Recognition and significance of multiple fluid inclusion generations in telogenetic calcites

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Abstract

Ferroan and non-ferroan calcites occur in fractures in the Lower Carboniferous of the Variscan foreland of southern Belgium. These fractures post-date the Variscan orogeny and the calcites have a telogenetic origin. The non-ferroan calcites formed by recrystallization of the ferroan calcites. Two types of monophase aqueous fluid inclusions are present in the ferroan calcite cement. Although both types of inclusions formed at a temperature not exceeding 50°C, one type contains a moderately saline fluid (3.6–16.3 eq. wt.% NaCl) and the other type fresh water (Tm ice = 0°C). The fluid inclusions in the non-ferroan calcite also contain fresh water.

Detailed petrography of the fluid inclusions indicate that the fresh water migrated through the crystals along opened cleavage planes and microfractures. Therefore, they have a secondary origin. The recrystallization of the ferroan calcites to non-ferroan calcites occurred in this fresh water. The saline fluid inclusions are not related to the above mentioned microstructures and although their origin remains unknown, they could represent the ambient fluid from which the ferroan calcites precipitated. The study of the relationship between the occurrence of fluid inclusions and the microstructures seems promising for the identification of fluid inclusions representing post mineral formation fluid and temperature conditions in calcite cements.

KEYWORDS: telogenetic calcites, fluid inclusions, fresh water, cleavage planes, microfractures.

Introduction

DURING the last decade fluid inclusion data have been increasingly used in studies of carbonate diagenesis. Microthermometry provides information on the composition, salinity and temperature of fluids from which diagenetic minerals precipitate (Aulstead and Spencer, 1985; Wojcik *et al.*, 1994). However, carbonates often contain several generations of fluid inclusions, partly reflecting the multiple fluids which migrated through the rock. During burial and tectonic deformation, resetting of primary inclusions and formation of secondary inclusions may occur. Secondary inclusions form later than the crystal in which they are found. Their origin is related to mechanical deformation of the crystal or to chemical reactions (Leeder *et al.*, 1987). Resetting of primary inclusions and formation of secondary inclusions occurs easily in carbonate minerals due to the presence of cleavage planes, twin lamellae and microfractures. Also secondary inclusions may reset after their formation. Likely mechanisms of resetting have been summarized by Goldstein (1986) as:

- stretching of the fluid inclusion cavities due to high positive pressures,
- hydrofracturing and opening of the inclusion cavities due to high internal pressures within the inclusions,
- the intersection of planes of crystal imperfections with the inclusions and hence the opening of the inclusions to exchange with an external pore fluid,
- neomorphism of the host mineral.

The first three processes often occur during burial and natural heating (Goldstein, 1986; Burrus, 1987; Prezbindowski and Larese, 1987). The fourth process can take place due to any significant change in the physico-chemical conditions of the environment.

The aim of this paper is to document the presence and composition of several fluid inclusion generations in fracture-filling calcites which formed after maximum burial and major tectonic deformation. By using a combination of detailed petrography and microthermo-

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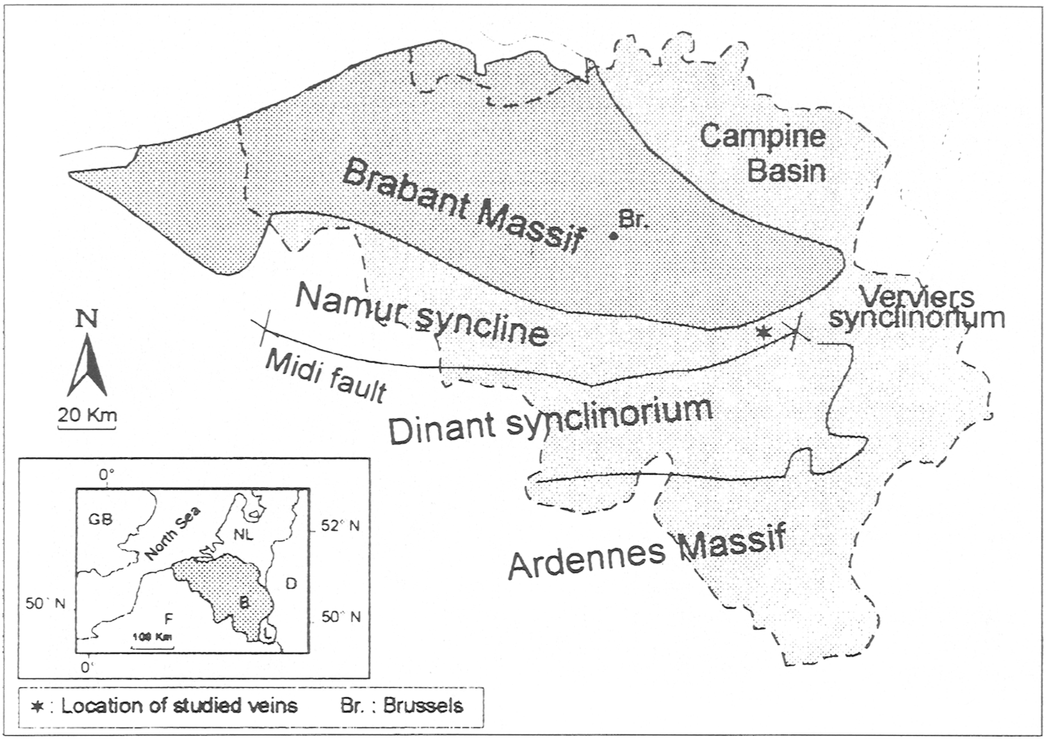

