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The tremolite veins of Campolungo and their genesis*

by *Ivan Mercolli*[1], *George Skippen*[2] and *Volkmar Trommsdorff*[3]

Abstract

Metasomatic veins in Triassic dolomite rocks at Costello, Campolungo, Switzerland, formed by the reaction:

$$5 \text{ dolomite} + 8 \text{ quartz} + H_2O = \text{tremolite} + 3 \text{ calcite} + 7\, CO_2.$$

Reaction occurred by infiltration of fluid along the contact between dolomite and preexisting quartz veins. Evidence from fluid and solid inclusions indicates that fluids of very high salinity were involved in the metasomatic process. The composition of the incoming fluid was not in equilibrium with quartz and dolomite and resulted in the formation of tremolite + calcite at the expense of the reactant minerals.

High salt contents may be related to an evaporitic source rock for the fluids as indicated by oxygen isotope measurements on carbonate minerals. Representative oxygen carbon and oxygen isotope data from coexisting calcite and dolomite in the veins are:

$$\delta^{13}C(PDB)\,(\text{dolomite}) = 0.04; \delta^{13}C(PDB)\,(\text{calcite}) = 0.02; \delta^{18}O(PDB)\,(\text{calcite}) = -8.1.$$

The structural position of the veins at Costello and postkinematic growth of tremolite are consistent with geothermometry and geobarometry which indicate that reaction occurred during uplift and cooling of the rock mass.

The heat necessary to drive the above devolatilization reaction to the right could have been provided by hot fluids entering the vein system or it could have been derived from wall rock adjacent to the veins. The presence of unreacted quartz and dolomite in close proximity suggests that H_2O was in limited supply. A wallrock source for the heat is therefore considered more likely.

Keywords: Tremolite veins, fluid inclusions, salinity, stable isotopes, Campolungo, Switzerland.

Introduction

Exactly two hundred years ago ERMENEGILDO PINI (1786) described a new mineral, later named tremolite, that he had discovered in dolomite rocks at Campolungo, Central Alps, Switzerland (Fig. 1). Campolungo subsequently became a classic mineral locality through the work of many scientists such as DE BELLEVUE (1792); VAUBERCHEM and STRUVE 1792); EBEL (1804); LAVIZZARI (1845); KENNGOTT (1866) and KOENIGSBERGER (1908). Historical reviews of the discovery at Campolungo are given by BIANCONI (1971, 1986) and a comprehensive description of the mineral occurrences is found in GÜBELIN (1939). The geological situation at Campolungo has been repeatedly studied, and the reader is referred for details to PREISWERK (1919), BIANCONI (1971), and KELLER (1968). The most detailed geological map of the area is by BIANCONI (1971). The petrology of tremolite from Campolungo has recently been studied by MERCOLLI (1979, 1982) and by WALTHER (1983). Both these authors used fluid inclusion microthermometry to evaluate the nature of the fluids involved in the formation of veins containing radiating tremolite. These same veins are the subject of the present study.

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* Dedicated to Professor Ernst Niggli on the occasion of his 70th birthday.

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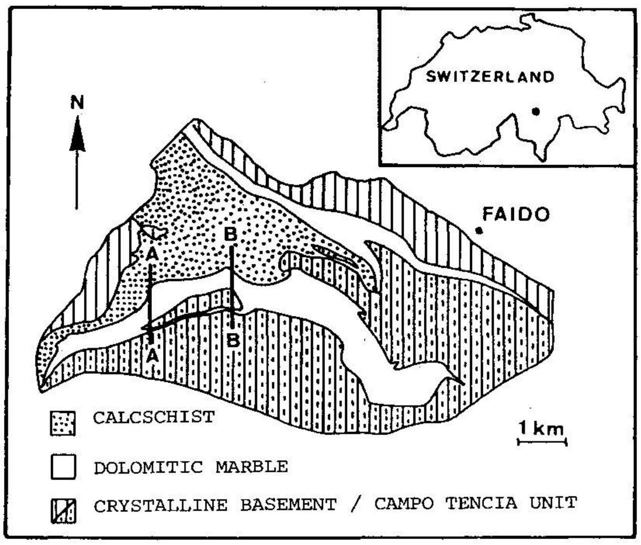

Fig. 1 Geological map of the Campolungo area. A–A and B–B refer to profiles at Passo Campolungo and at Passo Cadonighino respectively as shown in Figure 2.

Structure

The Campolungo area within the Lepontine Alps of Southern Switzerland exposes a Triassic sequence that forms the sedimentary cover of the Campo Tencia unit (Fig. 1). This Triassic sequence begins with a thin quartzite unit followed by various dolomitic lithologies. The dolomites are succeeded by an evaporite unit ("Rauhwacke") that has been modified both structurally and by chemical dissolution in near surface waters. A detailed description of the structural and lithological characteristics of the Campolungo area is given by BIANCONI (1971) and in part by KELLER (1968).

