

## Fluids in the Monte Pulchiana intrusion (N Sardinia): results on fluid inclusion study

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**ABSTRACT.** — The Monte Pulchiana leucogranite is one of the youngest post-tectonic shallow level Hercynian intrusions in Sardinia. The granite is a medium to coarse grained rock, consisting of quartz, microperthitic K-feldspar, sodic plagioclase and minor biotite. Within the pluton, especially in the border zones, some aplite dykes and pegmatitic pods are present. Sometimes the latter may contain euhedral quartz crystals in open vugs and cavities. Microscopic and microthermometric investigations on fluid inclusions in quartz from representative samples of granite and in one euhedral quartz from a pegmatitic vug give evidence of two fluid-rock interaction episodes. Two main types of fluids are recognized: the first one is present only in the granite, consisting of a highly saline aqueous fluid. Extremely variable NaCl content suggests oversaturation at the time of the final inclusion evolution (average salinity estimate around 30 wt.% NaCl;  $T_h = 200^\circ\text{--}310^\circ\text{C}$ , in absence of boiling); and the second, a much less saline aqueous fluid (5 wt.% NaCl;  $T_h = 100^\circ\text{--}260^\circ\text{C}$ ), probably of meteoric origin, is present in the granite as well as in the pegmatite.

These observations are consistent with the existence, in subsolidus conditions, of a first NaCl oversaturated brine, probably of initial magmatic origin. Absence of boiling imposes a pressure greater than 1.2 kb for the granite's depth of emplacement. At a later stage, homogeneous low salinity fluid (probably meteoric water) invaded the pluton. A post magmatic roughly isobaric trajectory is proposed starting at the inferred conditions of the granite end of crystallization ( $T \sim 700^\circ\text{C}$ ;  $P \sim 1.5$  kb).

**Key words:** Fluid inclusions, leucogranites, Sardinia, brines, P-T path.

### Introduction

Fluid inclusions in shallow intrusives have been widely studied mainly in order to characterize the physico-chemical features of associated fluid phases often responsible of ore deposition, see e.g. WEISBROD, (1981) and ROEDDER (1984). However, these techniques may also give more information on the postmagmatic evolution path and the episodes of fluid-rock interaction which have occurred in subsolidus conditions.

The Sardinia-Corsica Hercynian Batholith consists of several plutonites ranging from ultramafic rocks and gabbros to leucogranites; these latter ones crop out all over the two islands, constituting about 25% of all plutonites in Sardinia. Some characteristic features, including the Mo-mineralizations that are commonly associated with these granitic rocks, are reported in ORSINI (1980); BRALIA et al., (1981); GHEZZO and ORSINI (1982); GUASPARRI et al., (1984a, 1984b) and ROSSI (1986). In this paper we will discuss selected results on fluid inclusion study in the M. Pulchiana leucogranite intrusion, one of the largest in northern Sardinia (Fig. 1).

Observations have been made in quartz crystals of the primary mineral assemblage in

selected leucogranite samples and in euhedral smoky quartz crystal from a vug in an associated pegmatite.

Special attention has been given to the distribution and composition of fluid inclusions, in order to reconstruct the sequence of fluid-phase/rock interactions which affected the plutonite and are responsible for its widespread deuteric alteration.

Present results allow us to set an upper limit for the emplacement of the intrusion at about 4 km depth: at least two different fluid generations have been characterized. An idealized evolution path at low P and T (well in subsolidus condition) is proposed in Figure 9.

### Geological setting

The M. Pulchiana leucogranite intrusion is one of the youngest plutons of the intrusive sequence forming the Sardinia - Corsica Hercynian Batholith. This sequence, mainly late to post-tectonic, is dominated by two distinct associations:

- a calcalkaline association dominated by I-type biotite monzogranites and leucogranites, with minor ultramafic rocks, gabbros, tonalites and granodiorites.

- A subalkaline potassic association composed of sienomonzonites, monzogranites, alaskites, sienites, monzonites and diorites cropping out in northern Corsica.

In addition to these two main suites a minor part of the batholith is constituted by two-mica and/or cordierite bearing granites (S-type), syntectonic strongly foliated gneissic granites and sodic sienites.

The emplacement of the whole batholith took place in a time interval between 330 and 280 m.y., based on the Rb/Sr and K/Ar isotopic data (DEL MORO et al., 1975; COCHERIE, 1984).

In recent years, detailed mineralogical and petrological studies have led to a better understanding of the sequence of emplacement and crystallization history of these rocks (e.g. DI SIMPLICIO et al., 1974; ORSINI, 1976, 1980; BROTTU et al., 1978; BRALIA et al., 1981; GHEZZO and ORSINI,

1982; BROTTU et al., 1983; GIRAUD, 1983; COCHERIE, 1984; ROSSI, 1986).

