

## Crenulation cleavage differentiation: implications of solution-deposition processes

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**Abstract**—Metamorphic differentiation associated with cleavage development in crenulated anisotropic rock fabrics is commonly due to a redistribution of mineral phases within a certain volume of the fabric. Such a volume redistribution can be explained by solution transfer of soluble minerals from sites of high chemical potential, fold limbs, to sites of low chemical potential, fold hinges. The processes involved are dissolution, diffusional transfer via grain-boundaries and redeposition. The driving force for the diffusion, differences in chemical potential, is related to stress and fabric variations around the microfolds. The rate of transfer is influenced by the *initial solubility* of the mineral grains, the *kinetics of grain-boundary diffusion*, the nature of *grain contacts* and the *microfold wavelength*. The sense of migration of dissolved species is dependent on spatial variations in the magnitudes of *normal stress* and *mean stress* combined with *grain shape* and *grain orientation* changes around the microfolds.

### INTRODUCTION

CLEAVAGE development in crenulated metamorphic fabrics is due to a microfold-controlled differentiation process. The differentiation has been attributed to either: (a) a mass transfer-controlled redistribution, involving solution and deposition, of the pre-existing mineral components (Nicholson 1966, Roberts 1966, Ramsay 1967, Cosgrove 1972, 1976, Durney 1972a, Williams 1972, Trouw 1973, Gray & Durney 1976, Gray 1976, 1977, 1979); or (b) an introduction of micaceous components into the crenulated rock fabric (Rast 1966, Marlow & Etheridge 1977).

We suggest that mechanisms (a) and (b) are both responsible for crenulation fabric development, although one dominates (i.e. provides a faster strain rate) the other depending on the physical conditions at the time of the crenulation deformation. It is well known that during tectonic deformation the operative deformation mechanisms depend on mineralogy and grain size of the material and the prevailing physical conditions (Ashby 1972, Stocker & Ashby 1973, Atkinson 1976, Kerrich *et al.* 1977). Mechanism (b) accompanied by metamorphic grain-growth and recrystallisation presumably, dominates in a high temperature, high stress regime where dislocation flow mechanisms are active, for example at temperatures over 400°C). Mechanism (a) appears to dominate crenulation cleavage development in low temperature regimes (i.e. temperatures below 400°C). It is also favoured by low stress and low strain rates (Ashby 1972, Stocker & Ashby 1973, Rutter 1976). Because both mechanisms produce distinctly different microstructural features and relationships of crenulation cleavages (Marlow & Etheridge 1977, Gray 1979), the cleavage microstructure may be used to infer mode of fabric origin and approximate physical conditions at the time of origin.

In this paper however, we discuss some of the aspects and implications of a solution-deposition mass-transfer

process in terms of crenulation cleavage development. We maintain that an understanding of the development of crenulation cleavages in low to medium grade metamorphic rocks, that is the low temperature deformation regime, requires a recognition of the factors which affect differentiation, and an awareness of the implications of solution-deposition processes.

### SOLUTION - DEPOSITION PROCESSES

The combined processes of dissolution (pressure solution), diffusional transfer and crystallisation (free growth or deposition) of certain mineral phases in a rock undergoing deformation produce structural and chemical rearrangements such as changes in grain shapes and mineral proportions (Durney 1972b, Elliott 1973). In crenulated fabrics these fabric changes are spatially related to the microfolds (Cosgrove 1972, 1976, Gray 1976, 1979). It is implicit therefore that both microfolding, generally internal buckling, and solution-deposition processes, in that order, are necessary for crenulation cleavage development.

Solution and deposition, as discussed in this paper are considered part of a diffusive mass-transport process which involves diffusion via a grain boundary fluid-phase (i.e. grain boundary diffusion). This is in contrast to intragranular (also referred to as volume, lattice or self) diffusion (e.g. Stephansson & Berner 1971), free surface diffusion and pore fluid diffusion (Fletcher 1977). The grain boundary fluid is considered to be a dispersed phase of impurity ions trapped in the interatomic spaces, lattice vacancies, holes and channels which occur in the disordered zone between two adjacent crystals; that is along the grain boundary. This phase may not have all the properties of a free fluid, but we assume that the stress in this phase is approximately equal to the fluid hydrostatic pressure ( $p$ ).

