KINETICS OF METAMORPHIC LAYERING IN ANISOTROPICALLY STRESSED ROCKS*

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ABSTRACT. Evolution equations for rock texture and intergranular species concentrations are set forth to describe the development of differentiated layering in anisotropically stressed rocks. The processes of crystal growth/dissolution and intergranular species migration along grain boundaries are shown to account for the spontaneous development of metamorphic layers in an initially unbanded rock. They can also account for the development of metamorphic layering over and above a preexisting fabric. These effects are studied both by linear analysis (to demonstrate the conditions for spontaneous band formation) and by numerical solution of the nonlinear evolution equations. The Curie symmetry principle is used to put constraints on the functional form of the evolution equations for the texture and the intergranular species concentrations. The theory presented sets the stage for a number of very interesting questions on the nature of spontaneous generation of textural patterns in metamorphic rocks.

NOTATION

$A_\alpha$, $B_{\alpha,k}$, $C_{\alpha,kq}$ — vectorial coefficients in the functional Taylor series expansion of $\delta c_{\alpha}^{a}$; $k$ and $q$ are subindices denoting cartesian coordinates.

$C_\alpha$ and $S_\alpha$ — vectorial quantities defined respectively as $C_\alpha,11 = C_\alpha,22$ and $C_\alpha,33 = C_\alpha,11$. In the particular case of section III, all these quantities reduce to scalars and are then represented by $A_\alpha$, $C_\alpha$, $S_\alpha$.

$A$, $C$, $S$ — scalar quantities defined by eqs (III.19-21).

$c_\alpha$ — concentration of intergranular species $\alpha$, scaled to make it of the order of magnitude of $\rho_1$.

$\dot{c}_\alpha$ — concentration of intergranular species $\alpha$ in moles of $\alpha$ per unit of intergranular volume.

$D_X$, $D_Y$ — diffusion coefficients of species X and Y divided by the porosity $\phi$.

$G_i$ — rate of change of volume (in cm$^3$ s$^{-1}$) of an $i$ grain due to reaction.

$G_1$ — rate of volume change of an $i$ crystal due to reaction, made consistent with intergranular concentrations scaled to order $\rho_1$ (II.9a).

$\rightarrow J_\alpha$ — flux of $\alpha$ in moles/area time.

$\rightarrow j_\alpha$ — scaled flux of $\alpha$.

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$\mathbf{k}$ — the wave vector, normal to a set of (potential) bands; $2\pi/|\mathbf{k}|$ is the wavelength of the bands. $|\mathbf{k}|$ appears in some equations as $\mathbf{k}; \mathbf{k} \equiv k/|k|.$

$k_x, k_y, k_z$ — specific values of $|k|$ — see (III.26, 29, 30) and figure 1.

$\mathbf{K}_\alpha$ — a row vector with same number of components as the texture vector $\mathbf{T}.$

$L_i$ — dimension of (equant) grains of mineral $i.$ A function of $r$ and $t.$

$L_i^3$ — volume of one grain of mineral $i.$

$\{L_i^3\}$ — vector of all the grain volumes of the $N_M$ minerals at $r$ and $t.$

$n_i$ — number density of $i$, that is, number of grains of mineral $i$ per cm$^3$ of rock. A function of $r$ and $t.$

$\{n_i\}$ — vector of the number densities of the $N_M$ minerals at an $r$ and $t.$

$q_i$ — volume fraction of mineral $i$ ($q_i \equiv n_i L_i^3$).

$q^l$ — volume fraction of mineral $A$ in a preexisting banding in a bimineralic rock.

$q$ — mean value of the volume fraction of mineral $A.$

$q_c$ — critical value of $q_A$ defined by (IV.4); when $q > q_c,$ differentiated layering does not take place.

$Q$ — a quantity defined as $C + S (\sigma \cdot \mathbf{k})^2$ (eq III.25).

$r_x$ — short range of $\mathbf{K}_\alpha.$

$R$ — overall rate of all reactions in which $\alpha$ participates, in moles/rock volume $\times$ time.

$R_\alpha$ — rate of reactions involving $\alpha,$ scaled so that it is consistent with intergranular concentrations scaled to order $\rho_l$ (see II.9a).

$r$ — position vector.

$\mathbf{T}$ — texture vector.

$\mathbf{T}_u$ — mean texture vector.

$t$ — time.

$u$ — scaled velocity of rock plastic flow (II.13).

$v$ — velocity of rock plastic flow.

$W$ — thickness of a slab of rock.

$X, Y$ — (in sections II.E2(e) and III and figs. 2E,F) represent two intergranular species and also their concentrations (per unit of intergranular volume), scaled to the order of magnitude of molar densities of minerals, $\rho_l$ (that is, $X \equiv c_X, Y \equiv c_Y$).

$X_\alpha$ — concentration of intergranular species $\alpha,$ in moles of $\alpha$ per unit of rock volume.
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\(X_0^{eq}, Y_0^{eq}\) — constants (IV.2).
\(\delta x\) — a small deviation of variable \(x\) from its mean value \(\bar{x}\).
\(\Delta V\) — a volume small enough that \(\Delta V^{1/3} < \text{space of banding but large enough to contain enough grains to define the texture well.}\)
\(\phi\) — porosity of the rock (= pore volume over total rock volume).
\(\eta\) — quantity defined by eq (IV.4).
\(\zeta\) — inverse life time of texture perturbations, defined by (III.23) and plotted in figure 1.
\(\kappa_\alpha, \kappa_\beta\) — rate constants in growth rate eqs (IV.1), in cm\(^4\)/mole sec.
\(\lambda_\alpha, \lambda_\beta\) — constants, eq (IV.2).
\(\mu_i(T)\) — chemical potential of a grain of mineral \(i\) subjected to an external applied stress; it is averaged over all face orientations (Kamb, 1961a,b).
\(\nu_\alpha\) — number of moles of intergranular species \(\alpha\) in one formula unit of mineral \(i\).
\(\rho_i\) — molar density of mineral \(i\) (moles of \(i\) per unit volume of mineral).
\(\rightarrow\) — unit vector in the direction of the external applies stress.
\(\tau\) — scaled time (II.13).
\(\tau_c\) — time to dissolve a crystal of typical size \(L_c\).
\(\alpha\) — subscript denoting an intergranular species; it varies from 1 to \(N_i\), the total number of species.
\(i\) — subscript describing mineral species; it varies from 1 to \(N_M\), the total number of minerals.
\((n)\) — superscript identifying coefficients of Taylor expansions of \(c_\alpha, \vec{j}_\alpha, \vec{v}, R_\alpha, G_{ij}, n_i, L_i, s, u\).
\(\prime\) — superscript indicating a dimensionless variable (eqs IV.5).
\(\ast\) — superscript denoting re-scaled length, time, and velocity (IV.10).

1. INTRODUCTION

A. Background

Many foliations that can be found in metamorphic rocks of all grades consist of parallel, planar domains or bands that differ from their neighbors in their mineralogy and fabric (for a very good summary see Hobbs, Means, and Williams, 1976, chap. 5 and especially p. 213-252; following these authors, foliation is used here as a non-genetic term). Various types of these domainal foliations (cleavage, schistosity, layering) differ from each other by their scale and persistence and may grade into each other (Wood, 1974, p. 399). Where the domainal foliation is visible in hand specimen and “it can be shown that it developed by some process of dif-
ferentiation, it is called differentiated layering” (Hobbs, Means, and Williams, 1976, p. 229).

Differentiated layering is a kind of metamorphic differentiation, and as such “. . . implies redistribution of material by chemical reaction within a system without net addition or loss of material from the system” (Orville, 1969, p. 83). Criteria to recognize differentiated layering are:

1. Banding cuts across bedding (Williams, 1972);
2. Banding cuts across mineral grains that are themselves metamorphic (Vidale, 1974);
3. The metamorphic layering has a scale independent of the scale of bedding of the parent rock, which may be inferred in a general way from its chemical composition. “Thus fine differentiation lamination may be equally well developed in pelitic schists derived from shale (presumably finely bedded), in quartz-feldspathic schists derived from graywacke (presumably coarsely bedded), and in green schists derived from basalt.” (Criteria (3) through (6) and quotations are from Turner and Weiss, 1963, p. 99-100.)
4. “Individual layers [that] on close examination prove to be very flat discontinuous lenses.”
5. “Chemically there is no correspondence between many common types of layer and any ordinary sediments” (see also Orville, 1969).
6. “Microscopic examination may prove that a layered structure closely resembling bedding in the field crosses an earlier s-surface itself characterized by layering on a fine scale.”

