Canadian Mineralogist
Vol. 12, pp. 439-444 (1974)

LOW-GRADE METAMORPHISM: SOME THOUGHTS ON THE PRESENT SITUATION*

W. S. FYFE
University of Western Ontario

Abstract

The development of concepts concerning low-grade metamorphism is reviewed. The part these rocks play in the water inventory of the earth is stressed. The low-grade environment is one involving massive fluid flow and metasomatism. It is suggested that ocean volumes may be slowly decreasing with time due to the increasing volume of low-grade rocks.

Introduction

As the first speaker of a symposium on low-grade metamorphism, I felt that part of my task was to review a little of the history of the subject. I think I was chosen for this task partly because I came from New Zealand, and partly because I had the great good fortune of knowing, being taught by, or working with Douglas Coombs, F. J. Turner and the late Colin Hutton. It would certainly have been more fitting if Coombs had started this symposium, because, in large part, he initiated so many of our modern ideas on low-grade metamorphism.

As a student of chemistry I had become interested in zeolites at an early stage. I recall giving a seminar on zeolites and reviewing the elegant research of another New Zealander, R. M. Barrer, who had published a series of papers in the Journal of the Chemical Society on the synthesis of zeolites and a series of papers in the Trans. Faraday Society on molecular absorption by zeolites. Barrer had shown that one could take these low-density framework silicates, particularly those with continuous channels, remove the water and introduce many types of organic molecules into the holes. On account of the channels having a geometry which could be modified by cation exchange, he prepared useful "molecular sieves". Today zeolites and organic analogues are used on a large scale for such purposes. Recently, Munson & Sheppard (1974) have reviewed the occurrences and uses of natural zeolites. I suppose I was also fortunate in being born under the Southern Alps of New Zealand in the region of the Erehwon (nowhere) country of Samuel Butler, most of which involves low-grade metamorphism of a vast greywacke pile.

Modern metamorphic studies in New Zealand began with Turner (*e.g.* 1948) and Hutton. Hutton's 1940 N. Z. Geological Survey memoir must still be considered a classic in the description of the development of the greenschist facies. Turner was more concerned with higher grades and structural aspects of the problem. Then Coombs, whom I had first encountered as a demonstrator in physics, went to Cambridge with a very drab collection of rocks from the Taringatura district of Southern New Zealand. Under the supervision of the late C. E. Tilley, and with his meticulous eye and fertile imagination, he appreciated that these "unmetamorphosed rocks" were of considerable interest. Thus in 1954 he published the classic which was to establish the zeolite facies, the first major development since Eskola. Slightly before this, Steiner (1953) had described the significance of low-grade metamorphism in the Wairakei hydrothermal regions of New Zealand.

In essence, Coombs had found that the mineralogy of a thick (10 km+) section of volcanic debris was dominated by minerals of the zeolite-clay-chlorite-prehnite groups. His diagram (Fig. 1) summarizes much of the petrological reactions. One must conclude that if, in this region, the thermal gradient was of the order of 30°C km^{-1}, then this type of mineralogy must represent changes which join the surface rocks to the then well-known rocks of the greenschist facies. Further studies were to show the importance of prehnite-pumpellyite rocks between zeolite-dominated assemblages and the normal albite-epidote greenschists. With these studies (see Fyfe, Turner & Verhoogen 1958) a more or less complete metamorphic history could be described from the sediment interface into rocks with well-developed metamorphic fabrics (Fig. 2).

It is interesting to reflect on the situation which preceded the new developments. If one examines the classic work of Harker (1939), it is clear that the factors considered to be dominant in metamorphism were pressure

---

* Summary of a lecture delivered at the MAC symposium on low-grade metamorphism, May 1974.

