

A fluid-inclusion study of metamorphosed pelitic and carbonate rocks, south-central Maine

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ABSTRACT

According to Ferry (e.g., 1987), metamorphosed pelitic and carbonate rocks from south-central Maine record mineral equilibrium with a water-rich fluid; yet, the prograde reactions liberated a CO₂-rich fluid. This implies a large and pervasive H₂O influx during the initial stages of metamorphism. Fluid inclusions from twelve quartz segregations of variable metamorphic grade record a complex fluid history. Most of the first-generation fluid inclusions are CO₂-rich and have densities appropriate to the peak metamorphic conditions. These inclusions may contain the fluid released from prograde decarbonation reactions. In general, composition of the fluid inclusions changes with grade, reflecting limited circulation of the locally derived fluids in fracture networks. In one quartz segregation, the fluid-inclusion composition does vary with rock type in an outcrop. A later influx of H₂O is recorded by a late generation of fluid inclusions and is associated with hydrothermal alteration of granitic stocks. The late-stage fluids may have triggered retrograde reactions that resulted in the N₂, N₂-CO₂, and CH₄ observed in later generations of fluid inclusions.

INTRODUCTION

Ferry has proposed that metamorphism in south-central Maine is attributable to pervasive, voluminous influx of H₂O. A similar phenomenon of pervasive CO₂ streaming has been called upon to explain occurrences of granulite-facies rocks and charnockites (e.g., Newton et al., 1980; Kreulen, 1980; Janardhan et al., 1982; Glassley, 1983; Schreurs, 1984). This model of fluid flow implies that rocks are permeable to pore fluids under high pressure, either convecting or passing through the system (Etheridge et al., 1983). An alternative view is that the only fluid to move through (infiltrate) metamorphic rocks is that which is released by devolatilization reactions (e.g., Hewitt, 1973; Greenwood, 1975; Crawford et al., 1979; Thompson, 1983; Lamb and Valley, 1984). In reality, metamorphism may be an interplay of these two processes of channelized versus pervasive fluid flow, depending on the availability and quantity of infiltrating fluid.

The purpose of this study was to use fluid-inclusion observations, combined with previous studies of mineral equilibria and stable-isotope data, to constrain the relative importance of external influx and internal generation of fluids during progressive metamorphism, pluton emplacement, and retrograde cooling of south-central Maine. Our data show that these fluid inclusions do not preserve the H₂O postulated to have passed through the region during the early stages of metamorphism, as documented by mineral equilibria and stable-isotope studies. Instead, the earliest generation of fluid inclusions contain CO₂-

rich fluids. Later fluid-inclusion generations are mainly H₂O rich. In addition, other fluid compositions (e.g., N₂ and CH₄) were trapped during the later influx of H₂O-rich fluids that may be related to either pluton emplacement or retrograde reactions following the metamorphic maximum.

Geologic setting

Samples were collected from the Early Silurian Waterville and Sangerville (formerly Vassalboro) Formations in south-central Maine (Fig. 1). The formations are composed of metamorphosed, interbedded shale, argillaceous sandstone, and carbonate rocks. In general, the Waterville Formation has more pelitic interbeds than the adjacent Sangerville (formerly Vassalboro) Formation (Osberg, 1968, 1988). The area is a Buchan-type metamorphic terrane with metamorphic grade varying from the chlorite to the staurolite-andalusite zone into the sillimanite zone. In the southern part of the area, there are quartz monzonite stocks that intruded during the Devonian Acadian orogeny (Dallmeyer and Van Breeman, 1981). Osberg (1988) has interpreted the timing of pluton emplacement to have been prior to the regional metamorphism. Ferry (1981, 1987) has argued for only one metamorphic event synchronous with pluton emplacement. At least two metamorphic episodes are postulated for adjacent regions to the north and west of the area (Novak and Holdaway, 1981; Holdaway et al., 1982, 1988). Osberg (1988) has suggested that there were three events beginning with a greenschist-facies episode followed by the regional progressive metamorphism and finally a lo-

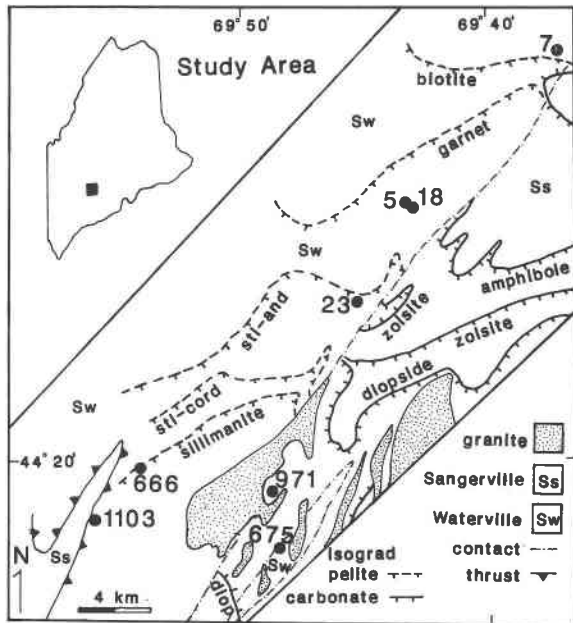


Fig. 1. Location map for fluid-inclusion samples in south-central Maine. Dotted pattern indicates quartz monzonite intrusions. Dot-dash lines show the boundary between the Waterville (Sw) and Sangerville (formerly Vassalboro) (Ss) formations. The barbed line shows a premetamorphic thrust fault with the teeth on the upper plate. The positions of the isograds are indicated by the hachured lines with hachures on the high-grade side. Stl-and = staurolite-andalusite isograd. Stl-cord = staurolite-cordierite isograd. Geology and isograds adapted from Ferry (1983d) and Osberg (1988).

calized retrograde metamorphism increasing in intensity to the southwest. These different interpretations of the timing of metamorphism and pluton emplacement suggest a need for more geochronological data. The stratigraphy, structural geology, and metamorphism of the pelitic schists in this region have been studied by Osberg (1968, 1971, 1979, 1988) and Ferry (1980a, 1982). Metamorphism of the carbonate rocks has been addressed by Ferry (1976a, 1976b, 1978, 1979b, 1980b, 1982, 1983a, 1983b, 1984, 1986a, 1987, 1988). Peak metamorphism was coeval with the final ductile deformation event (Osberg, 1988). Two periods of ductile folding and associated thrusting occurred prior to the third-generation folds. These early deformations are not expected to affect the fluid inclusions. Various aspects of the quartz monzonites have been discussed by Chayes (1952), Barker (1964), Ferry (1978, 1979a), Dallmeyer and Van Breeman (1981), Rumble et al. (1986), and Osberg (1988).