FIG. 1. Tectonic structure of the Variscan and location of the studied veins. B—Belgium, D—Germany, F—France, GB—Great Britain, L—Luxemburg, NL—Netherlands.

metry, we demonstrate the importance of fluid migration along opened cleavage planes and crystal imperfections for the composition of inclusions in fracture-filling calcites. This research shows that important information can be deduced from the relationship between microstructures in carbonates and the distribution of fluid inclusions and their characteristics.

Geological setting

Blocky ferroan and non-ferroan calcite cements, filling 1 cm and 140 cm wide fractures in the Visean (Lower Carboniferous) limestones on the northern flank of the Namur syncline, southern Belgium (Fig. 1), have been examined. The Namur syncline consists of Devonian and Carboniferous sediments which had been deformed at the end of the Carboniferous during the Variscan orogeny. In the eastern part of the Namur area, the Visean has been buried beneath 2300 to 3000 m of Upper Carboniferous strata. The maximum burial temperature was around 150°C (Helsen and Köningshof, 1994). No sedimentation took place during the Permian, Triassic and Jurassic. From the thickness distribution of the Cretaceous and Cenozoic rocks and from their palaeogeography, it can be deduced that the original thickness of the Cretaceous and Cenozoic did not exceed a hundred metres (Gullentops, pers. comm., 1995).

The 140 cm wide fracture has a NE–SW orientation with a dip to the NW (N30E72W) and cross-cuts the NE–SW trending Variscan folds and faults and Variscan non-ferroan calcite veins (see also Muchez *et al.*, 1995). The 1 cm wide fracture is located about 10 km from the former vein. Its orientation is unknown, however the vein cross-cuts and therefore post-dates an E–W oriented karst system of early Cretaceous age. The E–W orientation has been inherited from the Variscan orogeny. In the area studied, the karst has been filled with non-ferroan calcites and iron-oxides/hydroxides. To the west in the Mons basin, Wealdian continental sediments fill the karstic depressions (Vandycke *et al.*, 1991). Since the ferroan calcites formed after the early Cretaceous and since the Carboniferous strata have never been buried beneath more than 100 m after the Variscan, this cement has a telogenetic origin (*sensu* Choquette and Pray, 1970).