Solution is considered to occur on surfaces, generally either or both grain and layer boundaries, which are

statistically perpendicular to an axis ( $\sigma_x$ ) of tectonic compression. Migration of dissolved mineral phases is in response to chemical potential gradients which occur in differentially stressed geologic materials. The mechanics of such a diffusive mass transport process can be explained by non-hydrostatic crystal-solution thermodynamic theory. A brief discussion of this theory follows, but a more complete treatment is given by Durney (1972a, 1976, 1978) and Paterson (1973).

#### *An explanation from non-hydrostatic thermodynamics*

Consider the case of reversible equilibrium (i.e. equilibrium of Gibb's free energy) between a stressed crystal and a solution. If we assume that: (1) the crystal and its solute are one component and the solvent is a second component; (2) the crystal is not a solid solution and is approximately incompressible; and (3) temperature is constant (i.e.  $d = 0$ ), then equilibrium at any part of the grain boundary, following Gibbs (1878, equation 411), is given by:

$$\left(\frac{\partial\mu}{\partial x}\right)_{T,p} dx \approx v_1 d\sigma - v_2 dp \quad (1)$$

[equation (8), Durney 1976]

where  $\mu$  = partial molar chemical potential,  $x$  = mole fraction of solute in solution,  $T$  = absolute temperature,  $p$  = fluid pressure,  $v_1$  = molar volume of crystal phase,  $v_2$  = partial molar volume of solute in solution, and  $\sigma$  = normal stress. Since the differential fluid pressures then  $p$  can be treated as constant and uniform (i.e.  $dp = 0$ ). Equation (1) implies that both the chemical potential ( $\mu$ ) and solute concentration ( $x$ ), and therefore the solubility of a stressed mineral phase at any point along its boundary is directly related to the magnitude of the normal stress ( $\sigma$ ) at that point. Similar relationships have been used by Green (1970), Elliott (1973), Paterson (1973) and Weyl (1959). It is such normal stress related solubility variations around mineral grains which give rise to diffusive fluxes along their grain boundaries accompanied by either or both dissolution and growth. In the case of individual grains in a differentially stressed mineral aggregate, diffusive flux will be in the direction of normal stress decrease, causing dissolution at grain contacts under high normal stress and growth at contacts under normal stress.

The criteria for dissolution and growth at a crystal solution interface, assuming both  $T$  and  $p$  are constant, is given by:

$$RT \ln\left(\frac{a_e}{a_0}\right)_{T,p} \approx v_0(\sigma - \sigma_0) \quad (2)$$

[equation (10), Durney 1976]

where  $R$  = universal gas constant,  $a_e$  = local equilibrium activity,  $a_0$  = reference equilibrium at  $\sigma_0$ ,  $p$ , and  $T$ ,  $v_0$  = crystal molar volume at  $\sigma_0$ ,  $p$ , and  $T$ , and  $\sigma_0$  = reference normal stress. Equation (2) shows that the activity ratio

( $a_e/a_0$ ) is largely an exponential function of the normal stress. If the local activity is 'a' then:

1. where  $a = a_e$ , the solution is perfectly saturated with respect to the solid (i.e. local equilibrium).
2. where  $a < a_e$ , the solution is undersaturated (i.e. local pressure solution).
3. where  $a > a_e$ , the solution is supersaturated (i.e. deposition or growth).

The theory shows that any aggregate of stressed crystals, for example a stressed mineral fabric, is in a state of potential thermodynamic disequilibrium. Variations in normal stress ( $\sigma$ ) through this fabric can cause continuous dissolution, migration and growth such that either or both shape changes of crystals and a complete mineral redistribution may occur. It is also important to realise that these solution-deposition effects may occur at low differential stresses; lower than those required for dislocation flow (Rutter 1976).

#### DIFFERENTIATION OF CRENULATIONS

In crenulated fabrics the cleavage producing differentiation is intimately associated with the micro-folding. The cleavages coincide with the limbs, or former limbs, of the microfolds, and the degree of differentiation is clearly dependent on the tightness of the microfolds (Cosgrove 1976, Gray 1976, 1979). Crenulation cleavage development due to solution-deposition processes appears to involve solution of the more soluble minerals along the fold limbs concomitant with transfer of dissolved phases to the fold hinges, where they are deposited, presumably as overgrowths on existing grains and/or layers. This results in a complete mineral redistribution across the microfolds, associated with the development of a new layering parallel and coincident with their former limbs. A discussion of crenulation cleavage microstructure and textural evidence of solution-deposition processes is presented elsewhere (see Gray in press).