Two representative profiles after Bianconi are given in Fig. 2. The section at Passo Cadonighino (BB) shows the main regional phase of isoclinal folding (D1). The section at Passo Campolungo (AA) has been further deformed by a second and more locally recognized phase of deformation (D2).

Metamorphism

The Campolungo area lies within the kyanite zone of the metamorphic belt of the Central Alps (NIGGLI, 1960, 1970). It lies only a few kilometres north of the famous locality of Alpe Sponda (KELLER, 1968) with its staurolite-kyanite-paragonite schists. The mineral assemblage in dolomitic carbonate rocks is dolomite + tremolite + calcite + phlogopite, and in mafic schists, garnet + plagioclase amphibolites are typical. Two generations of tremolite that formed during the Tertiary Alpine Metamorphism can be related to the deformation history of the area. An older generation of tremolite prisms formed during the main stage of Alpine metamorphism and lies in the main schistosity (S1) with no preferred linear orientation (BIANCONI, 1971, p. 207). Its growth predates the second deformation, D2. A second generation of tremolite postdates the deformation, D2. In contrast to ductile behaviour during D1, the dolomite rocks deformed in a largely brittle manner during D2.

This suggests that the second generation of tremolite formed during the retrograde portion of the metamorphic history. Tremolite 2 is mainly developed around quartz nodules and veins. These quartz concentrations are confined to a stratigraphic layer of white dolomite and are therefore considered to be of Triassic age. They could have originated either as sedimentary chert layers that were pulled apart or as hydrothermal veins that existed prior to the first deformation. Evidence indicating the early existence of quartz is found at Passo Cadonighino where quartz nodules form boudins laying in S1 (Fig. 2b). The relationship of quartz at Costello near Passo Campolungo is different from that described above for Passo Cadonighino. At Costello, quartz veins are oriented in the axial plane of D2 (Fig. 2a) and are discordant to the first generation of tremolite which is aligned within S1. At Costello, therefore, quartz must have been remobilized during or after D2.

At both Passo Cadonighino and Costello, the second generation of tremolite grew postkinematically around the quartz nodules as clusters radiating out from a point source. The two generations of tremolite also have distinctive chemical characteristics. The Al/Na ratio and the F/OH ratio, for example, follow clearly separated trends in the two generations of amphibole (MERCOLLI, 1979, p. 54).

A considerable amount of quartz in close contact with dolomite persisted through the main period of metamorphism. Up to 3 percent disseminated quartz in the mineral assemblage of the dolomitic marbles (BIANCONI, 1971, p. 138) as well as the quartz concentrations de-

