

## Chlorite–mica aggregates: morphology, orientation, development and bearing on cleavage formation in very-low-grade rocks

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**Abstract**—Chlorite–mica aggregates in slates from northern Spain have been investigated in very-thin thin sections. Specimens from various parts of a fold show different morphologies that are dependent on the operating deformation mechanism. From the limb to the hinge rigid-body rotation becomes less important and folding and intragranular kinking are more common. This is associated with an increasing aspect ratio (width/length) for the aggregates.

Chlorite–mica aggregates were, in part, present before cleavage development and are statistically parallel to the bedding enveloping surface. Variation in degree of distortion of chlorite suggests continuous solution and growth during deformation. Muscovite, on the other hand, is generally more highly deformed, suggesting that it was all early. Splitting of micas along (001)-planes occurs, creating extension sites where chlorite growth can take place. Thus, an early diagenetic/metamorphic formation with mimetic growth of chlorite on detrital micas is favoured for the origin of the aggregates.

The behaviour of the aggregates during deformation provides information on a mechanism for cleavage development. As a result of intragranular kinking, high angle boundaries are created, with parts of the deformed grains at a low angle to the cleavage plane. Mechanical rotation appears to be the important mechanism for cleavage development in these rocks. Modification by recrystallization, solution and growth processes can account for the observed microstructure.

### INTRODUCTION

GREYWACKES of the Lechada formation from the Curavacas and Lechada synclines (Savage 1967, Kaars-Sijpesteijn 1981, van der Pluijm 1981) in the Cantabrian Mountains of northern Spain have been investigated by means of optical microscopy, using thin (<10  $\mu\text{m}$ ), highly polished, thin sections.

The principal constituents of these rocks are quartz, phyllosilicates (muscovite/illite, chlorite), calcite, feldspar, pumpellyite and heavy minerals (epidote, tourmaline, titanite, zircon). This mineral paragenesis indicates very-low-grade metamorphism, following Winkler's division (1979, p. 104). Illite crystallinity measurements confirm this, although caution has to be exercised with this method. Furthermore, the occurrence of mica beards, resulting in a typical texture for this grade (cf. Frey 1970), is consistent with this division.

This paper treats one typical phyllosilicate assemblage, chlorite–mica aggregates, present in most slates. Chlorite–mica aggregates, in the rocks described here, consist of stacked layers of white mica (muscovite) and chlorite which exhibit varying degrees of complexity. They are easily recognized by their relative coarseness compared to the average grain size of the other grains.

Various authors have recognized and discussed chlorite–mica aggregates. Some of these authors and the terminology used by them are listed below: Sorby (1853), Hoepfner (1956, 'Chlorite–Glimmer–Aggregate'), Voll (1960, 'stacks'), Attewell & Taylor (1969,

'mica porphyroblasts'), Williams (1972, 'inter-and overgrowth of chlorite'), Holeywell & Tullis (1975, 'intergrown white mica and chlorite'), Beutner (1978, 'chlorite grains with one or several interlayers of clear mica'), Roy (1978, 'chlorite–muscovite porphyroblasts'), White & Knipe (1978, 'chlorite rich pods'), Weber (1981, 'chlorite–mica aggregates'), Craig *et al.* (1982, 'chlorite mica stacks') and Woodland (1982, 'chlorite–muscovite composite grains').

Three fundamentally different origins for the aggregates can be proposed:

(i) Primary origin (detrital grains). The aggregates are clastic in origin, either (a) modified during weathering and transport (alteration of muscovite to chlorite) or (b) unmodified (e.g. Beutner 1978);

(ii) Primary-and-secondary origin. Mimetic growth of chlorite (with or without muscovite new growth) on detrital micas (nucleus) during a diagenetic or metamorphic stage (e.g. Voll 1960);

(iii) Secondary origin (porphyroblasts). Whole aggregate grows in rock during metamorphism (e.g. Weber 1981).

A more comprehensive description and discussion of these theories is given in Craig *et al.* (1982).

This study examines the morphology, orientation and behaviour of the aggregates during cleavage development. The aggregates were studied in four bedding-cleavage dihedral angles, 0, 30, 50 and 90°. The first three angles represent various positions in the limb of a fold, the fourth describes the hinge zone, for a fold with an axial plane cleavage.

Length and width of the aggregates were measured in thin section and therefore represent minimum values. Only aggregates with lengths of more than 30  $\mu\text{m}$  were considered, the reason for this will be discussed below. All sections were cut perpendicular to the bedding-cleavage intersection.

### GENERAL CLEAVAGE DESCRIPTION

Two generations of tectonic folding have been defined in the study area. The first ( $F_1$ ) shows tight to isoclinal folds with a well-developed axial-plane cleavage ( $S_1$ ). Second generation folds ( $F_2$ ) are related to faulting and are usually present as kinks, but locally folds occur. A cleavage ( $S_2$ ) is parallel to the axial plane of these second generation folds. A strong bedding-parallel fabric is present ('shaly cleavage'). In this paper only the cleavage parallel to the axial planes of  $F_1$  folds will be discussed from samples where little or no  $F_2$  influence was observed.

