

FLUID DISTRIBUTION IN THE LITHOSPHERE AS INDICATED BY FLUID INCLUSIONS

by

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Most petrological processes involve a fluid phase, either to initiate a mineral reaction, or to transport element during subsolidus recrystallization, to provide energy for the mass transport of molten magmas, etc. Recent years have seen a tremendous amount of work and decisive progress in the characterization of fluids of the lithosphere, but one problem is obvious: fluids are by definition changing and fugace, they vanish as soon as the process in which they participate - in many cases that they induce - comes to an end. Even for geological phenomena which are directly observable, like sedimentation of volcanism, the in situ analysis of fluids is impossible or at best very difficult. Therefore most of the knowledge derives from indirect sources: interaction with minerals, theoretical (mostly thermodynamical) calculations, comparison with experimental data. In some fields of geology, notably in metamorphic petrology, the theoretical prediction of fluids involved in mineral reactions has come to a high degree of precision, as aptly summarized in a recent book (Ferry 1982).

The direct approach: Fluid inclusions

Since the early days of modern petrography - in fact as soon as the microscope was applied to rock studies (Sorby 1858) - it is known that some rock forming minerals may contain samples of the "now missing" fluid phase in closed cavities, the so called fluid inclusions. The size and abundance are very variable, but upper limits are really spectacular: For the size, at least several liters in some giant crystals from pegmatites; for the abundance, Zirkel in 1873 (cited in Roedder, 1972), had estimated a number of 3.6×10^{11} (360 billions!) per cm^3 in an hayne from the Vesuve. Such an abundance leads evidently to an extremely small size: an average diameter of $1 \mu\text{m}$ or so, not enough to be studied correctly by present day microscope. But "good" inclusions, in the range $5-100 \mu\text{m}$, are present in almost any rock sample, sedimentary, igneous or metamorphic. At least on earth and in some stony meteorites (Fièni et al. 1978), as no fluid inclusions - but plenty glass inclusions - has ever been observed in any lunar sample despite an intense and systematic search.

One would expect that these direct samples, sometimes preserved for billions of years in archean rocks, would come as a prime evidence of fluid composition during petrological processes. Clearly, this is not the case. For many years, because of a lack of suitable equipment and a poor knowledge of fluid properties at high P-T, they have been completely ignored and they are still not even seen by many petrologists, who may confuse them with dust, artifacts, holes etc. The reverse is also true, and there are several well known examples quoting beautiful artifacts (air bubbles, droplets of epoxy) mistaken for natural inclusions. Until the late sixties, a handful of specialists, at least in the Western World, avoided the complete disappearance of the discipline, but their investigations were mostly confined to idiomorphic crystals in Alpine type veins and in mineral deposits.

A new situation came about 15 years ago, as a result of several independent advances: in the technology, notably when simple and efficient heating and freezing microscopic devices became available, in the knowledge of the theory and the physical properties (PVT) of natural fluid systems at high pressure and temperature, and in the prediction of fluid composition through mineral equilibria. Now fluid inclusions may be studied by a remarkable array of techniques,

from most simple (crushing stage) to very sophisticated: MicroRaman spectroscopy, Ion probe, Gas chromatography, Mass spectrometry, Electron microscopy, etc. The analytical results compare well with classical studies of solid phases, the small size of the samples being somewhat compensated by the easier analysis of fluid system. (For an up to date and complete review of the "state of art" in fluid inclusions studies, see Hollister and Crawford, 1981).

Fluid inclusion data: Possibilities and limitations

Fluid inclusions have in principle a decisive advantage on solid phases: they give an information, not only on the chemical composition as to the solid minerals, but also on the density, or molar volume, of the system. This last parameter remains constant for the solid phases, at least in first approximation, and is consequently commonly ignored as a variable of state. For fluids, we have a priori the choice between 3 variable of state, namely temperature, pressure, and molar volume, but of course only 2 are independent as they are related by the equation of state. But then, if the fluid trapped in the cavity is representative - no leakage, homogeneous fluid at the time of the trapping - we have an unique way to reach volumic properties during deep seated processes.

