Development of cleavage in limestones of a fold-thrust belt in eastern New York

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Abstract—Tectonic cleavage has developed in Lower Devonian limestones of the Hudson Valley Fold-Thrust Belt in New York State. Morphology and distribution of cleavage in these limestones is controlled by the amount of clay-quartz matrix present and by strain. Only limestones with greater than 10% clay-quartz matrix developed widespread cleavage. X-ray diffraction analyses indicate that the clay matrix was altered during deformation; the width of the 001 illite peak in cleaved lime wackestone is less than that in uncleaved lime wackestone from the same stratigraphic level. A minimum of 10% clay is necessary to provide interconnectivity between sites of dissolution and the free-fluid system. Preliminary analyses of bulk chemistry indicate that calcite is removed from the local rock system during cleavage development, supporting proposals that circulation of fluid through the rock plays a major role in the development of cleavage. Textural evidence suggests that both pressure solution and the ree-face dissolution contribute to the removal of ions at grain boundaries. Cross-cutting relations indicate that cleavage was initiated early during the development of the fold-thrust belt, but continued to develop during the late stages of folding.

INTRODUCTION

MANY of the questions posed by early students of tectonic cleavage (e.g. Sharpe 1849, Sorby 1853, 1856, Harker 1886) remain topics of active research today, despite a century of study. Debate continues concerning: (1) the factors that control distribution and morphology of cleavage (e.g. Wanless 1979); (2) the degree of rock alteration during cleavage development (e.g. Gray 1981a); (3) the timing of cleavage formation with respect to other structures in a deformed terrane (e.g. Powell 1974) and (4) the nature of rock-water interaction during cleavage development (e.g. Plessmann 1964, Williams 1972, 1977, Nickelsen 1972, Carannante & Guzzetta 1976, Platt 1976, Engelder *et al.* 1981, Gray 1981a, Borradaile *et al.* 1982).

This paper describes attributes of cleavage in limestones of the Appalachian fold-thrust belt in eastern New York State and reports new results that apply to the above questions. These results were obtained from field structural studies, calcite strain and X-ray diffraction analyses, and plasma-emission spectroscopy.

GEOLOGY OF THE STUDY AREA

The rocks considered in this study are part of a sequence of Lower Devonian shallow-marine limestones exposed near the town of Catskill in the Hudson River Valley (Fig. 1). These limestones, which were deposited



Fig. 1. Tectonic sketch map of the Hudson Valley. FB, Hudson Valley Fold-Thrust Belt; CM, Catskill Mountains; O, parautochthonous Ordovician flysch; TA, Taconic allochthon; K, City of Kingston; C, Town of Catskill. Circle encloses the study area.



Fig. 2. Stratigraphy of the lower Helderberg Group. Devonian limestones are separated from Taconic flysch by an angular unconformity and a thin unit of Silurian limestone. Brick pattern indicates units in which cleavage does not occur, and diagonal shading indicates units in which cleavage does occur. Carbonate-mineral contents (%CO₃) in weight percent of samples from this section are indicated on the right of the figure.

unconformably above Mid-Ordovician (Taconic) flysch, comprise the lower part of the Helderberg Group, and record transgression and regression of an epeiric sea (Rickard 1962, Laporte 1969, Sanders 1969). From the base, the formations are: Manlius Formation (laminated micrite; tidal-flat environment), Coeymans Formation (lime grainstone; beach environment), Kalkberg and New Scotland Formations (lime wackestone with varying proportions of clay and silt; sub-wavebase environment) and Becraft Formation (lime grainstone; beach environment). The aggregate thickness of these formations is less than 100 m, but in this interval there is a wide range of carbonate lithologies with different grain sizes, carbonate-mineral contents (Fig. 2) and detrital fabrics. Conodont coloration (Epstein et al. 1977), vitrinite reflectance (Friedman & Sanders 1982) and fission-track (Lakatos & Miller 1983) studies suggest that these rocks were heated to between 200 and 240°C, but it is not known if the time at which such temperatures were attained corresponds to the time of deformation.

Strata of the study area lie within a narrow (2 km wide) fold-thrust belt that runs the length of the Hudson Valley (Goldring 1943, Chadwick 1944, Babcock 1966, Sanders 1969, Murphy *et al.* 1980, Marshak 1982, 1983a,b). The style of deformation in this belt is similar to that of better-known fold-thrust belts such as occur in the Canadian Rockies or in the Appalachian Valley and Ridge, but the dimensions of structures in the Hudson Valley are smaller. Outcrops near Catskill expose the roof thrust of a duplex zone (cf. Boyer & Elliott 1982), and thus contain examples of fault-intersection zones and fault-bend anticlines.

