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# Fluid inclusions: tectonic indicators

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#### Abstract

During the first half of the 20th century fluid inclusions have been studied in ore geology in order to determine the chemistry and physical parameters (density) of the mineralising fluids. Apart from a few pioneering works in the middle of the century, fluid inclusions were not used as structural markers before the late 1980s. During the same time, experiments as well as observations of natural samples have shown that the volume, shape and density of fluid inclusions may change when submitted to pressure-temperature conditions different from their trapping isochores.

In view of these experiments, and based on natural examples from the Higher Himalaya metamorphic rocks, it is evident how fluid inclusions can be used as thermobarometric, structural and tectonic markers. Points of interest for further studies on fluid inclusions in nature and experiments are also stressed. © 1999 Elsevier Science Ltd. All rights reserved.

### 1. Introduction

"... and I argue that there is no necessary connection between the size of an object and the value of a fact, and that, though the objects I have described are minute, the conclusions to be derived from the facts are great." H.C. Sorby (1858)

Even with their minute size, fluid inclusions have been studied for a long time. Sorby's (1858) work may be considered as the first scientific work on these small traces of fluids circulating through rocks. Although these objects have been studied in several countries since that time, it is only during the early 1960s that experiments on fluid systems and development of new microthermometric stages have allowed considerable progress of research on fluid inclusions. As it can be seen by the chapter headings in the fluid inclusionist's bible (Roedder, 1984), applications of fluid inclusions exist in diverse geological disciplines such as ore deposits, sedimentary geology, metamorphic petrology, studies of crustal and mantle magmas, lavas and lunar geology. Tuttle (1949) and Wise (1964) were pioneers in using fluid inclusions (FI) as structural markers, but they were not imitated until the late 1980s. Moreover, high-pressure and temperature experiments have shown that fluid inclusions may change in shape and volume, as suggested by Skinner (1953), giving then some answers to the questions addressed by Roedder (1984, p. 576) as possible developments on fluid inclusion research.

This paper is in some way an extension of Crawford's paper (Crawford, 1992) and the questions I will address are: what can we learn from fluid inclusions with respect to the structural and tectonic history of rocks? How can recent experimental results be applied to natural samples? These questions will be addressed in this paper by using observations on natural samples, mainly from the Himalayas, and comparing them with experimental results to point out the many possible applications of fluid inclusion studies.

#### 2. Geological setting of the Himalayas

The Himalaya mountains result from a convergent orogeny where crustal shortening and thickening are

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Fig. 1. Orientations of fluid inclusion planes in quartz lenses from the Main Central Thrust pile. One hundred measurements in each stereogram. Lower hemisphere projection. Density contours at 1, 2, 5, 8 per cent of the data in a 0.55% area counting grid. NL10 is located below the MCT and NL116 and NL91 in the upper levels of the High Himalaya crystalline terrains. FIP in NL10 and NL116 are symmetamorphic  $H_2O-CO_2$  fluids within planes subperpendicular to foliation and lineation; their asymmetry relative to the lineation is consistent with a southwestward thrusting. FIP in NL91 are  $H_2O$  fluids perpendicular to the foliation plane and parallel to the lineation visible on the outcrop.

related to southward thrusting within the Indian plate (Le Fort, 1975; Molnar, 1984). The Main Central Thrust or MCT brought the high-grade High Himalaya crystalline terrains and the High Himalaya sedimentary series onto the low-grade metasedimentary Lesser Himalaya formations leading to an inverted metamorphic gradient (Gansser, 1964). P-T conditions calculated from mineralogical assemblages in the High Himalaya crystalline terrains above the MCT (Brunel and Kiénast, 1986; Hodges and Silverberg, 1988; Hubbard, 1989; Pêcher, 1989) indicate a clockwise P-T-t path due to crustal thickening followed by uplift and tectonic denudation resulting from back-folding in the High Himalaya sedimentary series and back-faulting (Le Fort, 1975; Burg et al., 1984). The whole pile has been ductily and strongly deformed (Bouchez and Pêcher, 1981; Brunel and Kiénast, 1986) and shows a schistosity subparallel to the MCT. Syntectonic and synmetamorphic quartz lenses and veins are frequent in the pile. The examples of fluid inclusions cited in this paper are from these quartz lenses.