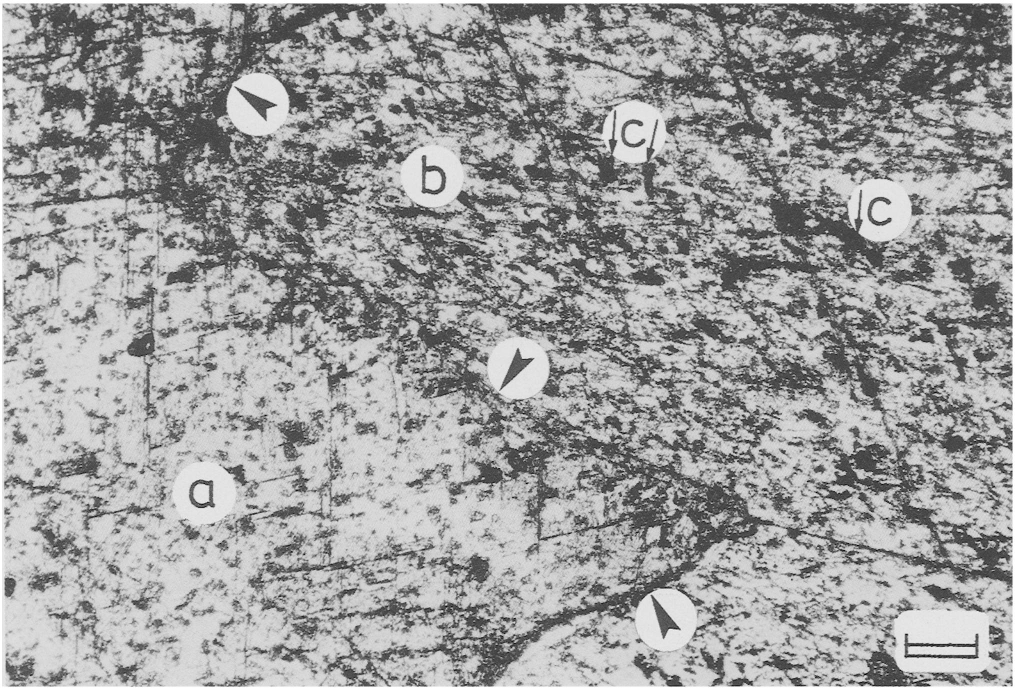

FIG. 2. Thin section photomicrograph showing ferroan calcite (a, clear) and an almost completely recrystallized crystal consisting of non-ferroan calcite (b) spotted with iron-oxides (c). Arrows mark the boundary between the ferroan and non-ferroan calcites. Scale bar is 170 μm.

Methodology

The calcite cements have been studied by optical and cold cathodoluminescence (CL) petrography. Polished slabs and thin sections were stained with Alizarin Red S and potassium ferricyanide. CL petrography was carried out with a Technosyn Cold Cathodoluminescence Model 8200 MkII. Operating conditions were 16–20 kV voltage, 0.42 mA gun current, 6.7 Pa vacuum and 5 mm beam width. Microthermometric analyses of fluid inclusions were performed on a Linkam heating-cooling stage. The fluid inclusions were selected by a petrographic study of one doubly-polished, ~150 μm thick section of each vein. Special attention was paid to a possible relation between the occurrence of each inclusion and the mineralogical characteristics. Therefore, photographs of the position of the inclusions within the crystal have been made under conventional and CL microscopy after the microthermometric analyses. A detailed description of the sample preparation technique and of the measurement procedure has been presented by Muchez *et al.* (1994). Reproducibility of the final melting temperature of ice (Tm_{ice}) was within 0.2°C.

Petrography and fluid inclusion analysis

Description and measurements. A solution of Alizarin Red S and potassium ferricyanide stains most parts of the calcites purple, indicating they are iron-rich (Dickson, 1966). Calcites in thin fractures (10–500 μm wide), cross-cutting the ferroan calcites, have a red colour and therefore are non-ferroan. The microfractures can also be filled with iron oxides (Fig. 3A). Some parts of ferroan calcite along these microstructures have been dissolved and filled with non-ferroan calcites and iron-oxides. The size of the corroded areas varies between a few tens and 200 μm. Also the outer parts of the ferroan calcites in the 1cm thick vein often stain red. This is due to recrystallization of the ferroan cement to non-ferroan calcite (Fig. 2). Ferroan calcites are blocky, show uniform extinction and do not contain twin lamellae. The crystals have a grey to brown colour and their sizes are between a few mm and several cm. Under CL they show a dull brown-orange luminescence. Cleavage planes are intensely developed (up to 10 per mm) and can be slightly curved. Non-ferroan calcites have a bright yellow to ochre luminescence.