The age of the M. Pulchiana pluton has not yet been defined but it must be close to 290 m.y., since the emplacement age of the younger leucogranite suite is referred to as 290-285 m.y. (with a  $^{87}\text{Sr}/^{86}\text{Sr}_i = 0.7085$ ) (DEL MORO et al., 1975). Its emplacement took place at shallow levels after the main tectono-metamorphic events and the uplift of the whole basement. This emplacement was likely to occur in extensional environment (GHEZZO and ORSINI, 1982). The pluton intrudes older late-tectonic plutonites. The contacts are sharp, truncating the magmatic foliation. The host rocks (Fig. 1) include medium-grained biotite granodiorites rich in dark microgranular magmatic xenoliths (at the northern contact), biotite granodiorites + biotite-amphibole tonalites (along the southern border of the pluton) and pinkish coarse-grained biotite monzogranites characterized by K-feldspar megacrysts. They all show discordant contacts toward the metamorphic metapelite country rocks which have a migmatitic character in the sillimanite + K-feldspar grade (FRANCESCHELLI et al., 1982).

A NE-SW striking fault separates the pluton in two segments and displays a lateral movement.

The biotite leucogranite is compositionally and texturally homogeneous; a coarse-grained equigranular pinkish granite consisting of quartz (~ 32%), oligoclase plagioclase with albitic rim (~ 30%), orthoclase + microcline microperthites (~ 35%) and minor iron-rich biotite (3-4%), with accessory magnetite, apatite, zircon, allanite. A strong pervasive deuteric alteration overprint is always present; and chloritization, albitization, sericitization, argillification are common alteration assemblages, often associated with minor fluorite. A network of microfractures is also present in the quartz crystals. Most of these are intercrystalline, but also intracrystalline cracks have been observed. Fluid inclusion trails underline the healed microfractures (DE VIVO et al., 1985). This overall pattern is similar to those described by many authors in granitic rocks (TUTTLE, 1949; PECHER et