## 3. Nature of the fluids

Pêcher (1979), Sauniac and Touret (1983) and Craw (1990) have studied fluid inclusions along sections across the MCT pile. They have determined the  $CO_2$ -H<sub>2</sub>O composition of the fluid inclusions with an increase of  $CO_2$  content toward the MCT. All these fluid inclusions are secondary, i.e. within healed microcracks. Density calculation of the fluid inclusions and the relative position of corresponding isochores and metamorphic mineral assemblages indicate that fluid inclusions rarely record the maximum P-T conditions

or the metamorphic peak but rather register lower pressure conditions. This has been interpreted by Pêcher (1981) as related to density changes of fluid inclusions during the uplift stage of the pile. The origin of the fluids was also questioned by Pêcher (1979): are they metamorphic or of lower crustal origin? A stable isotope study on the same quartz lenses and their wall rocks has shown that these  $H_2O-CO_2$  fluids are of metamorphic origin and result from the devolatilisation of the Lesser Himalaya formations below the MCT. These fluids have percolated throughout the whole MCT pile (France-Lanord et al., 1988).

The three studies cited above have also described a late generation of aqueous fluids in the MCT pile, which appears to be meteoric in origin (France-Lanord et al., 1988). The aim of the present paper is to demonstrate the tectonic signification of the orientation of fluid inclusion planes, the shape and density of inclusions with references to samples from the MCT pile and, then, to ask questions for the future.

### 4. Orientation of the fluid inclusion planes

As quoted above, fluid inclusions in quartz lenses from the MCT pile are secondary, i.e. they lie within cracks which have been healed by dissolution–crystallisation processes (Tuttle, 1949). Such an interpretation of the origin of fluid inclusion planes has been confirmed by experiments on crack healing or on synthetic fluid inclusions (Shelton and Orville, 1980; Smith and Evans, 1984; Brantley et al., 1990). The vast majority of observed microcracks in rocks, and therefore of healed microcracks (Fluid Inclusion Planes or FIP) appear to be extensional (Tapponnier and Brace, 1976;



Fig. 2. Initiation at high angle to the stretching lineation and on the grain boundaries of  $H_2O-CO_2$  FIP in a quartz lens located below the MCT. The section is perpendicular to the foliation plane and parallel to the stretching lineation (horizontal).

Kranz, 1983), i.e. mode I cracks (see Atkinson, 1987) within the  $\sigma_1$ - $\sigma_2$  plane.

Starting from that fact, people have used the orientation of healed microcracks (FIP) in guartz from granites or sandstones in order to determine regional paleostress histories (Lespinasse and Pêcher, 1986; Kowallis et al., 1987; Jang et al., 1989; Laubach, 1989; Ren et al., 1989; see the review of this aspect by Lespinasse, 1999). Similar studies have been made on rock samples from the metamorphic MCT pile of the Himalayan mountains and some examples are shown in Fig. 1. FIP are easily measured on a petrographic microscope using an XY stage and the calibrated focusing screw. The H<sub>2</sub>O-CO<sub>2</sub> fluid inclusion planes are generally oriented at a high angle to the stretching lineation and, therefore, are consistent with the synmetamorphic deformation of the pile (Fig. 1, NL10 and NL116 samples, Fig. 2) and generally with a southwestward thrusting. In contrast, the late FIP display a completely different orientation and are either vertical (H<sub>2</sub>O, Fig. 1, NL91 sample) or subhorizontal (late H<sub>2</sub>O-CO<sub>2</sub>, Fig. 3b; Boullier et al., 1991). It should be noted that aqueous fluid inclusions are restricted to the upper units of the pile, thus confirming their relation to the surface and their meteoric origin. Consequently, it is possible to distinguish the different generations of FIP from their orientation relative to the tectonic structures and to attribute fluids of different composition or different sources to different tectonic events.

The occurrence of inclusion planes also suggests that rocks may behave in a brittle manner in conditions where they are expected to behave ductily (Swanenberg, 1980). Fig. 2 shows initiation of  $H_2O CO_2$  bearing FIP at grain boundaries in an orientation which is consistent with the stretching lineation. Grain boundaries are the locus of stress concentration due to strain incompatibilities and sliding between neighbouring grains and this, therefore, enhances initiation of microcracks. Processes of wrapping fluid inclusions out of the grains by strain-driven grain boundary migration have been described in quartz by Kerrich (1976) and Wilkins and Barkas (1978). If they have not leaked, they may give the P-T conditions of the recrystallisation (Boullier et al., 1997). However, in other cases such as in Himalayan quartz lenses (Fig. 2), grain boundaries are sources rather than sinks for fluids. Looking at the large number of FIP (around 15 FIP/mm in Fig. 2), it seems that the amount of fluid within the grain boundaries may be important and, therefore, may have drastic consequences on the rheological behaviour of the rock.

