**OXFORD MONOGRAPHS ON GEOLOGY AND GEOPHYSICS NO. 21** 

# **Chemical Change in Deforming Materials**

**BRIAN BAYLY** 

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# **Chemical Change in Deforming Materials**

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New York Oxford OXFORD UNIVERSITY PRESS 1992

#### **Oxford University Press**

Oxford New York Toronto Delhi Bombay Calcutta Madras Karachi Kuala Lumpur Singapore Hong Kong Tokyo Nairobi Dar es Salaam Cape Town Melbourne Auckland Madrid

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Published by Oxford University Press, Inc. 200 Madison Avenue, New York, New York 10016

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Library of Congress Cataloging-in-Publication Data

Bayly, M. Brian, 1929– Chemical change in deforming materials / Brian Bayly. p. cm. — (Oxford monographs on geology and geophysics ; no. 21) Includes bibliographical references and index. ISBN 0-19-506764-9 1. Materials-—Compression testing. 2. Deformations (Mechanics)

3. Chemical equilibrium. 4. Geochemistry. I. Title. II. Series. TA417.7.C65B39 1992 620.1'1242—dc20 92-3822

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Printed in the United States of America on acid-free paper To Hans

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## PREFACE

Several years ago I found myself puzzled, and this book is an outcome of that puzzlement. The core of the puzzle lies, as does so much else, in the work of Willard Gibbs.

Gibbs considered a cube of a solid material; each pair of opposite faces was in contact with a fluid and the three fluids were at three different pressures. Gibbs proposed (1878, eqns. 393–395) that for equilibrium to exist, it was necessary for the chemical potential of the material of the solid in one fluid to be different from the chemical potential of the material of the solid in the second fluid. Saying nothing about the chemical potential of the material of the solid in the solid, he left a predicament that, for the science community as a whole, is still unresolved today. At equilibrium, one would expect the potential of the material of the solid to be the same in the solid as in any adjacent fluid, but how can it be the same as in three fluids at once? It was inability to walk away from that question that prompted the thoughts in the following chapters. Of course, many of the thoughts were picked up from other people and in this connection I should like to comment briefly first on policy, then on history.

The policy adopted has two threads: first, no attempt is made at a comprehensive review of what has been written on the topic by other people; second, in the main text, references to other writing have been kept to a minimum. Both of these derive from the wish to present some fundamental ideas in a plain and simple form. There is also the possibility or suggestion that, for all its high quality, earlier writing does not begin at the beginning. (Digressing briefly, I suggest that one needs to begin by taking a material component's potential as direction-dependent, thus being multivalued rather than single-valued at a point, and by emphasizing nonrecoverable deformation, where energy is dissipated, to the exclusion of recoverable or elastic deformation, where energy is stored. A runner in a short race uses two starting-blocks; if these two are indeed the two starting-blocks for the topic in hand, I know of no treatment that has got off to a good start by using both. Of course, my knowledge of what has been written is fragmentary and I apologize to any person who deserves mention at this point whom I am passing over.)

As regards history, let us look at the use of multi-valued potentials (I know too little to survey the extensive literature in which a component's potential is taken as single-valued). Key papers are two by Hans Ramberg: "The Gibbs' free energy of crystals under anisotropic stress, a possible cause for

#### PREFACE

preferred mineral orientation," Anais da Escola de Minas de Ouro Preto, 32 (1959) 1-12; and "Chemical thermodynamics in mineral studies," Physics and Chemistry of the Earth, 5 (1963) 225-252; and two by Ray M. Bowen: "Toward a thermodynamics and mechanics of mixtures," Archive for Rational Mechanics and Analysis, 24 (1967) 370-403; and "Theory of mixtures," in Continuum Physics, vol. 3, ed. A. C. Eringen, New York, Academic Press, (1976) pp. 1-127. In each pair, the second is easier to get hold of: in Ramberg's case, a more widely distributed journal and in Bowen's case a more complete and self-contained presentation. Ramberg introduces the idea of a component's potential at a point being multivalued; he mentions specifically the three principal values. Bowen introduces the idea of a component's potential being a tensor (with principal values matching those proposed by Ramberg).