The problem which we would like to discuss here is how and why solution-deposition processes produce cleavages in crenulated fabrics at low to medium grades of metamorphism. In the previous section it was shown that it is normal stress-related solubility variations around mineral grains which cause dissolution and growth, and give rise to diffusive fluxes along their grain boundaries. We suggest that a normal stress controlled variation in chemical potential ( $\mu$ ) across microfolds in stressed anisotropic fabrics is primarily responsible for solution-deposition type differentiation. Stress heterogeneity, and therefore variations in normal stress, occurs both at the scale of the grains (Raj & Ashby 1971) and at the scale of the microfolds (Dieterich & Carter 1969, Stephansson & Berner 1971) such that differentiation involving mass transfer will take place both around individual grains and across the microfolds. However, it is the transfer of dissolved species from the limbs to the hinges of microfolds in crenulated fabrics which produces crenulation cleavages.

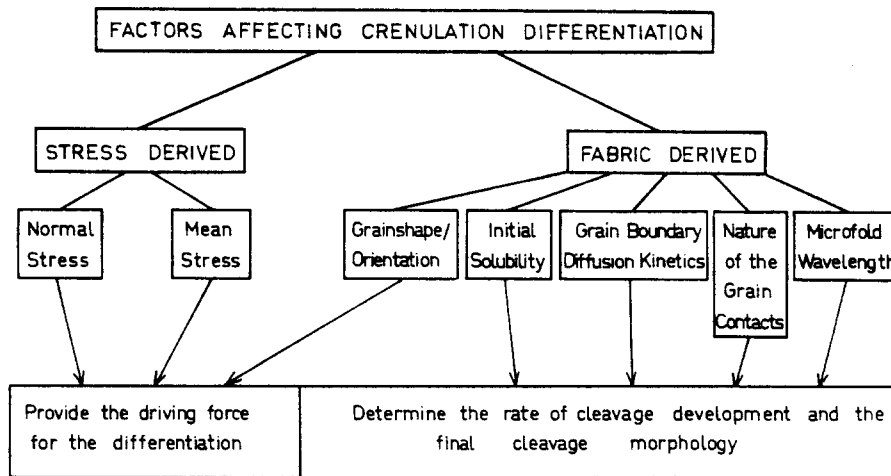


Fig. 1. Dendrogram showing the factors which affect the solution-deposition differentiation of microfolds in crenulated fabrics.

*Factors affecting solution-deposition differentiation of crenulations*

Other factors also affect such differentiation. These fall into two classes: fabric derived and stress derived (Fig. 1). A complex interrelationship of these factors during deformation determines the rate of cleavage development, and more importantly, the final cleavage morphology. The part each variable plays in the overall differentiation process will now be discussed.

1. *Normal stress.* Variations in normal stress ( $\sigma$ ) around microfolds produce differences in chemical potential ( $\mu$ ) which provide the driving force for solution-deposition type differentiation in crenulated fabrics (see previous section). The normal stress ( $\sigma$ ) acting on the fabric anisotropy along the microfold limbs will be higher than that acting in the hinge. This assumes the maximum principal compressive stress responsible for the folding is initially at a low angle to the anisotropy; i.e. a buckling type instability. Such variations in  $\sigma$  are exemplified in a simple fold model (Fig. 2), where a plane stress condition exists, stress across the fold is homogeneous, and the maximum compressive stress ( $\sigma_x$ ) responsible for folding is at a high angle to the fold axial plane. The component of normal stress ( $\sigma$ ) which acts at any point on the folded surface (Fig. 2b) is dependent on the magnitudes of the principal compressive

stresses ( $\sigma_x$  and  $\sigma_z$ ) and the angle  $\theta$  between the tangent normal to the layer at that point and the direction of the maximum principal compressive stress. (Note:  $\theta$  is also half the interlimb angle of the fold.) This relationship is:

$$\sigma = \sigma_x \cos^2 \theta + \sigma_z \sin^2 \theta \quad (4)$$

where  $\sigma$  = normal stress,  $\sigma_x$  and  $\sigma_z$  = the maximum and minimum principal stresses respectively, and  $\theta$  = half interlimb angle of the microfold, shown graphically in Fig. 2(c).