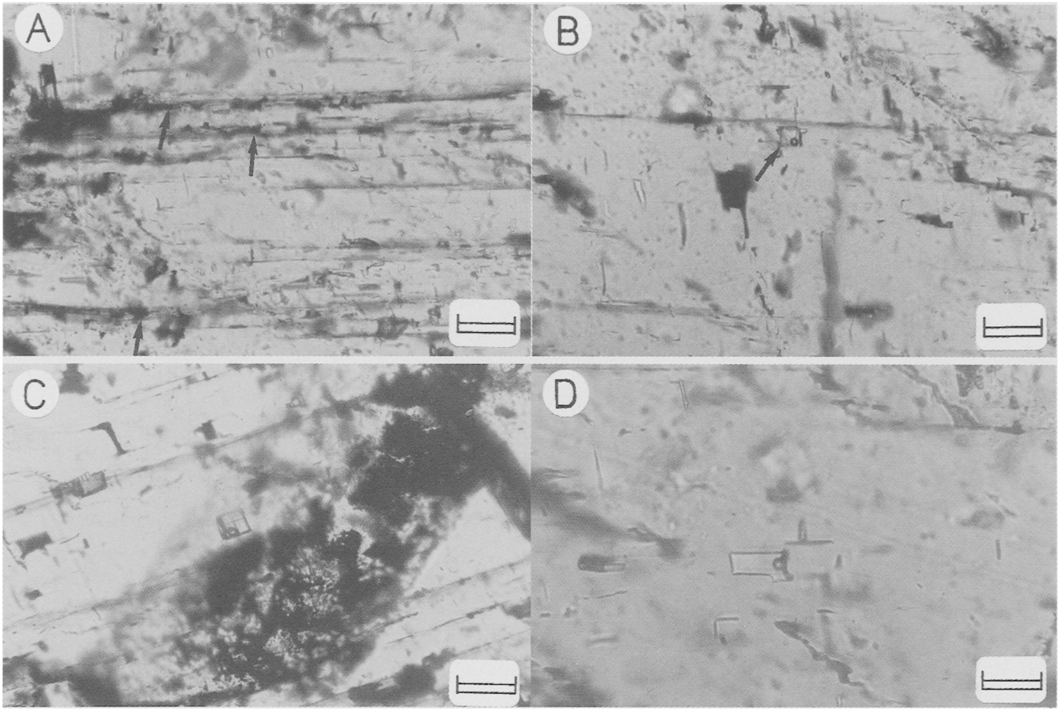

FIG. 3. Thin section photomicrographs of ferroan calcites. The two-phase inclusions were originally one-phase all-liquid inclusions which have been artificially stretched. A) Cleavage planes (arrows) filled with iron-oxides and liquid. Scale bar is 60µm. B) Inclusion (arrow) situated near cleavage plane (Tm_{ice} = 0°C). Scale bar is 60µm. C) Inclusion near fracture filled with iron-oxides (Tm_{ice} = −0.3°C). Scale bar is 60µm. D) Large type F1 inclusion with a Tm_{ice} of −10.5°C. Scale bar is 25µm.

Monophase aqueous inclusions are present in ferroan calcites. They occur isolated, in clusters or as trails. Trails of secondary inclusions occur throughout the ferroan calcite, but the size of most inclusions (<3 µm) does not allow a microthermometric study. Larger inclusions (5–60 µm) occur as part of a random distribution and in planes. No inclusion-poor and inclusion-rich areas delineating growth zones are present. Large inclusions are often present near cleavage planes and microfractures. Cleavage planes and microfractures are commonly healed and filled up with liquid, forming secondary inclusions.

Monophase aqueous inclusions also occur in recrystallized calcites and in non-ferroan calcites filling the corroded areas. In recrystallized calcites, fluid inclusions have a random distribution and represent conditions of recrystallization. In calcites filling corroded areas, inclusions occur in growth zones and have a primary origin. The size of inclusions measured is between 7 and 21 µm.

Despite the large size of some of the monophase aqueous inclusions in the calcites, no vapour bubble appeared after multiple periods of cooling. In the ferroan calcites, first melting temperature (Te) could be measured on more than a third of the artificially stretched (e.g. Goldstein *et al.*, 1990) monophase inclusions and lies around 20°C (type F1 inclusions). The final ice melting temperatures (Tm_{ice}) of the F1 inclusions in the 140 cm wide vein vary between −12.4° and −7.5°C and in the 1 cm wide vein between −3.4° and −2.1°C (Fig. 4). The inclusions with no obvious eutectic melting in both veins have final melting temperatures around 0°C (type F2 inclusions). All inclusions in the non-ferroan calcites have Tm_{ice} values around 0°C (type N1 inclusions, Fig. 4).