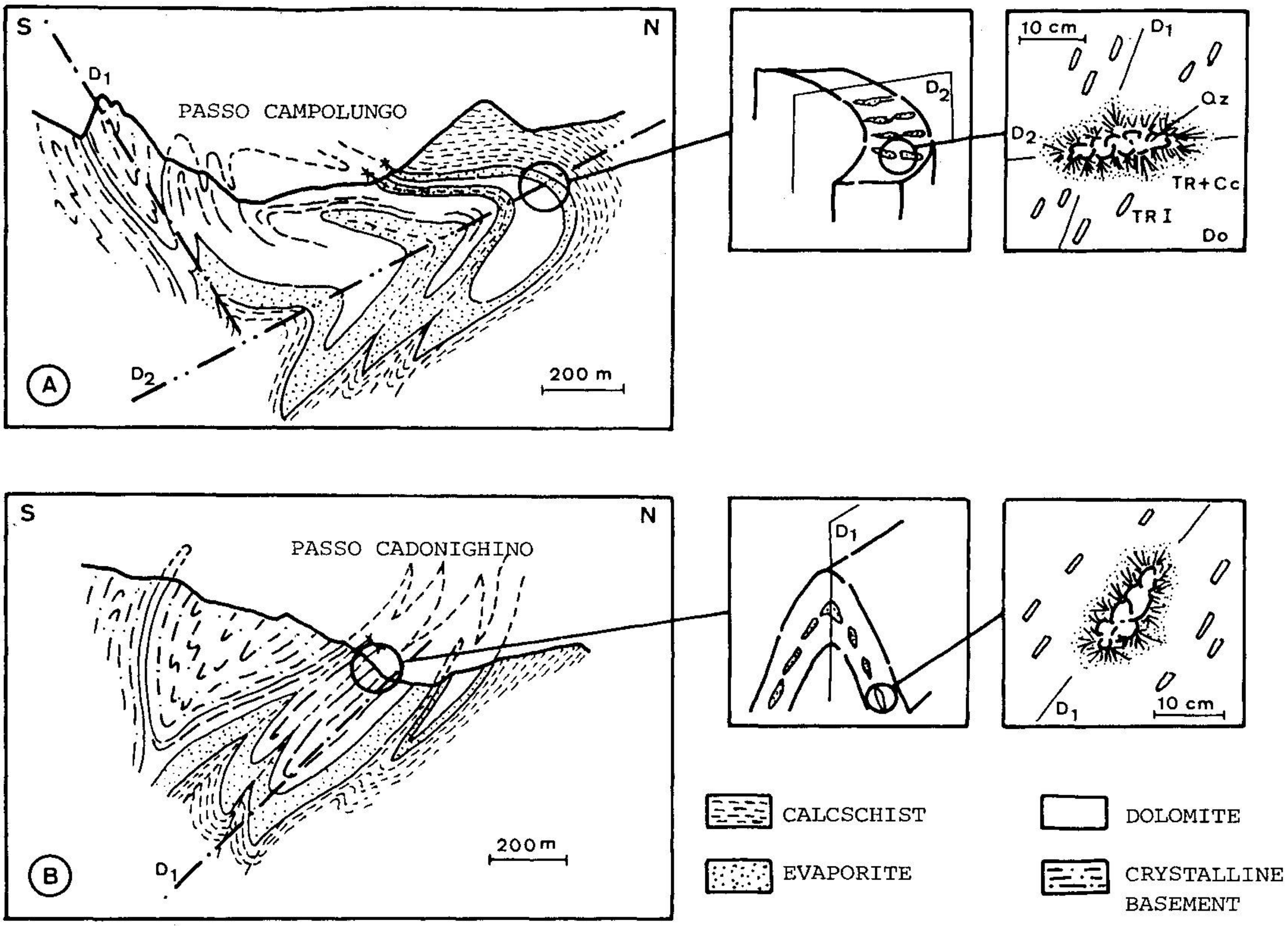

Fig. 2 Geological profiles from Figure 1 with north (N) and south (S) directions indicated. D1 and D2 refer to the axial plane surfaces of folds formed during the deformation phases 1 and 2 respectively. Expanded views of the circled areas are given to the right with the trend followed by quartz nodules and veins indicated. At Passo Campolungo the quartz concentrations lie within the plane defined by D2, whereas at Passo Cadonighino the quartz nodules are within S1, the compositional layering. Further enlargement of the fabric is seen further to the right with the orientation of tremolite 1 (TR I) parallel to D1 in both diagrams and of postkinematic radiating tremolite (Tr) + calcite (Cc) growing outwards from the quartz (Qz) nodules into the dolomite (Do).

scribed above survived the main phase of metamorphism. The failure of quartz to react could have been caused by CO_2-rich fluids resulting from the formation of phlogopite + calcite from potassic feldspar + dolomite (TROMMSDORFF, 1972). As discussed below, the common survival of quartz in close proximity with dolomite through the second stage of tremolite formation could have been due to insufficient H_2O (MERCOLLI, 1979).

Talc is widespread but not ubiquitous and occurs in at least two generations at the Campolungo locality. The talc commonly overgrows both generations of tremolite and is interpreted as a retrograde mineral formed at lower temperatures than both tremolite generations (MERCOLLI, 1979). Talc is not further considered in this paper.

P–T determinations from coexisting minerals and from fluid inclusions suggest much different temperatures and pressures for the main stage of metamorphism and for the later-stage veins. ROSENFELD (1969) and ADAMS et al. (1975) have used a piezometric technique to measure pressures during metamorphism. Rosenfeld reports 600 °C and 8 kilobars for the main stage of metamorphism at Campolungo. FREY et al. (1980) report a temperature of 600 °C at 6 kilobars for the region.

Temperatures based on oxygen isotope geothermometry of pelitic rocks from the area range from 500 to 680°C (HOERNES and FRIEDRICHSEN, 1980).

A detailed study of the fluids accompanying the development of the minerals in the quartz veins has been obtained by microthermometry. Details are available from MERCOLLI (1979, 1982) for Costello and from WALTHER (1983) for Passo Cadonighino.

Tremolite + calcite developed in the later veins at 500°C. This temperature is indicated by various techniques including calcite + dolomite geothermometry, microthermometry (MERCOLLI, 1982, WALTHER, 1983) and by (K,Na)Cl solid inclusions (TROMMSDORFF et al., 1985). Estimates of pressures for these veins have been obtained from the microthermometric studies and range from 1500 to 3250 bars.