The layering consists most often of a crude pattern of alternating light and dark bands (see descriptions by Ramberg, 1952; Williams, Turner, and Gilbert, 1954, p. 170; Kretz, 1961; Bowes and Park, 1966; Orville, 1969; Vidale, 1974; and many others). The pattern may consist of a few superimposed scales of repetition, as in the Scottish amphiboles described by Bowes and Park (1966). The light bands invariably contain high proportions of quartz and feldspars; the dark ones consist predominantly of phyllosilicates or amphiboles but also epidote, garnet, kyanite, sillimanite, ilmenite, or other minerals, depending on the grade.

Besides strictly mechanical contributions to banding involving rotation of platy minerals and plastic deformation of crystals, many workers have concluded that differentiated layering requires (at least local) diffusion of ions (for example, Eskola, 1932; Turner, 1941; Orville, 1969; Vidale, 1974; Wood, 1974, p. 397; Stephens, Glasson, and Keays, 1979; Gray, 1979). Proving diffusion, however, does not prove that a repetitive pattern of layering must result from it, and Robin (1979), Stephens, Glasson, and Keays (1979), Gray (1979), and Gray and Durney (1979) have recently attempted to explain layering (or crenulation cleavage) by invoking preferential dissolution of quartz in highly stressed regions of the rock and migration of silica to weakly stressed ones. The role of pressure solution in producing metamorphic differentiation has also been emphasized by Voll (1960, p. 534); dissolution of mineral grains raises the concentra-
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The phenomenon of solutes at and near the point of dissolution and must therefore be followed by diffusion of the solutes away from the site of dissolution. Furthermore, as shown by Bowes and Parks (1966) for some amphibolites from Scotland, many elements migrate toward the dark bands and simultaneously many others migrate toward the light ones.

There seems to be little doubt, therefore, that metamorphic differentiation involves pressure solution followed by diffusion of the solutes produced. Eventually, these solutes will reach points within the rock where they will recombine and yield crystals of the same or new mineral species. Our theory explores the kinetics of the interplay of these processes in metamorphic rocks.

The purpose of this contribution is to propose a reaction/diffusion theory that accounts for the development of the spatial periodicity of differentiated layering in an initially unbanded rock subjected to anisotropic stresses that are not spatially periodic. We show that the theory can also describe the formation of a new periodic banding superimposed on a preexisting one. The theory is macroscopic and phenomenological; in it, the periodicity of band spacing does not require a periodic distribution of the stresses themselves (Robin, 1979, fig. 2) but comes about through the kinetics of the processes of pressure solution, diffusion, and reprecipitation. We have presented preliminary reports of the theory (Ortoleva and Merino, 1980a and b). We shall assume that the diffusion of ions through the rocks is driven by chemical-potential gradients generated by local differences in texture in the presence of an overall anisotropic stress; we shall show that initially small omnipresent textural variations are amplified and evolve into a banding pattern.

Even though crenulation cleavage also involves pressure solution (preferentially of quartz, according to Gray, 1979; and Gray and Durney, 1979) and in all likelihood diffusion, we leave this type of cleavage outside our theory, because the usual equality of the spacing of the bands to the wavelength of the folds whose limbs end up being transposed indicates that the folding itself takes precedence as a cause of the crenulation cleavage over pressure solution/diffusion/reprecipitation.

Some justifications for linking diffusion to anisotropic stress are:

A. Field evidence that metamorphic banding and diffusion are often associated with nonhydrostatic stresses (for example, Stephansson, 1974 and references therein).

B. The predominant minerals of the light and dark bands have distinctly different elastic properties; this suggests that the minerals have been segregated on account of those differences. As can be seen in Clark's tables (1966, table 7-12), the elastic compliances of quartz and feldspar (which predominate in the light bands) differ significantly from those of the phyllosilicates and amphiboles (which predominate in the dark bands).

C. Banding (and this applies especially to the phyllosilicates of the dark layers) is most often accompanied by preferred orientation of mineral
grains, which is caused by nonhydrostatic stresses (see Merino and Ortoleva, 1980, and references therein).

D. The calculations of deVore (1969) suggest that a stressed rock can find it energetically more favorable to have its minerals segregated into bands than to have them uniformly mixed.

B. A Kinetic Theory of Texture

A description of the texture of a polycrystalline, multimineralic rock involves a number of parameters. This list of parameters—a "texture vector" $\mathbf{T}$—must be incorporated in a natural way into a kinetic theory in order that the evolution of the texture to a given state may be described. A typical vector is as follows:

$$
\mathbf{T} = \begin{bmatrix}
\text{Size distribution of all crystal types} \\
\text{Crystal orientation distribution of all crystal types} \\
\text{Shape distribution of all crystal types} \\
\text{Number density of all crystal types}
\end{bmatrix}. \quad (1.1)
$$

The texture is a macroscopic variable. It may vary from region to region in a rock. To be precise we must define the local texture as an average quantity in a volume $\Delta V$ about a point $\mathbf{r}$ at time $t$. As in other theories of macroscopic physics, the volume element $\Delta V$ must be large enough to contain a statistically significant number of crystals but small enough so that $(\Delta V)^{1/3}$ is much smaller than the length scale of the phenomena one wishes to describe—that is, the band spacing in the present problem. Note that if the band spacing is less than a few crystal diameters, then a macro-texture theory will be rather inaccurate. This provides a simple criterion as to how crude the description is. In many examples of banding a macro-texture theory is quite reasonable under this criterion. Note that the definition of texture embodied by eq (1.1) includes the conventional concept of texture in petrology and also the mineralogical mode.

A central goal of the present theory is to describe the evolution of banding. By its nature the banding brought about by metamorphic differentiation requires (Orville, 1969, p. 83) relocation of mineral constituents to concentrate certain minerals in their respective bands. Clearly this does not happen by moving whole crystals but is accomplished by the motion of solute species along grain boundaries. Thus to complete our theory we need to introduce concentrations of intergranular species and then obtain coupled dynamical equations for the texture and intergranular composition. We shall use these equations to study the evolution of metamorphic banding.

The connection between kinetics and the mechanics of an anisotropically stressed rock enters the theory via the rate laws for crystal dissolution and precipitation. We assume that the crystal growth rate is proportional to the affinity. Information on the dependence of the affinity on applied stress and local texture is obtained using the Curie symmetry principle (DeGroot and Mazur, 1962, p. 33-34, 57-64).
II. EQUATIONS OF TRANSPORT AND PHASE KINETICS

A. Assumptions

Because developing an equation of motion for the texture from fundamental statistical theory is a very complicated problem, at all stages of this study we will make the simplest assumptions consistent with retaining the essence of the banding phenomenon. With this in mind, we make the following assumptions:

1. In a volume element $\Delta V$, crystals of a given mineral species $i$ ($i = 1, 2, \ldots N_M$) are roughly equal in size and equant and have volume $L_i^3(r,t)$, where $r$ and $t$ denote position and time.

2. Elastic anisotropy and shape are not taken into account.

3. The intergranular space is negligible relative to total rock volume.

4. Nucleation of new minerals or additional crystals of the preexisting minerals is ignored.

5. A given crystal of mineral $i$ at $r$ may be described as having a chemical potential $\mu_i(T)$ that depends on local texture $T$ and the external applied stress; $\mu_i(T)$ is the facet-dependent potential (in the sense of Kamb, 1961a,b) averaged over all facet orientations.

6. The dissolution and reprecipitation take place only at crystal surfaces.

7. The intergranular species take part in no reactions other than dissolution and reprecipitation.

Let $n_i(r,t)$ be the number density (= number of crystals per unit volume) of mineral $i$. With this, the texture $T(r,t)$ is taken to be

$$ T = \begin{bmatrix} \{L_i^3(r,t)\} \\
\{n_i(r,t)\} \end{bmatrix} \quad (II.1) $$

We introduce the intergranular species $\alpha$ ($\alpha = 1, 2, \ldots N_I$) and denote their respective concentrations (in moles/rock volume) as $X_\alpha$. In general, the intergranular species are chemical entities, ionic or neutral and probably aqueous, different from the minerals. Thus our theory will consist of coupled evolution equations for $T(r,t)$ and the intergranular concentrations $\{X_\alpha(r,t)\}$. A mineral $i$ is related to the intergranular species $\alpha$ via a reaction of the form

$$ \text{Mineral } i \rightleftharpoons \sum_{\alpha=1}^{N_I} \nu_{i\alpha} \text{ (species } \alpha) \quad (II.2) $$

where the stoichiometric coefficient $\nu_{i\alpha}$ is the number of moles of solute species $\alpha$ in one formula unit of mineral $i$. Now that we have a set of macroscopic variables to describe the system and have delineated the available rate processes, we are in a position to derive the dynamical equations.
B. Equations of Motion

Mass balance equations are a standard bookkeeping device that leads naturally to an equation of motion (Fitts, 1962; DeGroot and Mazur, 1962). The mass balance for intergranular species $\alpha$ is

$$\frac{\partial X_\alpha}{\partial t} = -\nabla \cdot \vec{J}_\alpha + R_\alpha \quad (\text{II.3})$$

where $\vec{J}_\alpha$ is the $\alpha$-flux (moles/area-time) and $R_\alpha$ is the overall rate of reactions in which $\alpha$ participates (moles/rock volume-time). In this paper "reaction" refers to the processes of crystal growth and dissolution. It is the choice of $X_\alpha$ as the number of moles of $\alpha$ per unit rock volume that allows for the simple eq (II.3); $X_\alpha$ is related to the $\alpha$-concentration per unit intergranular volume by a factor equal to the fraction of the total rock volume that is intergranular space (see below).

