

Abstract wavy lines in black and grey, flowing across the top of the page.

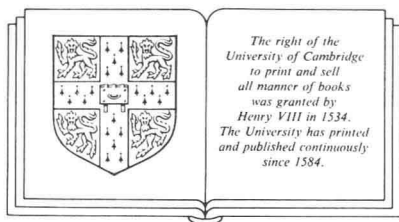
Flow and Reactions in Permeable Rocks

O. M. Phillips, F. R. S.

Flow and reactions in permeable rocks

O. M. PHILLIPS, F.R.S.

DECKER PROFESSOR OF SCIENCE AND ENGINEERING
AT THE JOHNS HOPKINS UNIVERSITY



CAMBRIDGE UNIVERSITY PRESS

CAMBRIDGE

NEW YORK PORT CHESTER MELBOURNE SYDNEY

Published by the Press Syndicate of the University of Cambridge
The Pitt Building, Trumpington Street, Cambridge CB2 1RP
40 West 20th Street, New York, NY 10011, USA
10 Stamford Road, Oakleigh, Melbourne 3166, Australia

© Cambridge University Press 1991

First published 1991

Printed in the United States of America

Library of Congress Cataloging-in-Publication Data

Phillips, O. M. (Owen M.), 1930–

Flow and reactions in permeable rocks/O.M. Phillips.
p. cm.

ISBN 0-521-38098-7

1. Fluid dynamics. 2. Rocks – Permeability. I. Title.

TA357.P48 1991

90-44208

552 – dc20

CIP

British Library Cataloguing in Publication Data

Phillips, O. M. (Owen Martin) 1930–

Flow and reactions in permeable rocks.

1. Hydrology

I. Title

551.48

ISBN 0-521-38098-7

To
Francis Pettijohn
and to the memory of
Hans Eugster

Preface

The aim of this book is to provide an account of the patterns of flow through permeable rocks and rock structures in the earth, patterns whose imprint is reflected in the distributions of mineralogical changes that have taken place over eons past. The nature of the changes themselves, including postdepositional formation of ore bodies and hydrocarbon deposits, is the proper concern of mineralogy and geochemistry, but the patterns, and in many instances the rates, of formation have been controlled by the flow, whether episodic and relatively rapid or continuing but slow, that occurred over these time intervals. Though the topic of flow through permeable media is rooted in the preceding century, its application to geochemical and mineralogical questions is fairly recent. Contributions to this area of inquiry are scattered in the geological, hydrological, and fluid mechanics literature. Some are important and penetrating; others, alas, needlessly obscure, misleading, or irrelevant. Part of the material presented here is being published for the first time, and much more remains to be discovered. This book is offered in the hope that it will provide a basis for future work or possibly a target for dissent; in either case understanding will be enhanced.

It is intended primarily for geologists, especially the well-trained graduate students who are emerging today, though I am conscious that it may present conceptual and technical difficulties for those whose background is more deeply geochemical than is mine. The subject necessarily involves a level of mathematics that may be uncomfortable to many geologists, but for those with a recent undergraduate background to the level of differential equations and vector calculus and with a spirit of mathematical adventure, I hope it will not be daunting. In the analytical developments, I have tried to eschew mathematical generality for its own sake, seeking instead to be specific to the matter at hand. Sufficient detail is given in the derivations that the logical steps can be followed with perhaps moderate effort or, for those prepared to trust, skipped over. Students of hydrology or the nascent area of geological fluid mechanics may, in contrast, find the discussion of basic flow properties elementary and unnecessarily detailed, but I must beg their patience until a common basis of understanding is developed. In an area such as this, the greatest rewards will come by a combination of insights from disciplines usually

studied separately, and what is elementary to some may be complex to others.

The broad subject of fluid mechanics has always involved the art of approximation, and this context provides no exception. Approximations must, however, be firmly based and clearly specified – there is little place for fanciful models that have sometimes intruded into the geological literature. In discussing the results, I have tried to explain in words not only what they mean, but also what geometrical simplifications and dynamical approximations have been involved and under what conditions they apply. To use results in an inappropriate context is to invite confusion, but if the context is clear, the ultimate reward can be the development of an understanding, an intuition, a feeling for the role that physical and chemical processes associated with flow have had, and are having, in remaking the rocks beneath us, what the flow can and cannot do, what patterns it can produce, and where to look for its manifestations.