Physical conditions during metamorphism

Temperature and pressure estimates of peak metamorphic conditions have been summarized by Ferry (1980a). Pressure was 3.5 ± 0.5 kbar (350 ± 50 MPa) in the staurolite-andalusite and sillimanite zones (Ferry, 1980a).

Pressure was assumed in Ferry's studies to be constant during metamorphism and during the intrusion of quartz monzonites. Peak metamorphic temperatures in the Waterville Formation have been recently reviewed by Ferry (1986b). Temperature ranged from ~ 375 °C in the chlorite zone to 590 °C in the sillimanite zone (Table 1). Temperatures given in Table 1 are either calculated from various geothermometers or extrapolated from known conditions at the various isograds (Ferry, 1987).

Estimates of the C-O-H-S fluid composition during metamorphism indicate that it was predominantly H₂O mixed with CO₂ (Table 1). Ferry (1987) assumed there was ideal mixing of all components in the metamorphic fluid. The fluid composition in equilibrium with the mineral assemblages was estimated by assuming $P_{\text{fluid}} = P_{\text{total}} = 3.5$ kbar. For pelitic samples, the value of $X_{\text{H}_2\text{O}}$ was calculated by assuming ideality and that $P_{\text{fluid}} = P_{\text{lithostatic}}$ and the value of X_{CO_2} was assumed by difference. The composition of the fluid may be more H₂O rich if there was nonideal mixing of the fluid (Baumgartner, personal communication, 1988).

A measure of how much fluid (fluid:rock ratio) the rock has interacted with can be derived from the previously estimated equilibrium-fluid composition combined with the progress of the reaction (e.g., Ferry 1983c, 1983d, 1986a, 1987; Tracy et al., 1983; Graham et al., 1983; Nabelek et al., 1984; Grambling, 1986). The fluid:rock ratio measures the amount of fluid that is out of equilibrium with the rock and reacted with the mineral assemblage and represents only the minimum amount of water that actually passed through the region. Published fluid:rock ratios range from 0.4 to 1.4 for the metamorphosed carbonates (Ferry, 1983a, 1987, 1988), graphitic sulfide-rich schist (Ferry, 1981), and at the biotite isograd (Ferry, 1984). Wood and Graham (1986) have contended that the fluid:rock ratio has been overestimated for the biotite isograd on the basis of uncertainties in the assumed temperatures of metamorphism, thermodynamic data, and solution assumptions. Ferry (1986a, 1987) has used both the relative fluid:rock ratios and integrated fluid:rock ratio to predict fluid flow during metamorphism of the Waterville Formation to make several generalizations: (1) fluid flow is channelized parallel to bedding at low metamorphic grade and becomes pervasive with increasing metamorphic grade, (2) as metamorphic grade increases so does the integrated fluid:rock ratio, and (3) for the rocks at the highest metamorphic grade, most of the fluid passed through during the early stages of metamorphism. These generalizations suggest a model of infiltration of aqueous fluid along channels early in the metamorphism followed by a decreasing amount of infiltrating fluid as the style of fluid flow changes to pervasive at high metamorphic grade. In contrast to the Waterville Formation, Ferry (1988) found that fluid flow was relatively pervasive through the adjacent Sangerville (formerly Vassalboro) Formation at all metamorphic grades, presumably reflecting a lithologic control of fluid flow.

TABLE 1. CO₂ content of observed versus predicted fluid compositions

Sample and ref.*	Rock type**	T (°C)	X _{CO₂}		Later fluids	Fluid : rock ratio‡
			Obs-erved†	Pre-dicted‡		
7-3	C	400	0.0	0.07	N ₂ , H ₂ O, N ₂ -CO ₂	0-0.04
5-b	C	450	0.7-1	0.14-0.16	CO ₂	0.1-0.8
5-d	C	450	1	0.14-0.16	CO ₂	0.1-0.8
18a	P	460	1	0.26	CO ₂	—
18d	C	460	1	<0.96	CO ₂	—
23c	P	505	0.8-1	0.31	CH ₄ , H ₂ O	—
23d	C	490	0.9-1	—	CH ₄ , H ₂ O	—
666	P	530	—	0.32	N ₂	—
971	C§	560	0.4-1	0.17	H ₂ O	1.5-2.5
675a	P	550	0.67-1	0.2-0.38	H ₂ O	—
675b	C	550	0.4-1	0.13	H ₂ O, CO ₂	3.6
1103	P	540	0-0.05	0.23	CH ₄	3.5

* References to Ferry's work on these samples are 7 (1987), 5 (1987), 18 (1976), 23 (1976, 1978), 666 (1980a), 971 (1980b), 675a (1980a), 675b (1980b), and 1103 (1981).

** P = pelitic host; C = calcareous host.

† X_{CO₂} as observed in the earliest generation of fluid inclusions. Dashed lines indicate that earliest generation was not a CO₂-H₂O mixture.

‡ X_{CO₂} and fluid : rock ratio as predicted by Ferry (same references as listed above for samples). A dashed line indicates that there is not a predicted composition or a fluid : rock ratio for the sample.

§ Sample from Vassalboro Formation; all others from the Waterville Formation.

Method of investigation

Twelve samples of quartz veins, pods, and segregations were studied from eight localities (Fig. 1). At some outcrops, quartz veins from more than one rock type were sampled. The host rocks have different relative proportions of pelitic and calc-silicate mineral assemblages (Table 1). All samples were chosen by textural and structural relationships to represent the earliest generation of quartz segregation at each outcrop. Some samples (5b and 18a) were of quartz-calcite veins. All samples but one (971) are from the Waterville Formation. Quartz veins in sample 1103 are hosted in a distinctive rock type—a graphitic sulfide-rich schist (Ferry, 1981).