In outcrop, this cleavage has the appearance of a slaty cleavage (Hobbs *et al.* 1976, p. 222) in the shale-siltstone units; it is a fracture cleavage (Hobbs *et al.* 1976, p. 216) in the more sandy units.

In thin section the cleavage generally is defined by small white micas, opaque minerals, oxides and calcitic material. Needle-shaped minerals, probably rutile, are present locally. The morphology of the cleavage varies from slaty cleavage to domainal cleavage to crenulation cleavage (Hobbs *et al.* 1976, p. 217), depending on the location in the fold and the ratio of quartz and feldspar to mica. On the concave side of a competent layer the slaty and domainal cleavage are predominant; on the convex side crenulation cleavage is more common. Higher ratios of quartz and feldspar to mica are associated with a generally more irregular-shaped cleavage. In the sand and siltstone units most quartz and feldspar grains are elongated parallel to the cleavage, possibly as a result of differential solution, and the grains are often linked by mica-beards.

Pumpellyite and tourmaline growth occurred after the formation of the cleavage. This is demonstrated by their relation to the cleavage lamellae which are overgrown by them. This mineral growth defines a post  $F_1$  metamorphic peak. The presence of the cleavage in felsic dykes indicates that development of the cleavage occurred after lithification of the rock.

### CHLORITE-MICA AGGREGATE DESCRIPTIONS

#### Morphology

Figure 1 shows photomicrographs of the aggregates for various positions in the limb of a fold. As the bedding cleavage dihedral angle increases, the aggregates become more deformed. Where the dihedral angle is low they are roughly rectangular or ellipsoidal with their

longest dimension approximately parallel to the crystallographic (001)-plane of both mica and chlorite and show little deformation (Figs. 1a-d). Where the angle is large they are more deformed and more irregular in shape (Figs. 1e & f). The observed deformation mechanisms are folding and intragranular kinking. In general, folds are less well developed in the aggregate boundary than in internal markers. In Fig. 1 (f) the white mica is folded with a thin layer of chlorite parallel to its (001)-plane. This aggregate is surrounded by more chlorite. The outer chlorite shows little deformation, even in the centre of the aggregate (the short limb of the folded white mica) and is locally at a high angle to both the white mica and the thin chlorite layer. Thus a rectangular shape is maintained even though the nucleus (white mica and chlorite) is folded.

Aggregates in the hinge of a fold show very complex patterns (Figs. 2 and 3). Folding and/or kinking are the most common deformation modes. Internal and external deformation are strong. Large aggregates (up to 120  $\mu\text{m}$ ) show multiple kinking and are usually cross-cut by the cleavage, with an average cleavage spacing of less than 30  $\mu\text{m}$  (e.g. Figs. 2c & d and 3). Often a new crystallographic orientation is present in these kink zones with the (001)-plane of the phyllosilicates at a low angle to the cleavage (Figs. 2a, c & d and Figs. 3b & d).

The chlorite-mica relationship can be relatively simple (e.g. Fig. 2a) or very complex (e.g. Figs. 2e & f and Fig. 3a), largely dependent on the amount of chlorite present and its position in the deformed aggregate. Increasing amounts of chlorite, located in hinges (or short limbs) of the deformed aggregates, usually result in a more complex character. Cross-cutting relationships between chlorite and mica are common (e.g. Figs. 3c & g). We find evidence for splitting of the white micas and infilling of the 'cavities' (extension sites) by chlorite. A good example is shown in Fig. 2 (b), Fig. 3 (h) is a sketch of this micrograph. The thickness of chlorite in the aggregates appears to be related to the angle between the (001)-plane and the cleavage. The larger the angle, the thicker the chlorite overgrowth (Fig. 2b and Figs. 3h & i). The thickness of the white mica, however, remains approximately the same. Thus, there is an increasing thickness of chlorite in a single folded aggregate from the limb to the hinge. This relationship is not as well demonstrated in more complex aggregates (e.g. Fig. 3f), since their deformation history involved various stages which obliterated this pattern.

Many generations of chlorite are present, as can be seen from cross-cutting relationships (Figs. 3a, c & g). It is not possible to correlate stages between individual aggregates, since different growth histories are found in neighbouring aggregates. Chlorite growth appears to have been active throughout the history of formation and deformation of the aggregate.

Another operating mechanism is solution of the aggregates (and single mica grains) at the contact with the cleavage lamellae, resulting in the truncated appearance of the aggregates (Fig. 2e), which is common in many slates (cf. Beutner 1978, Gray 1979).