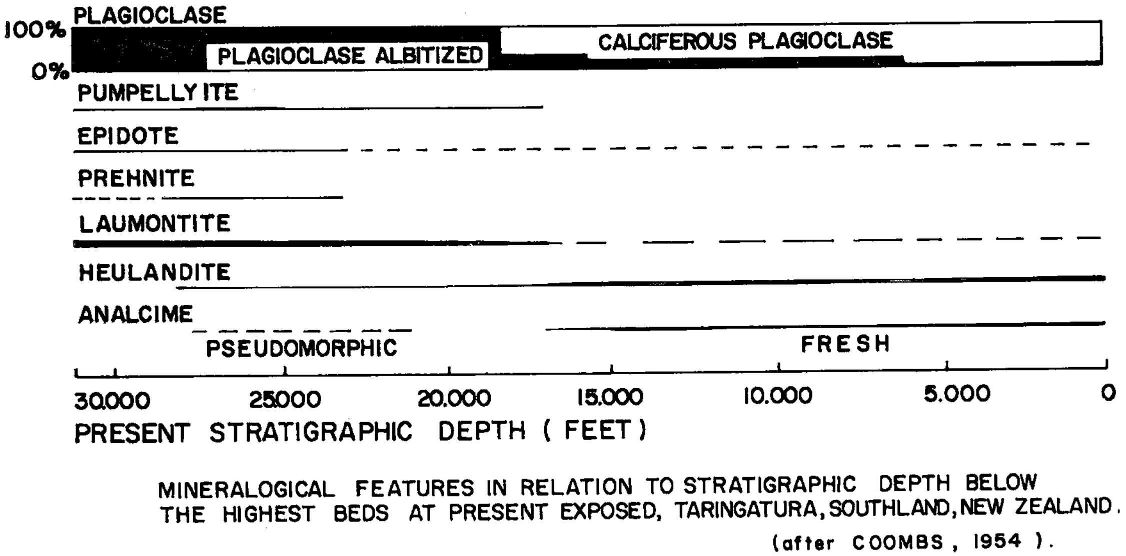

FIG. 1. The basic low-grade alteration pattern described by Coombs (1954).

(depth), temperature and stress. Stress was revealed in mineral and rock fabrics. Metamorphic intensity or grade was largely a function of temperature. The low-grade rocks were the greenschists. With the rocks described by Coombs, and equivalent more recent descriptions from around the world, the zeolite facies rocks frequently lack obvious metamorphic fabrics. The absence of fabric in otherwise fully recrystallized rocks is an observation requiring explanation (see below). In this discussion I shall consider low-grade rocks to be those crystallized below about 400°C. As the crust must partly melt in the range 700-800°C, low-grade rocks are thus involved in the upper 50% or so of the normal crust; too much to be ignored.

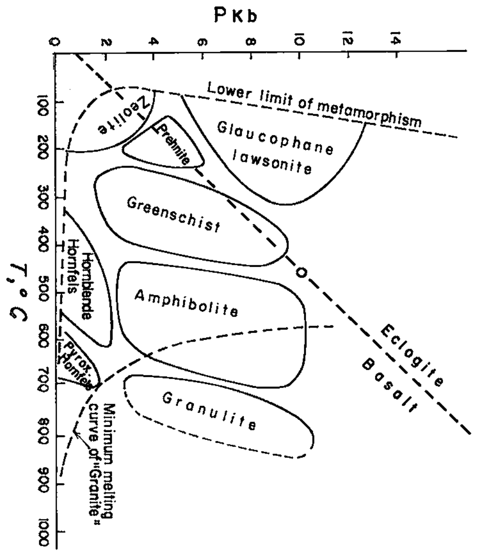

FIG. 2. Common conditions of formation of metamorphic facies (after Verhoogen *et al.* 1970). For most facies $P{=}P(H_2O)$. For eclogites and granulites $P{>}P(H_2O)$. The eclogite field is not known well at low temperatures.

Thermal gradients in metamorphic rocks range from 10°-1000°C km^{-1}. As pressure increases by about 250 bars km^{-1}, temperatures of 400°C may be encountered over a depth range of 40 km to much less than 1 km (the latter condition is probably common near the ocean ridges).