The ideal case is not often reached, as limitations are severe; multiple causes of error do exist, well known and abundantly described in the specialized literature: heterogeneous trapping, "necking down" (Roedder 1972), leakage, reaction with the enclosing mineral etc. The limitations are often quoted, explicitly or not, as a kind of excuse to ignore the fluid inclusions, starting from the point of view that their information will be certainly meaningless. This position is evidently not quite scientific: the inclusion are there, they are part of the rock, and since the techniques exist, they have to be studied as any other constituent. Would any petrologist accept to disregard zircons or any other accessory minerals in an igneous rock? After all, the distinction between "fluid" and "solid" is just a question of reference temperature: if observations were made near absolute zero, all fluid inclusions would indeed be solid. But this hesitation to engage in the tedious, time consuming, often frustrating observation of fluid inclusions - all those who have spent hours before finding a "good" inclusion which precisely decrepitates just before homogenization will understand me - is essentially due to a lack of education. With the possible exception of some places in the USSR, there is no university in the World where the basic principles of fluid inclusion observation, which requires nothing but the normal petrographic microscope, are taught with the optical mineralogy. As a result a great deal of information is lost: size, abundance, even nature of some fluids should be standard data in any rock description, and the inclusions remain mysterious to most researchers, who have not the experience to evaluate their information and to select the important from the trivial. I shall illustrate this comment by an example: most minerals from endogenous rocks are more or less altered, weathered. This might be a object of study, but normally the researcher will instinctively reject the most altered parts and concentrate on the fresher ones, without even mentioning it. The situation is exactly the same for fluid inclusions: many inclusions, sometimes by far the most abundant, may be very late and precisely related to this weathering. After some training, the observer will spot them immediately and concentrate on the deeper ones. This is just one aspect of a more serious problem that we are facing at the moment, namely the widening gap between specialists and not specialists. Things are going fast and the theory of fluid systems develops at an astonishing rate. We can now handle binary and even some ternary systems, through complicated phase diagrams which requires many hours of study (see e.g. Walther 1982). Fluid inclusionists constitute an active, relatively young group which obviously advances at great speed. There is a real danger of segregation which call for a fight on both sides: for the specialists, not being too semantic, too needlessly complicated; but also for the rest of the geological community, the need to assimilate

and to practice the basic principles of fluid inclusion observation and not to rely on the supposed expertise of a few individuals.

