micas are bent, kinked, broken and rotated into the cleavage domain (Figs. 7a-d and 8b), sometimes splitting and sliding along the basal cleavage plane into thin flakes. The domains are more closely spaced and continuous in the finer-grained horizons, and in the coarsest horizons may be inconspicuous and restricted to the margins of detrital quartz grains. Where a cleavage domain wraps around a quartz grain, the quartz tends to have a straighter truncated margin suggestive of pressure solution (Williams 1972, Elliott 1973, Groshong 1976, Rutter 1983), also indicated by the concentration of opaques along the seams. In some samples at the top of the section, closely-spaced cleavage domains occur in discrete zones separated by wider microlithons.



Fig. 3. Plot of frequency against log (spacing) for cleavage domains in selected samples, measured perpendicular to the cleavage. Other samples measured would have been plotted in intermediate positions and are omitted for clarity. Total number of domains measured per sample vary from 100 to 300. (Sample 1 has little detrital quartz, is finer-grained and has a greater degree of recrystallization compared with the other samples and so was generally omitted from the micro-structural measurements.)

Going down section, the cleavage domains become more closely spaced and continuous and although still anastomosing, exhibit a greater degree of parallelism. On a log-normal plot (Fig. 3), the frequency distribution of domain spacing is close to symmetrical with the mean spacing showing a systematic reduction with decreasing height in the section. This corresponds with the mesoscopic increase in cleavage intensity and the reduction in average grain-size and bed thickness. As the measurements were made on layers having approximately the same quartz grain-size in each sample (that of mediumcoarse silt, 20–30  $\mu$ m), the variation in domain spacing shown relates to the average grain-size of the outcrop rather than to the size of the detrital grains in the specific thin section (cf. Fig. 7).

Domain orientation exhibits an increasing degree of parallelism downwards in the section. The variability in orientation may reasonably be described by a normal distribution curve (Fig. 4). There is some indication of bimodality due to the anastomosing domains in places having a tendency towards a trapezoidal pattern (Borradaile *et al.* 1982).

Not only do the cleavage domains become closer spaced and more parallel towards the base of the section but they also become wider, more diffuse zones that can be resolved optically into films of oriented mica flakes forming an anastomosing network through the sample. The oriented phyllosilicate flakes within the domains increase in size as the domains widen until they are coarser than the original matrix grains. SEM photographs (Figs. 8b-d) clearly show the fine matrix micas bent and rotated into the domains and the development of long thin oriented aggregates within the domains (cf. Weber 1982). Coarser detrital micas also become rotated into the domains as thin flakes. Whereas in the higher samples the detrital micas parallel to bedding are commonly visibly kinked or crenulated at the domain margins (e.g. Fig. 8b), in the lower samples they are often truncated by the cleavage domains.



Fig. 4. Plot of frequency against orientation of cleavage domains for samples 10, 6 and 2. The zero orientation line is parallel to the mean cleavage trace as estimated by eye. The histogram in each case is drawn on a 2° interval. The thinner of the upper two curves for each sample is a 5° moving average of the histogram data, while the thicker curve is a normal distribution curve fitted to the data. All measurements were made in sections perpendicular to the bedding and cleavage. Number of measurements per sample: 250.

Quartz grains within the cleavage domains become lenticular and elongate parallel to the domain boundaries (Figs. 8b-d). The lack of internal deformation features such as deformation bands, deformation lamellae, or recrystallization, together with the preservation of large fluid inclusions and the presence of pressure shadows (Fig. 8a) strongly suggests pressure solution as the principal mode of deformation of the quartz grains (e.g. Durney 1972, Williams 1972, Elliott 1973, Kerrich 1978). With the decrease in spacing and increase in width of the cleavage domains downwards in the sequence, a greater proportion of quartz grains become incorporated. This is clearly demonstrated in Fig. 5 which is a plot of log (axial ratio) against orientation  $(R_f/\phi$  plot of Ramsay 1967, Dunnet 1969) for quartz grains from four representative samples. The quartz grains show a change in orientation pattern from one of weak parallelism with the bedding trace in samples near the top of the sequence to a strong preferred orientation parallel to the cleavage towards the base.