Rocks of the low-grade facies (zeolite, prehnite and greenschist) are now known from many places in the world (Munson & Sheppard 1974; Dobretsov *et al.* 1973). The best-known examples come from the more recent mobile belts, hydrothermal areas, and layer 2 of the ocean floor. They occur in rocks of all ages and it now appears are widely developed in the Archean (L. Jensen, personal communication and W. Jolly, this issue).

The most spectacular development of zeolite assemblages may be in igneous, partly glassy, volcanic rocks of all compositions. Zen (1961) has stressed that carbonate-rich sediments are frequently rather unreactive in the low-grade environment. Thus in a volcanic rock of low carbonate content, anorthite in plagioclase may react as:

$$\underset{\text{anorthite}}{CaAl_2Si_2O_8} + 2SiO_2 + 4H_2O \rightarrow \underset{\text{laumontite}}{CaAl_2Si_4O_{12}\cdot 4H_2O}$$

The equivalent sedimentary reaction:

$$Al_2Si_2O_5(OH)_4 + CaCO_3 + 2SiO_2 + 2H_2O \rightarrow CaAl_2Si_4O_{12}\cdot 4H_2O + CO_2$$

will rarely be more than incipient. The latter type of reaction may only proceed if CO_2 is continuously flushed from the system, and as the equilibrium $P(CO_2)$ of such a reaction is very low, a massive fluid volume would be required. Thus the clay-carbonate assemblages are the carbonate facies equivalent of the volcanic zeolite assemblage (see Ghent & Miller, this issue).

Such observations show that during metamorphism, gases diffuse ensemble, not as separate species. Minor species, or species at low partial pressures (CO_2, H_2, O_2, *etc.*) move with the bulk fluid which, except in rare cases, is dominated by water.

EXPERIMENTAL STUDIES

When it became obvious that low-grade rocks must record much of the metamorphic-tectonic history of the crust, Coombs, Turner and myself became interested in experimental studies concerned with the stability of the important phases. We were supported by a group of able students including A. J. Ellis, A. M. Taylor and later A. B. Thompson. In this work it became obvious that many of the established methods of studying phase relations simply would not work. Approaches based on the crystallization of unstable materials led to results that bore little relation to natural reality. Thermodynamic data were not available and in any case, too inaccurate to be useful.

All mineral reactions involve a series of rate processes dominated by steps of solution, diffusion, nucleation and growth. The slow steps tend to be nucleation and growth. But even at low temperatures, solution rates are measurable. We were thus forced to develop methods based on growth and solution. These proved to be extremely useful even at very low temperatures (Fyfe & Hollander 1964; Coombs *et al.* 1959; Thompson 1971). An advantage of these methods was that it was not necessary to use ultrafine unstable materials. Thus the study of low-grade reactions led to significantly improved methods for the study of many aspects of mineral equilibria. Further, the fact that the reactions could be studied at all in laboratory time, indicates that in nature there should be a reasonable approach to equilibrium.

WHY STUDY LOW-GRADE ROCKS?

As the general patterns of low-grade metamorphism have become well-established in rocks of all ages, why should studies still attract attention? What are the important problems now? To this writer, these rocks are of immense importance to our understanding of tectonic and geochemical history of the crust in that they are involved in the most massive dehydration (and hydration) processes in the crust. The low-grade rocks in the top 10 km or so of the crust carry the bulk of crustal water and other volatiles and a mass which is comparable in magnitude to the hydrosphere.

Surface sediments and volcanic debris contain large amounts of water (20-90%) (Blatt *et al.* 1972) in pore fluids and hydrated minerals. The same rocks may fix large amounts of carbon dioxide and sulphur in sulphides and even sulphates and because of atmospheric influences tend to be more oxidized than primary igneous rocks.

In the upper zones of low-grade or burial metamorphism, few rocks will contain less than 20% water. Thus for each km^3 of rock (about 2.2×10^{15} g) something like half a km^3 of water will be lost by the time the rock becomes a greenschist. In general, pore fluids are salty and become even more saline as metamorphism proceeds through filtration equilibria.