In writing this book, I have relied heavily on the guidance and advice of many colleagues, friends, and students who listened with patience, criticized thoughtfully, and pointed in new directions. In particular, I must thank Lucas Baumgartner, Edwin Clawsey, John Ferry, Grant Garven, Gu Daifang, Lee Hagee, Lawrence Hardie, Eugene Ilton, Barry Lester, Sakiko Olsen, Haydee Salmun, Dimitri Sverjensky, and Edith Wilson. It was James R. Wood who, quite some time ago, suggested the need for such a book and who gave generously of his time and his deep insights in reviewing an earlier draft. I am greatly indebted to Elizabeth Hendrickson for her skillful preparation of the manuscript and for her endless patience in revision, revision, and revision again. Most of all, I am grateful to Francis Pettijohn and the late Hans Eugster, from whom I have learned so much and to whom, and to whose memory, this book is dedicated with affection and respect.

Baltimore
September 1990


O.M.P.

Contents

Preface

page ix

1	Introduction	1
2	The governing principles	5
2.1	The physical nature of permeable rocks	5
2.2	Geometrical characteristics	11
2.3	The motion of fluid elements through the medium	13
2.4	Dispersal and diffusion of dissolved species	18
2.5	The transport velocity and the incompressibility condition	22
2.6	Darcy's law	25
2.7	Permeability k	29
2.8	The thermal energy balance	34
2.9	Dissolved species balance	35
2.10	Equations of state	38
3	General flow characteristics: patterns of flow	41
3.1	Stream functions	41
3.2	Natural or intrinsic coordinates	43
3.3	Boundary conditions for flow	45
3.4	Vorticity	47
3.5	Dynamical scaling	53
3.6	Uniqueness and minimum dissipation theorems	59
3.7	Geometrical scaling	61
3.8	Flow patterns near isolated lenses or cracks	67
3.9	Long and multiple lenticular layers	74
3.10	Flow transients	80
3.11	Diffusion-induced flow in fissures or fractures	87
4	General flow characteristics: patterns of reactions	90
4.1	Simple reaction types	90
4.2	The approach to equilibrium at dissolution or reaction boundaries	96
4.3	Isothermal reaction fronts	100
4.4	Gradient reactions	107
4.5	Mixing zones	112

4.6	Mixing and reaction patterns from faults and fractures	115
4.7	Enhancement and destruction of porosity	125
4.8	Isotherm-following reactions	130
5	Instabilities	140
5.1	Rayleigh–Darcy instability	140
5.2	Salinity stabilization	148
5.3	Double-advective, double-diffusive instabilities	152
5.4	Brine invasion beneath hypersaline lagoons by double-diffusive fingering	159
5.5	Stability in the presence of flow	161
5.6	Saffman–Taylor instability	162
6	Pressure-driven flows	166
6.1	Flow in confined aquifers	166
6.2	Flow in unconfined aquifers	176
6.3	Computation of three-dimensional basin-wide flows	182
6.4	Temperature distributions	185
6.5	Mixing zones	201
6.6	Trapping of hydrocarbons	205
6.7	Spatial distributions of mineralization associated with flow	211
7	Thermal convection 	220
7.1	The occurrence of thermally driven flows	220
7.2	Submerged banks of slowly varying thickness with small Rayleigh number and scale ratio	223
7.3	Convection and diagenesis in sloping isolated permeable strata	232
7.4	Compact platforms or reefs at low Rayleigh numbers	236
7.5	High Rayleigh number plumes	244
7.6	Flow, temperature, and gradient reaction patterns at intermediate Rayleigh number and scale ratio	252
7.7	Paleoconvection and dolomitization in the Latemar Massif	265
	<i>References</i>	271
	<i>Index</i>	279

1 Introduction

It has been evident since the days of James Hutton in the late eighteenth century that this solid earth has been in upheaval throughout a history longer than could easily be visualized. Recurrent and overlapping cycles of igneous intrusion and deposition, deformation and folding, with erosion by air and water, redistribution and sedimentation have shaped the surface of the earth and hinted at great tectonic forces within. The grandeur of the canvas painted by geology, the science of the earth, has excited the imagination of those who have the capacity to wonder and has drawn in the talents of biologists, physicists, and chemists such as Darwin, Rayleigh, Jeffreys, Elsasser, Urey, and many more in the preceding century and this. New discoveries in the basic sciences and new techniques of observation have again and again revolutionized our view of the earth – it has been barely twenty years since measurement of magnetic anomalies over midoceanic ridges forced the acceptance as reality of the process of sea-floor spreading leading to continental drift, a notion previously regarded as close to heresy.

