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Textures and mechanisms of metamorphic reactions in the Cockeysville Marble near Texas, Maryland

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Abstract

The Cockeysville Marble at Texas, Maryland was regionally metamorphosed at about 600°C and 6 kbar. During metamorphism the reactions between dolomite, quartz, and microcline to produce phlogopite, tremolite, and diopside caused the formation of mineral segregations and reaction bands. Various textures, including radiating sprays of tremolite mantled by calcite in dolomitic rocks and reaction zones of tremolite + calcite and phlogopite between metadolomite and calcite marble, can be explained by local metasomatic reactions taking place near equilibrium by mass transfer down chemical potential gradients between local domains. Modal mineralogy and reaction textures are used to infer the relative magnitude of fluxes for the major components, *viz.* $CaO > MgO \geq SiO_2 > KAlSi_3O_8$. Experimental solubility data support the generalization that the flux of a fluid species is proportional to its solubility.

Introduction

Detailed study of mineral relations in metamorphosed carbonate and pelitic rocks has shown that chemical equilibrium is commonly not achieved on the scale of a thin section. Thompson (1959) considered the role of local equilibrium and transfer of material in the presence of activity gradients. The concept was successfully employed by Carmichael (1969), Fisher (1970), Eugster (1970), Loomis (1976), Weare *et al.* (1976), and Foster (1977, 1978) to interpret textures in pelitic rocks; by Vidale (1969), Hewitt (1973), Vidale and Hewitt (1973), Burt (1974), Joesten (1974, 1977), Thompson (1975), Brady (1977), and Hoersch (1979) to explain calc-silicate reaction bands; and by Brady (1977) and Sanford (1978) to interpret ultramafic reaction zones. Preliminary results of the present study were presented orally by Sanford and Eugster (1974).

This paper is a systematic analysis of the mineralogy and textures of the Cockeysville Marble at Texas, Maryland. The Cockeysville Marble in the Flintkote Stone Products (formerly, Harry T. Campbell Sons') quarry at Texas, Maryland, contains several varieties of mineral segregations, including prominent dark brown phlogopite bands between white calcite marble and tan metadolomite, and striking clusters of radiating, bladed tremolite mantled by calcite within massive metadolomite. These and related textures, formed during the growth of metamorphic minerals, were clearly caused by chemical reaction and transport during metamorphism.

It will be shown that assemblages of minerals whose grains are in mutual contact reveal the equilibrium metamorphic conditions of particular samples, and that the sequence and modal relations of minerals in a set of reaction zones yield information on the stoichiometry of local metasomatic reactions and the nature of the dissolved, transported species. In most cases mobile and inert components can be distinguished unambiguously from observed reaction textures. The method of determining relative mobilities consists of proposing limiting, alternative combinations of mobile and immobile components in each reaction and predicting the various possible reaction textures. These predicted textures are clearly distinguishable from one another, and the choice of which one represents the actual mechanism is usually obvious by comparison with the natural rock. An important check on the mass transfer can be obtained by calculating the chemical potential gradients of individual species in the fluid.

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Geologic setting

Figure 1 shows the location of the quarry and of the samples. The Cockeysville Formation occurs near the base of the Glenarm Series, a group of Precambrian or Early Paleozoic metasediments resting unconformably on the Precambrian Baltimore Gneiss (Higgins, 1972; Crowley, 1976). The Cockeysville Formation is underlain by the Setters Formation, which comprises muscovite–microcline quartzite with subordinate mica gneiss and mica schist, and overlain by the thick Wissahickon Formation, which is composed mainly of pelitic schists (Higgins and Fisher, 1971; Crowley, 1976). These three formations make up the Glenarm Series, which mantles a group of elongate gneiss domes. The Texas Quarry is located in the Cockeysville Formation on the west flank of the Texas Dome. A detailed study of the Cockeysville Formation was carried out by Choquette (1957, 1960).

The quarry is located on the nose and SW limb of an anticline plunging gently NW (Choquette, 1957). Compositional layering, interpreted as bedding, strikes approximately NW and dips from 20° to 40° SW with a general steepening toward the SW. Fisher (1971) has demonstrated that the Cockeysville Formation exposed at Texas is right-side-up, as indicated by a bore hole which terminates in the Setters Formation. The upper beds of the Setters Formation underlying the Cockeysville Marble at Texas and much of the Wissahickon Formation, which overlies the Cockeysville, are composed of kyanite-, staurolite-, and garnet-bearing schists (Fisher, 1971).

Rock types

The principal rock types found in the Texas Quarry are metadolomite, calcite marble, and calc-schist (for definitions see Choquette, 1960). Medium-grained, bluish-gray metadolomite, calcite marble, and calc-schist, all interlayered on a fine scale and containing the assemblage dolomite + quartz, characterize the rocks in the northwest end of the quarry. The main part of the quarry contains coarse-grained, white calcite marble grading into medium- and fine-grained bluish-gray and tan varieties of metadolomite. The tan color is imparted by phlogopite. In this part of the quarry, tremolite occurs in place of the assemblage dolomite + quartz. Rare thin (1–2 cm) layers of microcline–quartz rock also occur. Modes for typical samples of each of these rock types are presented in Table 1.

Effects of sedimentary and diagenetic processes

The Paleozoic orthoquartzite–carbonate sequence, of which the Cockeysville Marble is a part, resting uncomformably on Precambrian gneisses, is typical

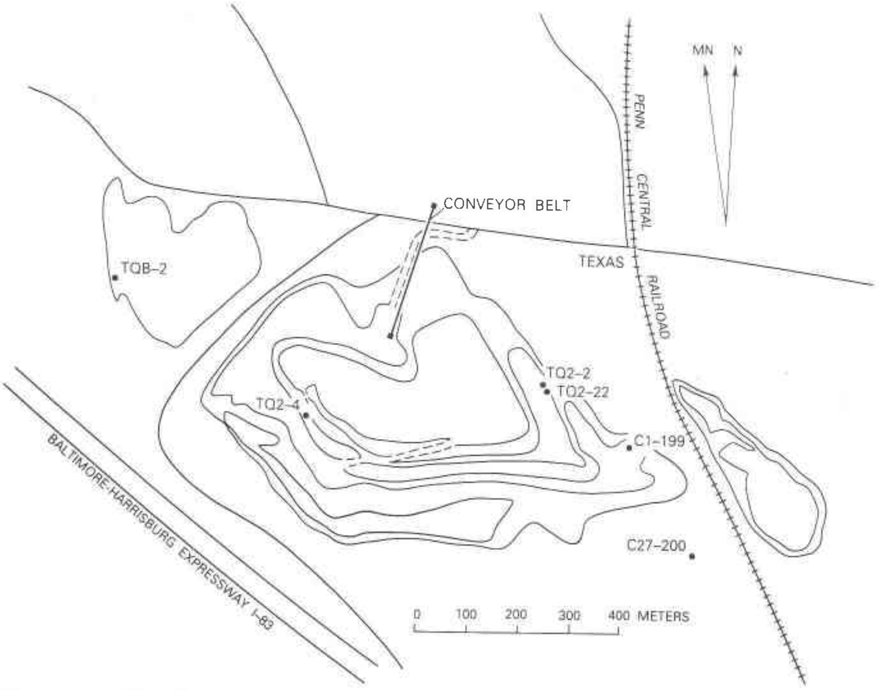

Fig. 1. Map of Flintkote Stone Products quarry at Texas, Maryland with locations of samples cited in text.

Table 1. Representative modes of rock types in Cockeysville Marble at Texas, Maryland

	Cc	Do	Tr	Ph	Mc	Q	Others*	No. Points
(1)	14.0	77.3	0.0	6.2	0.0	2.2	0.2	1318
(2)	16.7	72.9	2.8	7.4	0.0	0.0	2.8	2230
(3)	11.4	80.8	0.8	6.8	0.0	0.1	0.1	3023
(4)	85.3	0.0	0.0	0.4	4.8	9.1	0.4	963
(5)	94.5	0.0	0.0	4.3	0.4	0.6	0.2	4000
(6)	5.1	31.1	0.0	21.0	19.5	2.1	1.3	967
(7)	28.6	1.6	28.7	34.1	5.5	0.5	1.1	1141
(8)	6.2	0.1	0.1	5.3	66.0	19.9	2.4	1681

*Mostly pyrite, rare tourmaline.
(1) Metadolomite, C27-200
(2) Metadolomite, CI-199
(3) Metadolomite, TQ2-2 and TQ12-3
(4) Calcite marble C27-200
(5) Calcite marble, TQ2-22
(6) Calc-schist, TQB-2
(7) Calc-schist, TQ2-4
(8) Microcline-quartz rock, CI-199

of sedimentation at the edge of a stable craton having low relief (Crowley, 1976). Dolomitic rocks, which are the primary object of this study, probably represent deposition in a restricted intertidal, marine basin or a supertidal sabka environment (Selley, 1970, chap. 8). Such conditions would be expected to produce evidence of sea-level fluctuations, periodic flooding and evaporation, pore water replacement reactions, and occasional deposition of land-derived clastic material. Many features of the Cockeysville Marble at Texas can be attributed to these sedimentary and diagenetic processes.