Since crystal dissolution in a given region could in principle produce holes, which are never seen in metamorphic rocks, in our theory the crystalline aggregate under stress is allowed to flow plastically so as to prevent the formation of the voids that might result from dissolution. Let this flow have velocity $\vec{v}(r,t)$. With this, simple balance considerations lead to

$$\frac{\partial n_i}{\partial t} = -\nabla \cdot (n_i \vec{v}) \quad (\text{II.4})$$

$$\frac{\partial L_i}{\partial t} = -\vec{v} \cdot \nabla L_i + G_i \quad (\text{II.5})$$

where $G_i$ is the rate of change of volume of an $i$-crystal due to reaction. Eq (II.5) was derived by noting that, except for reaction, $n_i L_i$ (and not $L_i$) is a conserved variable. This explains why (II.5) contains no $L_i$ terms, unlike (II.4) which contains a $n_i \nabla \cdot \vec{v}$ term.

Stoichiometric considerations lead to the following relation between $R_\alpha$ and the $G_i$'s of all minerals:

$$R_\alpha = - \sum_{i=1}^{N_M} \nu_{i\alpha} G_i n_i \rho_i \quad (\text{II.6})$$

where $\rho_i$ is the molar density (= number of moles per unit volume) of pure mineral $i$. Given the initial texture and intergranular concentrations and boundary conditions, the above eqs (II.3-5) would constitute a complete theory if it were not for the fact that we still need an equation for the velocity $\vec{v}(r,t)$ caused by the deformation.

C. The Space-Filling Constraint and Flow

The statement made above that plastic flow fills in all voids that might develop because of dissolution implies a constraint on the velocity
To see this, we first note that if the intergranular space is negligible we must have

\[ \sum_{i=1}^{N_M} n_i L_i^3 = 1. \]  \hspace{1cm} (II.7)

This supplies one more equation to our original set (II.3-5) and hence can completely determine the velocity for one-dimensional cases (see computer simulation of metamorphic layering of sec. IV below). For 2- and 3-dimensional problems the present theory must be complemented by equations of creep (see, for example, Hult, 1966).

A simple equation for the velocity may be obtained from (II.7) and the dynamical eqs (II.4,5) by taking \( \partial / \partial t \) of both sides of (II.7) and then using (II.4,5) to eliminate \( \partial n_i / \partial t \) and \( \partial L_i^3 / \partial t \) to obtain

\[ \nabla \cdot \mathbf{v} = \sum_{i=1}^{N_M} n_i \mathbf{G}_i. \]  \hspace{1cm} (II.8)

This equation is supplemented by specifying the velocity on the boundary of the system. Note that since the system is incompressible, it may change shape but at essentially constant volume (of course elastic deformation does lead to some small volume changes, but we do not take this into account here). Physically (II.8) states that the divergence of the velocity is the net rate of change of volume caused by the reactions.

**D. Intergranular Volume Fraction Scaling**

The space between the grains in the rocks of interest here is very small. Thus as soon as any appreciable dissolution of a grain takes place the pore fluid becomes saturated locally, and further reaction progress is limited by the speed at which migration along the intergranular space can take away dissolution products. This picture breaks down, of course, when reaction rates are exceedingly small or when diffusion coefficients are large. Effective diffusion coefficients for migration of species along the intergranular space of metamorphic rocks have been measured by Brady (1978) to be on the order of \( 10^{-9} \) cm\(^2\)/sec at 1 to 2 kb and 650°C. Diffusion data are also given by Fisher (1978, table 1). The formulation here and in section III, but not that in section IV, rests on the assumption that diffusion is at most as fast as reaction. We shall examine the formal limit of the dynamics of the system as the porosity \( \phi \) approaches zero.

To assess properly the relative orders of magnitude of terms in (II.3) as \( \phi \to 0 \), we change our formulation to one in terms of concentrations \( \tilde{c}_\alpha \) in moles per intergranular volume. The concentration \( X_\alpha \) (moles per rock volume) is smaller than \( \tilde{c}_\alpha \) by a factor \( \phi \). The relative insolubility of

\[ \text{Later, however, we will invoke elastic deformation as the driving force for pressure solution. This does not contradict our assumption that plastic flow precludes the formation of holes by dissolution, because rocks behave plastically on the time scale of banding but elastically with respect to their instantaneous state of stress.} \]
most minerals makes \( \bar{c}_\alpha \) much smaller than the solid density, typically by a factor also, roughly, on the order of \( \phi \) for metamorphic rocks. Thus we introduce a scaled concentration \( c_\alpha \) such that

\[
X_\alpha = \phi^2 c_\alpha
\]  

(II.9)

where \( c_\alpha \) is an intergranular concentration on the order of the molar density of the solid; the real intergranular concentration of \( \alpha \) is thus of order \( 10^{-4} \text{--} 10^{-5} \), a factor that has been taken out via the definition \( \bar{c}_\alpha = \phi c_\alpha \).

The flux \( \mathbf{j} \) of intergranular species \( \alpha \) will be separated into a rock flow term \( \mathbf{v} X_\alpha \) and a diffusion term \( \phi^3 \mathbf{j}_\alpha \). A factor \( \phi^3 \) is introduced into the latter because as \( \phi \to 0 \) so do diffusion coefficients. \( G_i \) can be scaled into a rate \( G \) consistent with intergranular concentrations scaled to be of order \( \rho \) [see (IV.1)], and, because of the linear relation (II.6), so can \( R_\alpha \), yielding

\[
G_i = \phi G_i
\]

(II.9a)

\[
R_\alpha = \phi R_\alpha
\]

(II.9b)

Thus, the continuity eq (II.3) becomes

\[
\phi^2 \frac{\partial c_\alpha}{\partial t} = -\phi^3 \nabla \cdot \mathbf{j}_\alpha - \phi^3 \nabla \cdot (\mathbf{vc}_\alpha) + \phi R_\alpha.
\]

(II.10)

As \( \phi \to 0 \) the processes of interest do not proceed at infinite rate! Hence in this limit reaction must proceed constrained to the equilibrium manifold

\[
R_\alpha^{(0)} = 0
\]

(II.11)

where for any quantity \( \Psi \) we assume the existence of a Taylor series in \( \phi \),

\[
\Psi = \sum_{n=0}^{\infty} \Psi^{(n)} \phi^n.
\]

(II.12)

(II.11) implies that \( G_i^{(0)} = 0 \) (see eq II.6), and this in turn implies from eq (II.8) that \( \mathbf{v} \) is at most of order \( \phi^3 \). The right side of (II.4,5) is of order \( \phi^2 \), implying that the time scale for evolution of texture is of order \( \phi^{-2} \) or greater. The left side and the second term of the right side of (II.10) are thus of order \( \phi^4 \). Therefore \( R_\alpha^{(1)} \) must also vanish, and the last term of (II.10) is in fact at most of order \( \phi^3 \). Repeating these arguments one finds that the processes of interest evolve in fact on a time scale of order \( \phi^{-3} \), since the rate is proportional to \( \phi^3 R_\alpha^{(2)} \). We introduce an appropriate time \( \tau = \phi^3 t \). Examination of the velocity divergence formula (II.8) shows that \( \mathbf{v} \) is of order \( \phi^3 \), and hence, in summary, we have the new appropriately scaled variables

\[
t = \tau / \phi^3
\]