#### 5. Shape of fluid inclusions

In a first stage of crack healing, fluid inclusions are thin and sheetlike, but in more advanced stages, they acquire a negative-crystal shape which corresponds to their 'lowest possible energy level' (Tuttle, 1949). It is possible then to deduce the aperture of the crack from the geometry and arrangement of fluid inclusions. Onasch (1990) has used the fluid inclusion average diameter and separation in FIPs, and the FIP spacing in order to estimate the finite strain in quartz arenites. We have calculated the aperture of initial microcracks in Himalayan quartz lenses by analysing microphotographs of fluid inclusion planes using the public domain NIH Image program (U.S. National Institutes of Health, 1998). The volume of fluid has been calculated by summing the products of the surface of each fluid inclusion by its smallest diameter (cylinder geometry), and the initial opening of microcracks by dividing the fluid volume by the surface of the analysed image. It is generally one order of magnitude less than the average diameter of the fluid inclusions after healing. As a  $10^{-2}$  aspect ratio is generally observed for FIP (Boullier et al., 1991), a  $10^{-3}$  aspect ratio may be deduced for microcracks before healing. This value is similar to the  $10^{-3}$ - $10^{-4}$  aspect ratio measured for experimentally stress-induced microcracks (Tapponnier and Brace, 1976).

Subsequently, this low-energy configuration (negative crystal shape) may be destroyed if the fluid inclusions are not in equilibrium with their physical environment, i.e. if the pressure and temperature do not correspond to the isochoric line of the inclusions. This has been experimentally reproduced by Pêcher (1981), Gratier and Jenatton (1984), Pêcher and Boullier (1984), Boullier et al. (1989), Sterner and Bodnar (1989) and by Vityk and Bodnar (1995). Changes in shape may be of two types: rapid or slow.



Fig. 3. Schematic post-trapping evolution of  $H_2O-CO_2$  fluid inclusions. Densities decrease from isochoric line 1 to 3. (a) P-T-t path representing a nearly isothermal loading path; fluid inclusions are transformed into annular fluid inclusions (different grey levels correspond to inclusions within different planes of focus) or implosion halos and their density increases (from isochoric line 3 to 1). (b) P-T-t path representing a nearly isothermal decompression path; fluid inclusions display slow change of shape with stretching within the foliation plane, or rapid change of shape with decrepitation into fluid inclusion planes displaying decreasing densities and different orientations relative to the foliation and lineation. The sample described by Boullier et al. (1991) has followed the P-T-t path A (annular fluid inclusions) and then B (FIP issued from the decrepitation of large annular fluid inclusions).

### 5.1. Rapid change of shape due to decrepitation

If the pressure difference  $\Delta P$  between the confining pressure  $P_c$  and the internal fluid pressure within the fluid inclusion  $P_i$  is greater than the strength of the host mineral, then the fluid inclusion decrepitates. A microcrack appears around the previous larger fluid inclusion, then heals and leads to an intragranular fluid inclusion plane (Pêcher, 1981; Pêcher and Boullier, 1984) or to a 'decrepitation cluster' (Swanenberg, 1980). This process contributes to an instantaneous reequilibration of the fluid inclusion relative to its environmental P-T conditions. In quartz,  $\Delta P$  has been calibrated to be about 85 MPa for large fluid inclusions (Leroy, 1979) and is inversely related to the inclusion diameter D (in micrometres) by the relation  $\Delta P(MPa) = 4.26 \times 10^{-2} \times D^{-0.423}$  (Bodnar et al., 1989).

Such decrepitation structures are frequently observed in natural samples (Swanenberg, 1980; Roedder, 1984; Hurai and Horn, 1992). However, they are generally disregarded by most inclusionists because they are not representative of the initial conditions, although they may trace the P-T-t path as well as the stress orientation during the uplift history. This may be illustrated in one sample from the High Himalaya sedimentary series (Fig. 3b) which displays three types of inclusions showing decreasing densities and different orientations, thus constraining the P-T-t and stress path.