The minerals calcite, dolomite, quartz, and microcline are considered to be premetamorphic. Dolomite may have formed by direct precipitation from seawater in a lagoon-like basin or by replacement of preexisting calcite sediments. Quartz is probably detrital or recrystallized from diagenetic chert. Microcline may be detrital, authigenic, or a product of clay–pore fluid reaction. Detrital microcline is common in the underlying Setters Formation, though rare in most carbonate rocks. Authigenic feldspar replacing limestone is reported by Pettijohn (1975, p. 325). Of the possible clay precursors of microcline, glauconite is the closest in composition, particularly with respect to its K/Al ratio. Other clay minerals (*e.g.* illite, kaolinite, montmorillonite) would require metasomatic addition of K or removal of Al. The distribution of phlogopite and tremolite probably reflects the premetamorphic distribution of microcline and quartz, respectively.

The dominant foliation at Texas is a compositional layering associated with variations in the proportions of calcite, dolomite, phlogopite, quartz, and feldspar. Prominent color variations from white to bluish gray to tan on the scale of meters are caused by locally abundant calcite, dolomite, or phlogopite, respectively. Dolomite and calcite marbles very commonly contain thin (1–2 mm thick) partings of calc-schist spaced from 2 to 10 mm apart. Phlogopite gives the prominent tan to dark brown coloration to the calc-schist layers, but microcline, quartz, dolomite (in calcite marbles), and opaques are also concentrated in these layers.

Calc-schist layers are similar to seams described by Barrett (1964), and interpreted by him to be the result of intrastratal solution moving along planes parallel or subparallel to bedding, removing calcareous constituents and concentrating insoluble, non-carbonate minerals. Vertical spacing between layers, broad horizontal extent of layers, close general parallelism, the merging and diverging of layers, and structures resembling cross-bedding all suggest a similar origin. The Cockeysville Marble at Texas differs in that partings are thicker, grain size is coarser, and phlogopite or microcline is present instead of glauconite and illite. These effects can be attributed to regional metamorphism. Less abundant than these thin partings are layers (1–2 cm thick) of quartzite and microcline–quartz rock. These layers may be relict sedimentary beds of quartz and clay, possibly metamorphosed glauconite-rich hardground (Bathhurst, 1975, p. 394–412), they may be concentrations due to locally intense pressure solution, or they may be due to chemical differences generated during regional metamorphism. The last possibility is discussed later.

Taken together, these layers are probably parallel or nearly parallel to original bedding surfaces because (1) they parallel formation contacts elsewhere in the Cockeysville formation (Choquette, 1960); (b) they wrap around boudinaged dolomite and quartzose layers; and (c) they represent concordant compositional variations of widely different magnitude, from less than a millimeter to tens of meters. Primary compositional differences have probably been accentuated by stylolitic pressure solution and metamorphic reactions. Studies of stylolites and other pressure solution features in limestones (reviews by Pettijohn, 1975, p. 340–342; Bathhurst, 1975, p. 468–471) have shown that non-carbonate minerals can be concentrated along planes subparallel to bedding due to non-hydrostatic pressure during burial and diagenesis.

While calcite marble commonly occurs in concordant beds, many irregular lenses and stringers of calcite marble clearly transect compositional layering and grade laterally into metadolomite. Isolated fragments of metadolomite are commonly surrounded by calcite marble. Streaks or bands of "impurities," graphite, pyrite, or tourmaline, in the marble are parallel to the local bedding. These textures suggest an *in situ* reaction relationship between calcite and dolomite. Such calcite bodies represent limestone masses untouched when the original limestone was dolomitized, or they are the result of incomplete dedolomitization (similar to that discussed by Shearman *et al.*, 1961; DeGroot, 1967; Friedman and Sanders, 1967; Bathhurst, 1975). In either case, the absence of phlogopite in the calcite bodies indicates that they existed prior to the metamorphic growth of phlogopite in dolomite-bearing rocks. If dolomite had been present where the calcite bodies are now, then microcline would have reacted to form phlogopite according to reaction (1), discussed below. For this reason the discordant bodies of calcite not associated with abundant metamorphic tremolite and phlogopite are considered to be a product of diagenesis. Calcite lenses and segregations associated with the formation of phlogopite and tremolite during metamorphism are discussed below.

Mineral chemistry

Minerals were analyzed with the Harvard University Department of Geological Sciences Applied Research Laboratories model EMX-SM electron microprobe, and data reduced by the method of Bence and Albee (1968) and Albee and Ray (1970). Standards were natural and synthetic minerals whose compositions approached those of the unknowns.

All analyzed minerals in the rocks studied have nearly ideal, end-member compositions. Microcline, which shows characteristic twinning, has compositions in the range $Or_{96}Ab_4$ to $Or_{92}Ab_8$. Tremolite contains very little FeO, but Al_2O_3 varies from 0.21 to 2.57 mole percent. A typical analysis yields the formula:

$$K_{0.01}Na_{0.02}Ca_{2.00}Mg_{4.93}Fe_{0.07}Al_{0.09}Si_{7.92}O_{22}(OH)_2$$

Phlogopite deviates most from its end-member composition. A typical phlogopite analysis yields the formula:

$$K_{1.76}Na_{0.02}Ca_{0.005}Mn_{0.01}Mg_{5.49}Fe_{0.18}Ti_{0.12}Al_{2.44}Si_{5.78}O_{20}(OH)_4$$

Excess Al_2O_3 (over two Al per formula unit) plus small amounts of FeO and TiO_2 are present. Dolomite is nearly stoichiometric. A typical analysis yields the formula:

$$Ca_{0.99}Fe_{0.01}Mg_{1.0}(CO_3)_2$$

Calcite compositions vary from $Ca_{0.99}Mg_{0.01}CKO_3$ to $Ca_{0.94}Mg_{0.06}CO_3$. FeO and MnO are insignificant. Calcite coexisting with dolomite is typically more Mg-rich than calcite without nearby dolomite. Diopside is rare and was not analyzed.

All the above-mentioned minerals except calcite will be treated as having ideal end-member compositions. Compositional variations in calcite vary systematically with mineral assemblages in the rocks.

Chemical reactions are written with the abbreviations for components in their respective states: Q, SiO_2 in quartz; Cc, Ca CO_3 in calcite; Do, $CaMg(CO_3)_2$ in dolomite; Mc, $KAlSi_3O_8$ in microcline; Ph, $KMg_3AlSi_3O_{10}(OH)_2$ in phlogopite; Tr, $Ca_2Mg_5Si_8O_{22}OH)_2$ in tremolite; and Di, $CaMgSi_2O_6$ in diopside. Components dissolved in the fluid are written without abbreviation, *e.g.* SiO_2, for SiO_2 in the fluid.

Stable mineral assemblages

Although many thin sections contain all of the minerals calcite, dolomite, microcline, quartz, phlogopite, and tremolite (a few also contain diopside), only a limited number of possible combinations actually occur with the minerals in mutual contact. These are:

Dolomite + Quartz + Phlogopite + Calcite
Zone I
Tremolite + Quartz + Phlogopite + Calcite
Zone II
Tremolite + Quartz + Microcline + Calcite
Zone III
Tremolite + Diopside + Microcline + Calcite
Zone IV

Furthermore, in any given thin section only one of the above assemblages is observed. It will be shown that these assemblages represent mutually exclusive fields in temperature–pressure–fluid composition (X_{CO_2}) space, and that the sequence from zone I through zone IV represents stable equilibrium assemblages related by systematic variations in X_{CO_2} or metamorphic grade.