The other starting-block is emphasis on deformation processes that are dissipative rather than elastic. Links between a material's chemistry and elastic deformation have been widely sought, but the simpler links between change of chemistry and creep or dissipative or viscous deformation have been treated only more recently by G. B. Stephenson ("Deformation during interdiffusion," *Acta Metallurgica et Materialia*, 36 (1988) 2663–2683). This book is an attempt to combine the ideas of Ramberg with those of Stephenson.

Two more policies should be mentioned: one concerns language and the other individuality. Regarding language, we note the difference between legal and plain language. Each has virtues, each is needed in its proper place. When A wishes to introduce a new concept to B, to begin with plain language is often efficient. It may be necessary later to switch to more sophisticated language to increase the precision with which the concept is expressed, but to *start* by means of statements expressed with a high level of exactness (and with the sophisticated phrasing that that requires) is not necessarily efficient. Practice of course varies with the group addressed and the speaker's or writer's intention. Here an effort is made to express the ideas in plain language, with diagrams and analogies. A reader may benefit more from a complete set of ideas roughly sketched than from an incomplete set of ideas more precisely expressed.

So we come to the last prefatory remark, which returns us to the opening. What follows is not authoritative; the intention is to lay some ideas before the reader that he or she may cogitate about independently.

He that made us ... gave us not ... reason to fust in us unused.

SHAKESPEARE, Hamlet IV, 4

The authority of a thousand is not worth the humble reasoning of a single individual. GALILEO, Letters on sunspots (Rome, Lincean Academy 1613)

It is important that students bring a certain ragamuffin, barefoot irreverence to their studies; they are not here to worship what is known but to question it.

BRONOWSKI, Ascent of Man (London, BBC 1975)

PREFACE

At some points in the text, wording is chosen for terseness and at some points for clarity; in both instances the effect may be to imply definiteness or to seem didactic, but this is not the intention. I have tried to put the ideas in a form that the reader can grasp; it is the reader's business to decide how much or how little to accept. I would be happy if the reader would treat what follows as raw clay, and use it to mold something more durable or substantial for himself or herself.

#### Acknowledgments

It is a pleasure to record the part played by other people, both in general encouragement and in more technical discussions.

In the first group I wish to recognize my parents, my wife, Geoffrey Crowson and the Provost and Fellows of King's College, Cambridge. The value of individual people and the value of having people who are different from each other: in upholding these, the people named directed my course in life.

Colleagues who helped at various times with patience and good humor are Eric Wild, Don Drew, Ray Fletcher, Florian Lehner and George Fisher; many students, of whom I would like to name Sharon Finley, Frank Florence, Thomas Menard and Ray Donelick; and also Joseph Kestin, Jean Cogné, Peter Cobbold, Enrique Merino, Bob Wintsch and Graham Borradaile.

In both roles Hans Ramberg stands alone. He has enriched the sciences for all of us and, by a happy chance, plays a role in my life for which I never cease being glad. We are all in his debt.

Rensselaer Polytechnic Institute May 1992 B.B.