Even if the released fluids carry dissolved species at the ppm level, the geochemical effects may be massive. Many studies (*e.g.* Nriagu & Anderson 1970) have shown the transport of the minor elements by salty fluids. Such effects will be best seen in the very rare elements; metals like silver, gold *etc.* which form stable halide complexes. At this time, we are only beginning to appreciate the importance of such geochemical influences of massive low-temperature water flow back to the surface. Much of the evidence on the entire subject of crust-hydrosphere interaction must be contained in the low-grade rocks.

THE FLUID PRESSURE REGIME

As noted above most of the rocks involved in low-grade metamorphism are wet and most of the water must be removed by the time the rocks attain a temperature of 400°C. Porosity and permeability are rapidly lost with burial, partly due to solution of grains at regions of high stress concentration. Data from drilling indicates that at small depth (ρ = density):

$$P_{\text{fluid}} / P_{\text{rock}} \simeq \rho_{\text{fluid}} / \rho_{\text{rock}}$$

whereas at depths of a few kilometers

$$P_{\text{fluid}} \simeq P_{\text{rock}}$$

The latter condition would be anticipated during the dehydration of a rock with very low permeability. In a sense, water "fights" its way out of the rock by lifting the load.

Many lines of evidence support the general conclusion that during most prograde metamorphic processes (but not those involving granulites and eclogites) conditions are such that:

$$P_{\text{fluid}} \simeq P_{\text{load}} \simeq P_{\text{water}} = f(\text{depth}) = f(\text{temp.})$$

where f=function. Without such dependence of variables, metamorphic patterns would be chaotic.

Certain mineral equilibria are particularly sensitive to variation of fluid pressure and total pressure. Among low-grade reactions, the reaction:

$$\text{analcime} + \text{quartz} \rightarrow \text{albite} + H_2O$$

provides an excellent example. The conditions of this reaction have recently been considered by Thompson (1971). Because of the low density of analcime and the large entropy of water in the zeolite, the stability field with respect to albite is virtually eliminated if water pressure and total pressure are not similar. The observed occurrence of analcime (Coombs 1954) and the experimental data can only be reconciled if fluid pressures are large (see Fyfe 1973).

During dehydration processes in rocks with very small porosity, the general conditions will be:

$$\text{Volume}_{\text{hydrate}} < \text{Volume}_{\text{product}} + V_{H_2O}$$

Local transient expansion must occur. If the process is fast, a high local stress may be developed and this may lead to development of hydraulic fractures (Jaegger & Cook 1969). A rock body in the process of massive dehydration may thus develop a high microfracture density and may be envisaged in the mechanical sense as a quicksand. The large water content of low-grade rocks and the tendency to develop fractures may well have some bearing on the problem of the lack of mineral fabrics and schistosity.

OCCURRENCE OF LOW-GRADE ROCKS

The common situations of pervasive low-grade metamorphism are well-known and include:

(a) Mobile belts associated with the rapid accumulation of volcanic debris in a subduction environment. Examples can be drawn from most modern mobile belts (*e.g.* the Pacific margin). The key factors involved include large contributions from volcanic materials of a wide range of compositions and rapid burial and uplift. Rapid burial may cause a lower-than-normal thermal gradient.

(b) The ocean flow. Basaltic rocks of layer 2 materials of the ocean floor frequently contain rocks in the zeolite, prehnite and greenschist facies. The hydration reactions involved may be induced by convective water flow through the upper layers driven by shallow intrusions (Spooner & Fyfe 1973). In the near-ridge environment thermal gradients may be very steep and all low-grade facies may be represented in a small vertical thickness. As rocks leave the ridge environment, reactions may continue (Salisbury & Christensen 1973). The dominant reactions include hydration, addition of CO_2, O_2 and S to basaltic material. If fluid motion is slow, the reactions are highly exothermic and may contribute to oceanic heat production (Fyfe 1974). Studies of oxygen isotopes have shown that very large water volumes may be involved in the low-temperature alteration (Spooner *et al.* 1974). The product hydrated and metasomatized basalts (spilites) normally show massive chemical changes (see Hyndman 1972). In these rocks, metamorphic fabrics are seldom well-developed.