Chlorite–mica aggregates in very-low-grade rocks

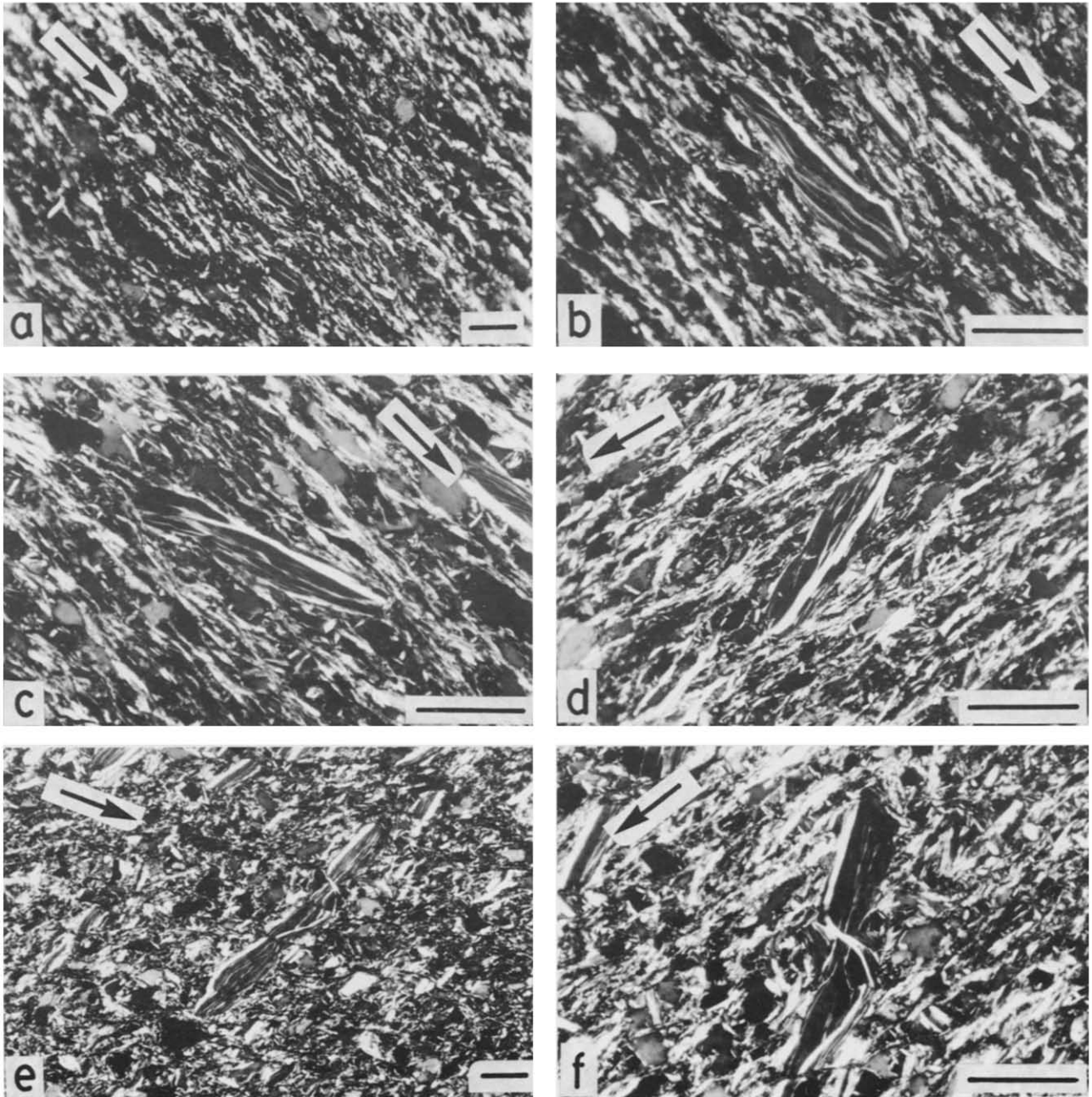


Fig. 1. Micrographs of chlorite–mica aggregates for bedding-cleavage dihedral angles of zero degrees (a & b), thirty degrees (c & d) and fifty degrees (e & f), representing different domains on the limb of a fold with an axial-plane cleavage. See text for discussion. Scale bars are 30  $\mu\text{m}$ ; cleavage orientation is indicated by the arrows.