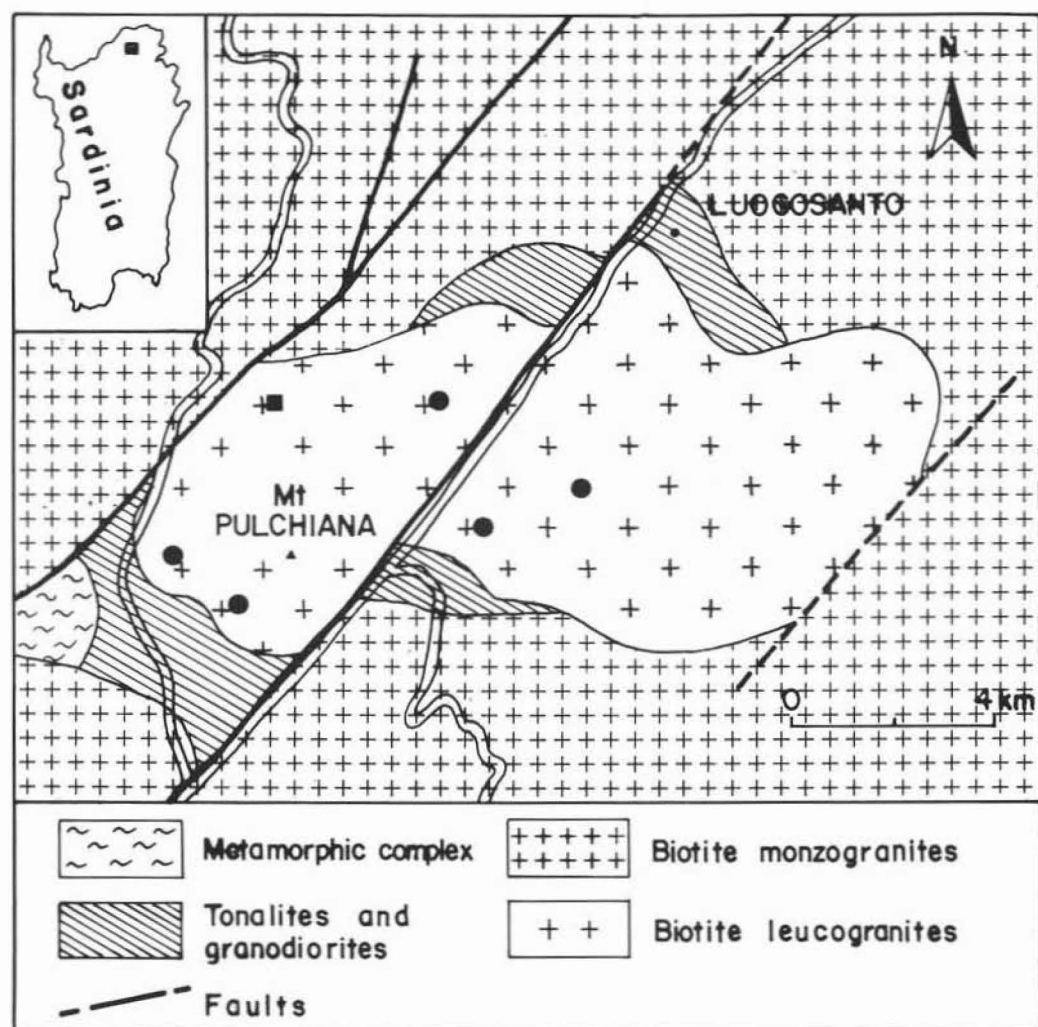


Fig. 1. — Sketch map of the M. Pulchiana pluton and location of the investigated samples. Circles: leucogranite samples; square: euhedral smoky quartz crystal from a pegmatite.

al., 1985; LESPINASSE and PECHER, 1986; KOWALLIS et al., 1987).

In the border zones of the pluton both aplitic dykes and pegmatitic pods are present. They constitute only a minor part of the pluton (no more than 5%).

Aplites occur as irregular, discontinuous dykes or lenses (10 to 100 cm thick) with fine-grained sugary texture. They often show a layered structure due mainly to different mineral grain size and biotite content, and contain small pods with pegmatitic texture. The mineralogy is analogous to that of the

granite itself.

The pegmatite pods have an irregular shape, a 20-150 cm diameter and are often intimately associated with the aplite dykes. They consist of milky quartz, reddish orthoclase-perthite, albite, minor biotite and other phases such as muscovite and garnet. These pegmatite pods are massive, but some of them have vugs lined by euhedral smoky quartz crystals (miarolitic pegmatites). Sometimes a moderately zoned pattern occurs; moving from a border with graphic texture, the grain size increases (up to 5-15 cm) to a core with

pegmatitic texture.

The following aplite + pegmatite petrogenesis will be proposed, mainly by referring to the JAHNS and BURNHAM's (1969) model:

— the pegmatite pods are the result of a closed-system crystallization when the granite reached the water-saturation conditions and locally a hydrous silicate melt crystallized until reaching its solidus, which is well below the granite solidus (JAHNS, 1982).

— The small quantity of pegmatites implies a late vapour saturation in the crystallization history of the granite.

— The rare vugs seem to testify a local «segregation and accumulation of aqueous fluids as crystallization of granitic magma proceeds...» (BURNHAM and NEKVASIL, 1986, pg. 260; JAHNS, 1982; LONDON, 1986).

— The aplite dykes are most likely the result of rapid nucleation and crystallization from a residual melt intruded in extensional fractures during the last stages of solidification of the pluton. This coincided with a local sudden drop in fluid pressure and hence in the volatile content of the magma.

## Fluid inclusions in the Monte Pulchiana granite complex

### Sample selection and methodology

Chosen among many of similar looking specimens, five representative samples of leucogranite and one euhedral smoky quartz

crystal (length: more than 10 cm, diameter about 5 cm) from a vug in a pegmatite, have been selected and studied in detail. Some samples from the pegmatite itself were also collected, but they appeared to be useless for both the scarcity and the small dimensions of fluid inclusions. In all of the samples, doubly polished plates of about  $2 \times 2$  cm were prepared. The thickness was  $100 \mu\text{m}$  for granite samples, much higher ( $200 \mu\text{m}$ ) for the smoky quartz crystal which was cut perpendicular to the «c» axis. These sections were first observed with a conventional microscope for the characterization of fluid inclusion dimensions, distribution and relative chronology (basic principles are described in WEISBROD, 1981; TOURET, 1977; and ROEDDER, 1984; pg. 150 sgg.).