(II.13)

\[
\mathbf{v} = \phi^3 \mathbf{u}.
\]
The dynamical equations with $G_i = \phi G_i$ then take the form
\begin{align}
\partial \mathbf{c}_a / \partial \tau &= - \nabla \cdot (\mathbf{u} \mathbf{c}_a) - (1/\phi^2) \nabla \cdot \mathbf{j}_a + (1/\phi^3) \mathbf{R}_a \tag{II.14} \\
\partial L_{i}^{3}/\partial \tau &= - \mathbf{u} \cdot \nabla L_{i}^{3} + G_i / \phi^2 \tag{II.15} \\
\partial n_{i}/\partial \tau &= - \nabla \cdot (\mathbf{u} n_{i}) \tag{II.16} \\
\nabla \cdot \mathbf{u} &= \sum_{i=1}^{N_M} n_{i} \frac{G_i}{\phi^2}. \tag{II.17}
\end{align}

Our procedure is next to substitute the expansion (II.12) for all quantities into (II.14-17) and collect terms. For a system of $N_M$ minerals this implies that evolution is constrained by $N_M$ equilibrium relations (equivalent to (II.11))
\[ G_i^{(0)} = 0, i = 1, 2, \ldots N_M. \tag{II.18} \]

To the next order we find $G_i^{(1)} = 0$. The interesting results come from the order $\phi^0$ terms:
\begin{align}
\nabla \cdot \mathbf{j}_a^{(0)} &= - \sum_{i=1}^{N_M} \nu_{i0} \rho_{i} n_{i}^{(0)} G_i^{(2)} \tag{II.19} \\
\frac{\partial L_{i}^{3(0)}}{\partial \tau} &= - \mathbf{u}^{(0)} \cdot \nabla L_{i}^{3(0)} + G_i^{(2)} \tag{II.20} \\
\partial n_{i}^{(0)}/\partial \tau &= - \nabla \cdot (\mathbf{u}^{(0)} n_{i}^{(0)}) \tag{II.21} \\
\nabla \cdot \mathbf{u}^{(0)} &= \sum_{i=1}^{N_M} n_{i}^{(0)} G_i^{(2)}. \tag{II.22}
\end{align}

These equations appear at first to lead to an open ended hierarchy, that is, we appear to need the higher order $G_i^{(2)}$ terms to calculate the lower order $0$-terms. This is not, however, the case. One first solves for the $N_M$ quantities $G_i^{(2)}$ in terms of the $0$-terms from any of (II.19,20) and substitutes them in the other equations. Thus for a system of $N_t$ intergranular species we are left with $N_t + 2N_M + 1$ unknowns and $N_t + N_M + 1$ equations. The $N_M$ excess unknowns can, however, be eliminated in terms of the remaining unknowns via the quasi-equilibrium relations (II.18). We shall apply this technique in the next section. We now turn to the determination of the form of the equilibrium relations.

In the remainder of the mathematical analysis of this paper we shall make the simplifying assumption that $N_t = N_M$, and that, in principle, we may solve the $N_M$ quasi-equilibrium relations (II.18) for the $c_0$ in terms of the remaining variables. As we shall discover, however, the relation is nonlocal.
E. The Equilibrium Functional: Relation Between Stress and Band Orientation

1. The equilibrium phenomenological functional.—Intuitively we expect that the locally averaged intergranular species equilibrium concentration at a point will depend on the number and configuration of the crystals in the vicinity of that point. For example, in a rock with minerals A and B, which have different elastic properties, we expect that if, say, a B crystal has A crystals above and below it (the anisotropic stress being applied in the vertical direction), then equilibrium intergranular concentrations should be different from what they would be if it had B crystals above and below. In our macroscopic theory these effects are included by regarding the equilibrium concentration $c_{\alpha}^{eq}$ at a point $\mathbf{r}$ as a function not only of the texture at $\mathbf{r}$, $\mathbf{T}(\mathbf{r},t)$, but also of $\mathbf{T}(\mathbf{r}',t)$ for points $\mathbf{r}'$ near $\mathbf{r}$. We assume that there are no time delay effects — that is, $c_{\alpha}^{eq}$ depends on $\mathbf{T}(\mathbf{r}',t')$ for $\mathbf{r}'$ near $\mathbf{r}$, and, in contrast, $t'$ only at $t' = t$. Mathematically a general relation between one quantity, here $c_{\alpha}^{eq}$, and a spatial distribution of values of another, $\mathbf{T}(\mathbf{r},t)$, is called a functional and is written $c_{\alpha}^{eq} = c_{\alpha}^{eq}[\mathbf{T}]$.

2. The Curie principle.—Symmetry and smoothness assumptions allow us to gain much insight into the functional $c_{\alpha}^{eq}[\mathbf{T}]$.

a. Linear response: Consider the small deviation $\delta c_{\alpha}^{eq}(\mathbf{r},t)$ due to a small deviation in texture $\delta \mathbf{T}(\mathbf{r},t)$ from a mean value $\mathbf{T}^u$ of a uniformly textured rock. A Taylor series of a function $f(x)$ for small $x$ may be written $f(x) = f(0) + f^{(1)}(0)x + \ldots + f^{(n)}(0)x^n/n! + \ldots$ where $f^{(n)}$ is the $n$th derivative of $f$ with respect to $x$. In a similar manner a functional may be expressed as a functional Taylor series. Expanding $c_{\alpha}^{eq}$ in $\delta \mathbf{T}$ we get, neglecting quadratic terms in $\delta \mathbf{T}$,

$$
c_{\alpha}^{eq}[\mathbf{T}^u + \delta \mathbf{T}] = c_{\alpha}^{eq}[\mathbf{T}^u] + \delta c_{\alpha}^{eq}
$$

where $\mathbf{K}_{\alpha}$ is a row vector with the same number of components as the texture and a "#" implies a sum over components (that is, a dot product). The kernel $\mathbf{K}_{\alpha}$ measures the degree to which a spatial distribution of the deviation of texture from uniformity affects the equilibrium concentration. $\mathbf{K}_{\alpha}$ must not depend explicitly on time, because it represents the functional analogue of the derivative evaluated at the (assumed) time independent uniform texture $\mathbf{T}^u$. We now focus our attention on the properties of the kernel $\mathbf{K}_{\alpha}$.

b. The short range ansatz: We do not expect that a deviation in texture at a point $\mathbf{r}'$ far from $\mathbf{r}$ should affect $\delta c_{\alpha}^{eq}(\mathbf{r},t)$. Such an independence implies that $\mathbf{K}_{\alpha}(\mathbf{r},\mathbf{r}')$ should decay rapidly as $|\mathbf{r}-\mathbf{r}'|$ increases much
beyond a few crystal diameters. This property we term the short range of \( K_\alpha \). Let \( r_g \) be the range of \( K_\alpha \), that is, we assume that \( K_\alpha (r,r') \) is negligible for \( |r-r'| \gg r_g \).

c. Smooth texture variations — the derivative expansion: In the present theory we have assumed that \( T \) varies on a length scale longer than a few crystal diameters — otherwise the texture becomes rather ill defined because of the poor statistics of crystal counting. Thus we shall limit our considerations to the case where \( T(r,t) \) varies on a length scale greater than a few crystal diameters — that is, greater than \( r_g \).

The smoothness of \( \delta T \) and the short range of \( K_\alpha \) imply that we only require \( \delta T \) and its first few derivatives at \( r \). Hence we expand \( \delta T \) in a Taylor series of powers of \( r-r' \) and to good approximation retain only the first few terms when inserted in (II.23). To carry out this procedure we introduce Cartesian coordinates according to \( r = (x_1, x_2, x_3) \). With this we have

\[
\delta T(r',t) = \delta T(r,t) + \sum_{k=1}^{3} \frac{\partial \delta T}{\partial x_k}(x'_k-x_k) + 1/2 \sum_{k,q=1}^{3} \frac{\partial^2 \delta T}{\partial x_k \partial x_q}(x'_k-x_k)(x'_q-x_q) + \ldots
\]

(II.24)

For smooth \( \delta T \) the higher order derivatives are small, and, since \( K_\alpha \) is short range, we may neglect them. Hence (II.23) becomes

\[
\delta c_{\alpha,eq} = A_\alpha \delta T + \sum_{k=1}^{3} B_{\alpha,k} \left( \frac{\partial \delta T}{\partial x_k} \right) + \sum_{k,q=1}^{3} C_{\alpha,kq} \left( \frac{\partial^2 \delta T}{\partial x_k \partial x_q} \right)
\]

\[
A_\alpha = \int K_\alpha(r,r')d^3r'
\]

\[
B_{\alpha,k} = \int K_\alpha(r,r')(x_k-x_k')d^3r'
\]

\[
C_{\alpha,kq} = (1/2) \int K_\alpha(r,r')(x_k-x_k')(x_q-x_q')d^3r'
\]

(II.25)

Symmetry considerations may now be used to put strong constraints on the phenomenological coefficients \( A_\alpha, B_{\alpha,k}, \) and \( C_{\alpha,kq} \) as follows.

d. Symmetry and the Curie principle: Because we have linearized about the uniformly textured state, \( K_\alpha(r,r') \) can only depend on \( r-r' \) and not \( r \) and \( r' \) independently. From this we see that a change of integration variable from \( r' \) to \( r-r' \) in (II.25) implies that the \( A_\alpha, B_{\alpha,k}, \) and \( C_{\alpha,kq} \) are independent of \( r \).