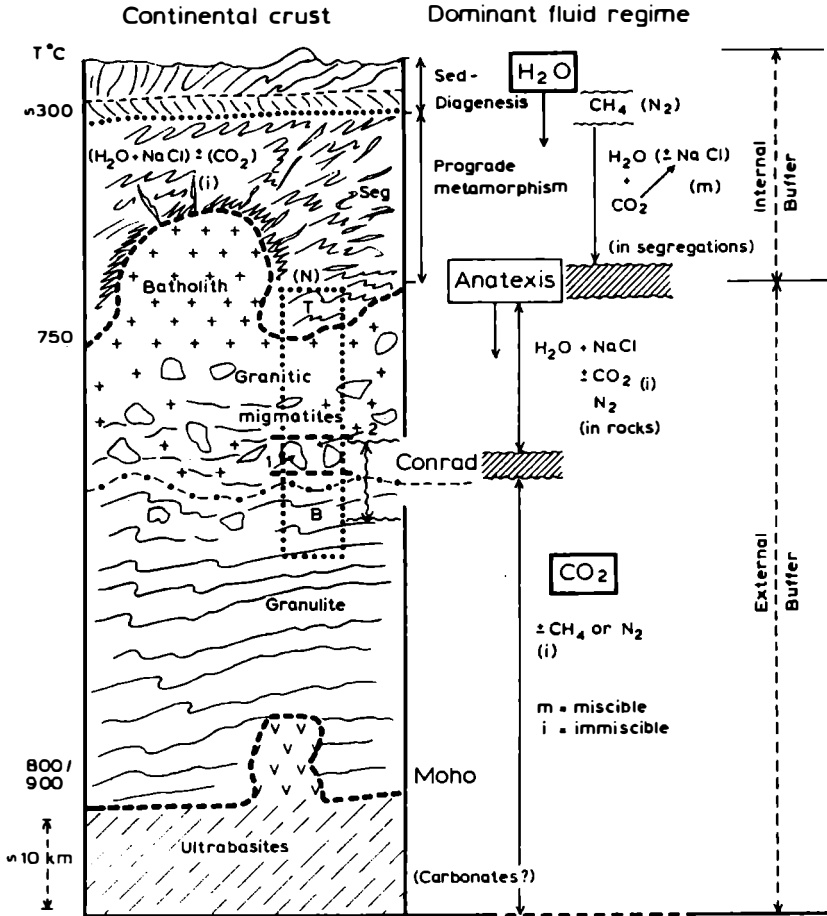


Fig. 1. Model of the fluid distribution in the continental crust and in the Upper Mantle as inferred from fluid inclusions studies (see Touret and Dietvorst 1983). m and i refer to the miscible/immiscible state of different fluids at the time of the trapping. T and B: supposed position of the Telemark (T) and Bamble (B), both in Norway (N) as they are exposed today (See Touret, 1981).

Some general results: a model of the fluid distribution in the Earth's Crust and Upper Mantle

I have sufficiently insisted on the difficulty and on the limitations of fluid inclusion studies to feel now at ease to present some general results which show the interest of this type of research. They deal with a) the nature of the fluids involved in deep seated processes and b) their distribution within the earth's crust and upper mantle.

a) Nature of the fluids

The total number of pure fluids encountered in inclusions is remarkably small. Only 4, namely H_2O , CO_2 , CH_4 , N_2 are common and the 2 first ones are by far the most abundant. Sulfur bearing species are exceptional (SH_2 , SO_2), H_2 and noble gases could be more abundant than normally assumed, but they are difficult to detect, and O_2 is only found in Uranium deposits, when water is destroyed by radioactive radiation (Dubessy, pers. comm.). These pure fluids may be mixed in any proportion and the aqueous fluids commonly contain dissolved ionic species, notably chlorides, often in such a quantity that the result is closer to a molten salt than to an aqueous solution. Among the unexpected results which could only be obtained by fluid inclusion studies, we can mention the high salinity of most aqueous inclusions in many igneous rocks - almost all alkali granites contain inclusions with NaCl as daughter minerals - and the discovery of fluids which, for some reason (pressure or temperature conditions, absence of necessary cations to build up a mineral structure), cannot be incorporated in the enclosing mineral: this relates essentially to CO_2 , but also CH_4 and N_2 , in lower crustal and upper mantle rocks (granulites, alkali basalts, ultrabasites).

b) Distribution of the fluids

The repeated observation of similar (or comparable) inclusions in rocks having been submitted to the same P-T regime leads to the conclusion that fluid distribution obeys to some simple and general rules. Practically all surface inclusions are water rich, while CO_2 , to a lesser extent CH_4 and N_2 , grossly dominates in the lower levels. In the Alps, prograde metamorphism is marked by a progressive and regular change in fluid composition in alpine type veins (increase of CO_2 content with increasing metamorphism). All these observations have led to a model of the distribution of fluids in the lithosphere based on an antipathetic behaviour between H_2O at the surface, and CO_2 at depth (Fig. 1). As this model has been previously described in some detail (Touret, 1974, Touret and Dietvorst 1983), its elaboration will not be discussed here, but some comments are appropriate concerning some key aspects of the model, and the mechanisms or processes which might be involved.

Fluid/Solid interaction in metamorphic rocks: Internal versus external buffering

Any large scale model supposes some oversimplification, as compositions remain constant for, at least, some kilometers. The validity of this hypothesis depends obviously from the buffering mechanism: either "internal buffer", the fluid composition being controlled by the local mineral assemblage, of "external buffer" (infiltration, Rice and Ferry 1982), large masses of fluids connected to an external source being able to flow through the rock.

Since Greenwood (1975), it is well known that most metamorphic assemblages have a large buffering capacity for small quantities of fluids, roughly the initial pore fluids plus the volatiles (H_2O , CO_2) liberated during prograde mineral reactions. But there are also cases where large amounts of fluids must percolate through the rock, as indicated by mass transfer calculations for most metasomatic processes (alteration in porphyries, formation of skarns or greisen etc.).

Both types of buffers must therefore exist in nature and they have indeed been inferred from thermodynamical calculations in fluid/mineral interaction (Rice and Ferry, 1982, p.289).