Fig. 5. Plot of log axial ratio  $R_f$  against orientation ( $\phi$ ) for 100 quartz grains from each of samples 8, 6, 5 and 3. The zero orientation line is parallel to the mean cleavage direction determined by eye, and the orientation is measured in degrees either side of this.

Although the range of  $R_f$  values remains approximately constant for all samples, the mode shifts to a higher value with increasing cleavage development (Fig. 6), reflecting a change of shape of the grains. This suggests that grains with low aspect ratios tend to be deformed more easily than more highly elongate grains, a conclusion consistent with current models of pressure solution (cf. Elliott 1973, Lisle 1977, Rutter 1983).

#### MINERALOGY

## X-ray diffraction

X-ray diffraction analysis shows the major mineral phases present over most of the samples to be quartz,



Fig. 6. Histograms showing frequency of  $R_f$  values for samples 8, 6, 5 and 3.

muscovite and chlorite, with quartz and chlorite decreasing and muscovite increasing downwards in the sequence. Peaks corresponding to the mineral pyrophyllite appear in sample 5 and increase in height down section. Smaller peaks, particularly those at 27.9° and 27.5° ( $2\theta$ ) on the flanks of the muscovite 006 peak are consistent with the presence of paragonite and a paragonite-muscovite interlayered mineral (cf. Frey 1969, 1970, 1978). No peaks corresponding to any clay minerals were found. Thus the rocks must be considered as metamorphic as any clays presumably have reacted to form micas or pyrophyllite.

Pyrophyllite is most probably a reaction product of kaolinite + quartz. Experimental investigation of this reaction (Thomspon 1970, Haas & Holdaway 1973, Day 1976) suggests temperatures in excess of about 270°C. The upper stability limit of pyrophyllite is around 350°C at low to moderate pressures (Day 1976). Dunoyer de Segonzac & Millot (1962) (see discussion in Day 1976) have suggested the production of pyrophyllite from feldspar. In the present case however, kaolinite is the more likely precursor since feldspar would be expected to be a detrital phase occurring with the quartz, and so its breakdown products should be more abundant in the upper part of the section. Kaolinite abundance on the other hand would be expected to increase in the clay-rich lower part of the mudstone.

The origin of the paragonite and paragonite-muscovite interlayered mineral may be similar to that proposed by Frey (1978) for these minerals in Upper Triassic and Lower Jurassic rocks from the Western Alps, namely metamorphism of sodium-bearing irregular mixed-layer illite-montmorillonite.

#### Electron microprobe analysis

Analyses of 27 coarser detrital micas were obtained from samples 3 and 5, and 15 strongly oriented domainal



Fig. 9. Plots of probe analyses of micas from samples 5, 3 and 1. Open triangles: large detrital micas; closed squares: oriented fine-grained, newly crystallized micas from cleavage domains. Analyses were carried out on the University of Otago microprobe using standards and operating conditions described in Nakamura & Coombs (1973) and correction procedures of Bence & Albee (1968).

micas from samples 1 and 3, where they were coarse enough for analysis. No significant differences between analyses from the various samples were noted. However, distinct differences in composition between the detrital micas and the recrystallized micas are apparent (Fig. 9). The recrystallized micas show an appreciable sodium content suggesting the presence of a paragonite phase. A scan across a larger area of newly crystallized micas from sample 1, where they are coarsest (Fig. 10), shows a general sympathetic variation in sodium and potassium contents suggesting the intergrowth of a paragonite-rich and a muscovite-rich phase on the scale of a few  $\mu$ m. The detrital micas on the other hand have a much lower sodium content, suggesting that the paragonite and the paragonite-muscovite interlayed mineral identified on the X-ray diffractograms, are located within the matrix mica and the oriented micas within the cleavage domains. The fact that the X-ray peaks for the paragonite phases are present in all samples indicates that the production of paragonite and the paragonitemuscovite interlayer mineral has occurred throughout







Fig. 8. (a) Photomicrograph, sample 3, showing elongate quartz grains with quartz-mica pressure shadows and straight margins. Note broad diffuse cleavage domains with oriented fabric (in extinction) (crossed polarizers); (b) SEM photomicrograph, sample 5. Cleavage is running from top right to bottom left. Note wide zones of phyllosilicates at high angle to the cleavage and narrow cleavage domains with oriented grains. In some places non-domainal micas are bent into domains, in others they are truncated; (c) SEM photomicrograph of sample 3. Cleavage is subvertical. Note large quartz grains with straight sides and cleavage domains with thin, oriented micas; (d) SEM photomicrograph of sample 1, showing well-developed fabric of oriented phyllosilicates but still preserving a fine-scale domainal structure. Prominent quartz grain shows surprisingly little elongation although the smaller one on the left does.