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# SYMBOLS

A	(i) an area
	(ii) a factor in an exponential term $Ae^{kx}$ , etc., especially, the
	governing stress magnitude in a stress that varies exponentially
Α	an atomic species in a binary material (A, B) or a compound
	(A, B)X
$A_i$	activity coefficient of component i
a	volume fraction of component A in a material
$a^{\mathrm{a}}$	volume fraction of an imaginary additive
В	the length-scale factor in an exponential term $Ae^{x/B}$
В	an atomic species accompanying species A (q.v.)
b	volume fraction of component B in a material
$C^i$ , $C_i$	concentration of species $i = (number of atoms of i in sample)/$
-,-,	(total number of atoms in sample). In a compound (A, B)X,
	ca number of atoms of A
	$C^{-1} = \frac{1}{\text{total number of } A + B}$
$C_{0}$	a concentration magnitude that is uniform through space
c	the extreme value of a fluctuation in composition from point to
	point
D	a diffusion coefficient: see Note following this list
$\tilde{D}^i$	a diffusion coefficient; see Note following this list
Ē	an elastic property
$E^* E^a$	a coefficient for interdiffusion of two atomic species
E E E	strain rate invariants
$L_{\rm I}, L_{\rm II}, L_{\rm II}$	linear strain rate
e	nical strain rates at a point
$e_1, e_2, e_3$	(i) mean strain rates at a point $a = (a + a + a)/3$
$e_0$	(i) mean strain rate at a point, $e_0 = (e_1 + e_2 + e_3)/5$
	(ii) a strain rate that is arbitrarily fixed and is uniform through space (Chapter 13 on)
e <sub>ii</sub>	linear strain rate along direction <i>i</i> (no summation convention)
eii	shear strain rate on a plane normal to <i>i</i> along direction $j$ ( $i \neq j$ )
e,	linear strain rate along direction n
$e'_n$	the difference $e_n - e_0$ , $e_n - (\text{mean strain rate})$
<i>F</i>	force
G	(i) free enthalpy or Gibbs free energy
-	(ii) elastic shear modulus $\sigma'_{\alpha}/2\varepsilon'_{\alpha}$

xvi	SYMBOLS
g, h	two coexisting phases
H	the governing stress magnitude in a term $He^{kx}$ that describes an exponentially varying stress
j	(i) amplitude of a fluctuation in pressure (Chapter 8) (ii) wavenumber as in $C = C_0 + c \sin ix$
Κ	a diffusion coefficient: $\partial(\text{vol})/(\text{vol}) \partial t = K \nabla^2 P$ . See Note follow- ing this list
Ka	coefficient for interdiffusion of two components
K <sup>i</sup>	diffusion coefficient for component $i$ in a composite material
<i>K</i> *	joint diffusion coefficient for two or more species moving together
$K_1$	diffusion coefficient: flux = $K_1 \partial (\text{conc})/\partial x$ . See Note following this list
<i>K</i> <sub>2</sub>	diffusion coefficient: particle velocity = $K_2 \partial \mu / \partial x$ . See Note following this list
Κ	elastic bulk modulus $\sigma_0/3\varepsilon_0$
k	(i) a general-purpose proportionality constant
	(ii) a diffusion coefficient: $\partial(\text{vol})/(\text{vol}) \partial t = k \nabla^2 \mu$ . See Note following this list
	(iii) inverse length in a term $e^{kx}$
	(iv) a factor relating viscosity to composition
L	the characteristic length of a material $2(NK)^{1/2}$
$L_i$	the characteristic length of material <i>i</i> in a composite material
$L_{\rm m}$	a mean value formed from two lengths of type $L_i$
$L_0$	the length of a circular arc that is a quarter-circle
1	a small length
l, m, n	a set of orthogonal directions
m	(i) a mass of material
	(ii) one of the set l, m, n
m <sub>i</sub>	mass of component i
Ň	viscosity $\sigma'_n/2e'_n$
Ni	the viscosity of part <i>i</i> of a composite material
N*	the viscosity of a mixture
n	(i) the extreme value of a fluctuation in viscosity from point to point
	(ii) one of the set l, m, n
n	a direction or unit vector
$n_1, n_2, n_3$	components of <b>n</b> along directions 1, 2, and 3
P	pressure
$P_0$	a uniform reference pressure
P <sub>e</sub>	one of the pair $(T_e, P_e)$ at which two phases are in equilibrium
Pi	a pressure imposed on a sample formerly in equilibrium at $P_e$
Q	quantity, for example quantity of heat or mass
R	(i) the gas constant
	(ii) in Chapters 12 and 13, the radius of a quarter-circle with arc-length $L_0$ , = $2(NK)^{1/2}$ . From Chapter 14 onward, L is used in place of $R$ for this quantity.
	used in place of K for this qualitity