Fluid inclusions provide a qualitative way to check out which mechanism predominates: For internal buffering, the composition of the fluid inclusion will depend strictly from the surrounding rocks and it will remain relatively constant in a homogeneous rock. For external buffering, fluid inclusions might remain the same for different rock compositions, but they may also be very variable at a given spot if several independent fluid pulses went through the rock.

The results are extremely instructive, as they show a constant competition between the two trends. But, first of all, they indicate a strong difference between a relatively low-grade (roughly, from diagenesis to the beginning of anatexis) and a "high-grade" metamorphic domain which comprises migmatites, granulites and ultrabasic rocks from the Upper Mantle. In the first domain, internal buffering is the rule for massive rocks and for locally derived metamorphic segregations. Inclusions in rocks seem to be strictly controlled by the rock composition, but they are in general so small that they can hardly be investigated in some detail. The situation is more complex for segregations, where most workable inclusions are to be found. In intermediate (caledonian) and, more characteristically, in high pressure (alpine) type of metamorphism, the fluid composition evolves regularly from H_2O (+/- NaCl) to $H_2O + CH_4$ (heavier hydrocarbons in the very early stages) and $H_2O +$ increasing CO_2 content, up to about 20/30 moles % near the beginning of anatexis (2nd sillimanite isograd). This evolution corresponds exactly to the "graphite" buffer at increasing P and T; it suggests that:

- variations in rock compositions (e.g. limestones/pelites) are minor compared to the influence of graphite, most likely derived from organic matter.
- the fluid composition depends from the P-T conditions at or near peak metamorphic conditions.

The low pressure type of metamorphism (hercynian, notably pyrenean) seems to be very different, at least in the few cases which have been investigated so far (see references in Hollister and Crawford 1981). Most segregations contain a fluid, generally CO_2 rich, independent from the nature of the host rock and from the metamorphic grade. This composition possibly indicates a distant control by anatectic masses (see below).

In high grade metamorphic rocks, the situation changes drastically in two ways:
- "Segregation" are now quartzo-feldspathic, approaching granitic composition. They correspond to "mobilisates", which indeed are richer in inclusions than the surrounding rock (Touret and Dietvorst 1983), but only relatively: Workable inclusions are present in many rock forming minerals, notably quartz.

- External buffering seems to be the rule, as
 - a) the same fluid - CO_2 rich, "carbonic" fluid (Touret 1974) - occurs in rocks from varying composition
 - b) CO_2 is most abundant in or near rocks which obviously cannot generate it during progressive metamorphism: anatectic melts, metagabbros or metabasalts, ultrabasites etc.

There are indeed exceptions and we find more and more examples of rocks in which CO_2 does not predominate: CH_4 and especially N_2 in metasediments and in migmatites (Kreulen and Schuiling 1982), highly saline brines in metasediments and in some granulites of the lower crust etc. As more documented studies become available, the situation is thus more complex than initially assumed (Touret 1974), but the general conclusion remains: Most fluids encountered in high grade metamorphic domains (all in granulites and related rocks) tend to lower drastically the activity of water and CO_2 , at world scale, is by far the most abundant constituent.