Thermometric properties of fluid inclusions were obtained with a Fluids, Inc., U.S. Geological Survey-type gas-flow heating-cooling stage following the methods described by Hollister et al. (1981). Calibration curves were constructed from known standards. A heating rate of 0.1 °C/min was used during phase changes. Several measurements were made on each fluid inclusion. The measurements for melting points and for CO₂ and CH₄ homogenization measurements could always be duplicated within ±0.2 °C and for aqueous homogenization within ±1.0 °C.

RESULTS

Fluid-inclusion types

Fluid inclusions are more abundant in quartz veins and segregations than in the host rock, where the inclusions

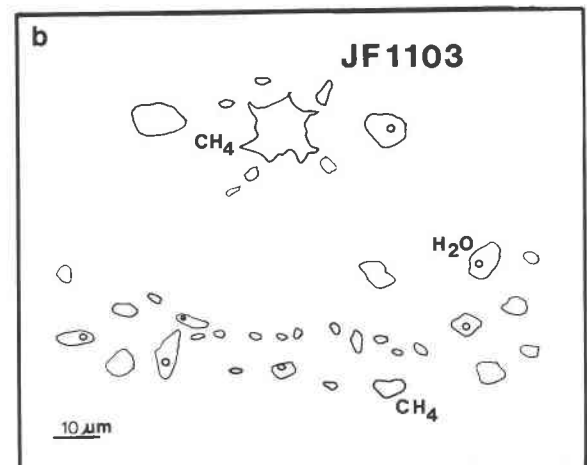
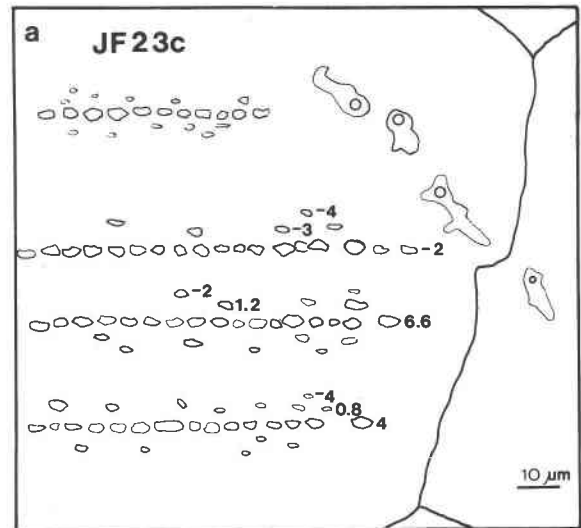


Fig. 2. Representative sketches of fluid-inclusion occurrences. (a) CO₂-rich inclusions occurring along healed fractures that do not cross grain boundaries. Later H₂O-rich inclusions cross grain boundaries and have vapor bubbles. Numbers given are CO₂ homogenization temperatures in °C for representative inclusions. Note the increase of T_h toward the center of the healed fracture. This increase is symmetrical on both sides of the fracture. (b) Distribution of CH₄- and H₂O-rich inclusions from a vein in graphitic sulfide-rich schist. All H₂O-rich inclusions have vapor bubbles. One CH₄ inclusion shows decrepitation textures. Both types of inclusions occur along the same healed fracture that does not cross grain boundaries. Scale bar in both figures is 10 μm.

were only observed in quartz. Five different types of fluid inclusions were identified in both the quartz veins and host rock.

CO₂-rich inclusions. The shape of the CO₂-rich inclusions ranges from irregular to negative crystal shapes with the majority being subhedral. Their size ranges from <1 to 15 μm. They may occur as solitary inclusions, in clusters, or in groups along healed surfaces (Fig. 2a). The

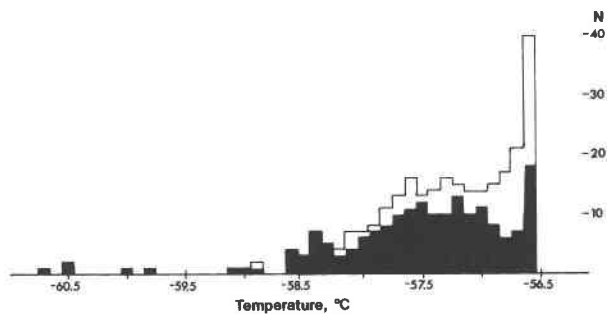


Fig. 3. Histograms for final melting temperatures (T_m) of CO_2 -bearing inclusions. N = number of inclusions measured. Black indicates carbonate-hosted samples and white indicates pelite-hosted samples.

healed surfaces were either grain boundaries or fractures that may or may not cross grain boundaries. These different types of occurrences were used to establish a relative trapping sequence.

H_2O -rich inclusions. Aqueous inclusions range from <1 to $30 \mu\text{m}$. The larger inclusions generally have amoeboidal shapes whereas the smaller ones are subhedral. Most of the aqueous inclusions are found on late healed surfaces that cross grain boundaries (Fig. 2a). Solitary aqueous inclusions occur only at greenschist facies in sample 7. No daughter crystals were found in any of the aqueous inclusions.

Mixed CO_2 - H_2O . This inclusion type is not abundant. Early generations of mixed CO_2 - H_2O inclusions are associated with clusters of CO_2 -rich fluid inclusions with no optically visible water. The intimate association of mixed CO_2 - H_2O inclusions with early generations of monophase CO_2 inclusions suggests there may be water that is visually undetectable in the early CO_2 -rich inclusions. Generally, it is difficult to detect less than 15–20 mol% water in dense CO_2 -rich inclusions (Roedder, 1972). Secondary generations are found at the intersection of late aqueous healed fractures with planes of CO_2 -rich inclusions.

CH_4 inclusions. These inclusions are characteristically rather dark and monophase at room temperature. Their shapes range from irregular to negative crystal. Some have features characteristic of decrepitated inclusions (Fig. 2b). They may occur along healed fractures, sometimes with coexisting aqueous inclusions, or as clusters of inclusions (Fig. 2b). CH_4 inclusions are generally larger than the aqueous or carbonic inclusions, ranging from 10 to $40 \mu\text{m}$.

N_2 -rich inclusions. Their shape is generally irregular, with sizes similar to the CH_4 -rich inclusions. These occur either ubiquitously throughout the sample or along discrete healed fractures that cross grain boundaries. There are no correlations among rock type, metamorphic grade, and occurrence of N_2 -rich inclusions.