SYMBOLS

R (cont.)	(iii) a stress difference $\frac{1}{2}(\sigma_{zz} + \sigma_{yy}) - \sigma_{xx}$ (Appendix 13A only)
R	a diffusing component (Chapter 4 only)
S	(i) entropy
	(ii) the stress difference $\sigma_{zz} - \sigma_{xx}$
$S_0$	a stress magnitude that is uniform through space
$S_{\rm I}, S_{\rm H}, S_{\rm HI}$	stress invariants
S	the extreme value of a fluctuation in stress from point to point
<i>s</i> ′	$= s(1 + j^2 R^2/2)$
Т	(i) temperature
	(ii) a stress difference $\frac{1}{2}(\sigma_{zz} - \sigma_{yy})$ (Appendix 13A only)
$T_0$	a uniform reference temperature
$T_{e}$	one of the pair $(T_e, P_e)$ at which two phases are in equilibrium
t	time; $\delta t$ , a short period of time
U	internal energy
V	(i) volume of a specified sample
	(ii) volume of 1 kg or 1 kg-mole of material, according to context
$V_i$	volume of 1 kg or 1 kg-mole of species <i>i</i> , according to context
$X^i, X_i$	concentration of component <i>i</i> , same as $C_i$
$X_{\mathrm{I}}, X_{\mathrm{II}}$	concentration invariants
x	volume-fraction of component X in a material
x, y, z	coordinates of a point
$x_1, x_2, x_3$	a set of orthogonal axes
1. 2. 0	
α	an angle in the xz plane or $x_1x_3$ plane
α α <sub>1</sub> , α <sub>2</sub>	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes
α α <sub>1</sub> , α <sub>2</sub> α, β, γ	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$
$ \begin{array}{l} \alpha \\ \alpha_1, \alpha_2 \\ \alpha, \beta, \gamma \\ \alpha_i \end{array} $	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i_i = A_i X_i$
$ \begin{array}{l} \alpha \\ \alpha_1, \alpha_2 \\ \alpha, \beta, \gamma \\ \alpha_i \\ \varepsilon \end{array} $	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i_i = A_i X_i$ linear strain, = (change in length)/(original length)
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i_i = A_i X_i$ linear strain, = (change in length)/(original length) principal strains at a point
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, = (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, = (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention)
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$ $\varepsilon_{ii}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ )
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_n$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$ $\varepsilon_i$ $\varepsilon_n$ $\varepsilon'_n$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_n$ $\varepsilon'_n$ $\theta$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle
$\alpha$ $\alpha_1, \alpha_2$ $\alpha, \beta, \gamma$ $\alpha_i$ $\varepsilon$ $\varepsilon_1, \varepsilon_2, \varepsilon_3$ $\varepsilon_0$ $\varepsilon_{ii}$ $\varepsilon_i$ $\varepsilon_n$ $\varepsilon'_n$ $\theta$ $\theta_k$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, = (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i>
$\alpha$ $\alpha_{1}, \alpha_{2}$ $\alpha, \beta, \gamma$ $\alpha_{i}$ $\varepsilon$ $\varepsilon_{1}, \varepsilon_{2}, \varepsilon_{3}$ $\varepsilon_{0}$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_{n}$ $\varepsilon'_{n}$ $\theta$ $\theta_{k}$ $\theta_{1}, \theta_{2}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i> angles measured away from a line in two orthogonal planes