But no more. Abundant evidence now indicates that the crust of the earth is spreading from the midocean ridges that gird the globe and is disappearing in the subduction zones of the deep ocean trenches carrying the continental blocks in a grandly and gradually changing pattern. Deep-mantle convection is presumably responsible, with material solid enough to transmit seismic S-waves behaving also as a gradually deforming fluid. The worldwide distribution of volcanism is intimately associated with these tectonic motions. In recent years, the fluid behavior of magmas, igneous intrusions, and melts has drawn the attention of such imaginative and powerful scientists as Herbert Huppert, Dan McKenzie, and Stewart Turner, and this field is sparkling with new ideas.

As applications of physics and chemistry brought new insights to geology, they also posed new questions. The development of geochemistry in the past fifty years has led to the clarification of many aspects of metamorphism, the term used to describe the transformation of minerals from one form to another at depth under conditions of high temperature and pressure. Volatiles may have had to escape so that the reaction would not reverse itself, but there has been plenty of time to accomplish this. Yet a number of mysteries remain. Natural processes are, in the large, dispersive, as the second law of thermodynamics insists. How is it that local

concentrations of minerals aggregate deep inside the earth, leading to the accumulation of economically valuable resources? The Mississippi Valley lead–zinc deposits were clearly formed after the lithification of the host rock, but where did they come from, how did they get there, why did they deposit where they did and not somewhere else? The dolomitization of limestone requires the importation of large quantities of magnesium; in these and other geochemical processes there is an evident need to account for the transport of minerals, presumably dissolved, through existing assemblages in quantities much larger than can be associated with compaction of a fluid-saturated matrix and at rates much faster than can be produced by diffusion. No reader of Francis Pettijohn's *Sedimentary Rocks* can remain unaware of the puzzle, but only fairly recently has the idea gained acceptance that these chemical transports must be associated with fluid flow through rocks that a hydrologist may consider hardly permeable, over time intervals of millions or hundreds of millions of years. When pressure or buoyancy forces are present, the latter usually associated with temperature gradients, interstitial fluid may flow through the matrix if it is reasonably permeable or along cracks and fractures if it is less so. What are the large-scale and small-scale patterns of flow? In which direction does the fluid move, how fast, where from, and where to? What governs the distributions of reaction, of dissolution, and of deposition that, over eons, led to the distributions of mineralization that we see today?

The search for answers to these questions involves a combination of structural geology, which defines the geometry and characteristics of the matrix; geochemistry, which specifies the nature of the reactions that can occur; and fluid mechanics, which governs the flow and transport of dissolved chemicals from one place to another. The fundamental rules of fluid percolation through rocks or other permeable media have been known since Darcy's time, but the variety of flow patterns in geological structures and their textural and geochemical consequences are far from having been fully explored. The ranges of pertinent parameters are often extreme – the permeability of an evaporite bed may be ten orders of magnitude smaller than that of a gravel bank. Even in one material, limestone, field measurements may indicate a permeability five orders of magnitude larger than that inferred from laboratory samples because of mesoscale fracturing or dolomitization. Chemical reactions may proceed almost instantaneously or take thousands of years to approach equilibrium. In many instances, the appropriate parameters are simply not known even to an order of magnitude, and this provides an opportunity and a trap – an opportunity in that careful analyses of the consequences of flow can reduce or place limits on the uncertainty, and a trap in that one can often “pick a number” within a wide range to give any desired numerical agreement.