Metamorphic textures in zone I

Disseminated phlogopite in metadolomite

Phlogopite, commonly occurring in metadolomite, is scattered randomly throughout the rock (Fig. 2a) in books 0.5–2 mm in diameter. Abundances are characteristically uniform (for modal analyses see Table 2), with phlogopite ranging from 6–8 percent. The texture of the rock is granular with interstitial anhedral calcite and euhedral phlogopite between subhedral dolomite grains.

The presence of microcline in dolomite-free rocks, coupled with its absence in metadolomite, suggests that phlogopite formed at the expense of microcline according to the reaction:

$$\mathrm{Mc} + 3\mathrm{Do} + \mathrm{H_2O} = \mathrm{Ph} + 3\mathrm{Cc} + 3\mathrm{CO_2} \qquad (1)$$

Reaction (1) produces calcite as well as phlogopite. For metadolomites, which have an average of 7 modal percent phlogopite, a maximum of 5 percent calcite can be produced by this reaction. This is about one third of the average calcite content of the metadolomites (see Table 1). The remainder was therefore originally present in the metadolomite. It also follows that the phlogopite reaction (1) produces a minor amount of calcite and cannot be responsible for the pure calcite marble lenses and veins discussed above under sedimentary and diagenetic features. Since no disseminated microcline remains in the metadolomite, reaction (1) has evidently gone to completion.

Phlogopite rims between metadolomite and calcite marble

Phlogopite also occurs in monomineralic rims up to several cm thick separating coarse-grained calcite marble from metadolomite (Fig. 2a). The shape of these rims is determined by the geometry of the marble–metadolomite bodies; the thickness of the rims appears to be directly proportional to the size of the bodies and to the phlogopite content of the metadolomite. In the simplest case, thick beds of marble and metadolomite are separated by planar rims roughly parallel to the bedding. Calcite marble also occurs as lenses or stringers of all sizes enclosed in metadolomite, with the lenses typically sub-parallel to the bedding. Phlogopite rims follow the shape of the marble lenses, but may not be continuous. Phlogopite is commonly concentrated at the ends of the lenses. In the most extreme case, coarse calcite marble bodies freely transect the bedding and enclose metadolomite blocks. The phlogopite rims again follow the marble–metadolomite interface. Quartz, pyrite, and tourmaline also are commonly concentrated with phlogopite in these rims.

Table 2. Modes of reaction zones between metadolomite and calcite marble, Sample No. C27-200

Rock Type	Cc	Do	Tr	Ph	Mc	Q	Others*	No. Points
Metadolomite	14.0	77.3	0.0	6.2	0.0	2.2	0.0	1318
Outer Rim	66.7	0.0	0.0	23.9	0.0	8.7	Tr	598
Inner Rim	59.7	0.0	0.0	20.3	15.3	3.4	1.4	295
Calcite Marble Core	85.3	0.0	0.0	Tr	4.8	9.1	Tr	963

* Mostly pyrite, rare tourmaline.

Where calcite marble containing disseminated microcline adjoins metadolomite, a double rim develops. Figure 2b shows one side of such a calcite layer including a phlogopite-rich layer adjacent to metadolomite. The phlogopite-rich layer consists of two zones. The zone closer to the metadolomite consists of an intergrowth of phlogopite and calcite, while the zone adjacent to the marble contains phlogopite, microcline, and calcite. In some cases, quartz is present throughout. Modes of a typical reaction zone of this type are shown in Table 2.

These textures are evidence of reaction (1) which has not proceeded to completion. As a consequence, the stable product assemblages microcline + phlogopite + calcite (in the calcite marble) and dolomite + phlogopite + calcite (in the metadolomite) separate the unstable reactant minerals microcline and dolomite.

Textures in zone II

Tremolite–calcite segregations

Tremolite occurs as individual fiber bundles 1–2 mm in diameter disseminated throughout much of the metadolomite (Fig. 2c), as radiating bladed knobs 5–10 mm long in calc-schist, and as fibrous masses 5–100 mm long within veins and lenses of calcite marble (Fig. 2d). All of these tremolite masses are partly or completely surrounded by pure calcite haloes, thus representing metamorphic segregations. Quartz is typically absent. The calcite layer of these segregations is commonly surrounded by a phlogopite rim. The overall net reaction responsible for these segregations is probably

$$5\mathrm{Do} + 8\mathrm{Q} + \mathrm{H_2O} = \mathrm{Tr} + 3\mathrm{Cc} + 7\mathrm{CO_2} \qquad (2)$$

With rare exceptions (those in which relict quartz remains in the segregation core) the reaction has gone to completion.

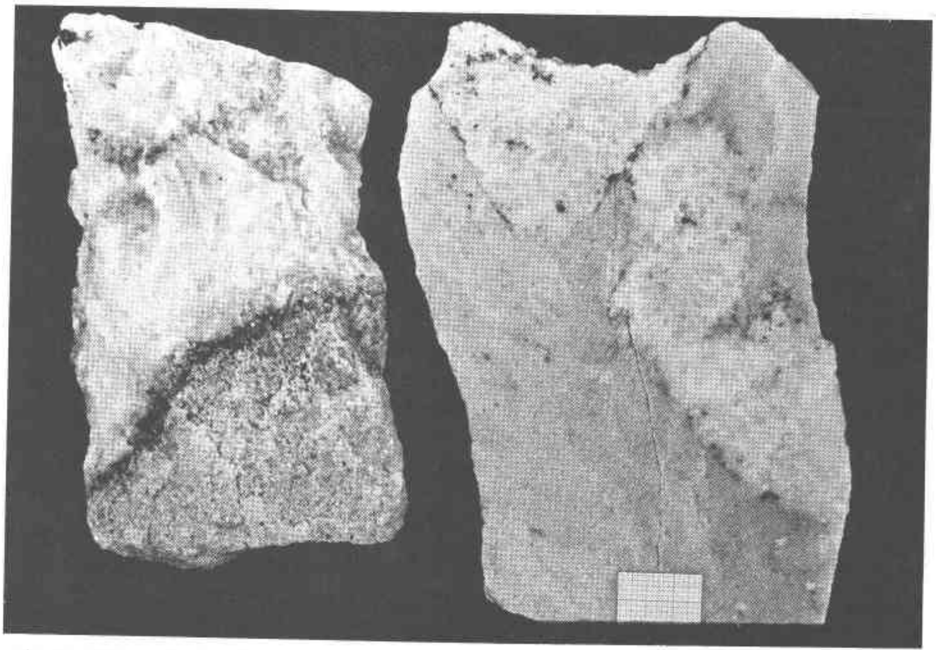

Fig. 2a. Phlogopite rims between calcite marble (white) and metadolomite (gray). Grid, 2 × 3 cm.

Phlogopite + tremolite rims between metadolomite and calcite marble

Thin (1–2 cm) layers of microcline–quartz rock occur in the Texas quarry, commonly at contacts between calcite marble and metadolomite; rarely with metadolomite on both sides of the layer. Figure 3a shows white calcite marble on the left and buff metadolomite on the right. The dark, banded layer in between consists of quartz, microcline, and pyrite. The white layer next to it is a zone of tremolite and calcite. Microcline–quartz layers are commonly fractured and folded. Layers in contact with metadolomite develop two reaction zones: a tremolite–calcite zone next to the metadolomite and a phlogopite zone next to the microcline–quartz rock (Fig. 3b). Table 3

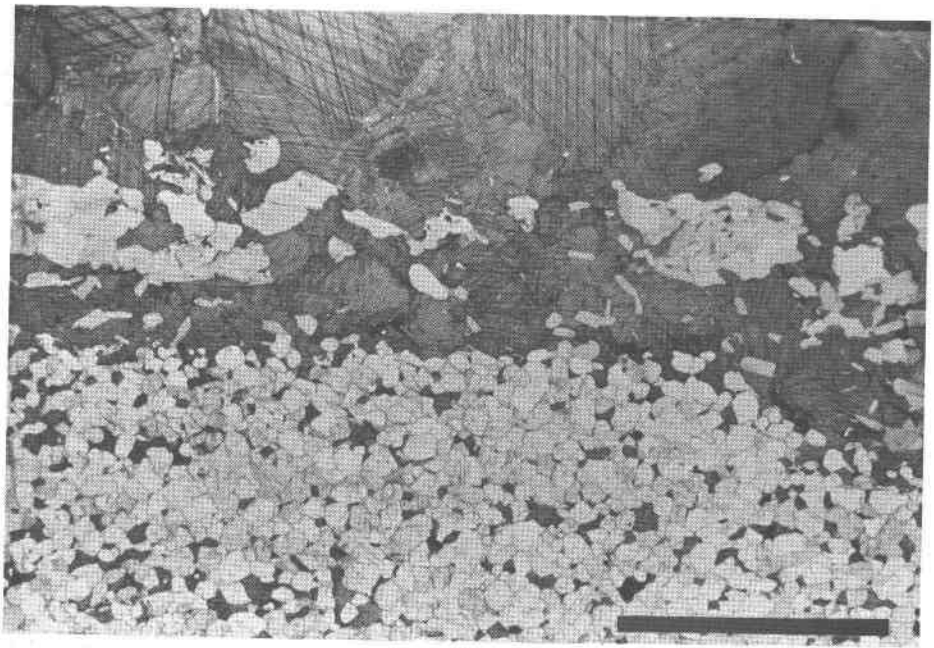

Fig. 2b. Photomicrograph of phlogopite rims in calcite marble (gray) at contact with metadolomite (white grains with interstitial gray calcite). Sample No. C27-200. Length of bar, 1 cm.