A particularly useful feature of the Catskill area for the study of cleavage development is a fault-intersection zone in which cleavage intensity of the Kalkberg Formation increases (i.e. spacing between cleavage domains decreases) within a distance of 45 m. This fault-intersection zone is on the northwest limb of the Central Anti-

cline (Figs. 3a & b) along Route 23 just to the southeast of Catskill Creek. On the southeast limb of the fold, the Kalkberg Formation is uncleaved, whereas in the wedge of rock caught between a ramp and a bedding-plane detachment on the northwest limb, cleavage intensity increases progressively from weak to almost slaty (see Engelder & Marshak 1985 for terminology). The exposures at the Central Anticline permit direct comparison of cleaved specimens with uncleaved specimens taken from the same beds. In the fault-bounded wedge. moderate cleavage begins at 4 m above the base of the unit, strong cleavage at 6-7 m, and near-slaty cleavage at 8 m. Beds thicken by 30% in the wedge, apparently as a consequence of cleavage-parallel extension. In our figures, therefore, the stratigraphic levels of samples from the cleaved interval are normalized with respect to those of the uncleaved interval.

CONTROLS ON CLEAVAGE MORPHOLOGY AND DISTRIBUTION

At low metamorphic grade, each carbonate lithology responds differently to stress during deformation (e.g. Wanless 1979). For example, impure limestone, such as the muddy lime wackstone of the Kalkberg and New Scotland Formations, characteristically develops spaced cleavage (Fig. 3c), whereas pure limestone, such as the micrite and grainstone of the Manlius and Becraft Formations, does not develop cleavage even where tightly folded. Pure limestone may, however, contain tectonic stylolites (see Engelder & Marshak 1985). The morphology of cleavage or stylolite domains also is strongly controlled by rock composition. Contrasts in deformational behavior among different carbonate lithologies is a manifestation of how strain is partitioned between two principal processes active in low-grade limestones: rockwater interaction (largely responsible for cleavage) and twin gliding. Similar statements have been made concerning deformation of low-grade quartzites during deformation (Heald 1956, Morris 1981). Cleavage distribution and morphology are also controlled by other parameters including structural position and strain.

Cleavage morphology

The following is a brief description of the attributes of cleavage and tectonic stylolites in the three principal limestone lithologies of the study area: laminated micrite, lime grainstone and lime wackestone.

Tectonic stylolites in micrite. Micrite beds, which occur in the Manlius Formation, are finely laminated, with 1-5 mm thick layers of pure calcite mud alternating with 0.5 mm thick clay-rich seams. Tectonic stylolites in this micrite are disjunctive (Powell 1979) and sutured (Wanless 1979, Marshak & Engelder 1985). In cross-section, individual stylolite domains range in length from 3 to 15 cm, thereby cutting across several bedding laminae, and the teeth are about 0.5 mm in amplitude. It is







Fig. 4. Photomicrographs of cleavage (or stylolite) domains in limestones. Field of view in all photos is 3 mm. (a) Sutured stylolite in micrite (Manlius Formation). Amplitude of teeth exceeds the dimensions of individual grains. (b) Sutured stylolite in grainstone (Becraft Formation). Amplitude of teeth is comparable with grain size. (c) Non-sutured cleavage domains in lime wackestone (Kalkberg Formation). (d) Interaction of cleavage domains with larger grains in a lime wackestone. Note that domains thicken adjacent to the large grain and that all faces of the grain are solution-pitted. Cleavage is parallel to the arrow.



Fig. 7. (a) Photomicrograph of lime grainstone from the Becraft Formation (field of view is 3 mm). Note the abundance of twins. (b) Photomicrograph of lime wackestone from New Scotland Formation (field of view is 3 mm). Note the relative lack of twins and the evidence of solution-pitting. (c) Scanning-electron micrograph of lime wackestone showing interconnectivity of matrix. Scale bar (top right) is 10 μ m. (d) Scanning-electron micrograph of lime grainstone showing lack of matrix and occurrence of cemented grain boundaries. Twin lamellae are emphasized by etching. Scale bar (top right) is 10 μ m.





common for individual stylolite domains to bifurcate so that for short intervals (1 cm), two parallel stylolite segments enclose a small microlithon. At their terminations, the stylolites commonly bend and disappear into the clay-rich bedding laminae, which themselves have a sutured morphology. Examination under an optical microscope indicates that the amplitude of stylolite teeth exceeds the grain size of the rock, that there are small offsets of domains at stylolite-bearing intersections, and that tiny pyrite framboids cluster along the domains (Fig. 4a). Scanning-electron microscopy emphasizes that the boundaries of stylolite domains can be very sharp, and that vein fill occurs in the cores of some domains.