Even the basic three-dimensional geometrical structure of a basin is

seldom known precisely. Much of the information in structural geology has been derived from two-dimensional exposures and surficial mapping. Drilling is expensive and cores give only widely separated vertical traverses. The techniques of seismic reflection now make it possible to obtain much greater detail, though even in well-explored regions, the disciplined imagination of the structural geologist is still a necessity. The difficulties are compounded as we reach farther into the past. Not only have the geological structures evolved with the relentless sequence of tectonic upheaval, erosion, and deposition, with the consequent evolution of the patterns of flow, temperature, and pressure, but also the physical and chemical nature of the rocks themselves has changed with metamorphism, dissolution, and cementation. The correct interpretation of these changes requires all the skill of the geochemist and the petrologist, and an appreciation of the influence of interstitial flow is but one of the tools that must be employed. It is an important one, though; flow provides fluxes of chemical species, slow but enduring throughout geological time, that allows reactions to continue and petrological changes to accumulate.

It is not the purpose of this book to construct a detailed model of the flow-associated changes that have occurred in any particular basin or other geological structure, but rather to develop, from the basic physical and chemical balances, a set of derived or secondary rules that can guide geological interpretation in a variety of contexts. We seek associations among the general characteristics of geological structures, the characteristic patterns of flow that they allow, and the patterns and rates of physical and chemical changes that occur as a consequence and are reflected in the distributions of mineralization. A number of examples that exhibit these connections are given, but they are far too few. Ideally, a development such as this would be guided by geochemical flow experiments in permeable media, but this would require extrapolation from achievable laboratory flow rates, time scales, temperatures, and pressures to those that occur deep in the earth over millions of years. A number of important experiments by Elder and others, however, have illustrated aspects of convective flow patterns in some simple geometries. The art of numerical simulation of basin-wide flow, temperature, and reaction patterns has developed rapidly in recent years, notable contributions having been made by Garven and his collaborators, but these inevitably require a specific configuration and choice of the numerical values of the parameters involved; it is not always clear how specific features of the solution depend on these choices. The results presented here can, nonetheless, provide insights by relating these features to the local or overall flow characteristics. The two approaches are, indeed, complementary. A conceptual framework elucidating the relations among flow characteristics, driving forces, structure, and reaction patterns enables us not only to understand the results of numerical modeling more clearly, but to check them. (Nu-

merical calculations *can* converge to a grid-dependent limit, and artifacts of a solution *can* be numerical rather than geological.) Numerical modeling provides a quantitative description and synthesis of a basin-wide flow in far greater detail than would be feasible analytically. The combination of the two techniques is a much more powerful research tool than either alone.

In the next chapter are specified the general geometrical characteristics and basic physical and chemical balances that underlie the developments in the remainder of the book. These are the rules by which the game is to be played, but within these rules, many kinds of play are possible. Chapters 3 and 4 develop a variety of these and illustrate the closeness of the relationships among structure (the distribution and characteristic geometry of permeability variations), flow within the structures, and distributions of chemical reactions of several types associated with this flow. When the interstitial fluid viscosity is uniform and buoyancy effects are negligible, the flow solutions are unique, but if temperature or salinity variations are significant, a given initial distribution may be dynamically unstable and evolve into some other. Some conditions for stability or instability are illustrated in Chapter 5 by typical geological examples. The two great driving forces for large-scale circulations through rocks are pressure differences, hydraulically induced, and variations in basement temperature or heat flux; their flow, thermal, and mineralogical signatures are described in the last two chapters, again by means of several examples in which direct observations or numerical experiments are sufficiently complete to make comparison possible. One again laments that there are not yet enough examples such as these; it is to be hoped that in future years more will become available to provide support (or otherwise) for existing results and point in new directions. Only thus can this area of geology advance.

2 The governing principles

2.1 THE PHYSICAL NATURE OF PERMEABLE ROCKS

The geological materials with which we are concerned usually lie at one extreme of the range of “porous media” encountered in nature and technology. The porosity, the volume fraction of connected voids that allow fluid movement, may approach unity in slag-wool insulation material; it may be as large as 0.4 or 0.5 in a well-sorted sandbank (Pryor, 1973). The skeletal remains of corals that abound in tropical reefs contain myriad interstices on scales of up to a centimeter or so and may have a similarly large porosity; Figure 2.1 shows a sample from Bermuda at approximately half-scale. This kind of structure, however, is at the high-porosity extreme of those generally encountered. Compaction by overlying sediments, the infilling of interstices by finer grains, and the precipitation of cements from solution can reduce the porosity by an order of magnitude or more and reduce the permeability, as we shall see, by four orders of magnitude or more.