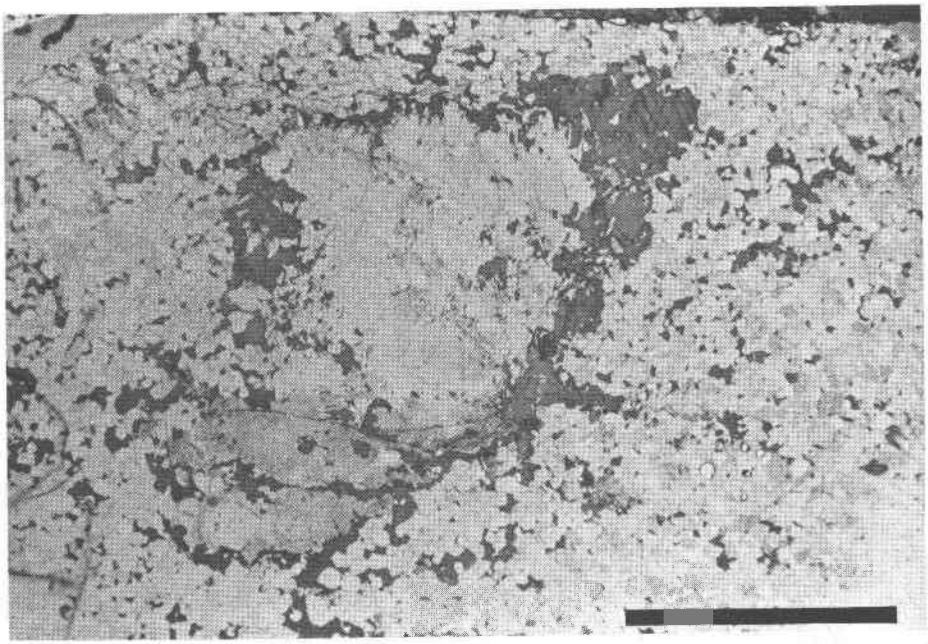

Fig. 2c. Photomicrograph of tremolite–calcite segregation with bundle of tremolite fibers (center, white) surrounded by calcite rim (gray) in metadolomite (white grains with interstitial gray calcite). Length of bar, 1 cm.

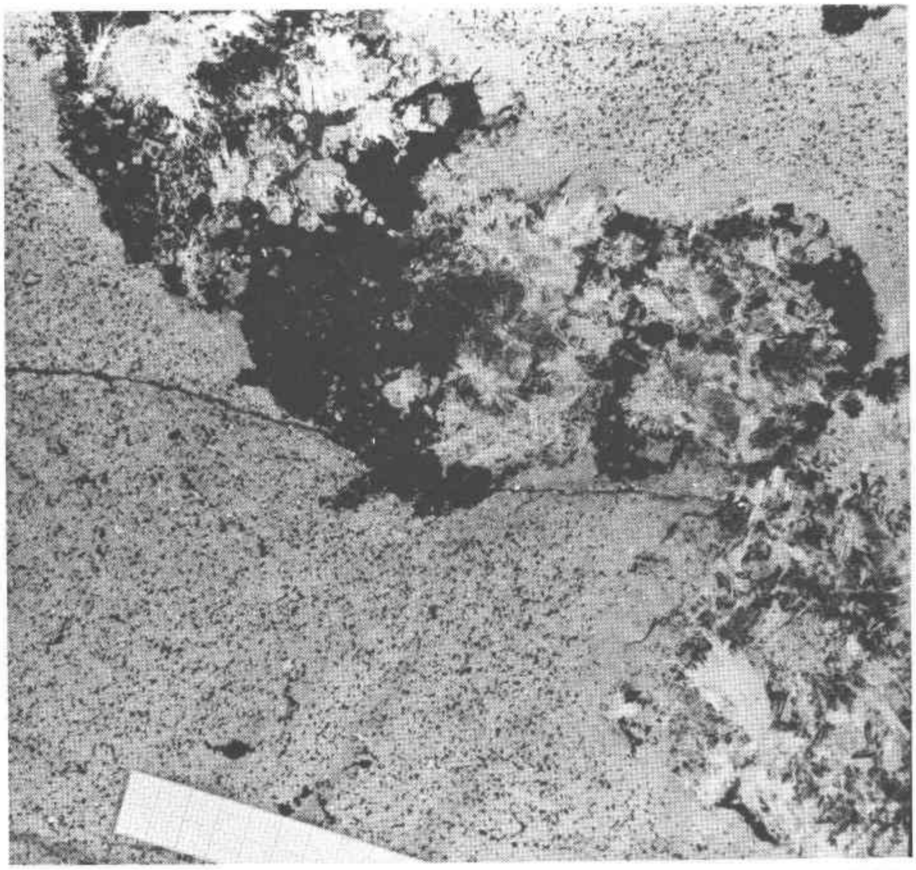

Fig. 2d. Calcite vein (gray) containing fibrous tremolite (white) in metadolomite (white grains with interstititial gray calcite). Grid, 1 cm wide.

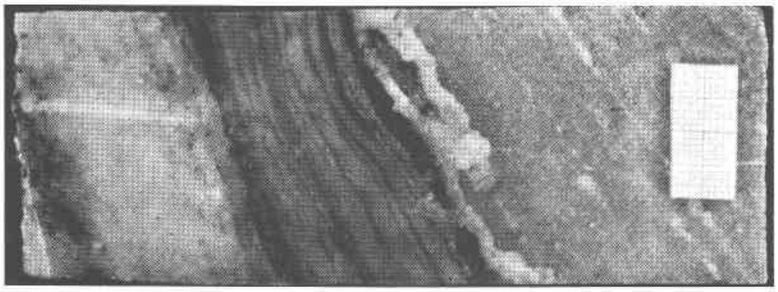

Fig. 3a. Microcline–quartz layer (banded dark gray) with calcite marble (white) on left and metadolomite (gray) on right. Between the microcline-quartz layer and metadolomite is a zone of tremolite and calcite (white) and a zone of phlogopite (dark gray edge on microcline–quartz layer). Grid, 1 × 2 cm.

gives modes for the four units. The tremolite–calcite zone is equivalent to the tremolite segregations discussed above. Phlogopite in the phlogopite zone contains small microcline inclusions, some of which exhibit optical continuity, indicating replacement of microcline by phlogopite.

The most significant features of the sequence are: (a) the absence of dolomite in all zones except in the metadolomite; (b) the presence of tremolite virtually limited to the tremolite zone; and (c) the concentration of phlogopite next to the microcline–quartz zone. Two reactions apparently involved in the formation of the mineral zones are (1) and (2).

Less common than microcline–quartz layers are angular quartzite fragments. These exhibit similar reaction rims except that the phlogopite zone is only partially developed (Fig. 3c).

Textures in zone III

Tremolite + microcline segregations

This class of segregations resembles the tremolite–calcite segregations in zone II except that the phlogopite crystals are commonly ragged and rimmed by clusters of small microcline grains. In these tremolite–microcline segregations, microcline is essentially absent in the host rock, which is either calc-schist or metadolomite. The tremolite–phlogopite–microcline aggregates are commonly surrounded by a calcite rim.