Fig. 10. Microprobe scan across a large area of oriented micas in a cleavage domain, sample 1. Grain boundary shown was prominent break; other boundaries are almost certainly present but were not clearly visible optically. Note sympathetic variation of sodium and potassium. Percentage values for Na<sub>2</sub>O and K<sub>2</sub>O are approximate only, as measurements were not calibrated directly against standards, reduction procedures were not employed, and corrections were not made for variations in specimen current. Scanning speed was 10  $\mu$ m min.<sup>-1</sup>.

the mudstone, irrespective of the stage of development of the cleavage domains, presumably by diagenetic and metamorphic reconstitution of matrix illite and montmorillonite. The presence of paragonite and muscovite as a strongly oriented fabric is therefore due to the recrystallization and enhanced growth of the matrix micas within the domains.

The newly crystallized micas tend to have higher alkali contents, closer to stoichiometric, than the detrital micas, and lower phengite contents. The large detrital micas are more variable in composition, presumably due to their more varied history both in the source rocks and during weathering and diagenesis.

#### DISCUSSION

The development of cleavage in the rocks under discussion is clearly related to the initiation and growth of discrete, spaced cleavage domains. As the cleavage domains are the principal deformational microstructure, their closer spacing, accompanied by increased flattening of detrital quartz grains towards the base of the unit indicates a greater strain on a thin-section scale, probably reflecting a more uniform distribution of the bulk strain. The relationships between the domain spacing and the average grain-size, bed thickness and mica content strongly suggests that the cleavage intensity and strain distribution is a function of the initial bulk lithology.

Processes that are involved in the initial development of the cleavage domains appear to be crenulation, kinking, bending and fracturing of detrital micas, accompanied by pressure solution of quartz. These are essentially the same as reported by other authors (e.g. Gray 1978, Knipe 1981) for similar features.

The development of a strongly oriented mica fabric takes place by the crystallization/recrystallization and

preferred growth of oriented micas within the cleavage domains. Suitably oriented mica nuclei are probably generated by the rotation of diagenetic matrix micas into the cleavage domains where their initially greater strain energy, finer grain-size and greater ease of diffusion within the domain leads to their enhanced growth (Knipe 1981).

The coarsening of the cleavage domains downwards in the section is related to the degree of recrystallization and the growth of the phyllosilicates within them. Since it is unlikely that the base of the mudstone unit was at a significantly higher temperature than the top, the degree of recrystallization within the domains appears to be related to their spacing, continuity and width.

Although difficult to quantify, the amount of mica incorporated into the domains by kinking, bending and sliding (cf. Means 1975) and by passive concentration by pressure solution may be critical to the development of the recrystallized mica fabric. This will be greater in more micaceous lithologies and in samples in which a greater amount of strain is absorbed by the development of domains. In samples with poorly developed domains, growth of an oriented mica fabric has not developed despite the replacement of original clay minerals by higher-grade phyllosilicates. We conclude that, in the rocks studied, the development of cleavage domains by kinking, bending and rotation of micas and by pressure solution is a necessary prerequisite for the development of an oriented, recrystallized mica fabric, and in turn is related to the initial lithology.

Acknowledgements—We would like to thank Tony Reay for advice on the X-ray diffraction work, Yosuke Kawachi for assistance with the microprobe analyses and Julia Williams for help with the SEM. Rob Knipe is thanked for making valuable comments on the manuscript.