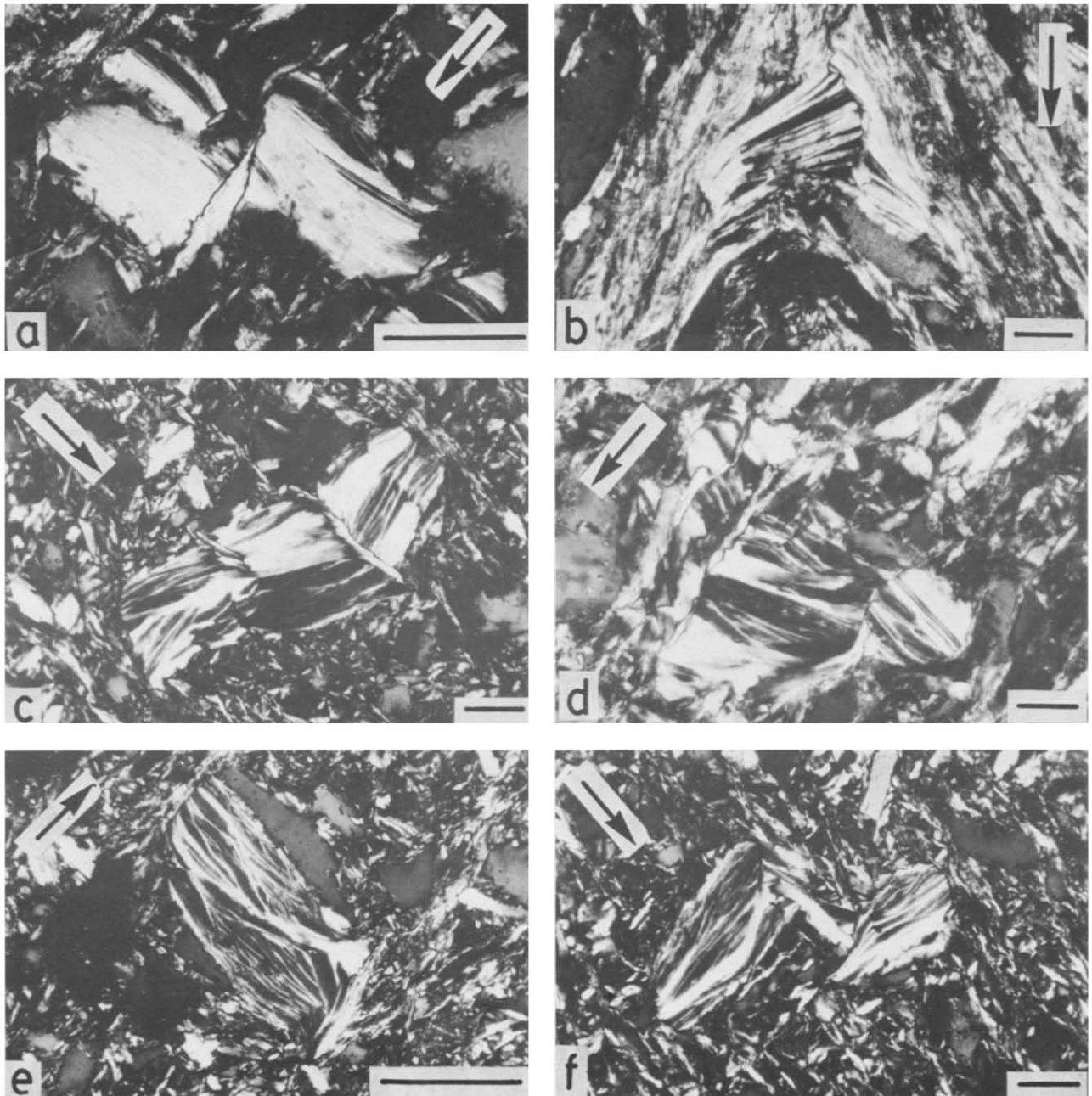


Fig. 2. Micrographs of chlorite-mica aggregates for a bedding-cleavage dihedral angle of ninety degrees, representing the hinge zone of a fold with an axial-plane cleavage. The basal planes of the aggregates are roughly parallel to bedding. See text for discussion. Scale bars are 10  $\mu\text{m}$ ; cleavage orientation is indicated by the arrows.