The growth of tremolite in the tremolite–microcline segregation probably took place by the reactions (2) and (3):

$$5Ph + 6Cc + 24Q = 3Tr + 5Mc + 2H_2O + 6CO_2 \quad (3)$$

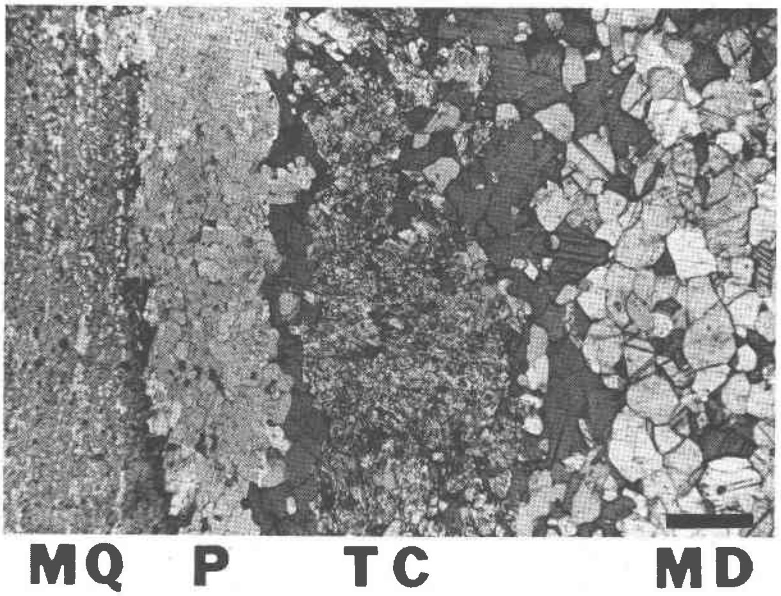

Fig. 3b. Photomicrograph of sample shown in Fig. 3a. Zones from left to right are microcline–quartz (MQ), phlogopite (P), tremolite–calcite (TC), and metadolomite (MD). Length of bar, 1 mm.

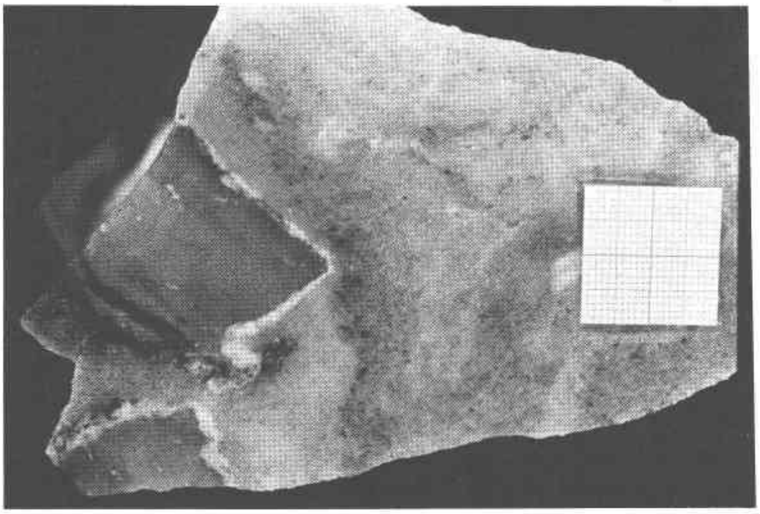

Fig. 3c. Quartzite fragments (dark gray) in metadolomite, tremolite–calcite zone (white) at contact. Grid, 2 × 2 cm.

Reaction (2) creates the initial tremolite–calcite segregation, while reaction (3) must have proceeded subsequently when phlogopite included in the core became unstable.

Textures in zone IV

Textures between metadolomite and microcline-quartz rock

In rare cases segregations like those previously described also contain diopside. Patches of small, deeply embayed diopside relicts occur in the tremolite zone (Fig. 3d). Individual grains exhibit a common optical orientation. Associated with the diopside are fine reddish-brown particles, presumably iron oxides, and rare sphene, suggestive of alteration products. Phlogopite in the neighborhood of the diopside is typically ragged, embayed, and surrounded by small microcline crystals. The phlogopite zone is discontinuous. A discontinuous zone of coarse-grained quartz between the phlogopite and tremolite zones and microfractures filled with quartz suggest late-stage production of quartz.

Diopside probably formed by the prograde reaction

$$\mathrm{Tr} + 2\mathrm{Q} + 3\mathrm{Cc} = 5\mathrm{Di} + 3\mathrm{CO_2} + \mathrm{H_2O} \qquad (4)$$

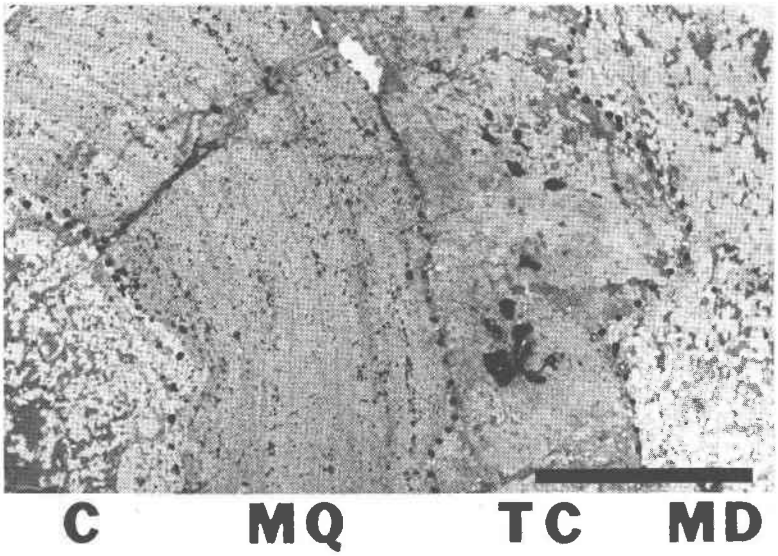

Fig. 3d. Whole thin section photograph showing zones (delineated by dots) from left to right, calcite marble (C), microcline–quartz (MQ), tremolite–calcite (TC), and metadolomite (MD). Dark patches in tremolite–calcite zone are altered diopside grains. Phlogopite zone is absent. Length of bar, 1 cm.

Table 3. Modes of reaction zones between metadolomite and microcline–quartz rocks, Sample No. C1-199

Rock Type	Cc	Do	Tr	Ph	Mc	Q	Others*	No. Points
Metadolomite	16.7	72.9	2.8	7.4	0.0	0.0	0.2	2230
Tremolite-Calcite Zone	14.7	1.9	45.1	7.5	0.0	0.5	0.3	588
Phlogopite Zone	3.7	0.0	0.0	72.5	1.5	18.6	3.7	363
Microcline-Quartz Zone	6.2	0.1	0.1	5.3	66.0	19.9	2.4	1681

*Mostly pyrite, rare tourmaline

The reaction

$$3\mathrm{Tr} + \mathrm{Mc} + 6\mathrm{Cc} = \mathrm{Di} + \mathrm{Ph} + 6\mathrm{CO_2} + 2\mathrm{H_2O} \quad (5)$$

may have occurred also.

Textures indicating production of quartz and microcline with consumption of diopside and phlogopite can be explained by reactions (4) and (5) proceeding to the left in a later stage of metamorphism. The assemblage phlogopite–quartz is clearly favored over the assemblage tremolite–microcline, which suggests that reaction (3) also proceeded to the left.

Equilibrium assemblages and metamorphic history

The observed reactions can be explained by progressive metamorphism of rocks originally composed of calcite, dolomite, quartz, and microcline. Mineral assemblages characterizing zones I through IV represent fields separated by divariant reactions in P–T–X_{CO_2} space. In Figure 4 these reactions are shown on a T–X_{CO_2} section at 6 kbar. The diagram is based on the conditions that (1) $P_{H_2O} + P_{CO_2} = P_t$, where P_t is total pressure; (2) the activities of $MgCO_3$ and $CaCO_3$ are fixed by coexisting calcite and dolomite; and (3) H_2O and CO_2 mix ideally. Thermochemical data for reactions in the system $CaO–MgO–SiO_2–H_2O–CO_2$ are from Skippen (1974). These data have been combined with experimental data on reaction (3) from Hewitt (1970) and Hoschek (1973), with corrections for the activity of $CaCO_3$ and $MgCO_3$ from Skippen, to yield thermochemical data for reactions (1) and (5). Data thus derived for reaction (1) agree well with experimental reversals of Puhan and Johannes (1974).