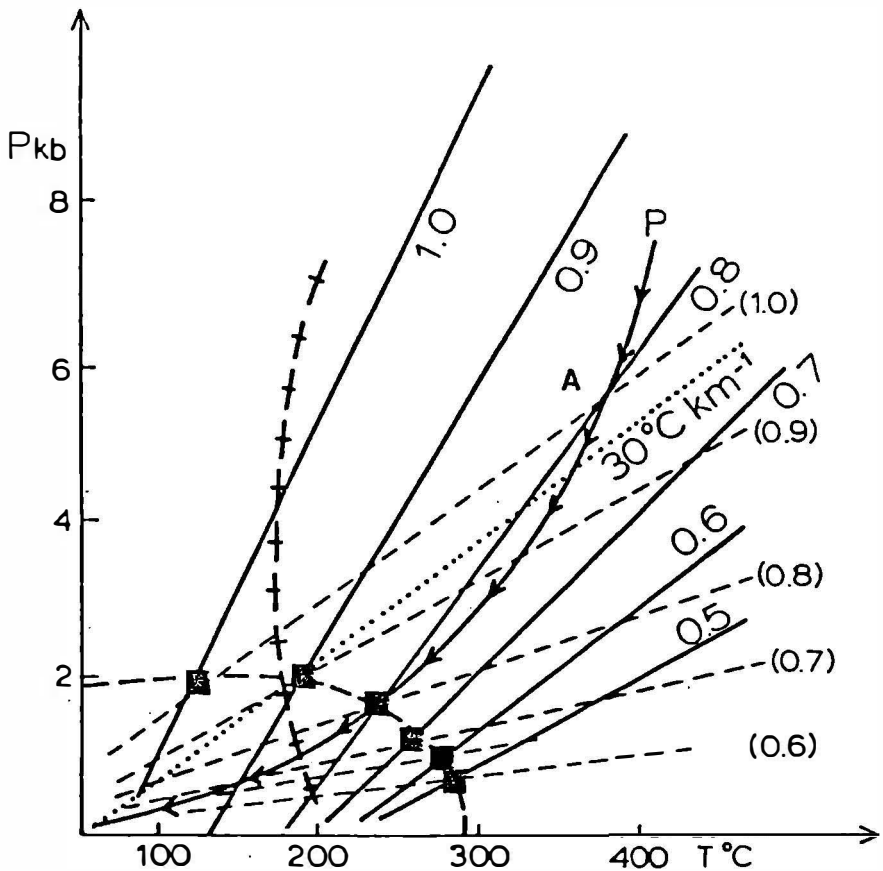


Fig. 2. H₂O (solid lines) and CO₂ (thin semi-solid) isochores. Numbers near the isochores = density in g/cm³ (solid numbers: H₂O, bracketted numbers: CO₂). Solid squares: Inversion density line: below this line (heavy semi solid), H₂O is denser than CO₂, above it the reverse is true. Example: at point A, intersection of a given, most usual (Hollister and Crawford, 1981) uplift path and the CO₂ isochore 1 g/cm³, the corresponding density of water is 0,8 g/cm³. Crosses: projection of the critical line in the system H₂O-CO₂.

Origin of the CO₂. Buffering mechanism in the lower crust

If the importance of the CO₂ of the lower crust cannot be overemphasised (Newton et al. 1980), there is currently a considerable debate concerning its origin, and, even if this point is not often explicitly stated, about the mechanisms by which a free CO₂ fluid phase can be maintained at these levels. CO₂ may be either juvenile, introduced by the synmetamorphic basic intrusives, which are so typical for some sectors of the lower crust (Touret 1969), or derives from the metamorphism of carbonate or carbon bearing sediments. Stable isotope data are not entirely convincing (Pineau et al. 1981, Hoefs et al. 1981), but the very low values (¹³C about -20 ‰) found for the CO₂ of fluid

inclusions seem to rule out a large scale participation of carbonates. However, different results are obtained from mass balance calculations (Glassley 1983) and the problem remains open. However, the major point is not so much the source of the CO₂ than the process by which pure CO₂ may be obtained and maintained at depth. For the first part, the obvious relation between CO₂ and anatectic melts - not with metacarbonates which contain commonly CH₄ or H₂O inclusions - immediately calls for a separation between CO₂ and H₂O by differential solubility in granitic melts. Details may be very complicated (Touret and Dietvorst 1983), but in general this process is certainly very efficient. For the second, namely how CO₂ can be maintained below H₂O fluids, we can only propose some working hypothesis which need to be checked and further elaborated: it seems that CO₂ density roughly increases with pressure, i.e. with depth: high pressure granulites contain denser fluids than low pressure granulite (Touret 1981). A simple glance to CO₂ and H₂O isochores shows that at depth CO₂ is denser than H₂O, while the reverse is true near the surface. Thus the antipathetic repartition of CO₂ and H₂O may simply result of gravity layered fluid masses in a relatively porous environment. (Fig. 2).

Conclusion

The difficulty of fluid inclusion studies, the many hypothesis which must be done for any interpretation must not hide the fact that several "unexpected" results, which later has proven to be very valuable for many petrological theories, could only be obtained from their observation. Only a handful of cases have been studied and the list of potential problems is almost endless. As methods and equipment are now routinely available, there is no excuse to ignore their information and, after an interruption of a good century, we have now to continue Sorby's work.

Acknowledgements

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