Microthermometric data and Raman analysis

The compositions of representative inclusions of most fluid-inclusion types have been checked by laser Raman spectral analysis. Carbonic inclusions have melting temperatures ranging from -56.6 to -60.7 $^\circ\text{C}$, with most inclusions melting close to -56.6 $^\circ\text{C}$, the melting temperature of pure CO_2 (Fig. 3). The depressed melting temperatures indicate the presence of another species, which is probably CH_4 as the other likely component, N_2 , was not detected by Raman analysis. There is no systematic regional difference in methane content as most samples have similar ranges of melting temperature (Fig. 3). The amount of CH_4 , as estimated by the techniques of Heyen et al. (1982), varies from $X_{\text{CH}_4} = 0.0$ to 0.20 with an average of 0.08.

Aqueous inclusions generally have melting temperatures below 0.0 $^\circ\text{C}$, indicating the presence of salts. The observed range in melting temperature corresponds to salinities of 0.5 to 10 wt% NaCl equivalent. All samples have an average salinity of 4 wt% NaCl equivalent (Fig. 4). The initial melting of ice was rarely observable. For most samples, it was approximately -21 $^\circ\text{C}$, indicating that the predominant salt is NaCl. The initial melting in the graphitic sulfide-bearing schist was approximately -33 $^\circ\text{C}$, suggesting that there is MgCl_2 in addition to NaCl (e.g., Hollister and Burruss, 1976).

The microthermometric data of mixed CO_2 - H_2O inclusions is similar to that for pure CO_2 and H_2O ; there is an identical range in X_{CH_4} in the CO_2 -rich phase and in salinity of the aqueous phase as determined from clathrate-melting temperatures.

Homogenization temperatures near -92 $^\circ\text{C}$ indicate CH_4 -rich fluid inclusions. However, some inclusions had homogenization temperatures above the critical temperature of CH_4 , possibly indicating the presence of another species. Only CH_4 was observable by laser Raman analysis. Mullis (1979) reported the presence of hydrocarbons in CH_4 -rich inclusions with homogenization temperatures above the critical point. Another possibility is that the additional species could be a S species as predicted by Ferry (1981).

Pure N_2 inclusions homogenize below -147 $^\circ\text{C}$. Their composition was verified by laser Raman analysis. Mixed N_2 - CO_2 inclusions have homogenization temperatures between -154 and -147 $^\circ\text{C}$ for the N_2 -rich portion, followed by CO_2 melting between -109 and -88 $^\circ\text{C}$, and finally total homogenization at temperatures ranging from -77 to -72 $^\circ\text{C}$. Most of these inclusions homogenized to a gaseous state, indicating low densities.

CHRONOLOGY OF THE FLUID INCLUSIONS

At least two distinct groups of fluid inclusions can be distinguished in almost all samples: (1) early inclusions and (2) late inclusions. This division is easily recognizable by a change in fluid composition as well as in fluid-inclusion occurrence.

Early inclusions, whether they are aqueous or carbonic,

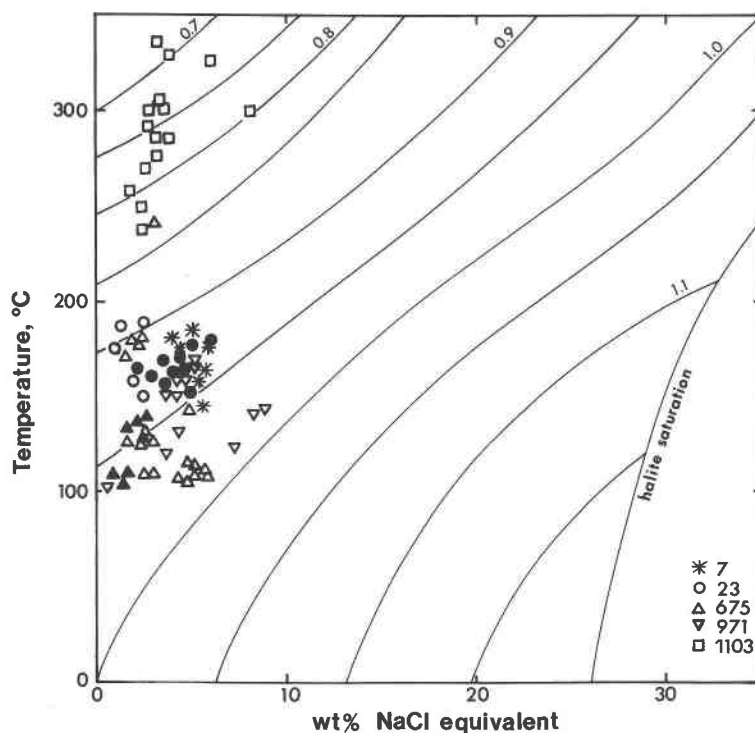


Fig. 4. Plot of homogenization temperature ($^{\circ}\text{C}$) versus salinity in wt% NaCl equivalent. Solid lines are isodensity and halite-saturation lines from Bodnar (1983). Open symbols are from samples of pelite. Solid symbols are from calc-silicate-bearing samples.

occur in clusters or in short linear trails that do not cross grain boundaries. In the high-grade, graphitic, sulfide-rich schist, aqueous inclusions occur on healed fractures that do not cross grain boundaries and occasionally with decrepitated CH_4 -rich inclusions (Fig. 2b).

Late inclusions occur on sharp trails that crosscut grain boundaries. Occasionally, mixed CO_2 - H_2O inclusions are found at the intersection of healed fractures bearing late aqueous and early carbonic inclusions.

Intermediate generations of carbonic inclusions are common in most samples. One type of intermediate CO_2 -rich fluids may have been trapped during hydrofracturing of the quartz veins (e.g., Burruss and Hollister, 1979; Etheridge et al., 1983). This type is identified as a set of parallel linear fractures with fluid inclusions along the main fracture all having identical homogenization temperatures. Fluid inclusions that occur off the main fracture show a symmetrical increase in homogenization temperature toward the center of a healed fracture (Fig. 2a). If this texture is associated with hydrofracturing, then the change in homogenization temperature corresponds to a drop in internal pressure of the fluid, assuming a constant temperature during hydrofracturing (Fig. 2a). For example, CO_2 -rich fluid inclusions in sample 23 record an increase in homogenization temperature of 2 to 9 $^{\circ}\text{C}$ from the edge to the center of the healed fractures (Fig. 2a). If these inclusions were trapped at a constant temperature ($\sim 505^{\circ}\text{C}$), the density decrease corresponds to

an internal fluid-pressure drop during crack healing of 269 bars. Vrolijk (1987) has documented fluid-pressure decreases of 1.5 kbar in quartz-calcite veins of the Kodiak accretionary complex. This change in internal pressure of the vein system may cause quartz precipitation because silica solubility decreases with decreases in pressure (Walther and Helgeson, 1977).