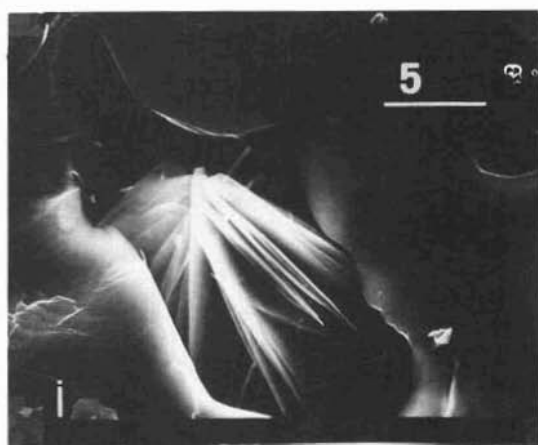
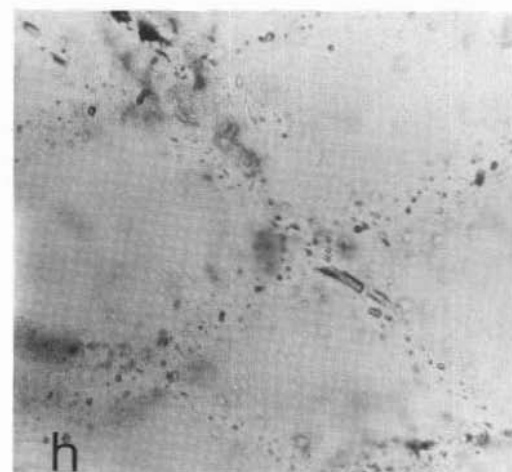
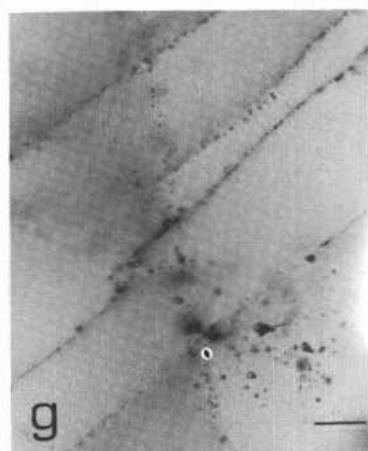
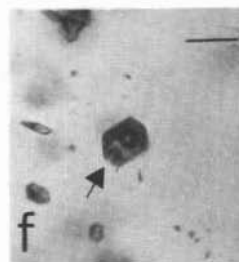
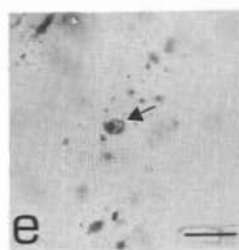
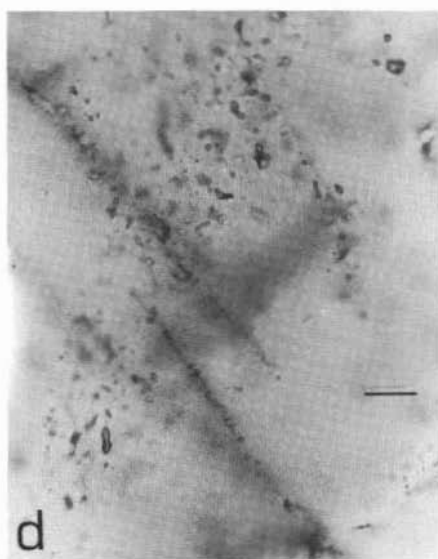
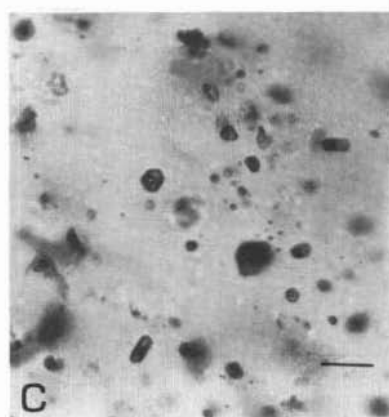
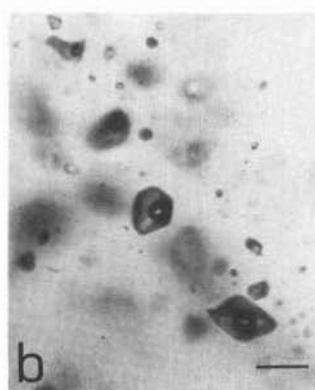
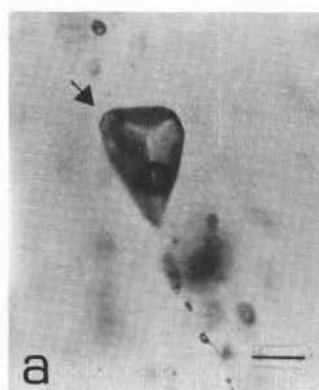
Microthermometric investigations have been performed with several heating-freezing stages: Chaixmeca (POTY et al., 1976) at the Free University of Amsterdam; and U.S.G.S.-type (ROEDDER, 1984) at the University of Rome. Calibrations were made using a number of organic and inorganic compounds as recommended in the literature (e.g. HOLLISTER et al., 1981).