Mechanism of vein formation

The distinctive texture of the amphibole and associated calcite in the veins suggests the possibility that these vein minerals could have formed by *precipitation* from a saturated fluid in open vugs and cavities. The vein mineral assemblages are, however, relatively free of oxides, sulfides, sulphates or other minerals that would have precipitated from the cooling of a large amount of introduced fluid. The textures of the vein minerals suggest that tremolite and calcite grew into the enclosing dolomite (WALTHER, 1983, Fig. 2) rather than filling an open cavity.

An alternative mechanism is a reaction among the mineral phases of the system as follows:

$$(1)\quad \underset{\text{dolomite}}{5\,CaMg(CO_3)_2} + \underset{\text{quartz fluid}}{8\,SiO_2} = \underset{\text{tremolite}}{1\,Ca_2Mg_5Si_8O_{22}(OH)_2} + \underset{\text{calcite}}{3\,CaCO_3} + \underset{\text{fluid}}{7\,CO_2}$$

WALTHER (1983) proposed reaction (1) to account for the phase relations in the veins and has demonstrated that the proportion of tremolite to calcite in the zone between dolomite and quartz is consistently close to that predicted by the stoichiometry of this reaction (1). Fluids of appropriate composition could react with the rock to produce an apparently prograde mineral assemblage, tremolite + calcite, during cooling and uplift provided a heat source is available.

Heat sources

The production of tremolite + calcite from dolomite and quartz is an endothermic process that requires the input of a significant amount of heat. This heat is given by the enthalpy of reaction for a process at constant pressure in a closed system. The enthalpy of reaction (1), according to data from ROBIE et al. (1979), is 388 kilojoules per mole of tremolite produced at 500°C.

The production of tremolite I (prismatic) during the main phase of metamorphism occurred by reaction (1) (BIANCONI, 1971, p. 143). This process is likely to have been relatively slow to allow time for regional heat transfer.

The necessary heat for the reaction in the *veins* must have been provided over a geological shorter period of time while veins remained open in brittle rock. Heat consuming reactions such as (1) must be short lived processes where declining temperature indicates overall heat loss. Two possibilities can be considered for the source of this heat: the rock adjacent to the vein provided the necessary heat with a resultant temperature drop, or fluid introduced along the veins carried heat to the reaction.

The geometry of the veins and the heat conductivity of dolomite can be used to determine the volume of rock available for the provision of heat. Typically, the veins are spaced 1 meter apart with 1 mole of tremolite estimated to occupy an average cross sectional area parallel to the vein wall of 10 × 10 centimetres (Fig. 3). A mixture of 5 parts dolomite (density 2.9 grams per cubic centimetre) to 8 parts quartz (density 2.65 grams per cubic centimetre) occupies a 5 cm thickness of vein for a cross sectional area of 10 × 10 cm. The product mixture of 1 part tremolite (density 2.9 grams per cubic centimetre) to 3 parts of calcite (density ~2.8 grams per cubic centimetre) occupies a 4 cm thickness of vein for a cross-sectional area of 10 × 10 cm. Deviation from the stoichiometric proportions of dolomite to quartz increases the cross sectional area occupied by one mole of tremolite for a given vein thickness. A volume of at least

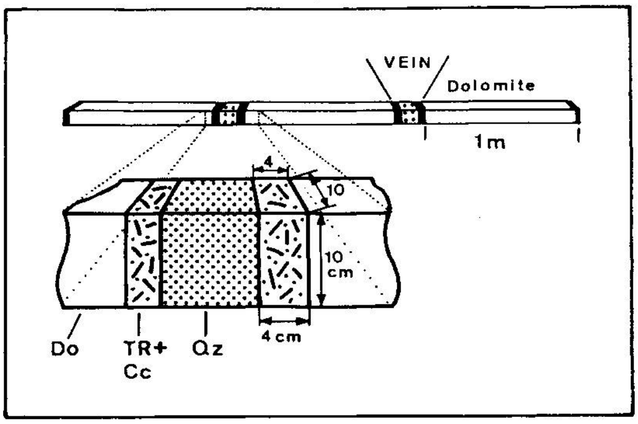

Fig. 3 Schematic representation of mineral relationships within metasomatic veins at Costello (Profile A-A of Fig. 1 and 2). Typically veins are spaced at one metre apart within the dolomite as indicated at the top of the diagram. Relationships within individual veins are further illustrated in the expanded view in the bottom part of the figure. A central dotted region of unreacted quartz (Qz) is bordered on each side by a zone of tremolite (Tr) plus calcite (Cc). The tremolite is of the second generation and has formed by reaction (1) between quartz and dolomite (Do). One mole of tremolite typically occupies the volume indicated on the figure i.e. 10 × 10 × 4 cm.