REGIONAL DISTRIBUTION

All different compositions and generations of fluids can be found throughout the entire region (Fig. 5, Table 1). Figure 5 shows the regional distribution of all fluid types in the samples studied. In Figure 5, the relative amounts of each different fluid type do not accurately reflect the abundance of the inclusion type, as we attempted to sample the entire range of compositions and densities but not necessarily in proportion to abundance. The small size of many of the inclusions precluded any efforts to estimate accurately the relative amounts of different fluids they contained. In almost all the samples, early inclusions contain CO_2 -rich fluids (Table 1; Fig. 5). The fluid inclusions in samples 7 and 666 consist of mostly late, low-density N_2 and N_2 - CO_2 mixtures. The presence of low-density N_2 -rich inclusions in both low- and high-grade samples indicates that its occurrence is independent of metamorphic grade. Another sample (1103) from a different host rock (graphitic, sulfide-rich schist) has a different early-fluid composition that consists of mixed aqueous and

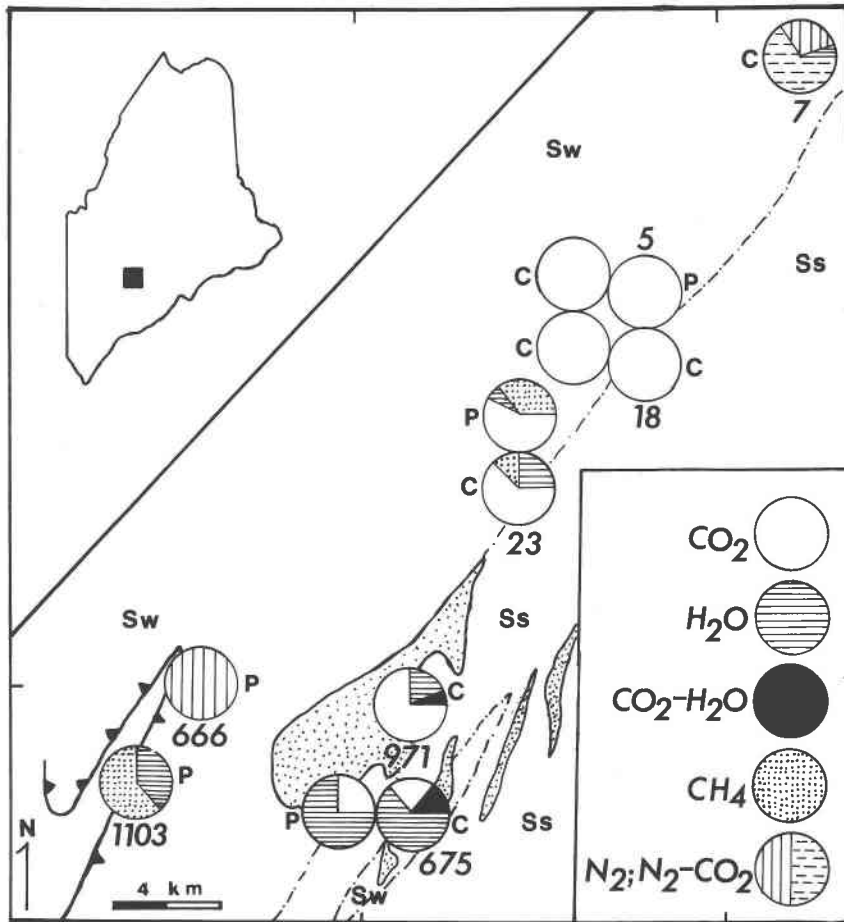


Fig. 5. Distribution map of fluid-inclusion types. C = metamorphosed carbonate, P = pelitic host rock. Numbers represent sample locality. Shading within each pie-diagram indicates the type and relative abundance of fluid compositions measured in each sample. As discussed in the text, the relative abundance may not reflect true abundance of each generation of fluid composition. Geologic symbols shown in Fig. 1.

methane-rich inclusions. Mixed $\text{CO}_2\text{-H}_2\text{O}$ inclusions were only found in the high-grade samples (971 and 675). Texturally late aqueous inclusions are most abundant in samples from near the quartz monzonite stocks (23, 675, and 971).

RELATION OF FLUID INCLUSIONS WITH METAMORPHIC GRADE

In general, homogenization temperature of early carbonic inclusions increases with increasing metamorphic grade (Figs. 1 and 6). The change in homogenization temperature can be related to a change in fluid-inclusion density. Fluid density was calculated for the earliest generation of fluid inclusions from their homogenization temperature and by applying the methods of Nicholls and Crawford (1985). Figure 7 shows isochores corresponding to densities calculated for the appropriate $\text{CO}_2\text{-CH}_4$ mixture in each sample. Using these isochores in combination with estimates of metamorphic pressure, it is possible to calculate the fluid temperature when the fluid

inclusion was formed. As estimated from the density, there is an increase of fluid temperature with metamorphic temperature (Fig. 7), although there is not an exact correspondence between the metamorphic-fluid temperature and the temperature calculated from mineral equilibria. However, the agreement is sufficiently good given the errors associated with each technique. Thus, in general we can assume that the fluid inclusions were equilibrated with respect to density near the peak metamorphic temperatures. At some outcrops, quartz segregations collected from different rock types have different homogenization temperatures (Fig. 6). This result leads to a discrepancy between the metamorphic temperature and the fluid temperature. This difference is less for samples hosted in metacarbonate than for those in metapelite (compare 675b with 675a, Fig. 7).