Still highly permeable is the partially cemented sandstone of Figure 2.2. The largest interstices are about 1 mm in diameter, and the granular texture, containing many finer pore spaces, is clearly evident. Many large pores are also apparent in dolomite from the Latemar Massif, northern Italy (Figure 2.3). Calcium ions from the original calcite mineral have been replaced by magnesium; the specific volume of the dolomite produced in the reaction is less by 3 to 13 percent than that of the original calcite, so that as the reaction proceeded, the porosity increased.

Networks of small cracks or fractures allow fluid percolation even when the matrix itself is relatively impermeable. Seepage from fractures can often be discerned in roadside rock exposures. Figure 2.4 illustrates a smaller-scale network, made visible by staining, intersecting a sandstone cleavage plane. The result of fluid movement along interlacing networks of pathways on a smaller scale again is shown in Figure 2.5. Natural preferential weathering on the face of the specimen has left micropathways of dolomites standing in relief against the unaltered calcite as a dense network on the left and along what was once a single crack on the right. The stained, thin-section photograph of Figure 2.6 also shows dolomitization along micropathways in a partially dolomitized transition

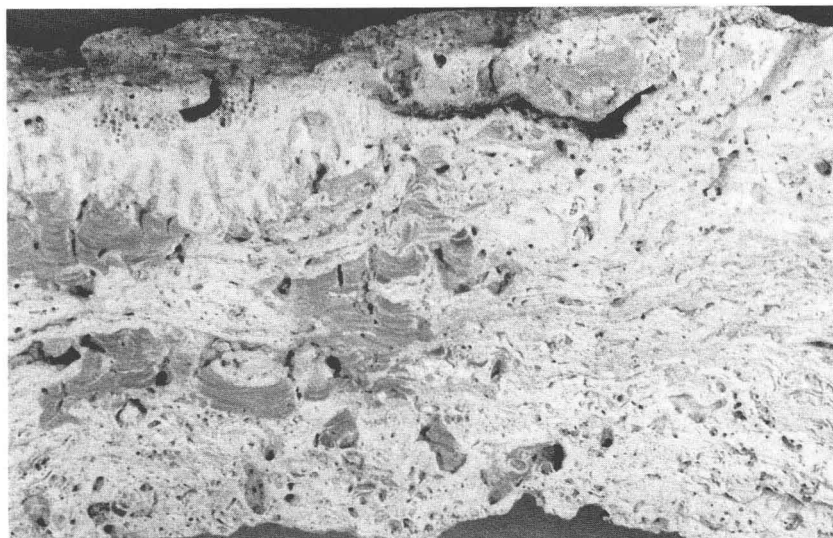


Figure 2.1. Extremely porous limestone from a Bermuda coral reef, approximately half-scale, containing many interstices with scales of up to a centimeter or so.

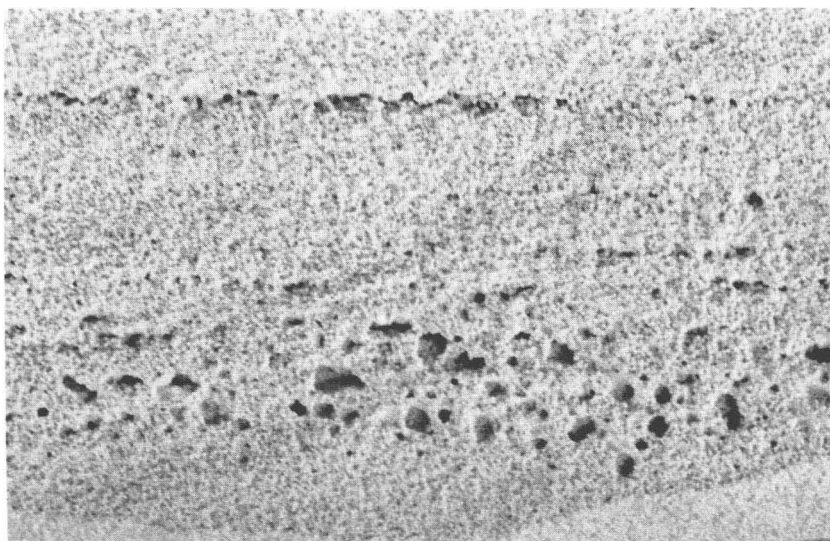


Figure 2.2. A highly porous, partially cemented sandstone, approximately full scale.