Tectonic stylolites in lime grainstone. The Becraft Formation contains pure, coarse lime grainstone composed of crinoid fragments in calcite cement. Tectonic stylolites in this lime grainstone are disjunctive and sutured. Domains commonly bifurcate and wrap around individual grains or clusters of grains (Fig. 4b), suggesting that the position of teeth on the stylolite surface is controlled by the grain-scale fabric of the rock.

Cleavage in lime wackestone. Lime wackestone of the Kalkberg and New Scotland Formations is composed of calcite fossil fragments in a matrix of clay, fine-grained quartz and calcite mud. Cleavage domains in this rock are non-sutured (Figs. 3c and 4c), and the spacing of domains depends on the proportion of clay in the wackestone and on strain. Variations in spacing that result from contrasts in lithology become less pronounced as strain increases. In zones of weak cleavage, traces of individual domains in cross-section tend to be wavy and are bounded by bedding planes, but in zones of very strong cleavage, domains tend to be more planar and their traces transect bedding planes. Commonly, however, it is impossible to trace an individual domain for more than 15-20 cm, both because domains are arranged in a relay pattern such that where one domain dies out, an adjacent one initiates, and because domains are anastomosing. Veining is uncommon in cleaved units.

Optical microscopy indicates that cleavage domains are composed of an intertwined network of microstylolites (Fig. 4d) and contain an accumulation of clay and quartz. Each microstylolite has a sutured morphology with very small-amplitude teeth, but the aggregate domain has a non-sutured appearance. Where the domain is thick, it is composed of many microstylolites, but at the terminus of the domain, the number of microstylolites decreases and the domain dies out as a horsetail. Constituent microstylolites of cleavage domains cluster more tightly along the margins of large relict calcite grains or along the boundary between zones of differing lithologies (Fig. 4c). Calcite grains in the lime wackestones typically have corroded grain boundaries (cf. Schwander et al. 1981, Gray 1981a, Oldershaw et al. 1982) and generally do not have calcite overgrowths. Though it is common to find calcite grains that are aligned with their long axes (in cross-section) parallel

to cleavage domains, microscopic pitting, typically, is not confined to crystal faces that are parallel to domains.

Microlithons between cleavage domains are lenticular in cross-section because of the anastomosing nature and consequent merging of cleavage domains. Fabric within the microlithons ranges from random to strong (Powell 1979) depending on spacing of the enveloping domains; microlithon fabric, defined by wispy microstylolites, is more pronounced where cleavage is stronger.

Cleavage distribution

Lithologic controls. Wanless (1979) noted that only carbonate rocks with greater than about 10% clayquartz matrix develop cleavage. This qualitative relation is apparent in the Helderberg Group, for cleavage occurs only in limestones with significant clay-quartz matrix (e.g. the Kalkberg and New Scotland Formations; Fig. 2). In order to evaluate the role that the clay-quartz matrix plays in making limestones susceptible to cleavage development, we documented quantitatively how deformation fabrics of naturally deformed limestones vary as a function of the proportion of clay-quartz matrix present in the rock.

Deformation of low-metamorphic-grade limestones of the Hudson Valley occurred primarily by twin gliding (Chapple & Spang 1974) and by rock-water interaction, with the latter process being responsible for cleavage (e.g. Gray 1981a). In practice, it is very difficult to quantify cleavage-related strain, but twinning strain is readily quantifiable (Groshong 1972). If it is assumed that a decrease in twinning strain in the Hudson Valley limestones corresponds to an increase in strain due to other processes, such as rock-water interaction, a plot of twinning strain as a function of clay-quartz matrix present should be roughly the inverse of a plot of cleavagerelated strain as a function of clay-quartz matrix content. Direct measurement of the proportion of clay-quartz matrix is also difficult, so the carbonate-mineral contents of our samples were determined instead, and thus twinning strain was plotted as a function of carbonatemineral content. Measurement of carbonate-mineral content can be done quickly by pulverizing samples and reacting the powder with an excess of hydrochloric acid; the partial pressure of the evolved carbon dioxide is proportional to the carbonate-mineral content.