There is no systematic relationship between the density of aqueous inclusions and metamorphic grade. Instead, the observed density of aqueous inclusions in each sample must be considered individually (Fig. 8). For instance,

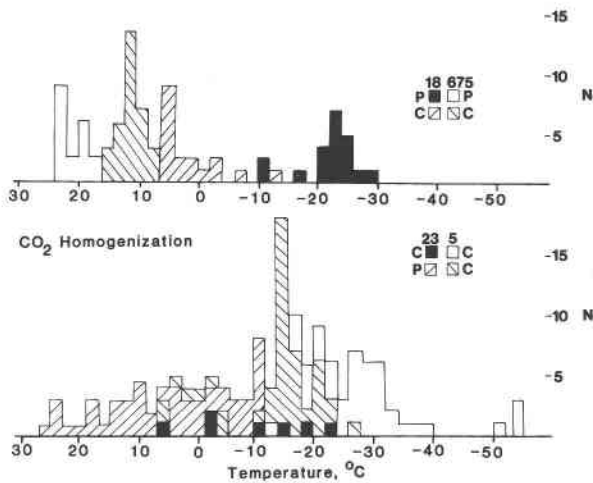


Fig. 6. Temperature of homogenization (T_h) for CO_2 -bearing inclusions. N = number of inclusions measured. P = pelitic host rock. C = metamorphosed carbonate.

the densities and corresponding isochores of early aqueous inclusions for the graphitic, sulfide-rich schist (1103) are consistent with metamorphic conditions whereas those in medium- and high-grade metacarbonates (23, 675, 971) are not (Fig. 8). The isochores for aqueous inclusions observed in the samples (675 and 971) near the quartz mon-

zonite stocks pass close to expected temperature conditions of late-stage alteration (Ferry, 1978). The isochore for the aqueous inclusions in a greenschist sample (7) passes close to the predicted metamorphic conditions. However, this isochore is also within the range of isochores for the other late secondary aqueous fluids (Figs. 4 and 8).

There is no obvious relationship between metamorphic grade and the distribution of low-density fluids (N_2 , N_2CO_2 , CH_4). This result is similar to other studies in regionally metamorphosed terranes by Swanenberg (1980), Kreulen and Schuiling (1982), Touret and Dietvorst (1983), Schreurs (1984), and Stout et al. (1986). These low-density inclusions were not trapped at the maximum metamorphic pressure.

DISCUSSION

Source of early fluids

Only one of the samples studied (1103) has H_2O -rich fluid inclusions that may record the H_2O -rich infiltrating fluid predicted by the mineral-chemistry data of Ferry. Any model of fluid evolution proposed for this region must account for the massive infiltration of water predicted by the mineral chemistry, stable-isotope data, and the ubiquitous CO_2 -rich inclusions in veins that were probably trapped at metamorphic conditions. This modeling problem is similar to that for many granulite-facies

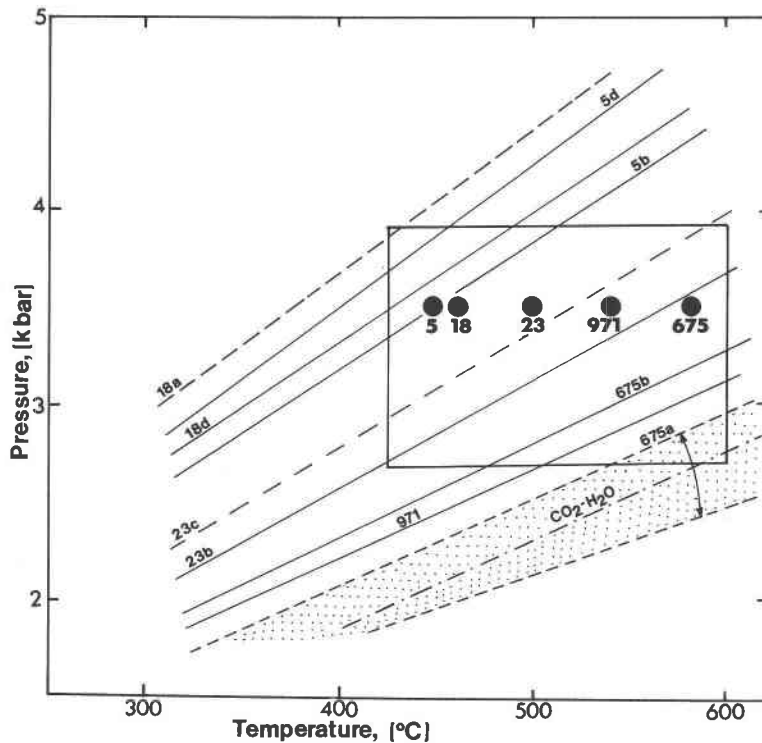


Fig. 7. Representative isochores for early generations of CO_2 inclusions. Dashed box and circles for P - T estimates of regional metamorphism from Table 1, assuming a constant pressure. Dashed lines are for samples from pelitic rocks, whereas solid lines are for metamorphosed carbonates. Dot-dash line is shown for a representative mixed CO_2 - H_2O inclusion in sample 675. Dotted area shows typical range of CO_2 isochores for one sample.

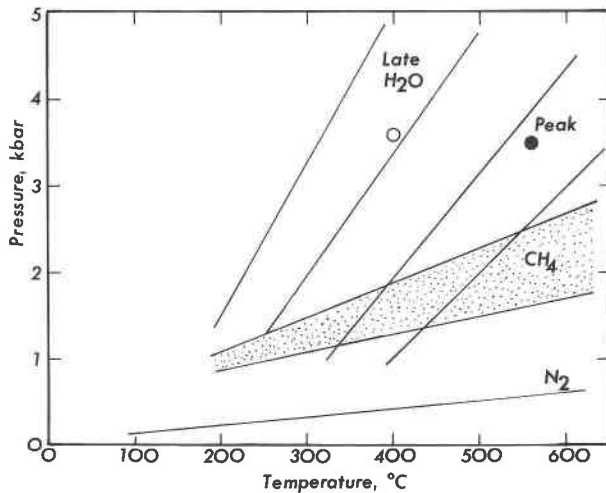


Fig. 8. Representative isochores for H_2O -, CH_4 -, and N_2 -bearing inclusions. Solid circle represents P - T estimates for peak metamorphic conditions for sample 1103. Open circle shows P - T conditions for alteration of quartz monzonite stocks. Dotted area encloses range of isochores for CH_4 -rich inclusions.

regions, as studies by Touret (1971), Kreulen (1980), Touret and Dietvorst (1983), Schreurs (1984), Hansen et al. (1984), Rudnick et al. (1984), Santosh (1984), Lamb et al. (1987), and van Reenan and Hollister (1988) all report CO_2 -rich inclusions. Two possible explanations appropriate for the overall discrepancy between predicted fluid compositions and observed composition seen in fluid inclusions in greenschist- and amphibolite-facies rocks are (1) fluid immiscibility and selective entrapment of CO_2 and (2) local derivation of CO_2 .