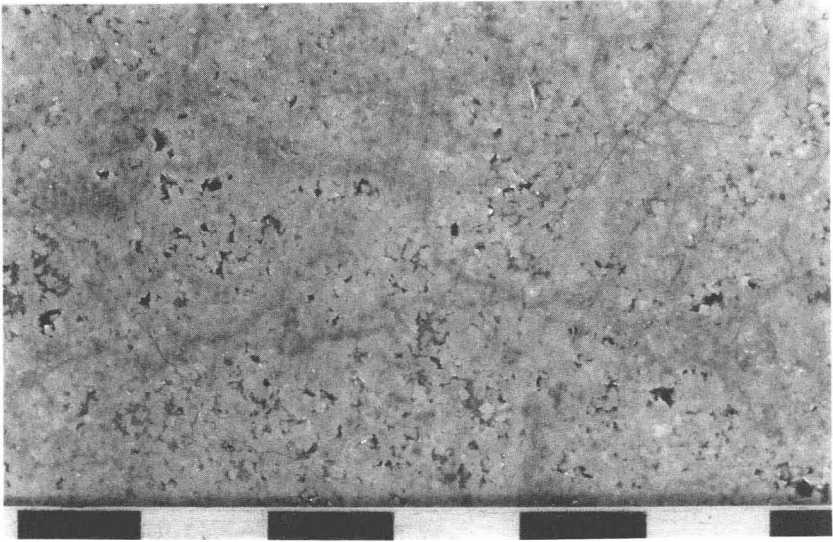


Figure 2.3. Pores in dolomite from the Latemar Massif in northern Italy. The blocks in the scale are 1 cm long. (Photograph courtesy of Dr. E. N. Wilson.)

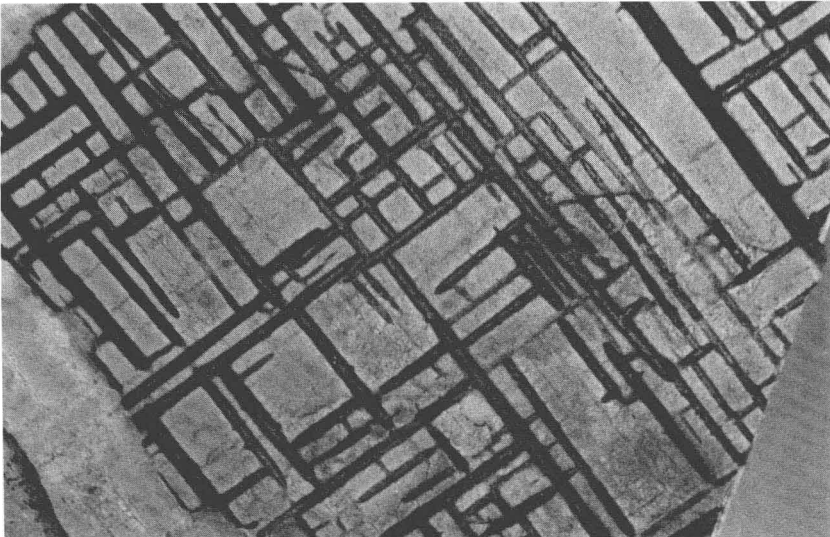


Figure 2.4. A network of cracks, made visible by staining, that provide pathways for flow along a sandstone cleavage plane. Approximately full scale.



Figure 2.5. The weathered face of a partially dolomitized calcite rock in which veinlets of dolomite produced along fluid pathways stand out in relief in a dense, fine tracery on the left and along a crack on the right. (Photograph courtesy of Dr. E. N. Wilson.)