# 5.2. Slow change of shape due to dissolutioncrystallisation processes

When P-T conditions differ slightly from that of the

isochoric line of a fluid inclusion, i.e. when there is an internal overpressure  $(P_c - P_i > 0)$  or underpressure  $(P_{\rm c} - P_{\rm i} < 0)$  in the inclusion, then dissolution-crystallisation processes occur (Gratier and Jenatton, 1984). Sterner and Bodnar (1989) and then Vityk and Bodnar (1995) have shown that the different shapes acquired by fluid inclusions depend on the sign of  $\Delta P$  or on the P-T paths which a fluid inclusion may follow after entrapment. They provide experimental examples that can be compared with natural examples, keeping in mind, however, that the stress configuration is never isotropic in nature, contrary to their experiments. The best similarity is given by annular fluid inclusions or implosion halos (Fig. 3a), which correspond to nearly isothermal compression or loading. It would also be interesting to understand the significance of 'stretched' fluid inclusions (Fig. 3b). They may be interpreted tentatively as due to dissolution-crystallisation processes comparable to those described by Sprunt and Nur (1977) around spherical cavities and induced by heterogeneous stresses around the inclusions. In that case, the shape of these fluid inclusions should be attributed to the uplift of the pile, as well as the subhorizontal microcracks described in Fig. 3(b). An important point is that the changes of shape are geologically rapid and obtained after a few days of experiments on aqueous fluid inclusions. Therefore, they are able to record very quick changes in P-T conditions which are not discernible by any mineralogical associations or zonations. It would be interesting to perform similar experiments on pure CO<sub>2</sub> fluid inclusions to compare the kinetics of morphological changes. As solubility of quartz is lower in  $CO_2$  than in water, we should expect slower morphological changes for CO<sub>2</sub> fluid inclusions.

## 6. Density of fluid inclusions

Morphological changes are due to dissolution-crystallisation mechanisms and may occur without any measurable volume change. However, they are generally accompanied by density changes of the fluid inclusions as experimentally demonstrated (Pêcher, 1981; Gratier and Jenatton, 1984; Pêcher and Boullier, 1984; Boullier et al., 1989; Sterner and Bodnar, 1989; Vityk and Bodnar, 1995). H<sub>2</sub>O leaking may also occur concomitantly in H<sub>2</sub>O–CO<sub>2</sub> (Bakker and Jansen, 1991, 1994) or H<sub>2</sub>O-NaCl fluid inclusions (Vityk and Bodnar, 1997). These density changes are probably related to dislocation gliding and water diffusion around the fluid inclusions (Boullier et al., 1989; Bakker and Jansen, 1994; Vityk and Bodnar, 1997). However, except in very simple and clear cases, these changes are rather difficult to prove in natural examples and some recognition and quantification criteria are still needed (Vityk and Bodnar, 1995).

#### 7. Conclusion: questions for the future

In the future, it would be interesting to develop experimental work on fluid-inclusion bearing quartz in nonhydrostatic conditions in order to complete the database initiated by Sterner and Bodnar (1989) and Vityk and Bodnar (1995). Then, it would be possible to compare the shapes and densities of natural fluid inclusions with identified examples of isobaric cooling, isothermal loading or decompression, or isobaric heating under stress. Numerical modelling could also be useful for a kinetic approach of changes in shape and density of fluid inclusions as a function of their size and physical parameters. The nature of the fluid appears to also be an important parameter controlling the solubility of the host mineral and, therefore, the kinetics of dissolution–crystallisation processes.

Another important question governing the permeability of rocks is the wetting angle of a fluid in a given mineral assemblage (Watson and Brennan, 1987) and the time-life history of microcracks (Smith and Evans, 1984; Brantley et al., 1990; Brantley, 1992). The latter is dependent on temperature, fluid pressure, fluid composition and wetting angle of the fluid in the considered mineral (Brantley, 1992). Wetting angle and microcrack time-life are also important parameters for dissolution-crystallisation mechanisms which occur in the upper crust, because they control the accessibility of fluids to the sites of dissolution or crystallisation, and then the dissolution-crystallisation flow laws (Gratier et al., 1999). As mentioned by Brantley (1992), the type of structures in which fluids are percolating in rocks (grain boundaries, microcracks and cracks) will control the rate of flow throughout the rocks or the rate of fluid-rock interactions (mineral transformations or reequilibration). It is therefore important to complete our knowledge on each type of these structures: supplementary experimental work and observation of fluid inclusions in natural rocks may contribute to that.

With a better knowledge of fluid inclusions it would be possible to investigate an important parameter in the upper crust: the fluid pressure. This parameter is important in fracturing and vein-forming mechanisms involved in many ore deposition processes and seismic faulting (Sibson, 1989; Parry and Bruhn, 1990; Cox, 1995; Robert et al., 1995). If we gain a better understanding of the post-trapping behaviour of fluid inclusions, these tiny objects will become a very powerful tool for understanding crustal-scale processes of deformation, as stated by Sorby (1858).

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