10 × 10 × 100 centimetres is therefore available, on average, to provide the heat for the production of one mole of tremolite. This volume corresponds to 157 moles of dolomite with a heat capacity of 234 joules per mole at 500 °C (Robie et al., 1979). For constant heat capacity, the heat provided by a temperature drop is given by the product of the heat capacity and temperature change. A temperature drop of 10.6 degrees in this volume of dolomite could therefore provide the 388 kilojoules of heat necessary to produce one mole of tremolite. This energy can easily be transferred within the available time since even average values of heat flow in the crust indicate that the necessary heat can be transferred from the dolomite into the vein in a few tens of years. The actual heat transfer would be even more rapid across high temperature gradients that could be locally created by reaction between rock and fluid.

The amount of fluid needed to carry the heat for the production of one mole of tremolite depends on the heat capacity of the fluid and on its temperature. For example it would require 1011 moles (approximately 26 litres with a specific volume of 1.45 cm^{3}gram^{-1}) of pure H_2O with a heat capacity of 38.36 joules mole^{-1}degree^{-1} at 500 °C to provide 388 kilojoules of heat through 10 degrees of cooling. This relatively large amount of fluid could be reduced by increasing the temperature range for cooling.

Water / rock ratio

The two heat sources described above have implications for the water to rock ratio. The fluid heat source requires water/rock ratios by weight in excess of 10 to pass 1011 moles of fluid through 5 moles of dolomite plus 8 moles of quartz. The superheated rock source requires water/rock ratios of only 0.01 to bring one mole of H_2O to the reacting minerals. Geologic evidence suggests that water/rock ratios were relatively low in the veins (Mercolli, 1979, p. 127). At many places in the veins, unreacted quartz remain in close proximity to dolomite indicating that reaction was being inhibited by factors such as H_2O influx. Water/rock ratios may have been low during vein development to account for this variable extent of reaction.

Fluid origin and evolution

Fluids that were introduced into the dolomitic rocks along the vein fractures were assumed by Walther (1983) to contain 5 weight percent NaCl equivalent. The source of these saline fluids is unknown; the importance of evaporites in the Triassic stratigraphy of the Campolungo has been emphasized above (e.g. Fig. 2). Stable isotopic measurements of the vein carbonates (Table 1) are consistent with typical data for Mesozoic sediments (Veizer and Hoefs, 1976). It is likely, therefore, that the incoming fluid that reacted with quartz and dolomite in the veins was either derived from or extensively interacted with the local mesozoic sequence. Baertschi (1957) reached a similar conclusion from isotopic measurements of vein carbonates in the Mesozoic Bündnerschiefer of Campolungo. The retention of the sedimentary signature in the vein carbonates is also consistent with limited mass transfer between the minerals and the fluid and thus with low water-rock ratios as suggested in the previous section.

Derivation of heat from the wall rock adjacent to the veins or, alternatively, from fluids

Tab. 1 Stable isotopic measurements.

	Carbon		Oxygen	
Sample No.	^{13}C	2σ	^{18}O	2σ
Do H4-1	1.20	0.017	-4.60	0.023
H4-2	1.54	0.016	-3.78	0.023
Cc H4-1	0.78	0.019	-5.46	0.036
H4-2	0.87	0.009	-5.39	0.018
Do TVC-2-1-1	1.39	0.014	-4.58	0.035
TVC-2-1-2	1.26	0.028	-4.94	0.049
Cc TVC-2-1-1	1.34	0.012	-8.21	0.018
TVC-2-1-2	1.51	0.011	-8.00	0.018
Do B7-1	0.44	0.018	-6.76	0.020
B7-2	0.61	0.027	-6.06	0.068
Cc B9-1	1.06	0.028	-6.55	0.019
B9-2	0.96	0.008	-6.17	0.023

Analyst J. Pika, ETH Zürich.
Data reported on PDB scale ‰. Do, dolomite; Cc, calcite.
All samples from Costello. For samples H4, B7, B9, see Mercolli (1982).

migrating along the veins also have consequences for the evolution of fluid composition during the metasomatic process. The large mass of H_2O that would flow through the veins to transport adequate heat precludes significant evolution of the fluid. Reaction progress could not cause a significant change in the composition of the vein fluid as only 7 moles of CO_2 are added by reaction of the 1000 or more moles of fluid needed to supply all of the heat. According to Mercolli (1979) early fluids in the Campolungo veins were very saline. If the fluid provided an important source of heat, then high salinity must have been reached at an earlier stage.

Cooling of the rock is also a source of heat for the reaction when quartz plus dolomite are at a temperature above the equilibrium (1) for the initial fluid composition. The 7 moles of CO_2 produced with each mole of tremolite are capable of causing significant change in the evolving composition of the fluid if the mass of the fluid is small.