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$\alpha$ $\alpha_{1}, \alpha_{2}$ $\alpha, \beta, \gamma$ $\alpha_{i}$ $\varepsilon$ $\varepsilon_{1}, \varepsilon_{2}, \varepsilon_{3}$ $\varepsilon_{0}$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_{n}$ $\varepsilon'_{n}$ $\theta$ $\theta_{k}$ $\theta_{1}, \theta_{2}$ $\lambda$ $\mu$ $\mu_{i}, \mu_{ii}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i> angles measured away from a line in two orthogonal planes wavelength of a sine-wave chemical potential chemical potential associated with direction <i>i</i> (no summation convention)
$\alpha$ $\alpha_{1}, \alpha_{2}$ $\alpha, \beta, \gamma$ $\alpha_{i}$ $\varepsilon$ $\varepsilon_{1}, \varepsilon_{2}, \varepsilon_{3}$ $\varepsilon_{0}$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_{n}$ $\varepsilon'_{n}$ $\theta$ $\theta_{k}$ $\theta_{1}, \theta_{2}$ $\lambda$ $\mu$ $\mu_{i}, \mu_{ii}$ $\mu_{n}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , = $A_iX_i$ linear strain, = (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i> angles measured away from a line in two orthogonal planes wavelength of a sine-wave chemical potential chemical potential associated with direction <i>i</i> (no summation convention) chemical potential associated with direction <b>n</b>
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$\alpha$ $\alpha_{1}, \alpha_{2}$ $\alpha, \beta, \gamma$ $\alpha_{i}$ $\varepsilon$ $\varepsilon_{1}, \varepsilon_{2}, \varepsilon_{3}$ $\varepsilon_{0}$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_{n}$ $\theta$ $\theta_{k}$ $\theta_{1}, \theta_{2}$ $\lambda$ $\mu$ $\mu_{i}, \mu_{ii}$ $\mu_{n}$ $\mu_{1}, \mu_{2}, \mu_{3}$ $\mu_{0}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i> angles measured away from a line in two orthogonal planes wavelength of a sine-wave chemical potential chemical potential associated with direction <i>n</i> principal values of $\mu$ at a point ( <i>i</i> ) chemical potential in a reference state
$\alpha$ $\alpha_{1}, \alpha_{2}$ $\alpha, \beta, \gamma$ $\alpha_{i}$ $\varepsilon$ $\varepsilon_{1}, \varepsilon_{2}, \varepsilon_{3}$ $\varepsilon_{0}$ $\varepsilon_{ii}$ $\varepsilon_{ij}$ $\varepsilon_{n}$ $\theta$ $\theta_{k}$ $\theta_{1}, \theta_{2}$ $\lambda$ $\mu$ $\mu_{i}, \mu_{ii}$ $\mu_{n}$ $\mu_{1}, \mu_{2}, \mu_{3}$ $\mu_{0}$	an angle in the xz plane or $x_1x_3$ plane angles measured away from a line in two orthogonal planes angles between a line and three axes x, y, z or $x_1, x_2, x_3$ activity of component $i$ , $= A_i X_i$ linear strain, $=$ (change in length)/(original length) principal strains at a point mean strain at a point, $\varepsilon_0 = (\varepsilon_1 + \varepsilon_2 + \varepsilon_3)/3$ linear strain along direction <i>i</i> (no summation convention) shear strain on a plane normal to <i>i</i> along direction <i>j</i> ( $i \neq j$ ) linear strain along direction n the difference $\varepsilon_n - \varepsilon_0, \varepsilon_n$ - mean strain a general-purpose angle an angle in the plane containing directions <i>i</i> and <i>j</i> angles measured away from a line in two orthogonal planes wavelength of a sine-wave chemical potential chemical potential associated with direction <i>i</i> (no summation convention) chemical potential associated with direction n principal values of $\mu$ at a point (i) chemical potential in a reference state (ii) mean value, $\mu_0 = (\mu_1 + \mu_2 + \mu_2)/3$