Also, because the expression (II.25) for \( \delta c_{\alpha,eq} \) holds for all \( \delta T \) we can selectively put all but one of the quantities \( \delta T, \frac{\partial \delta T}{\partial x_k}, \) and \( \frac{\partial^2 \delta T}{\partial x_k \partial x_q} \) equal to zero so as to investigate the consequences of the Curie principle.
(see DeGroot and Mazur, 1962, p. 33-34 and 57-64, for a detailed discussion of this principle). The Curie principle, for our purposes, states that since $\delta c_\alpha^{eq}$ is a scalar, its value must be independent of rotation or reflection about a given point of observation. For example consider the value of $\delta c_\alpha^{eq}$ resulting from a $\partial T$ which behaves like $(x_1-x_1^0)\partial T/\partial x_1$ near $r^0 = (x_1^0, x_1^0, x_3^0)$. Since $\delta c_\alpha^{eq}$ is a scalar it should not change its value upon a reflection $y_1 = (x_1-x_1^0) \rightarrow -(x_1-x_1^0) = y_1^R$. Thus we must have $B_{\alpha,1} \frac{\partial T}{\partial y_1} = B_{\alpha,1} \frac{\partial T}{\partial y_1} = - B_{\alpha,1}$. This can only be true if $B_{\alpha,1} = 0$. Hence we have

$$B_{\alpha,k} = 0, k = 1, 2, 3 \quad (II.26)$$

by carrying out the same argument for $k = 2, 3$. By a similar argument we can show that $C_{\alpha,kq}$ vanish for $k \neq q$. Next we assume that the applied uniaxial stress is along the $x_3$ coordinate. Symmetry then demands that $C_{\alpha,11} = C_{\alpha,22}$, but $C_{\alpha,33}$ is not necessarily equal to the other two nonzero $C_{\alpha,kk}$. With this $\delta c_\alpha^{eq}$ as expressed in (II.25) takes the form

$$\delta c_\alpha^{eq} = A_\alpha \frac{\partial T}{\partial x_1} + C_{\alpha,11} \left[ \frac{\partial^2 T}{\partial x_1^2} + \frac{\partial^2 T}{\partial x_2^2} \right] \delta T + C_{\alpha,33} \frac{\partial^2 T}{\partial x_3^2} \quad (II.27)$$

Next we define

$$C_\alpha \equiv C_{\alpha,11} = C_{\alpha,22} \quad (II.28)$$

$$S_\alpha \equiv C_{\alpha,33} - C_{\alpha,11}$$

and obtain

$$\delta c_\alpha^{eq} = A_\alpha \frac{\partial T}{\partial x_1} + C_\alpha \nabla^2 \delta T + S_\alpha \frac{\nabla \cdot \nabla}{\nabla^2} \delta T \quad (II.29)$$

where $\sigma$ is a unit vector along the special stress direction.

The phenomenological coefficients $A_\alpha$, $C_\alpha$, and $S_\alpha$ may in principle be determined by constructing rocks with various texture profiles and then measuring $\partial X_\alpha^{eq}$. In our approach they play a role analogous to diffusion or viscosity coefficients. Like the latter they may either be calculated theoretically or determined experimentally. Theoretically they present a most challenging problem in the analysis of random elastic media. This type of calculation is a difficult task just by itself and is beyond the scope of the present theory.

e. Microscopic interpretation of (II.29): From the microscopic point of view the expression (II.29) reflects the value of $\delta X_\alpha^{eq}$ averaged over an ensemble of crystal size, type, and orientation configurations. It would be interesting, for the purpose of modeling and interpretation, to understand what types of configurations contribute to various terms in the expression for $\delta c_\alpha^{eq}$. A rigorous treatment of this subject must be done via the theory of random media. Here we make what we believe to be reasonable conjectures for the purpose of adding to the intuitive picture.
Consider an aggregate of randomly distributed A and B crystals and assume that A, crudely speaking, is the less compliant mineral. Then the equilibrium concentration of Y, assuming Y ⇔ B, is expected to be higher if the B crystal is surrounded by nearest neighbors of mineral B rather than A, because an A shell would bear most of the applied stress. This type of effect is manifest in the \( A_\sigma \) term in (II.29). If there are A crystals on top and bottom of a B crystal but not on the sides (\( \sigma \) being vertical), then again \( Y^{\text{eq}} \) should be higher. Such effects are accounted for in the \( (\sigma \cdot \nabla)^2 \) and \( \nabla^2 \) terms in (II.29).

3. Stress and band orientation.—The preceding development shows explicitly how stress anisotropy enters \( \delta X_\sigma^{eq} \) and hence the evolution equations for the texture. In what follows we show that the state of uniform texture can become unstable to small (omnipresent) deviations of texture. One may write the general small inhomogeneity as a sum of terms corresponding to banded-like disturbances in various directions and with various wavelengths (that is, a Fourier analysis). Because of the \( \sigma \) term in (II.29) the amplitude of these components will grow at different rates, as will be shown in section IIIIB. Those that grow the fastest will eventually dominate, and a particular orientation of the bands relative to the applied stress is the long-time consequence. Some care must always be taken in drawing conclusions on a nonlinear set of equations using a linear analysis as implied here (see below). However, a full nonlinear analysis of this question is beyond the present work. Nonlinear effects on band formation in the context of the texture profile are accounted for in the numerical simulations presented in section IV.

4. Model calculation.—The structure of the theory set forth in this section is very complex. In the following section we consider a most simple example. Our goal is to demonstrate in more detail how the theory can be used to study the development of banding. Since there are so many effects going on simultaneously, it is important to start from a simple model so that all the interacting effects can be sorted out and interpreted.

III. PATTERN FORMATION IN A SIMPLE BIMINERALIC ROCK

A. The Model

We now apply our analysis to study the tendency for a bimineralic rock to band. The model to be analyzed is characterized by the reactions

\[
A \rightleftharpoons X \tag{III.1}
\]

\[
B \rightleftharpoons Y, \tag{III.2}
\]

relating the minerals A,B to mobile species X,Y (one example of such a simple dissolution chemistry is quartz). We assume a Fick diffusion in the form

\[
\vec{j}_X = -D_X \nabla X \tag{III.3}
\]

\[
\vec{j}_Y = -D_Y \nabla Y, \tag{III.4}
\]
where \(X,Y\) are the respective concentrations (per intergranular volume) times \(\phi^{-1}\) (that is, \(X = c_X\) and \(Y = c_Y\)), and \(D_X\) and \(D_Y\) equal the respective rock diffusion coefficients of \(X\) and \(Y\) divided by \(\phi\).

The quasiequilibrium constraints \(G_i^{(0)} = 0, i = A, B\) are written in the form \(\dot{X} = \dot{X}^{eq}[T], \dot{Y} = \dot{Y}^{eq}[T]\). With this and the stoichiometric coefficients \(\nu_{AX} = \nu_{BX} = 1, \nu_{AY} = \nu_{BY} = 0\), (II.19-22) become, dropping superscripts (0) for simplicity,

\[
G_A^{(2)} = \frac{1}{n_A \rho_A} \nabla \cdot (D_X \nabla X^{eq}) \tag{III.5}
\]

\[
\frac{\partial L_A^3}{\partial \tau} = -\dot{u} \cdot \nabla L_A^3 + G_A^{(2)} \tag{III.6}
\]

\[
\frac{\partial n_A}{\partial \tau} = -\nabla \cdot (u n_A) \tag{III.7}
\]

\[
\nabla \cdot \dot{u} = n_A G_A^{(2)} + n_B G_B^{(2)} \tag{III.8}
\]

and three more equations exactly in the form of (III.5-7) but with \(Y\) and \(B\) replacing \(X\) and \(A\) respectively. For this model we have

\[
T = \begin{bmatrix}
L_A^3 \\
L_B^3 \\
n_A \\
n_B
\end{bmatrix} \tag{III.9}
\]

so that we may eliminate \(G_A^{(2)}, G_B^{(2)}\) via (III.5) from (III.6-8) to obtain a closed set of equations in \(u, T\).