Alternative means of producing the variety of observed assemblages are shown in Figure 4. The dashed line represents progressive metamorphism with complete buffering of the fluid composition by the rock. The different assemblages could be explained by a gradient in temperature, from about 610° to 690°C. The observed assemblages could also be produced isothermally. If the fluid composition is buffered to varying degrees in different rocks due to variations in bulk composition and fluid/rock ratio, the fluid compositions would follow different paths, as illustrated by the dotted lines. All the observed mineral assemblages could be stable at a single temperature, *e.g.* 600°C. Because some zone I samples are less than 200 m from some zone IV samples, the second case is more plausible.

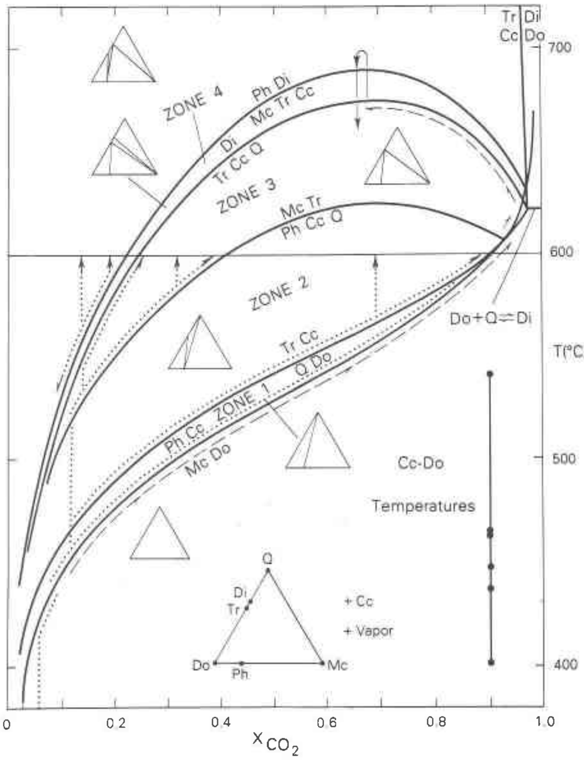

Fig. 4. T-X_{CO_2} diagram calculated for 6 kbar for equilibria in the system $CaO–MgO–SiO_2–KAlSi_3O_8–H_2O–CO_2$ with a fluid phase and calcite present with all assemblages. Data from Hoschek (1973), Skippen (1974), and Puhan and Johannes (1974). Arrows indicate possible fluid composition paths.

Also shown in Figure 4 are possible retrograde paths which could produce the observed zone IV textures associated with the reactions (4) and (5). Compositions of calcite coexisting with dolomite yield temperatures between 402° and 542°C, which are also plotted in Figure 4. Temperatures are based on the data of Goldsmith and Newton (1969). An average interchange energy (Ω) of 2655 cal/mole was used in the present calculations. Reaction textures and calcite–dolomite temperatures indicate that retrograde metamorphism caused minor alteration of zone IV silicate assemblages and that Ca–Mg exchange in carbonate minerals proceeded to lower temperatures than did discontinuous reactions involving silicates.

The zonation from I to IV is possible isothermally at a wide range of temperatures from 450 to 600°C. The assemblage staurolite–kyanite–biotite–muscovite–quartz in pelitic schists of the Setters Formation immediately below the Cockeysville Marble at Texas (Fisher, 1971) suggests minimum temperatures and pressures of 635°C and 6.4 kbar (Thompson, 1976; Holdaway, 1971). Therefore, actual peak pressures and temperatures were probably slightly higher than those of Figure 4.

Regardless of the actual temperature–pressure–fluid composition path, it is probable that the sequence of reactions encountered was the same, *i.e.*, from reaction (1) consecutively through (5).

In most thin sections adjacent minerals are in textural equilibrium and represent high-variance assemblages. Thus equilibrium has been achieved on the scale of the individual grains. However, in many cases equilibrium was not reached over distances larger than a few millimeters. These will now be examined in detail.

Reaction mechanisms

Equilibration of calcite compositions and concentrations of silicates and pyrite

Contacts between metadolomite and calcite marble are characterized by depletion of calcite and concentrations of phlogopite, microcline, quartz, pyrite, and tourmaline in all metamorphic zones I through IV. Reaction (1), which produces phlogopite, could conceivably cause the concentration of this mineral. However, this mechanism cannot explain the increased abundance of the other silicates and pyrite relative to the carbonate minerals. Nor is it likely that the components of these silicates diffused from the neighboring rocks toward the contact, since the chemical potentials of these components are fixed by the presence throughout of minerals with constant compositions (*e.g.*, SiO_2 in sample C27-200 has a constant chemical potential in both metadolomite and calcite marble because quartz is present throughout). A more plausible explanation involves dissolution of calcite from the contact with concentration of all other mineral constituents of the calcite marble.

The geometry and distribution of microcline–quartz layers suggest that they did not form by pressure solution, but that they are related to progressive metamorphic reactions. These layers are thicker (1–2 cm) than the thin (1 mm), stylolite-like partings of phlogopite, dolomite, quartz, and other sparingly soluble minerals in calcite marbles, and they are almost exclusively limited to metadolomite–calcite marble contacts in metamorphic zones II–IV.

A theoretical analysis of the progressive metamorphism of adjacent dolomite-rich and dolomite-free carbonate layers shows that such layers of "insoluble" minerals can be expected. At the low temperatures of diagenesis, equilibrium calcite dissolves little $MgCO_3$. We can assume that any original, metastable high-magnesium calcite breaks down to low-magnesium calcite and dolomite at relatively low temperatures and pressures. Thus, at the onset of regional metamorphism, calcite in the original dolomite rock is close to pure $CaCO_3$, like that in the predominantly calcite layers and bodies. With progressive metamorphism calcite grains adjacent to dolomite grains become richer in $MgCO_3$ by reaction with dolomite. In dolomite-free calcite marble there is no reaction, and calcite remains close to its end-member composition. Figure 5a is an idealized diagram of the initial configuration at the contact between metadolomite and calcite marble, each with a small uniform content of quartz and microcline. Differences in calcite composition between metadolomite and calcite marble are based on microprobe analyses (see above). Chemical potential gradients are thus created, as shown schematically in Figure 5b. The concentration of $CaCO_3$ is higher in calcite of the calcite marble (point A, Fig. 5b) than in calcite of the metadolomite (point B, Fig. 5b). For $MgCO_3$ the opposite is true. Therefore, $CaCO_3$ tends to diffuse from calcite marble to metadolomite and $MgCO_3$ from metadolomite to calcite marble.

Let us consider two limiting, alternative cases, either (1) the flux of $MgCO_3$ is much greater than that of $CaCO_3$, or (2) the flux of $CaCO_3$ is much greater than that of $MgCO_3$. In the first case $MgCO_3$ would be removed from the dolomite and migrate to the pure calcite with which it would react to form magnesian calcite. The metadolomite would show a local loss of volume and an increased concentration of insoluble material; the calcite marble would show a local increase of volume and a diminished concentration of insoluble material.

In the second case, calcite would dissolve and release $CaCO_3$ into solution. This $CaCO_3$ would migrate to the metadolomite where magnesian calcite would replace dolomite. The calcite marble would show a local decrease of volume and an increased concentration of insoluble material; the metadolomite would show a local increase of volume and a decrease in concentration of insoluble material.

The fact that the microcline–quartz layers are found within the calcite marble rather than at the border of the metadolomite suggests that the second case describes the actual situation, *i.e.*, that $CaCO_3$ flux exceeded that of $MgCO_3$. Microcline, quartz, and other relatively insoluble minerals responded passively to local volume changes. The result is shown diagrammatically in Figure 5c, and is similar to the effect described by Smigelskas and Kirkendall (1946). The greater solubility of $CaCO_3$ relative to $MgCO_3$ in aqueous solutions in equilibrium with calcite and dolomite (Rosenberg and Holland, 1964; Rosenberg *et al.*, 1967) supports the hypothesis of greater $CaCO_3$ flux.

The end product of this process would be the complete dissolution of calcite from original calcite marble lenses and layers, with the result that the minerals quartz, microcline, pyrite, and tourmaline, initially disseminated in minor quantities, would become major constituents.