This implies that folds with rounded hinges will have a continuous variation in  $\sigma$  from the limbs to the hinges, and therefore continuous variation in chemical potential ( $\mu$ ). With sufficient magnitude of  $\sigma$ , dissolution will occur along the microfold limbs with concomitant transfer to and redeposition in fold hinges. The diffusive flux is in the direction of normal stress decrease; that is from areas of high chemical potential, fold limbs, to areas of low potential, fold hinges.

With growth and propagation of the fold, the limbs become more normal to the applied principal stress  $\sigma_x$ , such that  $\theta$  decreases, with a resultant increase in normal stress ( $\sigma$ ). This indicates that solution-deposition differentiation should increase as microfolds develop in crenulated fabrics; that is as the folds tighten there should be increasing dissolution along their limbs.

A difficulty with this argument is the assumption of homogeneous stress. Computer simulations of multilayer folds (see Dieterich & Carter 1969, Stephansson & Berner 1971), have shown stress inhomogeneity with variations in stress orientation and magnitude across the folds. However, calculations (Gray 1976) of the normal stresses on the layering around the multilayer folds of Stephansson & Berner (1971, figs. 11a & b) showed a broad similarity in normal stress variation to the idealized homogeneous stress model presented above. There was a general increase in normal stress towards the limbs, and conversely a decrease towards the hinges around the folded multilayer. We believe therefore, that differentiation of crenulations due to solution-deposition processes in multilayer fabrics is largely the result of

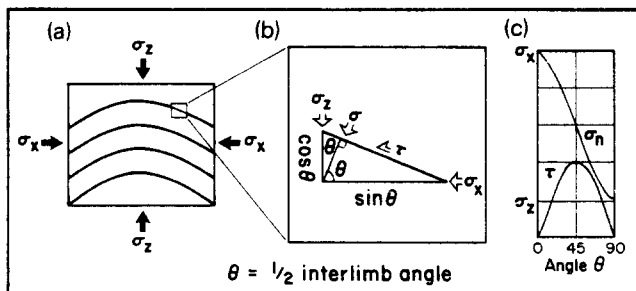


Fig. 2. Relation of normal stress to simple fold model under plane stress conditions. (a) Fold. (b) Resolution of stress along planar segment of fold limb. (c) Graph showing the relationship between shear stress ( $\tau$ ) and normal stress ( $\sigma$ ) with changing limb dip (or interlimb angle). ( $\sigma_x, \sigma_z$  = maximum and minimum principal compressive stress;  $\theta = 1/2$  interlimb angle.)

normal stress-controlled variations in chemical potential around the folds.

2. *Mean stress.* Mean stress ( $\bar{\sigma}$ ) may influence solute concentration, or solubility, in either or both differentially stressed aggregates and buckled anisotropic fabrics. For a single crystal in a stressed crystal aggregate where the average mean stress is  $\bar{\sigma}_a = (\sigma_x + \sigma_y + \sigma_z)/3$  and the deviatoric stress is  $\sigma_a = (\sigma - \bar{\sigma}_a)$ , equation (1) (this paper) can be rewritten as:

$$\left(\frac{\delta\mu}{\delta x}\right)_{T,p} dx = v_1 d\sigma'_a + v_1 d\bar{\sigma}_a - v_2 dp \quad (5)$$

[equation (2), Durney 1972b]

This indicates that stability differences may occur between crystals or separate segments of a folded fabric under different mean stress. Migration of dissolved species by grain boundary diffusion may occur from regions of high  $\bar{\sigma}$  to regions of low  $\bar{\sigma}$  in an attempt to achieve equilibrium. Mathematically modelled folds (Stephansson & Berner 1971, Hobbs 1972) suggest that mean stresses will be higher in the limb regions, thereby facilitating mass-transfer from the limbs to the hinges. However, Stephansson (1974) has shown that mean stress patterns in multilayer folds are complex, although he does state (p. 231) that the incompetent layers show pressure minima at the hinges.

Mean stress partially contributes to normal stress induced chemical potential differences around microfolds, and therefore would aid transfer of dissolved species around them.