#### REFERENCES

- Bence, A. E. & Albee, A. L. 1968. Empirical correction factors for the electron microanalysis of silicates and oxides. J. Geol. 76, 382–403.
  Borradaile, G. J., Bayly, M. B. & Powell, C. McA. 1982. Atlas of
- Deformational and Metamorphic Rock Fabrics. Springer, Berlin.
- Day, H. W. 1976. A working model of some equilibria in the system alumina-silica-water. Am. J. Sci. 276, 1254-1284.
- Dunnet, D. 1969. A technique of finite strain analysis using elliptical particles. *Tectonophysics* 7, 117–135.
- Dunoyer de Segonzac, G. & Millot, G. 1962. Pyrophyllite de diagenèse dans le Devonien inférieur du synclinal de Laval (massif armoricain). C.r. Acad. Sci. Paris 255, 3438–3440.
- Durney, D. W. 1972. Solution-transfer, an important geological deformation mechanism. *Nature*, Lond. 235, 315–317.
- Elliott, D. 1973. Diffusion flow laws in metamorphic rocks. Bull. geol. Soc. Am. 84, 2645–2664.
- Frey, M. 1969. A mixed-layer paragonite/phengite of low-grade metamorphic origin. Contr. Miner. Petrol. 24, 63–65.
- Frey, M. 1970. The step from diagenesis to metamorphism in pelitic rocks during Alpine orogenesis. *Sedimentology* **15**, 261–278.
- Frey, M. 1978. Progressive low-grade metamorphism of a black shale formation, central Swiss Alps, with special reference to pyrophyllite and margarite bearing assemblages. J. Petrology 19, 93–135.
- Gray, D. R. 1978. Cleavages in deformed psammitic rocks from southeastern Australia, their nature and origin. Bull. geol. Soc. Am. 89, 577-590.
- Groshong, R. H. Jr. 1976. Strain and pressure solution in the Martinsburg Slate, Delaware Water Gap, New Jersey. Am. J. Sci. 276, 1131–1146.
- Haas, H. & Holdaway, M. J. 1973. Equilibrium in the system Al<sub>2</sub>O<sub>3</sub>-

 $SiO_2$ -H<sub>2</sub>O involving the stability limits of pyrophyllite and thermodynamic data of pyrophyllite. *Am. J. Sci.* **273**, 449-464.

- Kerrich, R. 1978. An historical review and synthesis of research on pressure solution. Zentbl. Miner. Geol. Palaont. 1, 512-550.
- Knipe, R. J. 1981. The interaction of deformation and metamorphism in slates. *Tectonophysics* **78**, 249–272.
- Lisle, R. J. 1977. Clastic grain shape and orientation in relation to cleavage from the Aberystwyth Grits, Wales. *Tectonophysics* 39, 381-395.
- Means, W. D. 1975. Natural and experimental microstructures in deformed micaceous sandstones. *Bull. geol. Soc. Am.* 86, 1221-1229.
- Nakamura, Y. & Coombs, D. S. 1973. Clinopyroxenes in the Tawhiroko dolerite at Moeraki, northeastern Otago, New Zealand. *Contr. Miner. Petrol.* 42, 213–228.
- Ramsay, J. G. 1967. Folding and Fracturing of Rocks. McGraw-Hill, New York.

- Rupke, N. A. 1975. Deep-sea fan deposition in the Upper Carboniferous of the southern Cantabrian mountains, Spain. International Congress of Sedimentology, Nice, theme 5, 2, 359-367.
- Rupke, N. A. 1977. Growth of an ancient deep-sea fan. J. Geol. 85, 724-744.
- Rutter, E. H. 1983. Pressure solution in nature, theory and experiment. J. geol. Soc. Lond. 140, 725-741.
- Thompson, A. B. 1970. A note on the kaolinite-pyrophyllite equilibrium. Am. J. Sci. 268, 454-458.
- Weber, K. 1982. Microfabric of slates from the Rheinische Schiefergebirge. In: Atlas of Deformational and Metamorphic Rock Fabrics (edited by Borradaile, G. J., Bayly, M. B. & Powell, C. McA.). Springer, Berlin, 136–139.
- Williams, P. F. 1972. Development of metamorphic layering and cleavage in low-grade metamorphic rocks at Bermagui, Australia. Am. J. Sci. 272, 1–47.