Mechanism of reaction (1)

Reaction (1) results in the formation, in zones I and II, of layers and rims of phlogopite and calcite between metadolomite and microcline-bearing rock. Phlogopite produced by this reaction commonly is concentrated immediately adjacent to the microcline-bearing calcite marble or the microcline–quartz lay-

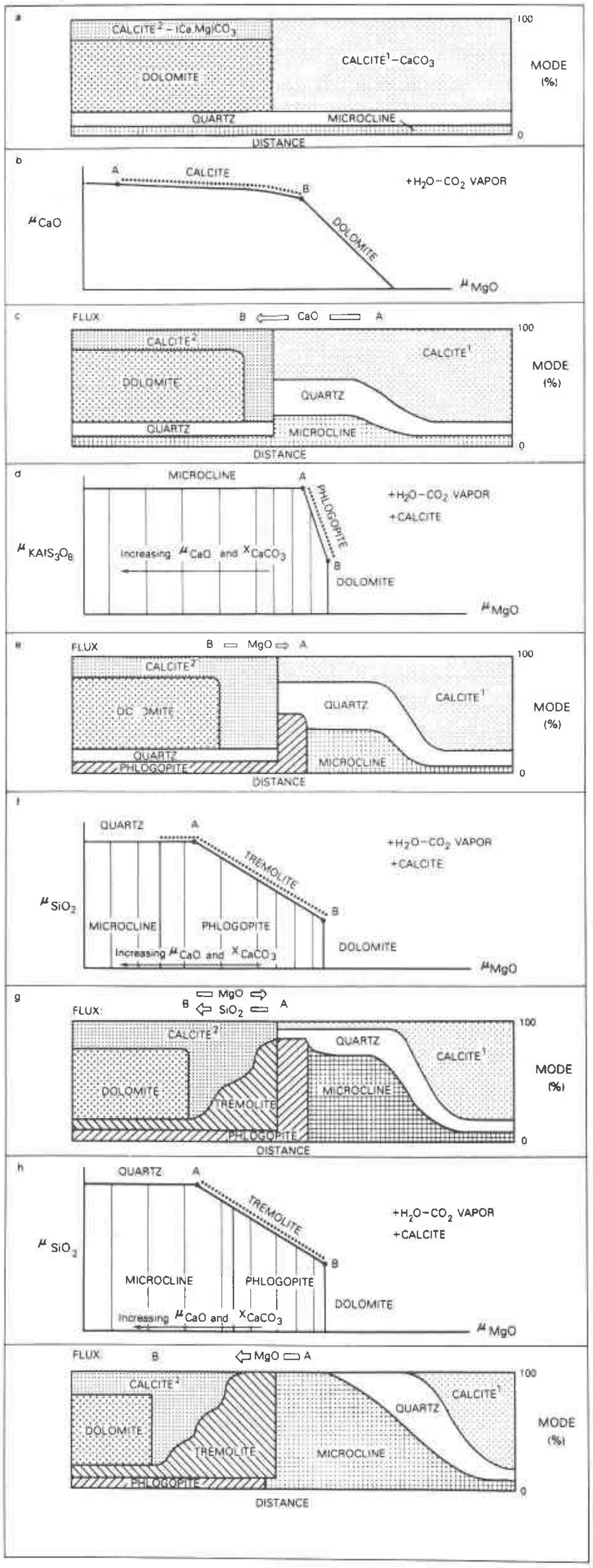

Fig. 5. Sequence of reaction zones between metadolomite and calcite marble: progression from diagenesis to intermediate metamorphic grade. Modal mineralogy at each stage is plotted schematically against distance (approximate length, 6 cm). Fig. 5a shows the hypothetical initial configuration. Figs. 5c, 5e, 5g, and 5i are idealized from observed mineralogy. For each metamorphic zone the appropriate chemical potential (μ) diagram is shown. Diagrams are constructed from Gibbs–Duhem relations among solid phases and fluid. Reaction paths of fluid are shown by dotted lines. (a) Initial configuration. (b) μCaO *vs.* μMgO, saturation surfaces of calcite (curved as a result of solid solution) and dolomite. (c) Reaction zone mineralogy at onset of reaction (1). (d) $\mu KAlSi_3O_8$ *vs.* μMgO projected along μCaO with calcite present. Topology in metamorphic zone I. (e) Reaction zone mineralogy in metamorphic zone I. (f) μSiO_2 *vs.* μMgO projected along μCaO and $\mu KAlSi_3O_8$ with calcite present. Topology of metamorphic zone II. (g) Reaction zone mineralogy in metamorphic zone II. (h) μSiO_2 *vs.* μMgO as in Fig. 5f. Topology of metamorphic zone III. (i) Reaction zone mineralogy in metamorphic zone III.

ers, whereas calcite forms a zone adjacent to the metadolomite. The reaction mechanism can be determined by considering two alternative limiting cases.

In Figure 5d chemical potentials in the system $CaO-MgO-KAlSi_3O_8-H_2O-CO_2$ at constant $P_t(= P_f$, fluid pressure), T, and X_{CO_2} are projected along the μCaO axis onto the $\mu MgO-\mu KAlSi_3O_8$ plane. Calcite is present in the area contoured for varying $CaCO_3$ concentration in the calcite (X_{CaCO_3}). Point A represents the calcite marble assemblage Mc + Ph + Cc, and B represents the metadolomite assemblage Do + Ph + Cc.

Reaction (1) takes place by two conjugate reactions. One occurs where microcline breaks down (A), and the other where dolomite breaks down (B). As one alternative we can assume that MgO is relatively immobile, and the reaction occurs solely by the mass transfer of $KAlSi_3O_8$. Coupled reactions for microcline and dolomite reacting are, respectively:

$$\mathrm{Mc} = \mathrm{KAlSi_3O_8} \qquad (1a)$$

$$\mathrm{3Do + KAlSi_3O_8 + H_2O = Ph + 3Cc + 3CO_2} \qquad (1b)$$

In this hypothetical case microcline dissolves, and $KAlSi_3O_8$ in solution (probably as a variety of separate species) diffuses to dolomite, with which it reacts to produce phlogopite and calcite. The resulting texture would have phlogopite + calcite in 1:3 molar proportions replacing dolomite.

However, the observed texture shows a zone of calcite replacing dolomite and another zone of phlogopite replacing microcline. This texture would be produced by the second alternative, that is if $KAlSi_3O_8$ is relatively immobile and MgO is the primary diffusing component. In this case the coupled reactions are:

$$\mathrm{3Do = 3Cc + 3MgO + 3CO_2} \qquad (1c)$$

$$\mathrm{Mc + 3MgO + H_2O = Ph} \qquad (1d)$$

Dolomite reacts to calcite, releasing MgO and CO_2. The MgO migrates to the microcline-bearing rock, where phlogopite forms. Two monomineralic zones of calcite and phlogopite, respectively, are formed. Because these predicted textures closely resemble the observed textures the second alternative is probably correct, that is, $KAlSi_3O_8$ is relatively immobile and reaction (1) occurs by MgO diffusion which couples the local reactions (1c) and (1d). Figure 5e shows the effects of reaction (1) on the idealized modal cross section between metadolomite and calcite marble. These effects are superimposed on the effects illustrated in Figure 5c.

Mechanism of reaction (2)

In zone II reaction 2 creates a zone of calcite + tremolite between metadolomite and quartz-bearing calcite marble or microcline–quartz rock. The reaction zone typically consists of both tremolite and calcite, but with tremolite more abundant next to the quartz-bearing rock and calcite more abundant next to the metadolomite. As in the previous section we can consider two alternative limiting pairs of coupled reactions.

In Figure 5f chemical potentials in the system $CaO-MgO-SiO_2-KAlSi_3O_8-H_2O-CO_2$ at constant $P_T(= P_f)$, T, and X_{CO_2} are projected along both μCaO and $\mu KAlSi_3O_8$ axes onto the $\mu MgO-\mu SiO_2$ plane. Calcite is present in the area contoured for varying $CaCO_3$ concentration in calcite. One of these contours coincides with the boundary between the stability fields of microcline and phlogopite. This boundary is drawn so that microcline, phlogopite, and quartz coexist rather than microcline, phlogopite, and tremolite (or dolomite), to be consistent with observed phase relations in zone II. Point A represents the calcite marble assemblage Q + Tr + Cc, and point B the metadolomite assemblage Do + Tr + Cc.