3. *Grain shape and grain orientation.* Variations in grain shape and grain orientation around microfolds cause bulk differences in average chemical potential, which should also facilitate the diffusive mass transfer of dissolved species. The concept of average chemical potential is related directly to the concept of average normal stress on a grain, that is, the sum of the normal force over the total surface of the grain divided by the total surface of the grain [Durney 1972a, equation (36),

p. 184]. Average normal stress will vary directly with homogeneous variations in mean stress over the grain, but it does not only depend on this. For inequidimensional

shaped grains it will be higher for grains with their long dimensions normal to  $\sigma_x$  than for similar grains oriented normal to  $\sigma_z$  in the same stress field (Fig. 3c). Also this orientation dependence will be greater the more inequidimensional the grain, but will be zero for perfectly equidimensional grains. In folded, statistically homogeneous, quartz-mica fabrics, the quartz grains, which are already elongate in the pre-existing cleavage, would therefore have a higher average chemical potential along the limbs where they lie at moderate angles to  $\sigma_x$  than grains in the hinges which lie parallel to  $\sigma_x$  (see Fig. 3b). Consequently the average chemical potential developed in the hinges of microfolds in these fabrics should be lower than that associated with the limbs, even if the stress field is homogeneous. This tends towards the same result predicted previously in the discussion of factors (1) and (2); that is a transfer of dissolved species from the limbs towards the hinges.

This difference in mean chemical potential due to grain shape and grain orientation variations between the limbs and hinges of such microfolds should increase as the structure tightens. This is because the grain orientation effect increases markedly with a decrease in fold interlimb angle. However, the effects of thinning of grains due to pressure solution on the limbs and thickening by overgrowth in the hinge may cancel each other because this would lead to increases in mean chemical potential in both areas.

4. *Grain boundary diffusion kinetics.* At low to medium grades of metamorphism diffusive mass transfer is considered to occur along grain boundaries via a fluid or solution medium (Durney 1972b, 1976, Fisher 1973, Rutter 1976, Vernon 1976). The kinetics of such diffusion must be influenced by the nature of the minerals in contact, that is their degree of lattice misfit, and by the nature of migrating ions. Although no quantitative data are available, it appears that in pelitic and semi-pelitic rocks most diffusion occurs along quartz-phyllsilicate contacts, since greatest dissolution occurs there (Heald 1955, Voll 1960, p. 534). We argue that dissolution is greatest here due to lower solute concentrations in the fluid medium which have arisen from faster diffusion of dissolved species from the site of dissolution via the quartz-mica interface. Because dissolution is dependent on solute undersaturation the diffusion rate must influence the dissolution rate, that is, the faster the dissolved solute is removed the faster the solute is dissolved. It appears that the degree of lattice misfit across a grain boundary is maximum, and therefore optimum for diffusion there, when the boundary is between a phyllosilicate and another mineral. At present, however, there are no analytical data to substantiate this.

Other variables, such as temperature ( $T$ ) and the activation enthalpy for diffusion ( $H_D$ ) also influence diffusion creep rates (Rutter 1978, p. 135). These are

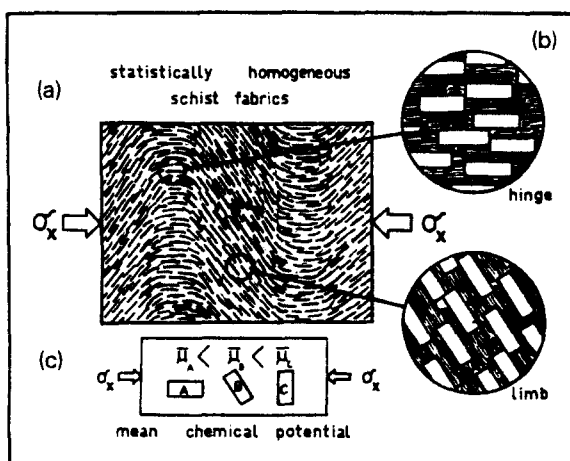


Fig. 3. (a) Grain shape and grain orientation effects in a statistically homogeneous mineral fabric. (b) Insets showing the grain orientation differences of the microfabric in the hinge and limbs. (c) Different mean chemical potentials of grains with different orientations with respect to  $\sigma_x$ .

related through a thin film diffusion coefficient ( $D$ ) defined by:

$$D = D_0 \exp(-H_D/RT) \quad (6)$$

where  $D_0$  = diffusivity at infinite temperature,  $H_D$  = activation enthalpy for diffusion,  $R$  = gas constant, and  $T$  = absolute temperature. When  $H_D$  is small, as it may well be in solutions,  $D$  may not be strongly temperature dependent to an extent which may be geologically significant.