The required temperature drop of 10.6 degrees in the wall rock to produce the necessary heat places limits on the composition of the incoming fluid. The composition of this fluid must deviate from the equilibrium fluid at 500 °C by an amount that is capable of sustaining reaction over the temperature interval of 10.6 degrees. Fig. 4 illustrates this requirement for a CO_2-H_2O fluid. This figure has been calculated using thermodynamic data from Berman et al. (1986) and CO_2-H_2O mixing from Kerrick and Jacobs (1981). The equilibrium fluid composition for the assemblage dolomite + quartz + tremolite + calcite at 500 °C, 2000 bars is mole fraction $CO_2 = 0.78$. A mole fraction of 0.69 permits the same assemblage to exist at a temperature lowered by 10.6 degrees. The maximum possible temperature drop in the reacting system is defined by the isobaric invariant point I, talc + calcite + tremolite + dolomite + quartz. A temperature decrease greater than the 40 degrees to this invariant point would have resulted in the production of talc rather than tremolite. Textural evidence indicates that the tremolite formed directly from dolomite and quartz without intermediate talc (see also Mercolli, 1982, and Walther, 1983). These constraints on fluid compositions and temperatures apply to pure CO_2-H_2O mixtures. Additional components in any of the phases would alter the given values. The presence of salt, for example, in the fluid has been indicated by both Mercolli and Walther. Trommsdorff et al. (1985) have presented evidence that indicates that at least in one instance saline aqueous brines at the reaction site were saturated in (Na,K)Cl. The amount of fluid