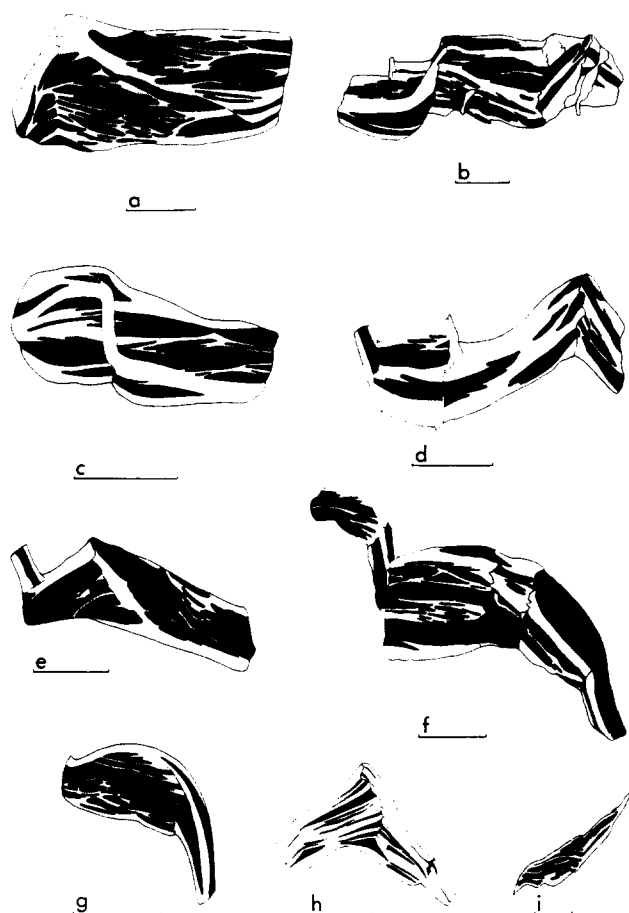


Fig. 3. Sketches of chlorite–mica aggregates drawn from photomicrographs. For (a) to (h) the bedding–cleavage dihedral angle is ninety degrees; for (i) it is fifty degrees. Scale bars are  $15\ \mu\text{m}$  and indicate the approximate bedding orientation. Cleavage is parallel to the long edge of page except in (i) where it runs from upper right to lower left. See text for discussion.

The relationship between aggregates and cleavage, where the cleavage lamellae are seen anastomosing around the aggregates or cross-cutting them, demonstrates a pre- and/or early syn-cleavage origin.

In summary, we find an increase in the folding of the aggregates, both internally and externally, from limb to hinge, resulting in the presence of the most complex aggregates in the hinge zone of a fold.

#### Orientation

In Fig. 4, four orientation histograms of chlorite–mica aggregate basal planes are given; (a), (b) and (c) representing the limb and (d) the hinge of a fold. In the limb we find that, as the dihedral angle between bedding and cleavage increases, so does the asymmetry of the distribution. Where cleavage and bedding are parallel (Fig. 4a), the distribution is normal with a large maximum (51%) within  $5^\circ$  of bedding/cleavage. Where bedding and cleavage are not parallel (Figs. 4b & c), the aggregates are only rarely parallel to the cleavage and the distributions are skewed. This pattern can be explained by rotation of the aggregates, from an initial bedding-parallel orientation, toward the cleavage plane during deformation. Some aggregates show large inclinations

(up to  $90^\circ$ ) to the cleavage. This is explained in terms of folded aggregates with the short limbs (or hinges) at a high angle to the cleavage, the long limbs are oriented nearly parallel to it, similar to parasitic folds. This is illustrated in Fig. 1 (f). The asymmetry of the aggregate is similar to the vergence of parasitic folds on this limb of the fold. In some cases the long limbs are removed by shearing, solution, recrystallization or a combination of them and only the short limbs remain.

In the hinge zone (Fig. 4d) we find most aggregates at an angle to both bedding enveloping surface and cleavage, due to intense folding of the aggregates. A small percentage (5%) is parallel to the cleavage, this fraction represents the limbs of isoclinally folded aggregates. This distribution is symmetrical around the bedding orientation.

Concluding, we find that a decreasing percentage of the aggregates is parallel to the bedding enveloping surface in going from the limb toward the hinge.

#### Aspect ratios

In addition to the greater complexity and increasing folding (both internal and external) of the aggregates from limb to hinge, we also find a change in aspect ratios (cf. Attewell & Taylor 1969). The aspect ratio is defined as the apparent width over the length, as measured in thin section.

In Fig. 5, the length and width of aggregates are plotted for specimens with a dihedral angle between bedding and cleavage of 0 and  $90^\circ$ , representing, respectively, the limb and the hinge of a fold.

On the limb there is a strong linear relationship between width and length (correlation coefficient,  $r = 0.91$ ) with an average aspect ratio of  $0.23 (\pm 0.004)$ . The hinge shows a slightly wider scatter of measurements and a weaker linear relationship ( $r = 0.70$ ); the average aspect ratio is  $0.57 (\pm 0.02)$ . There is no evidence for a variation in length of the aggregates prior to cleavage development. Measurements in various samples show that the length ranges from 10 to  $130\ \mu\text{m}$ , the largest percentage with a length of  $40\ \mu\text{m}$  for both limb and hinge. Therefore, variation in width is responsible for the variation in ratio. Primary thickness variation of the detrital micas has not been observed and we conclude therefore that the chlorite thickness is responsible for the variation. In the hinge zone the chlorite layers are thicker than on the limb. The same relationship has already been described for a single aggregate (Fig. 2b), demonstrating that the chlorite variation is present on different scales.

The thickness variation must be a result of preferential growth in the fold hinges if it is assumed that chlorite was not present in the original detrital aggregates. Alternatively, if it is assumed that both chlorite and muscovite are of diagenetic origin, then the variation must have developed by preferential removal from the limbs. A combination of both growth and solution at some stage in the aggregate development is probable.