Another aspect is that the kinetics of grain boundary diffusion not only determines the rate of diffusion of dissolved species, but also the pathway for diffusion. This is because diffusion along high diffusivity contacts will dominate, such that these contacts will be largely responsible for the transfer of the dissolved species. The nature of the mineral contacts in a rock fabric is therefore another important variable in the differentiation process (see factor 6).

**5. Microfold wavelength.** Solution transfer differentiation in crenulated anisotropic fabrics must be influenced by the wavelength of the associated microfolds. Each of the driving forces for diffusion discussed earlier in factors 1, 2 or 3 cause dissolved material to migrate from the limbs to the hinges of microfolds. The pathlength of diffusion, that is the distance from the fold limbs to the fold hinge, is therefore determined by the fold wavelength and fold amplitude. Any increase in the wavelength and amplitude of microfolds in a particular fabric would increase the pathlength of diffusion and decrease the steepness of the concentration gradients. Both these changes would effectively slow the rate of differentiation. Differentiation should therefore be most rapid in crenulated fabrics with small wavelength folds. This is because they have small diffusion pathlengths and steep concentration gradients.

**6. Nature of mineral contacts.** Mineral contacts refer to the interfaces between grains in granular and statistically homogeneous fabrics, and to the interfaces between layers, generally alternating quartzose and micaceous types, in multilayer fabrics. Because dissolution and diffusion are along grain boundaries then diffusional transfer of dissolved species must be influenced by the fabric anisotropy and in particular by the nature of the mineral contacts in the fabric (see factor 4). These fabric elements, that is the grain and layer contacts, provide the channelways for diffusion, and therefore determine the pathway for diffusion. Granular and statistically homogeneous fabrics may contain one or several mineral phases; mineral contacts may be monomineralic (i.e. between like minerals), or bimineralic (i.e. between two different minerals). As pressure-solution appears most active along quartz-mica contacts (see factor 4), then most dissolution and diffusion and therefore fabric modification should occur along bimineralic quartz-mica contacts. Other mineral contacts in the same rock fabric may show few or no changes. Similarly most fabric modification in multilayer fabrics should occur along interfaces between adjacent quartzose and

micaceous layers. However, solution-deposition processes in multilayer fabrics may operate simultaneously at the scale of the grains and at the scale of the layering.

**7. Initial solubility.** Minerals have different initial solubilities, or reference solute concentrations ( $c_0$ ), at any given reference pressure and temperature. Therefore for any specified physical conditions (stress, temperature and grain size) minerals with relatively low solubility, for example micas, have correspondingly low concentration gradients and therefore tend to migrate less rapidly than minerals having relatively high solubility, for example quartz and calcite. Differentiation in crenulated fabrics can therefore result from initial solubility differences between minerals along the limbs of the microfolds. It should be noted that solubility is not used here in the sense of solution in large fluid volumes, but in terms of a thin film, grain boundary fluid or dispersed phase of impurity ions.

Quantitative solubility data are scarce for low grade metamorphic minerals particularly within the  $P$ - $T$  regime of the greenschist facies (Barnes 1967, Currie 1968, Nickel 1973). However, petrographic observations based on the assumption that the cleavage minerals are less soluble and therefore less mobile than other minerals in the rock, have been used to establish a series of mineral mobility for crenulated low grade metamorphic rocks. This series in order of decreasing mobility is; (1) calcite, (2) quartz, (3) feldspar, (4) chlorite, (5) biotite, (6) muscovite, and (7) opaque minerals and carbonaceous matter. It indicates that all minerals which come after quartz may become part of the solution residue of a pressure solution zone in pelitic and psammitic rocks. A similar relative mobility series has been established from observations on stylolitic seams in sedimentary rocks (Heald 1955, Trurnit 1968).

Because quartz-mica-chlorite-feldspar assemblages are common in crenulated low grade metamorphic rocks, we consider it is their contrasting solubilities and mobilities, combined with the previous factors 1 to 6, which are largely responsible for the cleavage differentiation which occurs.