Strain due to twinning of calcite was measured in a suite of 15 samples collected from a gently dipping 10 m thick section of rock that straddles the Becraft/New Scotland Formation contact, a location at which there are no mesoscopic folds and at which the carbonate-mineral content of the rocks varies gradationally but rapidly. Based on the constant dip and the lack of internal folding, the interval appears mesoscopically to have been deformed homogeneously. The maximum shortening strain of each sample was determined from measurement of twin lamellae by the method of Groshong (1972), and the resulting strain measurements were plotted as a function of carbonate-mineral content.

The results (Fig. 5) indicate that twinning strain is a



Fig. 5. Magnitude of shortening due to twinning measured in rocks from the contact interval of the Becraft and New Scotland Formations plotted as a function of carbonate-mineral content in weight percent. Below 90% carbonate-mineral content, twin strain is less than 1.5% shortening. Above 90%, twin strain is greater than 1.5%, but shortening is variable.

function of carbonate-mineral content. In limestones with less than 90% carbonate-mineral content, shortening by twinning ranged between 0.4 and 1.3%, whereas in rocks with greater than 90% carbonate-mineral content, shortening by twinning ranged between 1.3 and 5.0%. In high-carbonate content limestones, the magnitude of shortening was quite variable, reflecting microscopic variations in strain. These results suggest that partitioning of strain between the processes of twin gliding and rock-water interaction depends on the amount of clay-quartz matrix present, and that a major change in the susceptibility of a limestone to strain by twin gliding occurs at a carbonate-mineral content of 90%. We interpret this result to indicate that 10% clay-quartz matrix must be present in a limestone for it to be susceptible to efficient deformation by rock-water interaction (and cleavage development) and thus, that Wanless' (1979) estimate is quite accurate. Where 10% clay-quartz matrix is present, the rock texture is such that an interconnecting network of matrix is present. Apparently, calcite grains distributed in the clay-quartz matrix of a wackestone were not subjected to sufficiently high deviatoric stresses to cause twinning (cf. Jamison & Spang 1976) as were grains in a tightly cemented grainstone. However, grains distributed in a wackestone were susceptible to deformation by rock-water interaction (Fig. 6). As discussed below, interconnectivity of the clay-quartz matrix provides an important link between sites of dissolution on grain boundaries and the free-fluid



Fig. 6. Synoptic diagram showing difference in behavior of stressed limestones as a function of the proportion of clay-quartz matrix. Upper row shows rock fabrics prior to deformation (arrows indicate compression direction) and lower row shows rock fabrics after deformation. Pure lime grainstone (low clay) deforms primarily by twinning, with formation of only isolated tectonic stylolites. Calcite grains in muddy lime wackestone (high clay) show solution corrosion but are only lightly twinned, and cleavage fabric develops.

system of the rock. When such a link exists, rock becomes susceptible to cleavage development.

The variation of twinning strain as a function of the proportion of clay-quartz matrix present in a limestone is obvious in thin section. Calcite grains in a lime grainstone of the Becraft Formation (Fig. 7a) are heavily twinned and do not have solution-corroded grain boundaries. In contrast, comparably sized calcite grains in lime wackestone of the New Scotland Formation (Fig. 7b) are only lightly twinned and do have solution-corroded grain boundaries. Matrix grains in this wackestone are embayed into the large grains, and an incipient cleavage fabric defined by wispy microstylolites is apparent. The contrast in textures is also visible in scanning-electron micrographs (Figs. 7c & d).

Structural controls. Cleavage intensity in a cleavagesusceptible unit such as the Kalkberg Formation is highly variable, even within the narrow width of the Hudson Valley Fold-Thrust Belt; lithology, though important, is not the only factor to control cleavage distribution. The contrast in cleavage development on opposite limbs of the Central Anticline (Fig. 3a), is a good example of this variation. In general, particularly strong cleavage occurs in fault zones (Fig. 8) and on the NW-overturned forelimbs of anticlines.

Cleavage intensity (defined by spacing of domains) depends on strain (e.g. Alvarez *et al.* 1978). Therefore, the variation in cleavage intensity within a unit, as described above, reflects variations in the magnitude of strain, and the relation between cleavage and structural geometry reflects the relation between strain and structural geometry. Distribution of cleavage may also reflect variations in stress magnitude dependent on structural geometry, for, as discussed by Rutter (1976, 1983), pressure solution, one type of rock-water interaction,



Fig. 9. X-ray diffraction analyses of clays from the Kalkberg Formation. (a) Typical X-ray diffractogram, indicating the presence of illite, chlorite and quartz. (b) Comparison of representative wide and narrow illite peaks, showing the method of measurement. Low peak to the right of the 001 peak is chlorite. (c) Results for measurements of illite-peak widths from cleaved and uncleaved suites. Left-hand chart shows measurements for glycolated samples, right-hand chart shows measurements for non-glycolated specimens. Each point represents the mean of from 4 to 12 runs on samples from a single bed.