We next make the conjecture that \(X^{eq}\) and \(Y^{eq}\) are functionals only of the volume fractions \(n_A L_A^3 \equiv q_A\) and \(n_B L_B^3 \equiv q_B\). But because the intergranular space is negligible for a rock with crystalline texture (eq II.7), \(q_A + q_B = 1\), and we can write

\[
X^{eq} = X^{eq}[q], Y^{eq} = Y^{eq}[q] \tag{III.10}
\]

where \(q \equiv q_A\). Combining (III.6,7) we find an equation for \(q\), and the model reduces to the following set of coupled equations:

\[
\frac{\partial q}{\partial \tau} = -\nabla \cdot (u q) + \frac{1}{\rho_A} \nabla \cdot (D_X \nabla X^{eq}[q]) \tag{III.10}
\]

\[
\frac{\partial n_i}{\partial \tau} = -\nabla \cdot (u n_i), i = A, B \tag{III.11}
\]

\[
\nabla \cdot \dot{u} = \frac{1}{\rho_A} \nabla \cdot (D_X \nabla X^{eq}[q]) + \frac{1}{\rho_B} \nabla \cdot (D_Y \nabla Y^{eq}[q]) \tag{III.12}
\]

Note the nice simplification that arises here because the \(q,u\) dynamics is independent of \(n_A, n_B\) (assuming \(D_A\) and \(D_B\) are constant or only depend on \(q\)).
B. Stability

One object of this paper is to examine the conditions for and evolution of banded patterns of metamorphic origin. Thus we must solve (III.10-12) for some initial texture $T(r,0)$ and boundary conditions. We could, for example, choose $T(r,0)$ to represent some initial banding — say of sedimentary origin — and watch banding arise at an angle to it due to some applied stress. Such “initial value” problems are describable within our theory. However, we prefer here to solve a simpler but more fundamental problem.

We investigate the situations under which an initially unbanded rock spontaneously generates bands. At first sight this seems impossible, because the driving force for evolution in (III.10,11) involves gradients; hence if $T(r,0)$ is initially constant (independent of $r$) then $\partial q/\partial \tau$, $\partial n_x/\partial \tau$, and $\partial n_y/\partial \tau$ are all initially zero, and no change can ever be generated. But this is an unrealistic situation, because no rock ever has perfectly uniform texture. Under some circumstances explored below, a small deviation $\delta T(r,0)$ from perfect uniformity, denoted $\delta_{b0}$, can grow into a large deviation. A good analogy here is the fact that a ball at the top of a mountain will eventually always roll down the side even though the “top of the mountain” position is a rest point of the noise free equations of motion. The noise comes from either molecular chaos or from flutter in the wind. In our problem the noise is omnipresent deviations, however slight, from uniform texture.

Thus our procedure is to apply the stability analysis used in other problems of self-organization. Once the tendency toward instability is demonstrated, one can attempt a large amplitude pattern analysis (Ortoleva and Ross, 1974; Aucmuty, 1979).

The instability toward pattern formation may be tested by letting

$$ q = \tilde{q} + \delta q $$

$$ \rightarrow \; \overrightarrow{u} = u + \delta u = \delta u $$

and dropping terms of quadratic or higher order in $\delta q, \delta u$ upon putting (III.13) into (III.10-12). We obtain

$$ \frac{\partial \delta q}{\partial \tau} = -\tilde{q} \nabla \cdot \delta u + \frac{1}{\rho_A} D_X \nabla^2 \delta X^q $$

$$ \nabla \cdot \delta u = \frac{1}{\rho_A} D_X \nabla^2 \delta X^q + \frac{1}{\rho_B} D_Y \nabla^2 \delta Y^q $$

Next we use the results of section IIIE to obtain $\delta X^q$ and $\delta Y^q$ in terms of $\delta q$. Eq (II.29) yields

$$ \delta X^q = \{A_X + C_X \nabla^2 + S_X(\nabla \nabla)\} \delta q $$

$$ \delta X^q = \{A_X + C_X \nabla^2 + S_X(\nabla \nabla)\} \delta q $$

$$ \delta X^q = \{A_X + C_X \nabla^2 + S_X(\nabla \nabla)\} \delta q $$

$$ \delta X^q = \{A_X + C_X \nabla^2 + S_X(\nabla \nabla)\} \delta q $$
\[ \delta Y^\text{en} = \{ A_Y + C_Y \nabla^2 + S_Y (\sigma \cdot \nabla)^2 \} \delta q \quad (\text{III.17}) \]

where we recall that \( \sigma \) is a unit vector in the direction of the special stress. Combining (III.14-17) we get

\[ \frac{\partial \delta q}{\partial \tau} = \{ A \nabla^2 + C(\nabla \cdot)^2 + S \nabla^2 (\sigma \cdot \nabla)^2 \} \delta q \quad (\text{III.18}) \]

\[ A = \frac{(1-\bar{q})D_X A_X}{\rho_A} - \frac{\bar{q} D_Y A_Y}{\rho_B} \quad (\text{III.19}) \]

\[ C = \frac{(1-\bar{q})D_X C_X}{\rho_A} - \frac{\bar{q} D_Y C_Y}{\rho_B} \quad (\text{III.20}) \]

\[ S = \frac{(1-\bar{q})D_X S_X}{\rho_A} - \frac{\bar{q} D_Y S_Y}{\rho_B}. \quad (\text{III.21}) \]

Eq (III.18) is easily solved in terms of Fourier modes in the form \( \delta q = q_k^0 \exp(ik \cdot r) \). These modes correspond to periodic variations (for nascent bands) of wavelength \( 2\pi/k \), \( k = |k| \), where \( k \) is normal to the bands. With this we obtain

\[ dq_k^0/d\tau = \xi_k q_k^0 \quad (\text{III.22}) \]

\[ \xi(k, k \cdot \sigma) = -A k^2 + C k^4 + S k^4 (k \cdot \sigma)^2 \quad (\text{III.23}) \]

where \( k = k/k, \ k = |k| \), and hence \( k \cdot \sigma \) is the cosine of the angle between \( k \) and the applied stress. The solution of (III.22) is \( q = q^0(0) \exp(\xi \tau) \)

\[ -A + k^2 \{ C + S (\sigma \cdot k)^2 \} > 0 \quad (\text{III.24}) \]

implies growth of bands perpendicular to \( k \) and with wavelength \( 2\pi/k \).

In figure 1 \( \zeta \) is shown schematically as a function of \( k \) for all combinations of the signs of the coefficients \( A, C \), and \( S \). If \( A < 0 \) very long length scale patterns (\( k \) small) will grow, regardless of orientation relative to the applied stress direction (that is, \( \sigma \cdot k \)). Shorter wavelength textural variations will grow at different rates depending on the orientation of \( k \) with respect to \( \sigma \). If \( S > 0 \) then the patterns will tend to line up perpendicular to \( \sigma \) (and hence \( k \) parallel to \( \sigma \), \( k \cdot \sigma = 1 \) when \( k \) is exactly parallel to \( \sigma \)). For systems where \( S \) is very small we expect no preferred orientation, and the texture will probably be mottled, provided that there is a range of \( k \) for which \( \zeta > 0 \).

Notice that the stability analysis does not involve the derivatives of \( D_X \) and \( D_Y \) with respect to texture. This is so because \( \partial D_X/\partial q \) brings in
terms like $|\nabla \delta q|^2$, which are negligible for small $\delta q$. However, these terms could be of interest when testing the stability of a preexisting banding to the genesis of a secondary banding. Because of the original banding $q^i(r)$ the stability theory would involve terms like $(\partial D_x/\partial q)\nabla q^i \cdot \nabla \delta q$ where $\delta q = q - q^i$ is the deviation of the volume fraction of A from the preexisting banding.