Immiscible fluids and selective entrapment of CO_2

The petrologic role of immiscible fluids has been recently reviewed by Crawford and Hollister (1986). Mixtures of water or aqueous brines with C-, S-, or N-bearing compounds are immiscible below 300–400 °C (Franck, 1977; Pichavant et al., 1982). The presence of salts will raise the consolute point and increase the range of fluid unmixing, as documented experimentally (Takenouchi and Kennedy, 1965; Naumov et al., 1974; Gehrig et al., 1979; Sterner and Bodnar, 1987; Zhang and Frantz, 1988) and observed in numerous fluid-inclusion studies (e.g., Pecher, 1979; Hollister and Burruss, 1976; Sisson et al., 1981; Hendel and Hollister, 1981; Mercolli, 1982; Stout et al., 1986). If there has been physical separation of an unmixed fluid, then it is possible to trap only one of the fluid components in fluid inclusions (e.g., Mullis, 1975). Several mechanisms have been proposed for selective entrapment including differential wetting properties of the two fluid phases (Mullis, 1975; Crawford and Hollister, 1986; Watson and Brenan, 1987) and density differences between the two fluid phases (Crawford and Hollister, 1986; Trommsdorff and Skippen, 1986). For most meta-

morphic terranes, CO_2 is denser than H_2O , which results in the possibility that the H_2O can segregate from CO_2 . On the basis of differences in the wetting properties of CO_2 -rich fluids and H_2O -NaCl fluids, Watson and Brenan (1987) concluded that the CO_2 -rich phase is relatively immobile compared to the H_2O -NaCl phase. Thus, it is reasonable to expect physical entrapment of only one of these phases.

If the aqueous fluid that infiltrated the region had a high salt content, then it is possible that this aqueous fluid would not mix with the CO_2 produced by metamorphic reactions. Ferry (1982, 1983a, 1983b), using whole-rock geochemistry, has suggested the initial fluid infiltrating the region was water rich and in equilibrium with the pelitic rocks. In addition, this fluid removed K and Na from carbonate rocks. Ferry (1983d) assumed that the aqueous fluid may have a chemistry similar to Salton Sea brines. If this is the composition of the inferred infiltrating fluid, its salinity would be too low to be immiscible with CO_2 at the metamorphic conditions.

The lack of any high-salinity fluid inclusions argues against fluid immiscibility. Sisson et al. (1981) noted that high-salinity inclusions are concentrated in matrix quartz and CO_2 -rich inclusions occur in quartz segregations. With this in mind, we searched matrix quartz for aqueous inclusions but found only CO_2 -rich inclusions similar to those in the quartz veins and pods. The distribution of mixed CO_2 - H_2O fluid inclusions may support fluid immiscibility, as they are only found at high metamorphic grade. This distribution would be expected because as temperature rises, more H_2O will be incorporated into the CO_2 -rich portion of the immiscible fluid (see Fig. 4; Sisson et al., 1981). However, the high-grade samples lack the H_2O -rich portion of the immiscible fluid. We lack the positive evidence (Ramboz et al., 1982) necessary to establish fluid immiscibility as an explanation for the occurrence of CO_2 -rich fluid inclusions.

Fluid immiscibility of H_2O and CH_4 probably resulted in the observed fluid inclusions in sample 1103. Experimental data on CH_4 - H_2O systems indicates that fluid immiscibility occurs below 400 °C at 3 kbar (300 MPa) (Franck, 1977; Welsch, 1973, in Pichavant et al., 1982). There are no high-temperature experimental data on the effect of salt on the immiscibility solvus in this system. The presence of both CH_4 and H_2O inclusions in the same cluster or on the same healed fracture argues for simultaneous trapping of the two fluid phases. The low density of most of the CH_4 -rich inclusions implies trapping at pressures and temperatures below peak metamorphic conditions. However, some of the CH_4 inclusions have decrepitated, indicating that the fluids were originally trapped at conditions of higher pressure and/or temperature. The coexisting aqueous inclusions do have isochores compatible with the metamorphic conditions (Fig. 8). These inclusions have salinities ranging from 4 to 10 wt% NaCl equivalent with initial melting temperatures indicating the presence of either MgCl_2 or CaCl_2 . These observations imply that moderate salinity (~7 wt% NaCl

equivalent) brines are immiscible with CH_4 at metamorphic conditions of 3.5 kbar (350 MPa) and 540 °C.

Local derivation of CO_2

As pointed out by Ferry (1986a), most of the reactions within the metacarbonates generate a CO_2 -rich fluid during metamorphism. The amount of CO_2 released depends on both the stoichiometry of the reaction involved and the progress of the reaction. Ferry (1983d) calculated the cumulative effects of several reactions and showed that the amount of evolved CO_2 decreases as the progress of the reaction increases in metacarbonate rocks of the Sangerville (formerly Vassalboro) Formation. Ferry (1976b) estimated that the composition of fluid released during metamorphism becomes progressively more water rich with increasing temperature. If we assume that the fluid-inclusion composition closely reflects the composition of the fluids generated by the host metamorphic rocks, then CO_2 observed in fluid inclusions in lower-grade samples may be derived from decarbonation reactions and/or reactions with graphite. Furthermore, the presence of mixed H_2O - CO_2 fluid inclusions only in upper amphibolite-grade samples (971 and 675, Table 1) may correspond to an increasing amount of H_2O produced (Ferry, 1976) or to increased progress of reaction as the amount of CO_2 evolved decreases from $X_{\text{CO}_2} = 0.9$ at biotite grade to $X_{\text{CO}_2} = 0.5$ to 0.8 at higher grades (Ferry, 1983d).