xviii	SYMBOLS
$\rho_i$	number of kg-moles of species $i$ in $1 \text{ m}^3$ of material
σ	a stress component
$\sigma_1, \sigma_2, \sigma_3$	principal stresses at a point
$\sigma_0$	mean stress at a point, $\sigma_0 = (\sigma_1 + \sigma_2 + \sigma_3)/3$
$\sigma_i, \sigma_{ii}$	normal stress component on a plane normal to $i$ (no summation convention)
$\sigma_{ij}$	stress component on a plane normal to $i$ and acting along direction $j$ ( $i \neq j$ )
σ <sub>n</sub>	normal stress component on a plane normal to n
$\sigma'_{\rm n}$	the difference $\sigma_n - \sigma_0$
$\sigma_t$	a shear stress component

#### Note on diffusion coefficients used

Material motion can be driven by a gradient in concentration or pressure or chemical potential. The coefficients mainly used for the three circumstances are  $D^i$ , K, and D, as follows. (For  $K_1$ ,  $K_2$ , and k, see below.)

$$D^{i} \quad \text{Flux} = D^{i} \frac{\partial}{\partial x} \left( \operatorname{conc} \frac{\operatorname{mol}}{\operatorname{m}^{3}} \right) \qquad \frac{\partial Q}{\partial t} = D^{i} \frac{\partial^{2}}{\partial x^{2}} \left( \operatorname{conc} \frac{\operatorname{mol}}{\operatorname{m}^{3}} \right)$$
$$\frac{\operatorname{mol}}{\operatorname{m}^{2} - \operatorname{sec}} \quad \frac{\operatorname{mol}}{\operatorname{sec}} \quad \frac{\operatorname{mol}}{\operatorname{m}^{4}} \qquad \frac{\operatorname{mol}}{\operatorname{m}^{3} - \operatorname{sec}} \quad \frac{\operatorname{mol}}{\operatorname{sec}} \quad \frac{\operatorname{mol}}{\operatorname{m}^{5}}$$
$$\frac{\partial X}{\partial t^{2}} = D^{i} \frac{\partial^{2} X}{\partial x^{2}} \left[ \begin{array}{c} X = \operatorname{mol} \\ \operatorname{fraction} \end{array} \right]$$
$$\frac{1}{\operatorname{sec}} \quad \frac{\operatorname{m}^{2}}{\operatorname{sec}} \quad \frac{1}{\operatorname{sec}} \\ \frac{\operatorname{mol}}{\operatorname{sec}} \quad \frac{\operatorname{mol}}{\operatorname{sec}} \\ \frac$$

SYMBOLS

D

 $e = \frac{\partial l}{l \partial t} = D \qquad \frac{\partial^2}{\partial x^2} \text{(potential)}$  $\frac{1}{\text{sec}} \qquad \frac{\text{mol}}{\text{Pa-m-sec}} \qquad \frac{\text{J}}{\text{mol-m}^2}$ = linear strain rate

For component A,  $K^{A} = 3D^{A}V^{A} = aD^{iA}V^{A}/RT$ , where a = volume fraction of component A.

 $K_1$  and  $K_2$  are used briefly in Chapter 3.  $K_1$  is the same as  $D^i$ ;  $K_2 = D^i/RT = K^A/aV^A = 3D^A/a$ .

k is used briefly in Chapter 10, = 3D = K/V; volume strain rate  $\partial(\text{vol})/(\text{vol}) \partial t = k \partial^2 \mu / \partial x^2$ .

Basis for the relation  $K^{A} = 3D^{A}V^{A}$ .  $\partial \mu^{A}/\partial P = V^{A}$ ; also in an isotropic process, volume strain rate = 3 × linear strain rate.

Basis for the relation  $K^{A} = aD^{iA}V^{A}/RT$ . For pressure-driven motion, influx of A in (m<sup>3</sup>/m<sup>3</sup>-sec)

$$= K^{\mathbf{A}} \partial^2 P / \partial x^2 = (K^{\mathbf{A}} / V^{\mathbf{A}}) \frac{\partial^2 \mu^{\mathbf{A}}}{\partial x^2}$$
(i)

Also, for composition-driven motion, influx of A in  $(m^3/m^3-sec)$ 

= (influx in mol/m<sup>3</sup>-sec)
$$V^{A} = D^{iA}V^{A} \frac{\partial^{2}(\operatorname{conc} \operatorname{mol/m^{3}})}{\partial x^{2}}$$
 (ii)

To compare (i) and (ii) we note that if conditions permit the term  $(\partial X^A/X^A \partial x)^2$  to be neglected,

$$\frac{\partial^2 \mu^A}{\partial x^2} = \frac{RT}{X^A} \frac{\partial^2 X^A}{\partial x^2}$$
$$= \frac{RT}{(\text{conc mol/m}^3)} \frac{\partial^2}{\partial x^2} (\text{conc mol/m}^3)$$
$$= \frac{RTV^A}{a} \frac{\partial^2}{\partial x^2} (\text{conc mol/m}^3)$$

Hence for composition-driven motion, influx of A in  $(m^3/m^3-sec)$ 

$$=\frac{aD^{iA}}{RT}\frac{\partial^2\mu^A}{\partial x^2}$$
(iii)

For equivalence of (i) and (ii),  $K^{A} = aD^{iA}V^{A}/RT$ .