When the quantity $Q$, 

$$Q(\sigma \cdot \hat{k}) = C + S(\sigma \cdot \hat{k})^2 \quad (\text{III.25})$$

is positive then we see that the larger $k$, the more rapidly perturbations grow (see fig. 1). For $A < 0$ also, $\zeta$ is monotonically increasing in $k$ (fig. 1), and hence the theory predicts that the very finest length scale banding is expected. Of course our theory is limited to length scales larger than the largest crystals. Thus we expect that for $A < 0$, $Q > 0$ banding should always be on the scale of a few crystal diameters. For $A < 0$, $Q < 0$, $\zeta$ has a maximum at $k_x$, 

$$k_x = \sqrt{A/2Q}. \quad (\text{III.26})$$

If $2\pi/k_x$ is shorter than a typical crystal size then again $\zeta$ is monotone in the physically relevant domain,

$$0 \leq k < 2\pi/L \quad (\text{III.27})$$

![Graph showing the stability of a uniform texture to the spontaneous formation of patterns of wavelength $2\pi/k$.](image)

**Fig. 1.** The stability of a uniform texture to the spontaneous formation of patterns of wavelength $2\pi/k$ is studied. Shown schematically here is $\zeta$, a function of $k$ for fixed orientation of the band with respect to the applied deviatoric stress. If $\zeta > 0$ the texture perturbation grows and if $\zeta < 0$ the perturbation regresses. If the pattern grows, it does so initially on a time scale of order $\zeta^{-1}$. $k$ is the length of a vector $\hat{k}$ that is normal to the nascent band (see sec. IIIB for a detailed discussion). $A$ and $Q$ are given by (III.19,25).
where L is the typical crystal size, and hence banding is again expected to be on a very fine scale. However, if $k_x$ is in the relevant domain (II.27) then the fluctuations that grow most rapidly are associated with $k_x (\sigma \cdot \mathbf{k})$.

The maximum value of $\zeta$ as a function of $\sigma \cdot \mathbf{k}$ is

$$\zeta_x (\sigma \cdot \mathbf{k}) = - \frac{A^2}{4} \left[ C + S (\sigma \cdot \mathbf{k})^2 \right]^{-1} = - \frac{A}{2} k_x^2$$  \hspace{1cm} (III.28)

which reaches a maximum for $C > 0, S \equiv 0$ when $\sigma \cdot \mathbf{k} = 0, 1$ respectively. Thus we expect that, for $k_x$ in the relevant domain, pattern formation should set in at a wavelength $2\pi/k_c$ where

$$k_c = \text{Max} \left\{ \frac{A}{2C}, \frac{A}{2(C + S)} \right\}.$$  \hspace{1cm} (III.29)

For the case $A > 0, Q > 0$ (see fig. 1), $\zeta$ becomes positive beyond $k_p$, $\zeta(k_p, \sigma \cdot \mathbf{k}) = 0$, where

$$k_p = \sqrt{A/Q}.$$  \hspace{1cm} (III.30)

If $k_p$ is in the relevant domain (III.27) then patterns are again on the fine scale, because the shortest length scale perturbations are growing fastest. For $k_p$ not in (III.27) or $A > 0, Q < 0$ there is no banding.

All conclusions on banding rest on neglecting nonlinear effects. Typically, such effects do not change the qualitative conclusions of the type discussed above. Numerical simulations of the full nonlinear equations for a simple model system are presented in the next section.

Finally, note that the instability criterion (III.24) is not just a thermodynamic one but, because of the diffusion coefficients, involves transport coefficients as well.

IV. A NUMERICAL SIMULATION

A. The Model

A numerical simulation of our texture dynamics is presented here. The study was carried out to investigate the nonlinear behavior of the system. A bimineralic system was modeled as follows. The reaction scheme was (III.1,2). Eqs (II.14-17) were simulated in one spatial dimension using (III.3,4) for $J_X$ and $J_Y$. We assumed that

$$G_A = \kappa_A (X - X_{eq}) L_A \frac{d}{dt}, \quad G_B = \kappa_B (Y - Y_{eq}) L_B \frac{d}{dt}$$  \hspace{1cm} (IV.1)

$$X_{eq} = X_{o,eq} (1 + \lambda_A q_A), \quad Y_{eq} = Y_{o,eq} (1 + \lambda_B q_B)$$  \hspace{1cm} (IV.2)

where $\kappa_A, \kappa_B, X_{o,eq}, Y_{o,eq}, \lambda_A,$ and $\lambda_B$ are constants.

Instability in this simple model occurs when $A$ of (III.19) is negative. We thus have instability to the creation of texture patterns when (letting $\bar{q}$ denote the uniform value of the volume fraction of $A, q = q_A = n_A L_A^3 = 1 - q_B$)

$$\frac{D_X \lambda_A X_{eq}}{\bar{q} \rho_A} + \frac{D_Y \lambda_B Y_{eq}}{(1 - \bar{q}) \rho_B} < 0.$$  \hspace{1cm} (IV.3)
From this we see that at least one of $\lambda_A$ or $\lambda_B$ must be negative for spontaneous pattern formation. If both $\lambda_A$ and $\lambda_B$ are negative, patterning is assured. If $\lambda_B > 0$ but $\lambda_A < 0$ then we have patterning for

$$\bar{q} < q_c \equiv \frac{\eta}{1 + \eta} : \eta = -\frac{D_X \rho_B \lambda_A X_0^{eq}}{D_X \rho_B \lambda_B Y_0^{eq}}, \lambda_B > 0.$$  \hspace{1cm} (IV.4)

This shows in a very transparent way that the condition for patterning depends on the initial uniform texture (through $\bar{q}$). The condition is dynamical, that is, stability depends on diffusion coefficients. Note also that the condition for instability does not depend on the magnitude of $\lambda_A$ and $\lambda_B$ but only on their ratio. Thus a weak dependence of $X^{eq}$, $Y^{eq}$ on texture (here on $q$) due to small $\lambda_A$, $\lambda_B$, slows down the evolution toward the patterned state but does not restrict the conditions for instability to occur.

**B. Dimensionless Equations**

To carry out our simulation we introduce characteristic quantities and define nondimensional variables as follows:

$$c_\alpha = \frac{L_e}{\rho_e} \kappa_\alpha, \ \kappa_\alpha = \frac{L_e}{\rho_e} \kappa_\alpha',$$

$$\tau = \tau/e, \ \tau = D_e D_\alpha',$$

$$r = \sqrt{D_e \tau}, \ u = \sqrt{\frac{D_e}{\tau}} u',$$

$$n_\alpha = L_e^{-3} n_\alpha', \ L_\alpha = L_e L_\alpha'.$$  \hspace{1cm} (IV.5)

The dimensionless variables are indicated with a prime; $\tau_e$ is the typical time to dissolve a crystal of typical size $L_e$; scaled intergranular concentration $c_\alpha'$ is in multiples of solid density. The typical diffusion coefficient $D_e$ is used with the typical time $\tau_e$ to form a typical distance $\sqrt{D_e \tau_e}$. This is then used to obtain a typical velocity $\sqrt{D_e / \tau_e}$; $n_\alpha'$ measures crystal number density in multiples of crystals per typical crystal volume, and hence $n_\alpha' \approx 1$. Finally since a given solid dissolves into a single component ($\Lambda \equiv X$) we use the same subscript ($\alpha = \Lambda, B$) to refer to the mobile species and mineral type.

The characteristic quantities defined in (IV.5) are used to put the equations of motion (II.14-17) in a dimensionless form suitable for computer simulation. We obtain, for a one dimensional system along the $r$ direction (dropping all primes for simplicity)

$$\frac{\partial X}{\partial \tau} = \frac{1}{\phi^2} \frac{\partial}{\partial r} \left( D_X \frac{\partial X}{\partial r} \right) - \frac{\partial}{\partial r} (uX) - \frac{\kappa_\Lambda}{\phi^2} L_\Lambda^2 n_\Lambda (X - X^{eq})$$

$$\frac{\partial n_\Lambda}{\partial \tau} = -\frac{\partial}{\partial r} (n_\Lambda u)$$

$$\frac{\partial L_\Lambda^3}{\partial \tau} = -u \frac{\partial L_\Lambda^3}{\partial r} + \frac{\kappa_\Lambda}{\phi^2} L_\Lambda^2 (X - X^{eq})$$  \hspace{1cm} (IV.6)  \hspace{1cm} (IV.7)  \hspace{1cm} (IV.8)
\[ \phi^2 \frac{\partial u}{\partial r} = \kappa_A n_A L_A \phi (X - X^e) + \kappa_B n_B L_B \phi (Y - Y^e). \] (IV.9)

Three more equations in the form (IV.6,7,8) with X and A replaced, respectively, by Y and B are also obtained.