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follows a linear viscous flow law. Thus, pressure solution occurs more rapidly (with consequently more intense cleavage) where stresses are higher. The high-strain zones observed in our field area could have been zones of stress concentration during deformation.

LITHOLOGIC CHANGES ACCOMPANYING CLEAVAGE DEVELOPMENT

Changes in clay minerals

As evident from the above discussion, clay-quartz matrix in a limestone plays an important role in the development of cleavage. Clay mineralogy changes during diagenesis of shales (e.g. Dunoyer de Segonzac 1970) and during cleavage development in slates (Bell 1978, Holeywell & Tullis 1975, Siddans 1977, Knipe 1979, 1981, Lee et al. 1983, Stephens et al. 1979, Gray 1981a), but quartz is largely inert (e.g. Gray 1981a). We used the cleavage gradient in the Kalkberg Formation of the Central Anticline to study changes in clay mineralogy accompanying cleavage development in limestone. This gradient occurs over an interval of only 45 m (measured parallel to bedding), so it is reasonable to assume that both cleaved and uncleaved samples have been subjected to the same P-T conditions. Our procedure for clay extraction and examination is presented in the Appendix.

The clay fraction in these limestones is composed of illite and chlorite. The width of the 001 illite peak (sometimes called the 'crystallinity') has been shown to be a sensitive indicator of diagenetic alteration of clays (e.g. Kubler 1968), and thus, we chose to focus on this parameter. Illite-peak width is a complex function of grain size, degree of preferred orientation on the slide, composition, and degree of perfection of the lattice. Peak widths of illite extracted from cleaved limestones of the northwest limb (Fig. 3a) were compared with peak widths of illite extracted from uncleaved limestone from corresponding (normalized) stratigraphic levels of the southeast limb.

The results (Fig. 9) indicate that there is a systematic variation in illite-peak width with stratigraphic level, both in the cleaved and uncleaved suites. This variation probably reflects the proportion of detrital illite in the samples. In addition, the peak width of illite from cleaved samples is slightly less than that of illite from uncleaved samples at the same stratigraphic level. Peak widths of all samples are affected by glycolation, but even after glycolation there remains a slight but noticeable difference between the peak widths of illite from cleaved and uncleaved samples.

The effect of glycolation on peak widths suggests that the illite contains interlayers of smectite. Measurement of the difference between 2θ values for illite 001 and 002 peaks suggests that 1–5% smectite is present in these rocks (cf. Reynolds & Hower 1970). The reduction of peak widths of illite in the cleaved suites, as compared with the uncleaved suites, could be the result of dewatering of residual smectite interlayers and consequent introduction, by ion-exchange reactions, of potassium into the clay structure during cleavage development. Peak narrowing may also result from improvement of the crystalline lattice or from an increase in clay-crystal size during cleavage development. Our data, while they indicate that illite does change during cleavage development in limestone, are not sufficient to distinguish between possible mechanisms that caused this change.

Chemical changes

The contrast in composition between cleavage domains and adjacent microlithons in limestone has been documented by several authors (Gray 1981a, Erslev et al. 1983, Schwander et al. 1981). Pronounced decrease in CaO with proportional increases in SiO₂, K₂O and Al₂O₃ indicates that the concentration of calcite decreases relative to clay and quartz in cleavage domains. It is not clear from these studies, however, whether ions removed from cleavage domains are redistributed in adjacent microlithons or are transported out of the local rock system. Volume-loss strain associated with cleavage (e.g. Wood 1974, Wright & Platt 1982, Engelder 1984) implies that transport of ions out of the local system does take place. Unless all elements are removed in proportion to their original concentration, volume-loss during cleavage formation will affect the bulk chemistry of the whole rock. We compared the bulk chemistry of a suite of limestones from the Kalkberg Formation where there is no cleavage (the southeast limb of the Central Anticline, Fig. 3a) with a suite from the northwest limb where there is strong cleavage, to determine if bulk chemical changes do occur during cleavage development (see Appendix for sample preparation technique).