(c) Hydration envelopes near high-level plutons. A common environment for extensive development of low-grade rocks is that well-illustrated by hydrothermally active regions like Wairakei, New Zealand. If a rising pluton comes close to the surface such that the fracture system caused by its upward motion intersects the hydrosphere, then, as at the ocean ridge, a convective cooling system may be induced. The general tectonic situation is tensional and rocks tend to have poorly developed mineral fabrics.

The energy from a pluton available to drive fluid flow is impressive. One km^3 of liquid granite at 800°C, can heat 5 km^3 of ground water to about 100°C. Many plutons have volumes of the order of 500 km^3. Just as in case (b), ore deposits are probable along with the extensive leaching of rocks.

(d) The subduction environment. The hydrated igneous materials of the new ocean crust must be almost quantitatively subducted or else the crust would thicken at a rate much faster than presently considered possible. As well, large volumes of sediment may be subducted or at least dragged down to form blueschists. Such subducted water must return, for if it did not, the oceans would be removed in a few hundred million years.

Because of the rapid rate of subduction of cold surface rocks, temperatures in the upper portions of the slab may not reach 400°C until depth of the order of 40 km. This is the environment for high pressure low-grade metamorphism. In such rocks zeolite facies assemblages may pass into those of the blueschist facies. Fluids liberated in this high-pressure environment may be relatively rich in many cationic species and promote for example, the intense soda metasomatism reported by Fyfe & Zardini (1967).

HYDRATION REACTIONS

Because many rocks of the surface environment are volcanic, their low-grade metamorphism may involve a massive hydration process. With many volcanic sediments it is likely that early alteration processes and pore fluids will be available to allow total development of hydrate phases in the greenschist facies (Hutton 1940). But with dense basaltic materials or massive basic and ultrabasic intrusions, hydration may be slow and not fully developed at shallow levels. It is from such dry rocks that eclogite facies mineralogy may develop within greenschist or blueschist environments. Spectacular examples are to be found in the Alps (Bearth 1959) where pillow lavas are partly recrystallized to eclogites. In these rocks hydrated minerals and eclogite may be in disequilibrium and the rate of attainment of equilibrium is diffusion-controlled (Fry & Fyfe 1971). From these rocks and an approximate knowledge of water pressures in the hydrated assemblages it is possible to derive water (or hydrogen) diffusion coefficients of the order of 10^{-15} cm^2 sec^{-1}, values similar to those for any inorganic species in crystalline materials. It would appear that rapid fluid penetration of rocks requires some degree of permeability, possibly by generation of fractures.

THE WATER INVENTORY

One of the old and difficult problems of geology is to ascertain how ocean volumes have changed with time. In this context I think we have overlooked the importance of metamorphic rocks. The present ocean mass is about 1.4×10^{24} g; the mass of the crust 2.3×10^{25} g. For the crust to contain a volume of water equal to that of the oceans, the average water content must be 6%. The real average for the crust might be more like 4% and much of that water will be in the rocks below 400°C!

While low-grade rocks are present in the Archean they are best-developed in the most recent mobile belts. In fact, the Archean is characterized by the common occurrence of very high-grade, dry rocks of the granulite facies. Modern-type plate tectonics, and possibly a cooler earth, appear to favor thick piles of low-grade rocks.

If the earth is cooling, and if the volume of low-grade rocks is increasing, then the water in the oceans may be diminishing. Further, in a cooling earth, the quantities of water held in the upper mantle in phases like amphibole and phlogopite could also be increasing. Such conclusions would be invalid if another water or hydrogen source is possible and it is not impossible that there could be a hydrogen source in the core. At present there is no evidence bearing on this question.