A comparison of the microthermometric data of inclusions with their position in the crystals showed that type F2 inclusions are characteristically situated near healed cleavage planes (Fig. 3B) or near healed microfractures (Fig. 3C). The longest dimension of the inclusions are not necessarily parallel to the direction of the cleavage planes and microfractures. They may occur parallel, perpendicular or randomly

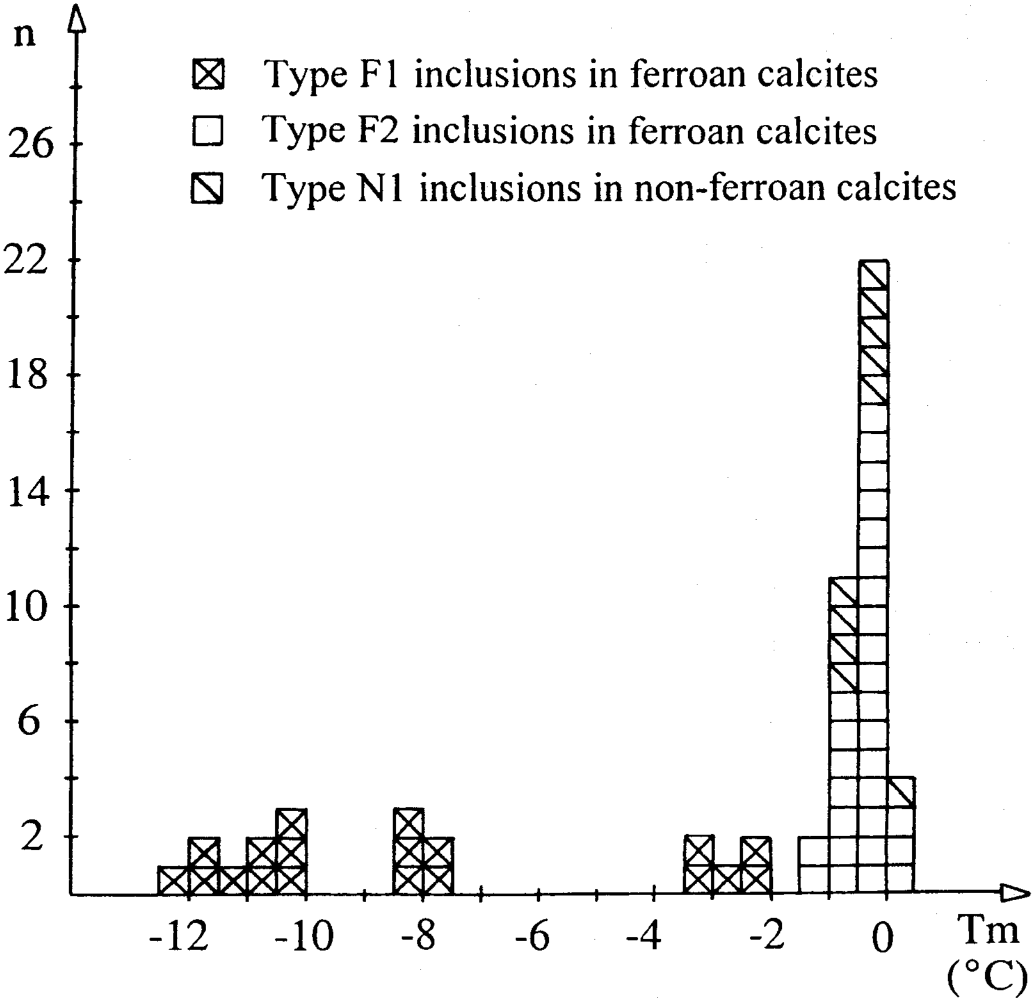

FIG. 4. Histogram of the final melting temperatures (Tm_{ice}) of the fluid inclusions in the ferroan and non-ferroan recrystallized calcites. The class intervals used are 0.5°C.

oriented to these directions. The size of the inclusions studied is significantly greater than the width of the cleavage planes and most microfractures. The type F1 inclusions are present throughout the crystals and are located near or at a distance from fractures and cleavage planes (Fig. 3D). The type N1 inclusions partly occur in growth zones and represent conditions of non-ferroan calcite precipitation.

Interpretation. No vapour bubble appeared in the monophase aqueous inclusions after temperature cycling, indicating that they are most likely not metastable (see also Muchez *et al.*, 1994). Monophase aqueous inclusions form at a temperature around or below 50°C (Sabouraud *et al.*, 1980). This low temperature is in agreement with the reconstructed geological history, which indicated precipitation of the ferroan calcites after the early Cretaceous and under a minimal post-Variscan burial.