Measurements from both cleaved and uncleaved suites follow the same trend when plotted as a function of normalized stratigraphic level (Fig. 10a). Moving averages for measurements from the interval of very strong cleavage (above the 7 m stratigraphic level) are shown in Fig. 10(b). The large standard deviation probably reflects the original inhomogeneity of the rock. Because of the large standard deviations, these results should be considered only preliminary and must be confirmed by further work. Figure 10 indicates that there is a decrease in CaO (on the order of 5%, corresponding to about 10% decrease of CaCO₃) and a comparable increase in K₂O, Al₂O₃ and SiO₂, suggesting that calcite was preferentially removed during the development of cleavage and that some of the CaO ions were transported out of the local rock system. Thus, cleavage development does not result simply in a redistribution of ions from domain to microlithon, but in a change in bulk chemistry of the rock.

A plot of CaO vs SrO (Fig. 10c) shows that SrO/CaO ratios for cleaved samples plot in a different field from those of uncleaved samples. This result is different from the result that would be expected if the process of cleavage development involved only removal of calcite.



Calcite contains a higher concentration of strontium than does illite; thus, preferential removal of calcite with respect to illite would result in SrO/CaO ratios that plot on a diagonal line with a positive slope. Figure 10(c) does not show this relation, and thus suggests that strontium was concentrated in the rock during the development of cleavage. Perhaps new strontium-rich carbonate phases, incorporating strontium derived from the underlying Taconic flysch, precipitated in the cleaved rock. These phases, indicative of incongruent solution (cf. Beach 1979), have not yet been identified petrographically.

CHRONOLOGY OF CLEAVAGE DEVELOPMENT

The timing of cleavage development during formation of other structures in the fold-thrust belt can be determined from cross-cutting relations. Cleavage occurs in nearly flat-lying strata of the basal units of the Catskill clastic wedge to the west of the identifiable folds and faults of the Hudson Valley Fold-Thrust belt (cf. Engelder 1979), an observation which suggests that cleavage was initiated very early in the development of the foldthrust belt, perhaps just prior to development of ramps and associated folds. This hypothesis is further supported by the observation that cleavage-fault intersection lineations are generally overprinted (covered) by linear calcite slip fibers that formed on fault surfaces during movement. Such an overprinting relation is not universal; at some localities, cleavage domains cut the slip fibers on faults, indicating that cleavage continued to form subsequent to some fault movement. Cleavage domains commonly bend asymptotically into fault surfaces, and cleavage intensity generally increases adjacent to fault zones (Fig. 8a), relations which suggest that early formed cleavage planes rotated in response to movement on the faults, and that there is an association between fault movement and an increase in cleavagerelated strain. This association indicates synchroneity between fault movement and cleavage development. Formation of intense cleavage in the fault-bounded wedge of the Central Anticline is a manifestation of cleavage-related strain in a fault-intersection zone. Cleavage in the fault-bounded wedge must have formed subsequent to the formation of the wedge geometry.

Map traces of cleavage in the Hudson Valley Fold-Thrust Belt generally parallel the traces of fold axial planes, though locally there is cleavage transection like that described by Gray (1981a) and Stringer (1975).

Fig. 10. Changes in rock chemistry accompanying the development of cleavage. All analyses are on samples from the Kalkberg Formation. The oxide proportions are given in weight per cent. (a) Analyses of four major elements. Each point is the mean of from one to five analyses for samples from the same bed. (b) Moving averages of chemical analyses. [Open circles are uncleaved samples (U), solid circles are cleaved samples (C).] Interval 'a' combines analyses from beds between stratigraphic levels of 7 to 8.5 m. Interval 'b' combines measurements from beds between levels of 8.6 to 9.5 m. Error bars are one standard deviation. (c) Plot of CaO vs SrO for samples of Kalkberg Formation from the Central Anticline. Open circles are uncleaved samples, solid circles are cleaved samples.



Fig. 11. Evolution of cleavage during formation of a fold-thrust belt. Cleavage tracks the advancing toe of the blind thrust, is rotated during detachment faulting and flexural slip, and locally intensifies in certain structural settings. T_1 , T_2 and T_3 are in time sequence.

Cleavage refraction is common on fold limbs; cleavage is oriented at a high angle to bedding in high-carbonate content beds and at a low angle to bedding in lowcarbonate content beds (Fig. 8b). Cleavage therefore fans around folds in high-carbonate content beds. Cleavage refraction is related in part to shear movement parallel to bedding during folding of sequences in which there is ductility contrast between layers (Helmstaedt & Greggs 1980, Treagus 1983). Observation of gross parallelism between cleavage traces and fold axial traces, and of cleavage refraction on fold limbs in the Hudson Valley, imply that timing of cleavage development overlaps with the timing of folding in the fold-thrust belt. This conclusion is anticipated, for the genetic relationship between folds and faults in fold-thrust belts (Suppe 1983) requires that if cleavage formed throughout the period of development of the faults, it also formed throughout the period of development of the folds.