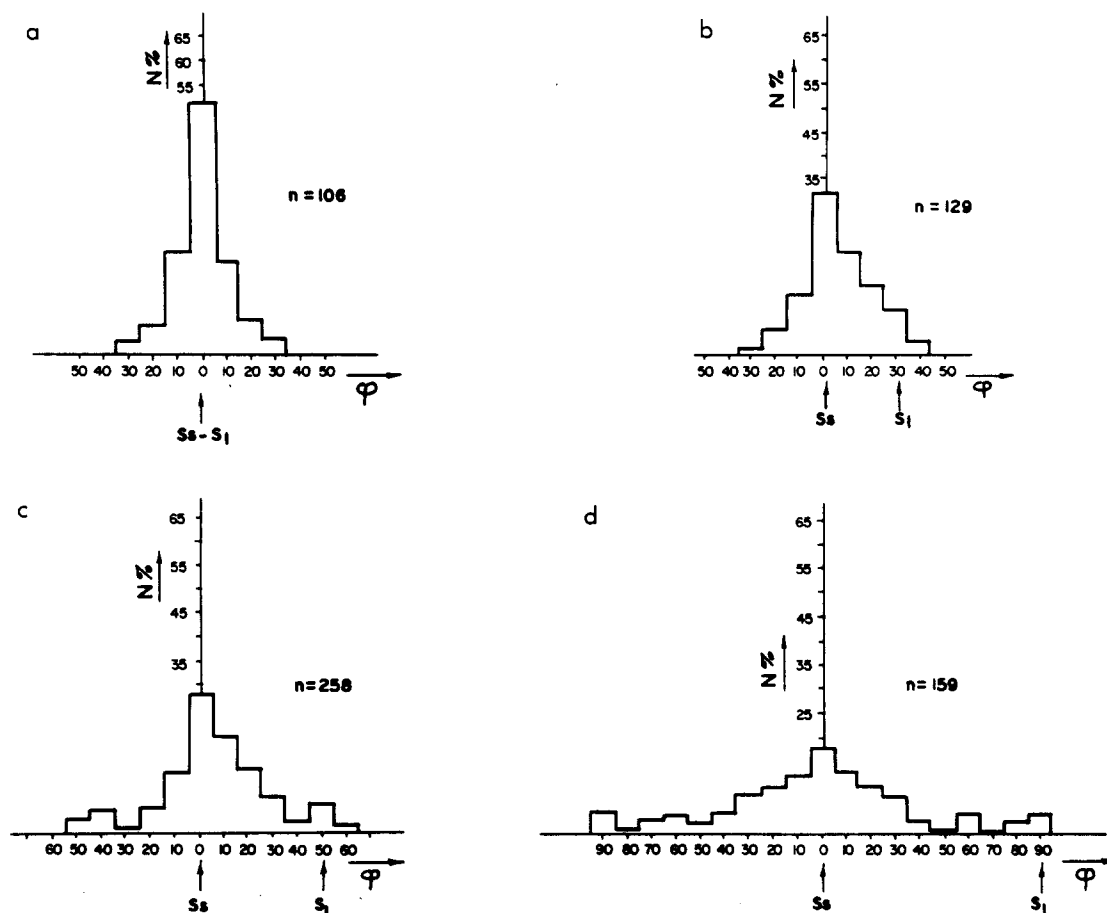


Fig. 4. Orientations of chlorite-mica aggregates (001)-planes, for bedding ( $S_s$ )-cleavage ( $S_1$ ) dihedral angles of zero degrees (a), thirty degrees (b), fifty degrees (c) and ninety degrees (d). 'n' is total number of measurements,  $N$  is percentage.  $\psi$  is angle with bedding. Note the skewness of the total distribution for (b) and (c) toward the cleavage orientation.

## DISCUSSION AND CONCLUSIONS

From the descriptions of the aggregates and their relationship with the cleavage it follows that they were present before, or early in the development of the cleavage, since both anastomosing and cross-cutting cleavage lamellae around and through the aggregates are found. Whether the chlorite in the aggregates origi-

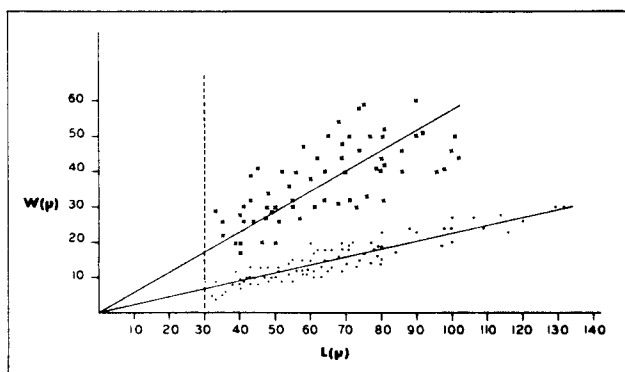


Fig. 5. Relationship between length ( $L$ ) and width ( $W$ ) for 64 aggregates in the hinge (crosses) and 87 aggregates on the limb (dots) of a fold, with a minimum length of  $30 \mu\text{m}$ . Two discrete areas can be recognized and their relationships are shown by linear regression lines:  $Y(\text{hinge}) = 0.57(\pm 0.02)X$ .  $Y(\text{limb}) = 0.23(\pm 0.004)X$ . There is a slightly wider scatter of points for aggregates in the hinge zone than for aggregates on the limb. This is reflected in the correlation coefficients ( $r(\text{hinge}) = 0.70$ ,  $r(\text{limb}) = 0.91$ ).