Two distinct fluid inclusion populations have been recognized in the Tm_{ice} data from the ferroan calcites. The final melting temperature in the type F2 inclusions indicates a very low salinity, likely representing meteoric water. The observation that the type F2 inclusions typically occur near cleavage planes, within secondary planes and near microfractures strongly suggests that the fluids in these inclusions were trapped after crystal growth and migrated along then open cleavage planes and microfractures. Such a migration is clearly indicated by the presence of non-ferroan calcites or iron oxides in the cleavage planes and microfractures. The presence of the low salinity N1 fluid inclusions (Tm_{ice} = 0°C) in the recrystallized calcites shows that recrystallisation of ferroan calcites to non-ferroan calcites (and iron-oxides) took place in fresh water. The large type F2 inclusions near cleavage planes or microfractures in the ferroan calcites could have formed during the opening of these structures but could also represent re-equilibrated inclusions, already present before the opening. According to the latter hypothesis, the earlier formed inclusions leaked and became refilled with a new fluid during the opening of the planes and the creation of fractures. However, tiny micofractures at the boundaries of inclusions, which would support this hypothesis, have not been recognised.

The Te data of the type F1 inclusions (~−20°C) indicate a NaCl–H_2O composition for the fluids. The final melting temperatures between −12.4° and −7.5°C, in the 140 cm wide vein, and between −3.4° and −2.1°C in the 1 cm wide vein, correspond respectively with a salinity range of 16.3−11.1 eq. wt.% NaCl, and of 5.6−3.6 eq. wt.% NaCl (Bodnar, 1993). In the samples investigated, the origin of the type F1 inclusions cannot be ascertained since no relationship with growth zones or healed fractures can be observed. They could represent either primary or secondary inclusions. In comparable post-Variscan ferroan calcite veins, primary inclusions have a high salinity (9.2−23.2 eq. wt.% NaCl; Muchez *et al.*, 1995). These ferroan calcites precipitated between 40° and 60°C from a meteoric water with a $\delta^{18}O$ between 0.6‰ and 2.0‰ SMOW, which underwent an intense water–rock interaction (Muchez *et al.*, 1995). These fluids migrated upward from the deeper subsurface (>1 km) along fractures and faults. A similar origin can be proposed for the F1 inclusions which formed at or below 50°C. Although no precise trapping temperature is available for the F1 inclusions, it could be speculated that moderate to low salinity fluids in the F1 inclusions formed at lower temperatures than the high salinity fluids in the previously investigated ferroan calcite veins. High-salinity inclusions have often been found in ferroan calcites (e.g. Dorobek, 1987; Muchez *et al.*, 1991). In addition, several ferroan calcite occurrences are interpreted to have precipitated from meteoric water (Emery and Dickson, 1989; Nelson and Read, 1990).

In summary, the following processes have been recognized in the fractures investigated:

– precipitation of ferroan calcites, perhaps from moderately saline fluids which migrated upwards along fractures and faults from deeper parts of the basin,

– infiltration of fresh water along opened cleavage planes and microfractures,

– recrystallization of ferroan calcite to non-ferroan calcite and iron-oxides in this fresh water, along with precipitation of non-ferroan calcites in dissolved parts of the ferroan cement.

Conclusion

Two types of large monophase aqueous inclusions are present in the telogenetic calcites in the Lower Carboniferous of the Namur syncline. The first has a moderate salinity (3.6−16.3 eq. wt.% NaCl) and the second a very low salinity (~0%). Petrographic relations show that low salinity waters migrated through the ferroan calcites along opened cleavage planes and microfractures. They also caused a recrystallization of ferroan calcites to non-ferroan calcites and iron oxides.

A detailed study of the relationship between microstructures in carbonates and the occurrence and characteristics of fluid inclusions can be very significant for the identification of post mineral precipitation fluid and temperature conditions in vein cements and host-rock. In future fluid inclusion research, such an approach could be used to distinguish primary from secondary (which do not distinctly occur in trails), or resetted inclusions in growth zones. The investigation of a small number of petrographically well-defined inclusions can give more useful information than that of a large group of randomly selected inclusions (see also p. 67 in Goldstein and Reynolds, 1994).

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