In the steeply dipping to overturned northwest forelimbs of anticlines, it is clear that cleavage formed at a late stage with respect to some fold development. Moderate to strong cleavage, which is oriented subparallel to bedding, is visible on the forelimbs of these folds. This cleavage cannot be traced around the hinge to the backlimb of the fold and, therefore, did not form in response to pre-tectonic stresses related to depositional loading, but must have formed subsequent to the development of the fold. Mitra & Elliott (1980) showed that late cleavages in the fold-thrust belt of the central Appalachians cross-cut early cleavage of the same overall deformation event, but we did not observe such a relation in the Catskill area.

The preceding discussion implies that cleavage developed throughout the evolution of the fold-thrust belt; it initiated prior to movement of the faults, formed synchronously with the folds and faults, and continued to form even during the final stages of deformation in the region (Fig. 11). During its long history of formation, cleavage may change orientation with respect to bedding, perhaps both as a consequence of mechanical rotation of early formed cleavage, which might necessitate shear movement on domains, and by rotation of the beds with respect to far-field stresses, thereby changing the local stress field (Dietrich 1969, Dietrich & Carter 1969). Cleavage domains, once formed, become planes of low shear strength, because they contain concentrations of clay. In some fault zones, imbrication of beds was accommodated by sliding on weak cleavage domains. The imbrication was not a consequence of shortening across cleavage domains oriented obliquely to bedding (cf. Groshong 1975), for bed segments were transported by as much as 15 cm over adjacent segments.

DISCUSSION: THE NATURE OF ROCK-WATER INTERACTION DURING CLEAVAGE DEVELOPMENT

Observations and results reported in this paper have application to understanding processes involved in rockwater interaction during cleavage development and to understanding the role of clay in these processes. In the last decade, pressure solution has been the focus of much study, and has been considered to be the principal type of rock-water interaction responsible for the development of cleavage in low-metamorphic grade rocks (e.g. Rutter 1976, 1983, Alvarez et al. 1976, Groshong 1975, Geiser 1974). Pressure-solution deformation involves three steps: (a) dissolution at stressed grain contacts, (b) diffusion in grain-boundary fluid films toward sites of lower stress and (c) precipitation at lower-stressed grain contacts (e.g. DeBoer 1977). For pressure solution to occur, a fluid film must exist along grain boundaries, and this film must have sufficient shear strength to transmit a deviatoric stress (Weyl 1959, Rutter 1976, 1983). It has been shown that such films can exist (Rutter 1983). Pressure solution is not the only type of rock-water interaction that occurs in rocks. Free-face dissolution (e.g. Weyl 1958, 1959, Berner 1978), by which minerals dissolve in an undersaturated fluid under hydrostatic conditions, may also take place (e.g. Engelder 1982). The observation that solution pitting on a small scale is not restricted to grain surfaces parallel to cleavage domains suggests that pitting does not always occur at locations where stresses are highest, and thus that distribution of dissolution activity is not strictly controlled by variation in stress magnitude. Apparently, some of the dissolution of crystals during cleavage development occurs by free-face dissolution, and thus both pressure solution and free-face dissolution contribute to cleavage development.

Theoretical calculations suggest that slow diffusion rates limit the extent to which dissolved ions can migrate out of the local rock system during deformation (Etheridge *et al.* 1984). The paucity of overgrowths and veins in cleaved units of the Hudson Valley and the change in bulk chemistry of the rocks indicate, however, that ions were removed from the local rock system during deformation. This observation implies that migration of ions does not occur solely by diffusion. Mimran (1977), Engelder (1984) and Etheridge *et al.* (1984) have suggested that circulation of free-fluid results in advective mass transfer of dissolved ions in rocks. Our observations suggest that advective transfer of ions occurs during cleavage development.

The process of cleavage formation may therefore involve three steps: first, dissolution of grains (by either pressure solution or free-face dissolution); second, diffusion of dissolved ions from the grain boundary into the free-fluid system, and third, advective transport of dissolved ions in the free-fluid out of the local rock system. High permeability, required for free-fluid circulation, can be maintained in rocks that have hydrostatic pressures approaching lithostatic pressures even under metamorphic conditions (Etheridge et al. 1984). Cleavage fabrics could develop either as a direct response to pressure solution, in that grains dissolve unequally in response to variations in grain-boundary stresses, or as a result of directed collapse of the rock matrix as the soluble grains are removed by free-face dissolution in a compressive environment ('house-of-cards collapse', to use the analogy of Gray 1981a). This latter process was implicit in a discussion of stylolites by Sorby (1908, p. 225).