nated in the early diagenetic/metamorphic stages or as a result of weathering (and transport) can not be concluded unequivocally from this work. In fresh samples of unconsolidated silty clay similar-looking aggregates are found at a depth of 7.45 m (P. Smart, pers. comm. 1982), which suggests a pre-deformation origin. Our descriptions show a relationship between chlorite layers and deformation indicating a pre- and/or syn-deformational history for the chlorite. The morphology and the general parallelism with bedding indicates a sedimentary origin for the white micas (cf. Williams 1972). The detrital white micas are generally more deformed than chlorite, indicating that the chlorite is mostly younger than the white mica. From the morphology and distribution of the aggregates (cf. Craig *et al.* 1982), we favour a primary-and-secondary origin for the chlorite-mica aggregates.

From the orientation histograms of Fig. 4 it can be concluded that a bedding-parallel fabric, defined by micas, was present before the onset of deformation. In all four fold domains we find a fraction of the aggregates oriented parallel to local bedding, reflecting the orientation prior to shortening and rotation. This is not only observed for the chlorite-mica aggregates, but even more apparent from the many large ( $>15 \mu\text{m}$ ), detrital, white mica grains. Bedding-parallel fabrics in shale have been documented by many workers (e.g. O'Brien 1970, Moon 1972 and Curtis *et al.* 1980). Bedding-parallel

fabrics in slates are also commonly described (Williams 1972, Etheridge & Lee 1975, Holywell & Tullis 1975, Knipe & White 1977, Beutner 1978, Maltman 1981 and others). With increasing intersecting angle between bedding and cleavage the aggregates become increasingly deformed, both internally and externally. This shows that the importance of folding and kinking increases relative to rigid body rotation from the limb to the hinge of a fold. The change in deformation mechanism and symmetry is consistent with a non-coaxial deformation history on the limb (responsible for the rigid body rotation) and a coaxial deformation history (developing symmetrically folded aggregates) in the hinges, proposed for fold development (Williams 1976, Knipe & White 1977). Woodland (1982) in a study on chlorite-mica aggregates in the Martinsburg formation, concludes that mechanical rotation was of little importance during foliation development, but also notes that in some specimens (Lehigh Gap) rotation of aggregates toward the cleavage did occur.

The observed variation in aspect ratio has been explained by shortening with addition and/or subtraction of chlorite. There is also a relationship between chlorite and folding in individual aggregates, demonstrated by:

(1) larger chlorite thickness in the more deformed samples;

(2) the relation between chlorite and white mica, where the chlorite and white mica show cross-cutting relationships, indicates that chlorite growth must have taken place at least after initial deformation of the white mica (with or without the presence of early chlorite);

(3) white mica is usually more deformed than chlorite. Chlorite exhibits a complex range in deformation intensity;

(4) the variation in chlorite thickness in a fold is also present on the scale of a single aggregate, illustrated in Figs. 2b and 3h. From limb to hinge of the aggregate, we find an increasing chlorite thickness, while the total thickness of white mica remains about the same.

From (4) we conclude that splitting of white mica along its (001)-planes occurs. Extension sites are created where the (001)-plane is at a low angle to the shortening direction. These extension sites are subsequently filled with chlorite (cf. White & Knipe 1978, Knipe 1981). This mechanism was rejected by Craig *et al.* (1982), largely because they did not observe a relationship between orientation of the aggregates and chlorite thickness. They proposed that the aggregates were produced by pre-tectonic replacement of clay minerals during burial. Although we do not exclude this as a possible origin for the aggregates (see above), we do find a relationship between aggregate morphology and chlorite thickness and deformation in these rocks.

The presence of mica, with its basal planes at a high angle to the cleavage and thus at a high angle to the extension direction, is also favourable to the replacement of muscovite by chlorite, since chlorite occupies a greater volume than mica. The reaction muscovite-chlorite involves addition of Mg, Fe and H<sub>2</sub>O with release of K (Cosgrove 1976). Furthermore, in some aggregates

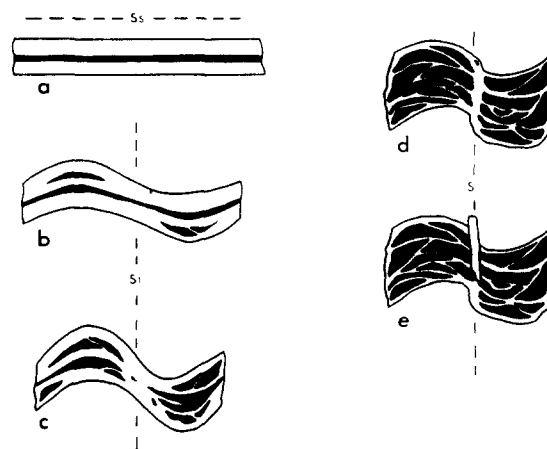


Fig. 6. Evolution of a chlorite-mica aggregate during cleavage development. An initial stacking of mica and chlorite (a) is deformed by (001)-plane parallel shortening. Extension sites arise from splitting along the basal planes and they are filled with chlorite. There is contemporaneous solution of chlorite at compressive sites (b & c). Continued shortening with growth and solution results in the complex aggregate of (d). In kink zones of the aggregate a new orientation develops (e).

quartz is present, possibly new growth also related to extension.