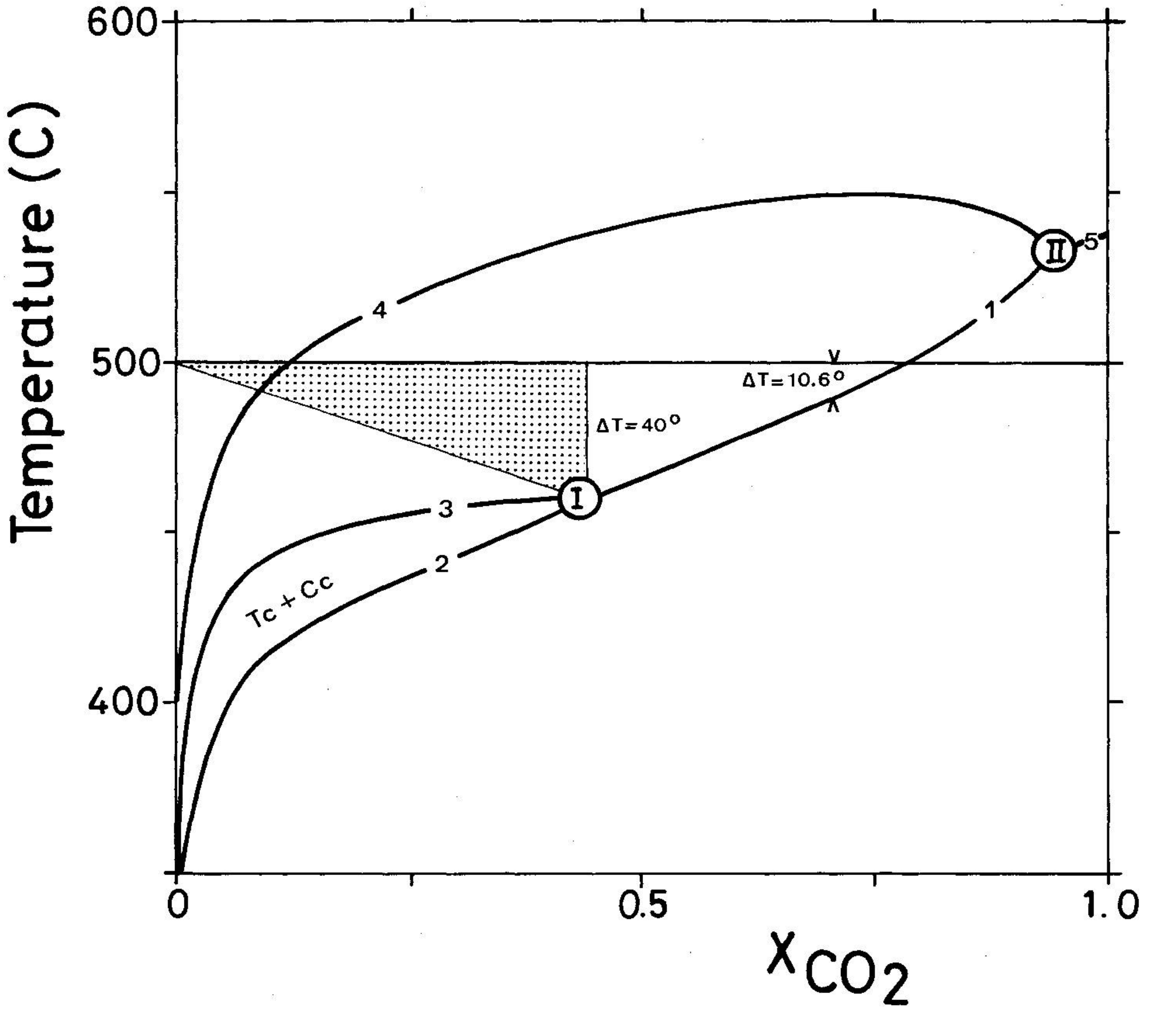

Fig. 4 Temperature versus mole fraction CO_2 at 2000 bars for five reactions commonly observed in metamorphosed dolomitic marbles. (1) Quartz + dolomite → tremolite + calcite. (2) Quartz + dolomite → talc + calcite. (3) Talc + calcite + quartz → tremolite. (4) Tremolite + calcite + quartz → diopside. (5) Quartz + dolomite → diopside. Point I indicates talc + tremolite + quartz + dolomite + calcite, point II indicates tremolite + diopside + quartz + dolomite + calcite. The 500 °C isotherm intersects reaction (1) at mole fraction CO_2 = 0.78. The heat required for the production of 1 mole of tremolite requires a temperature decrease of 10.6 degrees as indicated between the vertical arrows. The composition of the introduced fluid must therefore be more H_2O-rich than that indicated by the vertical arrows in order to produce one mole of metasomatic tremolite. The maximum temperature decrease expected in the Campolungo rocks is determined by the fluid composition at point I at which the production of metasomatic talc would occur. According to the data on figure 4 this temperature decrease is 40 degrees. Initial fluid compositions within the shaded area could result in tremolite production by reaction (1) without encountering the talc stability field.

evolution that may have occurred to bring the incoming fluids to saturation is unknown, as there is no reliable indication of the composition of the incoming fluids. An undersaturated fluid must, however, encounter the two-phase region in order to be driven to saturation by progress in reaction (1) according to TROMMSDORFF and SKIPPEN (1986). This requirement places a limit on the composition of the incoming fluid. Fig. 5 presents a composition section (BOWERS and HELGESON, 1983) through CO_2-H_2O-NaCl at 500 °C and 2000 bars with a fluid evolution line for the stoichiometry of reaction (1). Only fluids within the shaded area can evolve to subcritical conditions. An aqueous brine in NaCl-H_2O with a mole fraction of 0.35 NaCl (i.e. 11 weight percent NaCl) is therefore a limiting composition for fluids that could eventually evolve to saturation at 500 °C and 2000 bars.