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# CHEMICAL CHANGE IN DEFORMING MATERIALS

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# OVERVIEW AND PREVIEW OF CONCLUSIONS

The purpose of this book is to fill something of a gap. In general, thermodynamics has been a great success and has provided a means of understanding and predicting material behavior of almost all kinds at the macroscopic level. Even when thermodynamic statements were limited to equilibrium states they were widely useful, and with extension to nonequilibrium states almost all behaviors that a person might observe directly became accessible to theory. But there has been and is one resistive point: if a cylinder of material is more strongly compressed along its length than radially, it is in a nonequilibrium state no matter how ideal its condition in other respects, and the effect of this type of nonequilibrium has not been successfully explored.

The physical consequences are, of course, well known; the cylinder deforms in ways successfully described in almost all respects by the methods of continuum mechanics. But the chemical consequences are less well known. For example, suppose the cylinder contains iron and is surrounded by some second iron-bearing phase; suppose further that before the cylinder is compressed axially, the cylinder and its surroundings are in equilibrium. When the axial compression is imposed, how is the equilibrium disturbed and what processes begin to run? The purpose of the book is to provide the outline of a comprehensive approach to this question.

The question has been discussed extensively in technical journals and in complicated ways. The stimulus for this book is the belief that the topic need not be so complicated. There are two equations that describe the stresses in the cylinder that have up to now not been used; using these neglected equations provides a point of view not taken by other writers, and it is the fresh point of view that permits certain simplicities to be seen (the key equations are 6.3 and 8.10). Of course, we make headway only to a limited extent; not all problems are answered, not all complications are resolved. The existence of a central and unresolvable complication is recognized toward the end of this overview, in the section on Continuum Behavior and Atoms.

*Materials considered.* We shall give most of our attention to materials that are in the process of deforming. The material one imagines depends on the time scale envisaged: the ice of a glacier deforms appreciably in a year, whereas silicone putty deforms appreciably in a few minutes; water deforms

even more readily but we shall not consider anything as mobile as water because here forces related to the material's acceleration can be as large as the other forces present. We shall stay within circumstances where the material's acceleration can be neglected. Hot ceramic materials are considered, both crystalline and glassy; also the naturally occurring silicates and oxides known to geologists; also polymers, metals and semiconductors. We make gross simplifying assumptions so that the differences among these materials are submerged; the material treated in detail is in fact an imaginary material simpler than any of these. But the materials listed are the target at which the simplified discussions are aimed.

*Elastic behavior*. At least for a start, we shall also choose circumstances where elastic behavior can be neglected. A stressed elastic material differs from the same material when unstressed in respect of bond lengths and quantity of energy stored per gram or kilogram; but we shall assume that these changes hardly affect the way the material flows or creeps—at least for a start. After discussion of the flow behavior, the question of elastic behavior will be taken up.

Chemistry. We shall assume rather simple chemistry. In some of the discussion, the material will be as simple as ice, that is to say of a single, fixed composition; and at other times we shall consider binary mixtures such as the sulfide (Zn, Fe)S—which can be thought of as a mixture of ZnS and FeS—or a polymer mixture of trifluorethylene and tetrafluorethylene. With such a binary material, there is the possibility of variation in composition within a sample. One might say that the purpose of the book is to learn to understand how nonhydrostatic stress can affect such variations in composition.

Stresses. Inside a deforming continuum (glacier or putty lump, as before), if one could inspect a small area, one would find the material on one side of the area pressing and dragging on the material on the other side: normal to the element of area considered, there is either a compression or a tension, and in addition there is a stress tangential to the area considered. We shall give attention mainly to the normal stress and mainly to compressions (less emphasis on tensions). Among the circumstances to be considered is the special case of hydrostatic pressure, where, whatever plane one considers through a single point inside the continuum, one finds the same compressive stress across it; and also, most importantly, the more general case where, at a single point, some planes are at a higher level of compressive stress and some at a lower level.

Chemical potential. In classical thermodynamics, equilibrium states are emphasized. In a state of equilibrium, each component of a material has a chemical potential that takes the same single value at every point in the system considered. A component's chemical potential depends on the pressure and temperature and on the component's concentration. For