Our present studies of low-grade metamorphism have begun to show us the immense significance of atmosphere-hydrosphere-crust interaction. In geochemical detail, we as yet know very little.

REFERENCES

BEARTH, P. (1959): Über eklogite, glaukophanschiefer und metamorphe Pillow-laven. *SMPM Zurich* **39**, 267-286.

BLATT, H., MIDDLETON, G. & MURRAY, R. (1972): *Origin of Sedimentary Rocks.* Prentice-Hall, New Jersey.

COOMBS, D. S. (1954): The nature and alteration of some Triassic sediments from Southland, New Zealand. *Trans. Roy. Soc. N.Z.* 82, 65-109.

————, ELLIS, A. J., FYFE, W. S. & TAYLOR, A. H. (1959): The zeolite facies with comments on the interpretation of hydrothermal synthesis. *Geochim. Cosmochim. Acta.* **17**, 53-107.

DOBRETSOV, N. L., KHLESTOV, V. V. & SOBOLEV, V. S. (1973): The facies of regional metamorphism at moderate pressures. (Transl. by D. A. Brown). Australian Nat. Univ., Canberra, Dept. Geol. Pub. 236, 299 p.

FYFE, W. S. (1973): Dehydration reactions. *Amer. Assoc. Pet. Geol. Bull.* **57**, 190-197.

———— (1974): Heats of chemical reactions and submarine heat production. *Geophys. J. Roy. Astr. Soc.* **36**, (in press).

————, TURNER, F. J. & VERHOOGEN, J. (1958): Metamorphic reactions and metamorphic facies. *Geol. Soc. Amer. Mem.* **73**.

———— & HOLLANDER, M. A. (1964): Equilibrium dehydration of diaspore at low temperatures. *Amer. J. Sci.* **262**, 709-712.

———— & ZARDINI, R. (1967): Metaconglomerates in the Franciscan Formation near Pacheco Pass, California. *Amer. J. Sci.* **265**, 819-830.

HARKER, A. (1939): *Metamorphism.* (Second edit.). Methuen, London.

HUTTON, C. O. (1940): Metamorphism in the Lake Wakatipu region, Western Otago, New Zealand. *N.Z. Dept. Sci. Ind. Res., Geol. Mem.* **5**.

HYNDMAN, D. W. (1972): *Petrology of Igneous and Metamorphic Rocks.* McGraw-Hill.

JAEGGER, J. C. & COOK, N. G. W. (1969): *Fundamentals of Rock Mechanics.* Chapman & Hall Ltd., and Science Paperbacks.

MUNSON, R. A. & SHEPPARD, R. A. (1974): Natural zeolites: their properties, occurrences, and uses. *Minerals Sci. Engng.* **6**, 19-34.

SALISBURY, M. H. & CHRISTENSEN, N. I. (1973): Progressive weathering of submarine basalt with age: further evidence of sea-floor spreading. *Geology* **1**, 63-64

SPOONER, E. T. C. & FYFE, W. S. (1973): Sub-sea-floor metamorphism, heat and mass transfer. *Contr. Mineral. Petrol.* **42**, 287-304.

————, BECKINSALE, R. D., FYFE, W. S. & SMEWING, J. D. (1974): O^{18} enriched ophiolitic rocks from E. Liguria (Italy), Pindos (Greece), and Troodos (Cyprus). *Contr. Mineral. Petrol.* **47**, 41-62.

STEINER, A. (1953): Hydrothermal rock alteration at Wairakei, New Zealand. *Econ. Geol.* **48**, 1-13.

THOMPSON, A. B. (1971): Analcite-albite equilibria at low temperatures. *Amer. J. Sci.* **271**, 79-92.

VERHOOGEN, J., TURNER, F. J., WEISS, L. E., WAHRHAFTIG, C. & FYFE, W. S. (1970): *The Earth.* Holt, Rinehart & Winston Inc.

ZEN, E-AN (1961): The zeolite facies: an interpretation. *Amer. J. Sci.* **259**, 401-409.

Manuscript received June 1974.