Eqs (IV.6-9) are very difficult to solve in general because \( \phi \), the intergranular space volume fraction, is very small. The degree of this “stiffness” depends, however, on the initial data, particularly on their length scale. To make computations possible we restrict ourselves to a length scale of order \( \phi \). This leads to the following set of convenient rescaled variables:

\[ \tau = \phi^2 r^* \]
\[ r = \phi r^* \]
\[ u = \phi^{-1} u^* , \]

all other quantities retaining their meaning as in (IV.8,9). With this (IV.6-9) become (dropping the superscript “*” for simplicity)

\[ \frac{\partial X}{\partial \tau} = - \frac{\partial (uX)}{\partial r} + \frac{1}{\phi^2} \left\{ \frac{\partial}{\partial r} \left( D_X \frac{\partial X}{\partial r} \right) - \kappa_A n_A L_A \phi (X - X^e) \right\} \] (IV.11)

\[ \frac{\partial n_A}{\partial \tau} = - \frac{\partial (u n_A)}{\partial r} \] (IV.12)

\[ \frac{\partial L_A}{\partial \tau} = - u \frac{\partial L_A}{\partial r} + \kappa_A L_A \phi (X - X^e) \] (IV.13)

\[ \frac{\partial u}{\partial r} = \kappa_A n_A L_A \phi (X - X^e) + \kappa_B n_B L_B \phi (Y - Y^e) . \] (IV.14)

Again there are three more equations in the form (IV.11-13) with X and A replaced, respectively, by Y and B.

We choose a one dimensional slab of width \( W \) and hence

\[ 0 \leq r \leq W . \] (IV.15)

At the boundaries we have the conditions

\[ u(0) = u(W) = 0 \] \;(IV.16)

\[ \frac{\partial X}{\partial r} = \frac{\partial Y}{\partial r} = 0 \text{ at } r = 0,W , \] \;(IV.17)

the latter ensuring no loss of intergranular species through the ends of the system.

**C. Numerical Simulations**

The numerical simulations were carried out using the backward difference method dividing space up into a grid (Von Rosenberg, 1969). We have explored the development of banding in a rock with initially uniform texture (fig. 2) and initially random texture deviations (fig. 3). Since our theory also applies to any initial nonuniform texture, we show the development of metamorphic banding superimposed on a preexisting bed-
Fig. 2. Temporal and spatial development of a small texture deviation from uniformity showing growth of differentiated layering in a rock consisting of minerals A and B. A small local deviation is not only amplified (provided $\xi > 0$, see fig. 1) but also induces secondary extrema which in turn induce tertiary ones, et cetera. The bimineral model is described by (IV.11-14). All the quantities plotted here and in following figures are dimensionless as described in the paragraph above eqs (IV.11-14) of the text (sec. IV). Shown are: the volumes of A and B crystals ($L_a^n, L_b^n$) in (A) and (B), number densities of A and B crystals ($n_a, n_b$) in (C) and (D), scaled concentrations (X,Y) of X and Y (see (II.9)) in (E) and (F), volume fraction ($q$) of A in (G), and rock flow velocity ($u$) in (H). (I) is a blowup of (G) to emphasize the satellite bands induced by the primary band.

The initial texture was chosen so that the stability criterion (IV.3) indicates instability—that is, growth of small, omnipresent deviations from uniformity. Data chosen for the simulation were $\kappa_i = D_i = \rho_i = 1$, $\lambda_a = \lambda_b = -1/2$, $\phi = 0.1$, $X^{**} = Y^{**} = 0.1$.

From (IV.2), if $\lambda_a < 0$ then the chemical potential of a given A crystal is lowest when it is surrounded by other A's—hence X** decreases with increasing $q$, and similarly for $\lambda_b < 0$. Thus, $\lambda_a < 0$ and $\lambda_b < 0$ each favor segregation.
Fig. 3. Same as figure 2 except for a random initial texture deviation. The initial condition was taken to be a Fourier series whose coefficients were chosen at random such that the root mean square deviation from the average was only a few percent of the latter. Note that the system evolves toward banding of large length scale, with large maxima in \( q \) tending to annihilate smaller neighboring ones. For example, in frame c the second maximum from the right decreases as time goes on, losing A to the maximum to its left, where the free energy of A is lower. The initial small random deviations grow because \( q = 0.25 < q_e = 0.5 \).

These simulations are discussed further in the figure captions. They demonstrate that the model can predict a great variety of band forming phenomena.

V. CONCLUSIONS

We present a phenomenological kinetic theory that can account for the development of spatially periodic differentiated layering in many metamorphic rocks subjected to uniaxial stresses. Our treatment results from carrying accepted concepts and mechanisms (pressure solution, diffusion, and precipitation) to their ultimate consequences. No external, spatially-periodic stress or preexisting textural periodicity are needed to bring about differentiated layering — the formation of these patterns is a case of self-organization.

The theory proceeds by first quantifying the texture, which can be regarded as a vector whose components are the grain size, orientation, shape, and modal abundance for each mineral, and then setting forth evolution equations that couple its time variation to that of the intergranular species concentrations. In the theory, the driving force for the spontaneous development of differentiated layering is an applied aniso-

Fig. 4. Evolution of a rock consisting initially of two layers, one is coarse grained and predominantly mineral A, and the other is fine grained and predominantly mineral B. The rock undergoes differentiated layering. Otherwise the system is the same as described in section IV and the caption of figure 2, differing from the case shown in figure 2 only by the initial texture.
tropic stress acting via the texture dependence of the equilibrium concentrations of species in the grain boundaries. Information on this dependence is obtained by applying the Curie symmetry principle to the calculations of the crystal growth rate and the equilibrium concentration of the intergranular species. This approach leads to eq (II.29), which incorporates (in the third term on the right) the role of the direction of the applied stress in determining the orientation of the banding, and involves phenomenological quantities $A_0$, $C_0$, and $S_0$. We have not tried here to determine these coefficients, which could in principle be done by experiment or by the theory of random elastic media. The goal is to study the dynamics of the formation of banding patterns. This we have carried out in two ways for the case of a bimineralic rock with each mineral dissolving to only one species (III.1.2): firstly, by working out the condition for instability to banding (III.2.4), and secondly, by solving numerically the system of nonlinear differential equations. This numerical simulation (see figs. 2 and 5) shows that very small local departures from an initially uniform texture can grow into spatially periodic segregations of predominantly one or the other mineral. Other cases of metamorphic banding are simulated for rocks with initially non-uniform texture (figs. 3 and 4). The numerical simulations give both the texture (that is, in our simplified case, the grain size and mode of each mineral) and the composition of the intergranular phase (that is, the concentrations of the species $X$ and $Y$) at each point within the rock as time passes.

The ability of a rock to organize itself under particular conditions in response to anisotropic stress should not be surprising. Many inorganic systems are known to display this property under a variety of driving forces such as concentration gradients, surface tension, or heat flux (Nicolis and Prigogine, 1977; Ortoleva, 1978; and in the geological literature, Verhoogen, 1980, p. 123; Elder, 1976, p. 139 and following; Haase and others, 1980; Mc Birney and Noyes, 1979).

The theory presented here could be refined and extended in several ways. One would be to determine (experimentally) the form of the rates $\{G_i\}$ of (II.5). Another would be to consider more realistic situations than the $A = X$, $B = Y$ of sections III and IV — for example, dissolution reactions with some ions in common, which is the case for most silicate rocks. Other extensions would be to include nucleation of new mineral

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**Fig. 5.** Same as figure 2 except $\lambda_B = +1/2$. Comparison of frame c with figure 2G shows that growth of bands is much slower in the present case. This is expected because $\lambda_B > 0$ implies that a B crystal has lowest free energy when surrounded by A’s and therefore impedes the segregation that $\lambda_A < 0$ favors. See caption to figure 2.
species, crystal orientation, and shape as textural variables, and the effects of more spatial dimensions on the nonlinear dynamics. It will be best to add levels of complexity one at a time to separate out the consequences of competing effects. Sections III and IV deal only with the simplest self-organizing mechanism.

Given that our approach rests on well established mechanisms, we believe that the theory applies to many rocks subjected to anisotropic stresses during metamorphism provided that reaction/diffusion can keep up with strain rates. We plan to study banded rocks to try to develop specific petrographic criteria to test the theory.

Our theory does not preclude textural changes brought about by purely mechanical means such as slip and whole-body crystal rotation. If pressure-solution/diffusion/precipitation in a rock can keep up with the strain rate, then the texture of the rock will evolve only through the combination of reaction and diffusion described in detail in this article. If pressure-solution/diffusion/precipitation cannot keep pace with the strain rate, then the texture may be modified both as described in this paper and by bending, rotation, slip, and other mechanical means. In any case, whatever the strain rate, we conclude that true metamorphic segregation into evenly-spaced, alternating light and dark mineral bands (criteria for which are given in the Introduction) has to come about through the combined action of reaction and grain boundary diffusion, that is, through the feedback described here between texture and intergranular-fluid composition.

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metamorphic layering in anisotropically stressed rocks