#### *Discussion of crenulation differentiation by solution-deposition processes*

Each variable or factor involved in the solution-deposition differentiation of crenulations has been critically evaluated here. The ideas discussed represent a simplistic overview of the general aspects of the differentiation process. We consider these factors are important for crenulation cleavage development at low to medium grades of metamorphism. It is the interaction of these factors at conditions favourable for solution-deposition processes which produces the cleavage. The interrelationships between conditions, fabrics and processes are represented diagrammatically in Fig. 4. The necessary prerequisites are the stress, the pre-existing fabric and the deformation processes. Important considerations from the differentiation flow-diagram (Fig. 4) include:

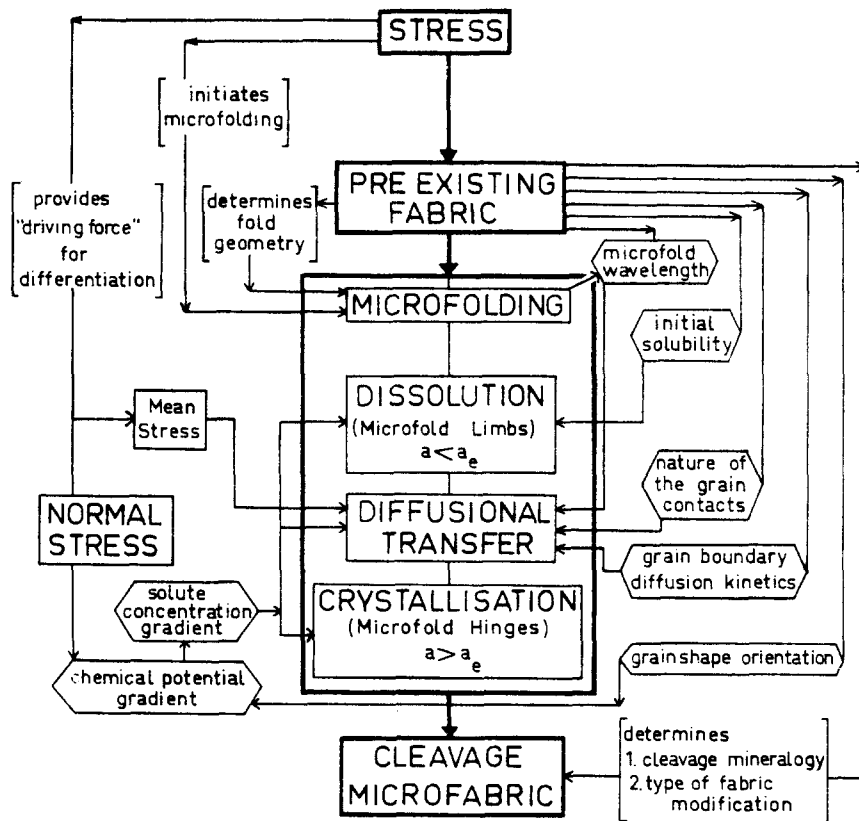


Fig. 4. Flow-diagram illustrating the interrelationships between the various stress and fabric induced parameters, and the solution-deposition processes of dissolution, diffusional transfer and crystallization.

(a) The stress initiates the folding and due to normal stress induced solute concentration and chemical potential gradients indirectly provides the driving force for the solution-transfer processes. The cleavage differentiation is therefore dependent on the variations in normal stress which occur around microfolds.

(b) The rates of the operative processes responsible for the differentiation are dependent on the physical conditions and properties of the pre-existing fabric, such as initial solubility, grain shape, orientation and grain size.

(c) The pre-existing fabric determines the geometry of the crenulation folds, the mineralogy of the cleavage and the type of fabric modification associated with development of the cleavage.

We maintain though that crenulation cleavage, by definition, will not develop unless the initial fabric is anisotropic, either statistically homogeneous or layered, and has mineral phases with different initial solubility and therefore mobilities.

## CONCLUSIONS

Solution-deposition processes are capable of producing crenulation cleavages at low to medium grades of metamorphism (Cosgrove 1972, 1976, Gray 1976, 1979). The differentiation is due to solution-transfer of the most soluble minerals, predominantly quartz in pelitic rocks, from the limbs of microfolds towards the hinges where they are deposited as overgrowths on existing grains and at layer interfaces. Cleavage

development therefore involves a redistribution of minerals depending on their relative solubilities and mobilities through a crenulated fabric. Such a mechanism implies that: (a) crenulation cleavages are either solution zones or solution surfaces along which relatively insoluble mineral residues have been concentrated and aligned; (b) that a proportion of the shortening normal to the cleavages in crenulated rocks is due to local volume reductions along the cleavages; and (c) that cleavage spacing in crenulated rocks is some function of the dominant wavelength of the initially formed microfolds and the amount of solution-shortening across their limbs. We maintain, unlike Cosgrove (1976, p. 166), that both microfolding and differentiation are essential for the development of crenulation cleavage *sensu stricto* (Gray 1977a, p. 299).