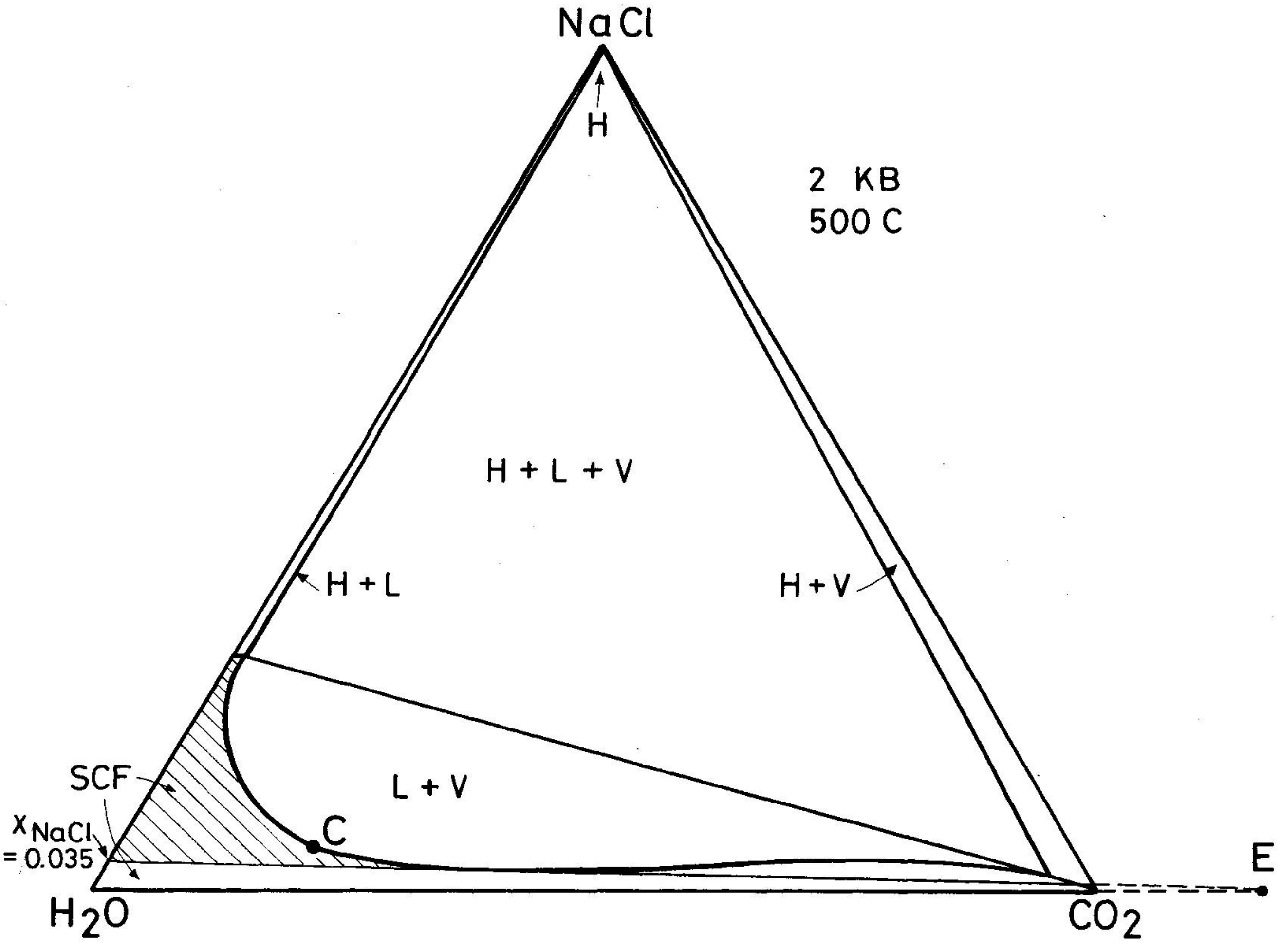

Fig. 5 Composition section for the ternary, CO_2-H_2O-NaCl at 500 °C. 2 kbar, as given by BOWERS and HELGESON (1983). H = halite, L = liquid, V = vapour, SCF = supercritical fluid. The consolute point (C) indicates the composition at which the tie-line joining liquid + vapour is reduced to a point. The region of supercritical fluids is divided into two parts by the fluid evolution curve from mole fraction 0.035 to the hypothetical endpoint (E) determined by the stoichiometry of reaction (1), see TROMMSDORFF and SKIPPEN (1986); that part of the supercritical region indicated by the ruled pattern contains those fluid compositions that can evolve to the two phase region, L + V, when reaction progress drives the fluid composition towards the endpoint E. The removal of the CO_2-rich vapour leaves a residual liquid in the system which evolves towards saturation in halite with continuing progress in reaction (1). The remaining part of the supercritical region involves fluids which do not encounter the two phase region as a result of progress in reaction (1).

Concluding statement

TROMMSDORFF and SKIPPEN (1986) consider a "boiling" system as an explanation for saturation of the product fluids at sector B (MERCOLLI, 1982, Fig. 4) of the locality Costello at Campolungo. This locality is at the highest vertical exposure in the veins and is approximately 100 m above the lowermost exposures at Costello. The term boiling is used here to suggest the separation of a CO_2-rich vapour from an aqueous NaCl-rich brine and the subsequent loss of the lower density vapour from the system through highly permeable rock. The heat consuming step in this process is the production of a large number of moles of fluid as a result of reaction. The segregation of CO_2 has a small effect on the heat budget of the system (see DRUMMOND and OHMOTO, 1985, p. 127; and WOOD and Spera, 1984).

Open system boiling as described, for example, by DRUMMOND and OHMOTO (1985) may have been facilitated at the comparatively high pressure and temperature of the Campolungo veins by the mechanical properties of dolomite rock. Dolomite exhibits greater strength than most common rocks under these conditions (Heard, 1976), and may have been capable of maintaining open fractures under conditions of less than lithostatic pressure on the fluids. Hydrostatic fluid pressures have been observed in the Kola borehole to depth of 8000 metres (Kozlowski, 1984). The possible depth of the Campolungo veins is from 6000 metres under lithostatic conditions of 2000 bars to much deeper levels under hydrostatic conditions.

The salt-saturated subcritical fluids associated with the product minerals are the earliest fluid inclusions that have been recognized in the veins. Later generations of lower temperature inclusions progressively decrease in salinity to nearly pure H_2O. This decreasing salinity suggests that salt from the evaporitic source rocks was no longer being dissolved by fluids passing through the Campolungo rocks, or that meteoric waters contributed significantly to the later fluids. The cessation of vein mineral development is expected when the amount of fluid passing through the veins decreased or the rock supplying heat had been cooled to the equilibrium temperature for the composition of the incoming fluid. With a decreasing amount of fluid, the buffering capacity of the rock would adjust the fluid composition to maintain equilibrium at the regional temperature.

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