We suggest the following history for the formation of chlorite-mica aggregates (Fig. 6). Assuming an aggregate with a layer of chlorite present before cleavage development (Fig. 6a) and (001)-plane parallel shortening, folding of the aggregate will create extension sites in the hinges by splitting along the mica basal planes and subsequent infilling with chlorite (Figs. 6b & c). Solution of chlorite in compression sites could be a contemporaneous process. Continued shortening and continuous chlorite growth result in a complex aggregate (Fig. 6d). The formation of high angle boundaries due to intragranular kinking of the white mica and growth results in a new orientation, approximately parallel to the cleavage (Fig. 6e); compare, for example, Fig. 6(e) with Figs. 2(c) & (d) and 3(d). Recent work by one of us (v.d.P.) indicates that the occurrence of the morphologies shown in Fig. 6(d) or (e) can be dependent on the metamorphic grade of the rock. Chlorite-mica aggregates in low-grade rocks (reaction epidote-clinozoisite occurred) generally have the morphology shown in Fig. 6(e) with small aspect ratios. It is noteworthy that the same configuration (Fig. 6e) can also originate from a detrital mica without initial chlorite being present before the onset of deformation. Folding of the white mica will create extension sites, as described above, filled with chlorite. Since no pre-deformational chlorite is present, solution of chlorite in the initial stages will be of lesser importance. In the later stages of the development the same morphology can be expected, since both growth and solution of chlorite can occur. The relatively simple chlorite-mica aggregates in Figs. 1(a) & (b), however, may indicate the presence of pre-deformational chlorite (cf. Craig *et al.* 1982). From this model it follows that chlorite growth and solution will take place throughout the development of the aggregates,

probably starting in the earliest stages of diagenesis and continuing after cleavage formation.

Material transfer from the aggregates takes place at the contacts with the cleavage planes (cf. Beutner 1978). Therefore, with a cleavage spacing not exceeding 30  $\mu\text{m}$ , in order to minimize the influence of solution on the ratios, only aggregates with a length greater than 30  $\mu\text{m}$  were considered. Further, smaller aggregates approach the grain size of quartz and feldspar grains and may therefore behave differently from the larger aggregates. Measurements on small aggregates resulted in aberrant dimensions (aspect ratios much greater than 1) and their orientations showed a much wider distribution compared to the samples with a length greater than 30  $\mu\text{m}$ .

The development of chlorite–mica aggregates in the hinge zone, where folding and intragranular kinking occurs, may provide information about the development of an axial-plane foliation. The features described in the aggregates can also be recognized in the smaller mica grains, but the large grain size of the chlorite–mica aggregates allows more detailed observations.

Formation of kink zones is very common (e.g. Fig. 2). Often a narrow zone defined by two high angle boundaries, with the (001)-plane of the intervening mica at a low angle to the cleavage plane is present, the zone usually coincides with a cleavage lamellae outside the aggregate (Figs. 2c & d). Some micas in this zone can be traced outside the aggregate, resulting in a nearly cleavage-parallel orientation for micas, which is further enhanced by growth of the mica parallel to the zone and the cleavage (Figs. 2 and 3).

Etheridge & Hobbs (1974) described microstructures from experimentally and naturally deformed micas. They suggest that nucleation and growth in kink zones may play a role in the reorientation of micas. Williams *et al.* (1977) proposed models for the development of axial plane foliations largely based on the formation of kink zones in micas. They demonstrated this from deformation experiments on salt–mica samples. Their observations in greenschist and amphibolite facies schists (low-grade and medium-grade rocks) possibly confirm this. Two aspects of the models are discussed:

- (1) formation of relatively strain free micas with the same orientation as the more undeformed micas in the kinks by kink band boundary migration and
- (2) nucleation and new growth of micas parallel to the kink planes (and to the pre-existing anisotropy defined by the kinked foliation).

Our observations are in accordance with the first model, evidence for the second is also observed and will be discussed in a future paper. Knipe (1981) also favours the explanation of early mechanical rotation for the presence of grains in the hinges of crenulations over recrystallization in very-low-grade slates. Thus, in very-low-grade rocks mechanical rotation of bedding parallel micas can result in a (nearly) cleavage-parallel preferred mica orientation by the creation of kink zones with the (001)-plane of the micas at a low angle to the cleavage and subsequent growth.

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