In general, fluid-inclusion composition is not controlled by rock type, as samples of quartz segregations in pelite and metacarbonate from the same outcrop have similar early and retrograde fluid inclusions (Fig. 5). This finding implies limited movement of locally derived fluids on an outcrop scale. Another example of restricted fluid mobility is the graphitic sulfide-rich schist (1103), which has a distinct early fluid composition ($\text{H}_2\text{O} + \text{CH}_4$). Presumably, this fluid composition represents a locally derived fluid that had not infiltrated other rock types in the region. For most of the region, metamorphic grade controls the compositional differences between the early fluid inclusions. Control by metamorphic grade contrast to the results of Ferry (1984, 1987), who has documented differences in fluid:rock ratio between metapelites and metacarbonates within the Waterville Formation that he attributed to differences in permeability of the host rock. So, fluid flow patterns recorded by mineral equilibria in different rock types are different from those observed as fluid inclusions within fracture networks of quartz segregations. Preliminary stable-isotope data on quartz-calcite veins and their wall rocks also record different types of fluid flow (D. Rumble III, personal communication, 1988).

Infiltration of H_2O -rich fluids may have transported heat into the region, with the mechanisms of H_2O transport governed by metamorphic grade and rock type, as suggested by Ferry (1984, 1987). However, the fluids generated by the metamorphic reactions in response to the increased heat were dominantly CO_2 rich and may be preserved as the fluid inclusions in quartz segregations.

The lack of preservation of the infiltrating H_2O as fluid inclusions might be caused by differences in transport properties of CO_2 versus H_2O (Watson and Brenan, 1987) or rate of fracture healing for different fluids (Brantley, 1988).

Source of retrograde fluids

There are two types of retrograde fluid related to different processes during cooling of the region: (1) fluids associated with intrusion of the quartz monzonite stocks and (2) fluids involved with re-equilibration and retrograde breakdown of minerals. The former type of fluids are H_2O rich and the latter consist of mixtures of N_2 , CO_2 , and CH_4 .

Ferry (1978, 1979a) and Rumble et al. (1986) have documented hydrothermal alteration of the quartz monzonite stocks. The alteration involved circulation of H_2O and CO_2 at 425 ± 25 °C and reactions producing chlorite, titanite, muscovite, calcite, and epidote (Ferry, 1978, 1979a). These alteration assemblages were in equilibrium with fluids with low but variable amounts of CO_2 ($X_{\text{CO}_2} = 0.0$ –0.13; Ferry, 1978; Rumble et al., 1986). The stable-isotope data of Rumble et al. (1986) are consistent with the hypothesis that the metasedimentary host rocks are the source of the water-rich fluid responsible for the hydrothermal alteration. In two samples (675 and 971) near the quartz monzonite stocks, there are abundant secondary aqueous inclusions (Fig. 5). As mentioned previously, the isochores for these late aqueous inclusions pass near the pressure-temperature conditions estimated for the hydrothermal alteration. Two samples (7 and 23) away from the quartz monzonite stocks also contain similar secondary low salinity inclusions. Thus, the fluid circulation associated with the hydrothermal alteration is fairly extensive as it occurs sporadically over 20 km. This interpretation depends on the relative timing of intrusion. If the quartz monzonite stocks are premetamorphic (Osberg, 1988), then the late aqueous fluids may be related to retrograde metamorphism and not hydrothermal alteration. Further work is needed to determine if the regional distribution of secondary aqueous inclusions is spatially related to the intrusions.

Late fluid inclusions with CH_4 and N_2 - CO_2 mixtures occur scattered throughout the area (Fig. 5). This regional distribution may suggest that retrograde processes may be responsible for the occurrence of CH_4 - and N_2 -bearing fluids across the entire region. Osberg (1988) has documented late chlorite associated with a retrograde metamorphic episode. The occurrence of N_2 - CO_2 mixtures in low-grade rocks and N_2 -rich inclusions in high-grade samples is similar to the distribution of N_2 -bearing inclusions in the Dome de l'Agout (Kreulen and Schuiling, 1982). The introduction of water associated with the hydrothermal alteration may have triggered the retrograde breakdown of biotite to chlorite, releasing any NH_4^+ from the K^+ site (cf. Duit et al., 1986). In addition, if this late water infiltrated regions with graphite, it may have re-

acted with the C to produce CH₄, observed as late fluids in sample 23.

CONCLUSIONS

From this study of fluid inclusions from rocks of south-central Maine, the following conclusions can be made:

1. There are five compositional types of fluid inclusions present in quartz segregations: CO₂-rich, H₂O-rich, mixed CO₂-H₂O, CH₄, and N₂ fluid inclusions.

2. CO₂-rich and CO₂-H₂O fluids were equilibrated during the peak metamorphism, as the density of the inclusions correlates with increased metamorphic grade. The origin of CO₂-rich fluid is enigmatic. Arguments were presented that it was derived from local metamorphic reactions.

3. CH₄ and H₂O were simultaneously trapped in a sample of graphite sulfide-rich schist. These fluids may have been immiscible at metamorphic conditions.

4. Late aqueous fluids penetrated the region near the quartz monzonite stocks and possibly other areas. These inclusions have isochores consistent with the alteration conditions of the intrusions. This aqueous fluid could possibly trigger retrograde reactions in the adjacent metamorphic rocks.

5. The final fluid trapped was N₂, N₂-CO₂, and CH₄, which may have been released from hydration of biotite and graphite in local, late retrograde reactions as postulated by Duit et al. (1986) for the Dome de l'Agout region.

In summary, the fluid-inclusion observations record a complicated fluid history in this region. Ferry (e.g., 1987) has hypothesized that initially water-rich fluids infiltrated the area and triggered metamorphic reactions as recorded by mineral equilibria. This aqueous fluid may represent one portion of an immiscible fluid. However, there is no conclusive evidence for fluid immiscibility that can explain the observed CO₂-rich fluid inclusions. Metamorphic reactions released CO₂-rich fluids, which may have been preserved as CO₂ and CO₂-H₂O fluid inclusions. The absence of primary H₂O-rich and presence of CO₂-rich fluid inclusions probably reflect different mechanisms of fluid transport for the infiltrating versus locally derived fluids. Probably the infiltrating fluid is penetrative, flowing along grain boundaries. In contrast, fluid-inclusion observations imply that flow of the locally derived fluid was channelized into fracture networks and trapped as synmetamorphic fluid inclusions in quartz veins. The presence of fluid inclusions with similar compositions in different rock types supports fracturing as the method of transport for CO₂-rich fluids on outcrop scale; however, the composition of the earliest generation of fluid inclusions changes with metamorphic grade, which implies that the fracture networks do not extend over the entire region.

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