If it is assumed that MgO is relatively immobile and SiO_2 is the diffusing component, we have the coupled reactions:

$$\mathrm{8Q = 8SiO_2} \qquad (2a)$$

$$\mathrm{5Do + 8SiO_2 + H_2O = Tr + 3Cc + 7CO_2} \qquad (2b)$$

which occur next to the quartz-bearing rock and metadolomite, respectively. The predicted texture is a single zone of tremolite and calcite in a 1:3 molar ratio replacing dolomite.

The second alternative is for SiO_2 to be immobile relative to MgO with the coupled reactions:

$$\mathrm{5Do = 5Cc + 5MgO + 5CO} \qquad (2c)$$

$$\mathrm{8Q + 5MgO + 2Cc + H_2O = Tr + 2CO_2} \qquad (2d)$$

In this case two monomineralic zones, one consisting of calcite and the other tremolite, would replace dolomite and quartz respectively.

The observed textures are intermediate between the two predicted textures, as illustrated in Figure 5g. Tremolite and calcite occur together (as predicted by the first case), but tremolite tends to be concentrated near the quartz-bearing rock and calcite is more abundant next to the metadolomite (as in the second case). Thus both MgO and SiO_2 are significant diffusing components. The relative abundances of calcite and tremolite suggest that MgO diffusion predominates over SiO_2 diffusion.

Mechanism of reaction (3)

Textures in zone III resemble those in zone II, except that phlogopite is ragged and embayed by microcline. Hypothetical limiting cases for the mechanism of reaction (3) are for MgO, SiO_2, or $KAlSi_3O_8$ to be mobile.

Figure 5h shows the chemical potential relations with the same axes and constraints as Figure 5f. Comparison with Figure 5f shows that reaction 3 produces a shift in the relationship between chemical potentials defined by the assemblage Mc + Ph + Cc and those defined by Q + Tr + Cc.

Coupled local reactions which conserve MgO are:

$$5Ph + 6Cc + 24SiO_2 = 3Tr + 5KAlSi_3O_8 + 2H_2O + 6CO_2 \quad (3a)$$
$$5KAlSi_3O_8 = 5Mc \quad (3b)$$
$$24Q = 24SiO_2 \quad (3c)$$

In this case phlogopite would be rimmed by tremolite as reaction 3a proceeds. Silica consumed by 3a would be supplied by the dissolution of quartz (reaction 3c). Excess $KAlSi_3O_8$ produced by 3a would diffuse down its chemical potential gradient toward the metadolomite and precipitate as microcline (reaction 3b).

Local reactions which conserve SiO_2 are:

$$24Q + 6Cc + 15MgO + 3H_2O = 3Tr + 6CO_2 \quad (3d)$$
$$5Ph = 5KAlSi_3O_8 + 15MgO + 5H_2O \quad (3e)$$
$$5KAlSi_3O_8 = 5Mc \quad (3f)$$

Tremolite would replace quartz (reaction 3d), phlogopite would dissolve congruently (reaction 3e), and microcline would grow by precipitation (reaction 3f).

The third case, with $KAlSi_3O_8$ conserved, is illustrated by the reactions:

$$5Ph = 5Mc + 15MgO + 5H_2O \quad (3g)$$
$$24SiO_2 + 6Cc + 15MgO + 3H_2O = 3Tr + 6CO_2 \quad (3h)$$
$$Q = SiO_2 \quad (3i)$$

Phlogopite would be replaced by microcline (reaction 3g), tremolite would grow by precipitation and replacement of calcite (reaction 3h), and quartz would be dissolved (reaction 3i). This is clearly the actual case. Microcline replaces phlogopite, indicating $KAlSi_3O_8$ is conserved. Quartz must be dissolved to supply SiO_2 for tremolite growth in the tremolite–calcite zone. MgO is transported from the zone where microcline replaces phlogopite to the zone of growing tremolite. The cumulative effect is shown in Figure 5i.

Discussion

The relative transport or flux of a particular component is a function of concentration (in the intergranular medium), diffusion rates, and concentration gradients. The fluxes determined from textural evidence are consistent with known solubility data. At elevated temperatures aqueous solutions in equilibrium with calcite and dolomite dissolve more $CaCO_3$ than $MgCO_3$ (Rosenberg and Holland, 1964; Rosenberg *et al.*, 1967). Therefore, greater transport of $CaCO_3$ is to be expected, all other factors being equal. The component $KAlSi_3O_8$ probably exists in solution as a variety of species. On the scale of a thin section there appears to be no gain or loss of K_2O, Al_2O_3, or SiO_2 which would cause additional phases to appear. Thus the transport of K_2O and SiO_2 is limited by the concentration and mobility of Al_2O_3 in the fluid. Because of the known low solubility of Al_2O_3 even in corundum-saturated solutions (Anderson and Burnham, 1967; Roberson and Hem, 1969), it is not surprising that MgO is the major diffusing component for reaction (1).

Silica in aqueous solutions probably occurs as hydrated complexes such as H_4SiO_4 (Alexander *et al.*, 1954; Engelhardt *et al.*, 1975). In carbonate rocks with an intergranular fluid rich in CO_2, the activity of H_2O is much less than unity. Therefore, the solubility of silica in equilibrium with quartz is considerably lower than in CO_2-free aqueous solutions (Shettel, 1974). The solubility of MgO could be relatively higher in carbonate-rich fluids if it occurs as an associated carbonate species such as $MgCO_3$. Therefore, it is possible for MgO flux to exceed that of SiO_2.

The few experimental kinetic investigations relevant to reactions in the Cockeysville Marble tend to support these interpretations. In the presence of an aqueous fluid the reaction calcite + quartz = wollastonite + CO_2 proceeded by means of wollastonite rim growth on calcite and dissolution of quartz (Gordon, 1971). In contrast, wollastonite rims grew on quartz and calcite dissolved in experiments open to CO_2 and with no water added (Kridelbaugh, 1973). These data are consistent with lower SiO_2 solubility relative to that of CaO in more CO_2-rich fluids. Reaction zones between pressed pellets of various oxides and hydroxides were produced in experiments with water present (Ildefonse and Gabis, 1976). Although not directly comparable because of differences in fluid composition, these experiments illustrate a principle relevant to the present study. Quartz and brucite separated by graphite reacted to talc and olivine. Quartz dissolved leaving a void on that side of the graphite. The reaction proceeded by means of silica dissolution and diffusion through the graphite to the side originally containing brucite. Thus, the total volume of solids decreased on the side with the more soluble component, and the volume of solids increased on the side with the less soluble component.

In the rocks of the present study, this principle accounts for the decrease of volume (suggested by concentration of quartz, microcline, phlogopite, and accessory minerals) due to the preferential dissolution of calcite, diffusion of CaO from calcite marble to metadolomite, and the relative immobility of MgO. Unlike the experiments, the natural rocks contain no voids, because of lithostatic pressure. Instead, the initial contact, marked by relatively insoluble minerals, moves with respect to a volume-fixed reference frame in the direction opposite to that of the major diffusing species (as if, in the experiment of Ildefonse and Gabis, the growing talc on the brucite side expanded and pushed the graphite marker along the experimental capsule to fill the void on the quartz side).

Conclusions

Simple concepts of progressive metamorphism and relative chemical mobility are sufficient to explain a variety of complex reaction textures found in the metamorphosed carbonate rocks at Texas, Maryland. Alternative choices of "immobile" and "diffusing" components in each reaction imply alternative pairs of coupled local reactions and lead to predictions of diagnostic textures. It has been shown that calcite-bearing dolomite will react with pure limestone when each are metamorphosed to temperatures where calcite in equilibrium with dolomite accepts significant $MgCO_3$ in solid solution. The greater transport of CaO from the calcite marble over that of MgO from the metadolomite leads to concentrations of silicates and pyrite in layers and rims at the metadolomite–calcite marble contacts. Layers of nearly pure quartz and microcline may result. Textures of phlogopite formed by reaction (1) indicate that MgO transport greatly exceeds that of $KAlSi_3O_8$. Textures of the tremolite in reaction (2) suggest that both MgO and SiO_2 are important diffusing components, with MgO possibly being the dominant one. Microcline forming at the expense of phlogopite indicates that MgO and SiO_2 are the dominant mobile components for reaction (3) and that $KAlSi_3O_8$ was relatively inert.

The available evidence suggests that for carbonate rocks metamorphosed at moderate metamorphic grade, the fluxes of the components decrease in the order $CaO > MgO \geq SiO_2 > KAlSi_3O_8$. The relative fluxes are consistent with available solubility data, that is, the magnitude of the flux of a particular species is proportional to its solubility in the fluid.

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