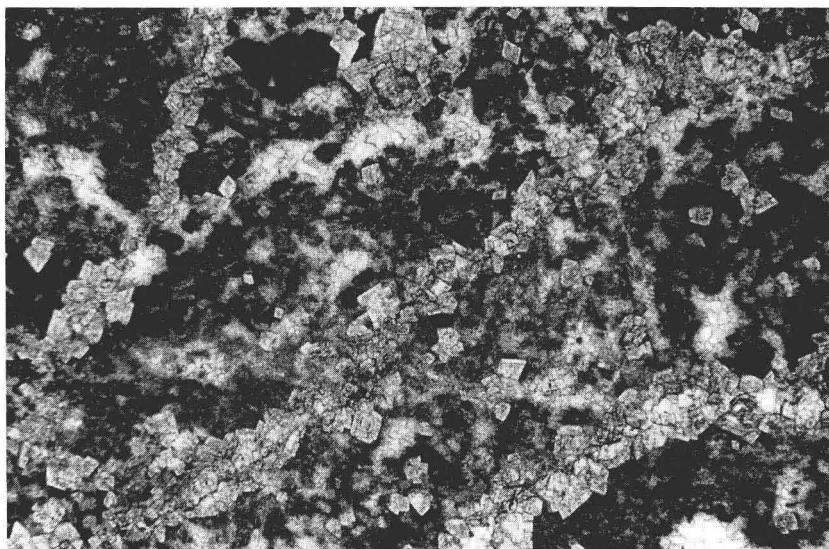


Figure 2.6. Dolomitization along linear pathways (the strings of lighter rhombic crystals) in a calcite matrix. Magnification $\times 20$. (Photograph courtesy of Dr. E. N. Wilson.)

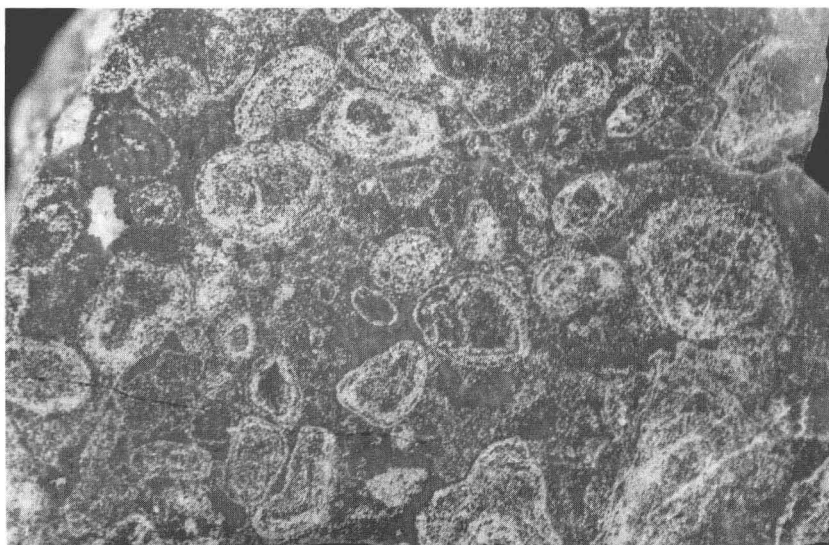


Figure 2.7. Dolomitization around the edge of grains (light) in a partially dolomitized grainstone, indicating a generally pervasive fluid flow. Full scale. (Photograph courtesy Dr. E. N. Wilson.)

zone – the lighter rhombic dolomite crystals form a continuous network against the darker calcite background.

When the interstitial seepage is more pervasive, dolomitization (and fabric alteration in general) becomes more uniform and ultimately more complete. Figure 2.7 shows a polished and stained slab of subtidal grainstone in which only the edges of the grains have been dolomitized (again they show as light). In the photomicrograph of Figure 2.8, the lighter dolomite has replaced the darker calcite almost completely in the region on the right, though calcite central cores remain unaltered in many of the grains. Figure 2.9, at higher magnification, shows a tiny fracture, now filled with calcite cement, in a matrix of dolomitized grainstone. A pore, intersected by the fracture, has also been filled with calcite. The porosity of some of these samples is now extremely small, but evidently, during a previous time interval, it was sufficient to allow the movement of solutions from which the cements precipitated.

Fault systems also provide conduits for fluid motion. Sibson (1987, 1989) has pointed out that dilatational fault jogs in particular can open up a network of fractures that immediately draws in surrounding interstitial fluid and subsequently provides pathways for continuing fluid motion. If cementation occurs, a network of veins through the original fabric ultimately results in a fabric typically characterized, according to Sibson,