Theoretical and experimental studies (Ashby 1972, Stocker & Ashby 1973, Atkinson 1976, Rutter 1976, Kerrich *et al.* 1977) indicate that solution-deposition processes will dominate in cleavage development at low to medium grades of metamorphism. Non-hydrostatic thermodynamic theory and some aspects of the kinetics of grain boundary diffusion indicate those variables and factors which determine firstly, where solution and deposition will occur in crenulated fabrics, and secondly the direction of mass-transfer. The thermodynamics (see Green 1970, Durney 1972a, 1972b, 1976, Elliott 1973, Paterson 1973, Rutter 1976) suggests that differences in normal stress ( $\sigma$ ), and therefore chemical potential ( $\mu$ ), between the limbs and hinges of developing Ramberg-Biot type buckle folds in aniso-

tropic mineral-fabrics, should result in continuous dissolution, migration and deposition at different points around the folded anisotropy. Solution will occur along fold limbs which are regions of high  $\sigma$  and therefore high  $\mu$ , whereas deposition will occur in the fold hinges which are regions of lowest  $\sigma$  and therefore low  $\mu$ . Migration of dissolved species will occur from sites of high  $\mu$  to sites of low  $\mu$ , that is from fold limbs to fold hinges, along normal stress-controlled concentration gradients which parallel the folded anisotropy. This involves diffusive-transfer via grain boundaries associated with the pre-existing structural anisotropy; that is the anisotropy represents a preferred diffusion path. If the folds develop sufficiently then a complete mineral redistribution will occur with the formation of a fine-scale compositional layering or crenulation cleavage.

Crenulation cleavage development discussed in this paper is fundamentally different to the approaches of Cosgrove (1976) and Fletcher (1977). Both consider that mean stress is greater in either or both micaceous minerals and layers than in either or both quartzose grains and layers, because of a higher assumed competency of micaceous minerals over quartz. It is to such differences in mean stress between the minerals in different parts of a fold that they then base explanations for the migration of minerals in folded anisotropic fabrics. This is quite different to the explanation put forward by us, which assumes no differences in stress in different minerals within the fabrics. We believe their initial assumption that the micas are stiffer than quartz is incorrect. The mechanical behaviour of single crystals seems to be sufficiently well known from experimental measurements (Clark 1966) to say that quartz is much stiffer than micas both elastically, and in terms of their respective systems of easiest slip. Furthermore, in terms of aggregate layers this assumption runs counter to well established field and thin section observations that quartz-rich layers behave relatively more competently than mica-rich layers (Ramsay 1967, Sherwin & Chapple 1968, Bayly 1970, Huddleston 1973). Although Cosgrove (1976) introduces the concept that the chemical potential of a solid is directly related to the normal stress on its surface (p. 168), this is not specifically used in his discussion. Fletcher (1977) uses a different concept (cf. 'Riecke principle', Durney 1978) but this requires the existence of open pore channels.

Another approach to crenulation cleavage development relates the mass transfer necessary for cleavage formation to metamorphic driving forces associated with chemical reactions and their related volume-change anisotropies (see Marlow & Etheridge 1977). Microscopic and analytical investigations of crenulated rocks of low to medium metamorphic grade (Cosgrove 1972, 1976, Gray 1976, 1977b, 1979) indicate that these are not important factors in producing a crenulation cleavage anisotropy at low pressures and temperatures. Such processes, reflected by growth of new grains and changes in mineral chemistry, are more important at higher grades of metamorphism, that is above the biotite zone in the greenschist facies. For example, the rocks

described by Marlow & Etheridge (1977, p. 874) are characterized by andalusite-staurolite-garnet-biotite-muscovite-quartz-plagioclase ( $An_{20-30}$ ) assemblages, implying lower to middle amphibolite facies metamorphism and temperatures up to 600°C.

In conclusion, we emphasize once again that cleavage development in crenulated fabrics of low to medium metamorphic grade is due to a complex interaction of the pre-existing fabric with both microfolding and solution-deposition processes. Variables such as the type of fabric anisotropy, mineral contacts, mineral proportions, grain shape, grain and layer orientation, and the amplitude and wavelength of the microfolds are important influences in the differentiation process.

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