Several authors have emphasized the importance of clay in controlling mechanisms of quartzite and limestone deformation (e.g. Heald 1956, Weyl 1959, Rutter 1976, 1983, DeBoer 1977, Wanless 1979, Morris 1981, McEwen 1978). Quartz and calcite dissolve under different pH conditions, so the fact that clay affects the susceptibility of both quartzite and limestone to solution deformation suggests that the role of clay is textural. Weyl (1959) suggested that grains hold water films which act as channel ways (cf. Williams 1972) through which ions diffuse away from the site of dissolution. In part, clay grains may be important because their highly charged surfaces can hold onto fluid films and provide grain contracts access to water even under the stress conditions that cause cleavage development. In light of the role that fluid circulation plays in cleavage development, we suggest that a minimum 10% clay is necessary to permit interconnectivity of dissolution sites at grain boundaries with the free-fluid system. Without this interconnectivity, dissolved ions will not be removed from the local rock system and a cleavage fabric cannot develop. Random variation in distribution of clay in the original rock, causing variations in degree of interconnectivity, may therefore control the location at which cleavage domains initiate.

CONCLUSIONS

(1) Cleavage distribution is strongly controlled by limestone composition; limestones containing greater than 10% clay-quartz matrix develop widespread cleavage, whereas limestones with less than 10% clay-quartz matrix develop isolated tectonic stylolites. This dependence appears to be textural, and may reflect the role that clay plays in providing interconnectivity between the sites of dissolution along grain boundaries and the free-fluid system. Cleavage morphology is also controlled by limestone composition; tectonic stylolites in pure limestones have a sutured morphology, whereas cleavage domains in impure limestones have a non-sutured morphology.

(2) Solution-corrosion of grain boundaries is not restricted to faces of crystals parallel to cleavage domains, suggesting that free-face dissolution plays a role in cleavage development.

(3) Peak width of the illite component of lime wackestones decreases during cleavage formation. This decrease may result from dewatering of residual smectite, increase in crystal size, or improvement of lattice structure.

(4) Bulk chemistry of limestone appears to change during cleavage development, reflecting preferential removal of CaO from the local rock system, perhaps by advective transport in a free fluid. Participation of fluid circulation in cleavage development is also indicated by the lack of veins and grain overgrowths in cleaved units.

(5) Cleavage development occurs throughout the evolution of the fold-thrust belt, resulting in variation of the geometric relations between cleavage and bedding.

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APPENDIX

Clay analysis

The clay fraction from limestone samples was extracted by digesting the limestones in dilute acetic acid buffered to a pH of 5.5. Iron oxide was removed from the product with sodium hydrosulfite, and the final product was saturated with magnesium chloride. We separated the >2 μ m fraction by settling in water, and then made a smear slide of this fraction. The smear was analyzed with an X-ray diffractometer. During the analyses, the goniometer was set to run at $1^{\circ} \min^{-1}$ and the chart at 60 inches h^{-1} . At our machine settings, $1^{\circ} 2\theta = 25.5$ mm of width measured on the chart. A straight sloping baseline was drawn from 5.3 to 10.5° at the base of the chart trace, and the peak width at half height was measured with a micrometer. Our machine settings differ from those of Kubler (1968); thus, our peak widths cannot be directly compared to his crystallinity index.

Chemical analysis

The bulk chemistry of a suite of limestones from the Kalkberg Formation where there is no cleavage (the southeast limb of the Central Anticline) was compared with a suite from the northwest limb where there is cleavage. For each sample, we collected about 0.5 kg of rock, including a representative proportion of both cleavage domains and microlithons. Many duplicate samples were collected from the same stratigraphic level. The samples were washed, dried, and pulverized, but because of the friability of these rocks, it proved impossible to avoid losing some of the tiny fragments during processing. The rock powders were baked overnight at 900°C to decarbonate them and to remove free water. Weight loss during this baking was in the range 15-20%. The resulting powder was mixed with lithium metaborate and melted at 1250°C, and then the molten rock was dissolved in dilute nitric acid. The chemistry of the acid solution was analyzed with a plasma-emission spectrometer. When making our runs, we did not have access to a rock standard with high CaO content. Thus, while comparisons within our sample suite are valid, the absolute numbers cannot be considered to be highly accurate representations of the actual compositions of the rocks.