Each measurement was repeated at least once to ensure reproducibility; the same objective (Leitz UTK 50,  $32 \times$  long frontal distance) has been systematically used.

Some fragments of one euhedral quartz crystal have also been cemented to sample holders, carbon coated and analyzed (SEM/EDS, University of Siena) in order to characterize qualitatively some daughter phases which could have remained in opened fluid inclusions.

Fig. 2. — Fluid inclusions in M. Pulchiana granite complex. a) Primary muscovite bearing (arrow) inclusions in the core zone of the euhedral quartz crystal in the pegmatite vug. b) Primary, crystal negative shaped two-phase (L + V) aqueous inclusions in the euhedral quartz crystal (vug in the pegmatite). c) General view of fluid inclusion distribution in the euhedral quartz crystal (vug in the pegmatite). d) Late trails of two-phase (L + V) aqueous inclusions in a quartz crystal from the leucogranite ( $G_{II}$ , see text). e) A typical early three-phase (L + S + V) halite bearing inclusion ( $G_I$ ), (arrow), in a quartz crystal in the leucogranite. f) Large primary inclusion, negative crystal shaped and muscovite bearing (arrow), from the euhedral quartz crystal (vug in pegmatite). g) Intersection of  $G_{II}$  different trails (see text) in quartz from the leucogranite. In the lower right end of the photograph a group of  $G_I$  inclusions remains visible between two generations of intersecting, late  $G_{II}$  trails (one diffuse and relatively early, several (five) others very late and sub-parallel). h) Network of two-phase (L + V) aqueous inclusions ( $G_{II}$ ) within a leucogranite quartz crystal. i) SEM microphotograph of an opened fluid inclusion in the euhedral smoky quartz crystal (vug in pegmatite); assemblage of platy crystals identified as muscovite on the basis of both morphology and composition (see text).

Figures from a) to h) were photographed with plane polarized transmitted light and bar scale correspond to  $20 \mu\text{m}$ . Bar in photo i) gives scale in  $\mu\text{m}$ .





### Main types of fluid inclusions

Fluid inclusions are the result of trapping of aqueous solutions, both in quartz from the granite samples and in the smoky quartz from the vug. From the number of phases present at room temperature, several types can be recognized:

— *Two phase aqueous inclusions* ( $L + V$ ): they are by far the most common type of fluid inclusions present in all samples. Their dimensions vary from a few to 40–50  $\mu\text{m}$ , with the biggest inclusions in the smoky euhedral quartz crystal (Fig. 2b). Liquid/vapour volume ratios vary widely but the vapour phase never exceeds 30% vol. of the whole inclusion.

— *Halite bearing aqueous inclusions* ( $L + S + V$ ): in the plutonite, they are often present in clusters associated with two phase aqueous inclusions (Fig. 2e). They contain a cubic isotropic daughter mineral, which from its appearance and refractive index (identical to  $\omega$  of quartz), is assumed to be halite. The relative volume of the salt cube is quite variable. The shape of these three-phase inclusions is often somewhat irregular but no typical features of «necking down», thin capillary at one end of the cavity (ROEDDER, 1981), have been ever observed.

— *Muscovite bearing inclusions* ( $L + S + V$ ): in some fluid inclusions of the smoky quartz crystal, radial aggregates of hairlike crystals, strongly anisotropic, have been observed (Fig. 2 a, f). From the data in the literature and their distinctive appearance, they were first assumed (FREZZOTTI et al., 1986) to be dawsonite ( $\text{NaAl}(\text{CO}_3)(\text{OH})_2$ ) (COVENEY and KELLY, 1971). However, despite great technical difficulties, some crystals could be photographed and qualitatively analyzed with the SEM/EDS (Fig. 2i).

The following procedure proved to be successful;

1) breaking small fragments (about 2 mm in diameter) of the doubly polished quartz wafer, after microscopic observation.

2) Fixing vertically the broken piece on the S.E.M. sample holder, so that the broken face, 200  $\mu\text{m}$  thick, can be directly observed under the S.E.M. Three or four chips can be assembled in a single holder.

Most cavities are empty but workable crystals occur in some of them (less than 5%).

Qualitative analysis (major K, Al, Si) and the appearance of the crystal indicate muscovite.

Our results suggest that the recognition of dawsonite in fluid inclusions when based only on optical appearance is doubtful and it should be systematically confirmed by direct analysis (Raman probe, microprobe, etc.).

*Fluids in the leucogranite:  $G_I$  (early, saline aqueous fluids) and  $G_{II}$  (late low salinity aqueous fluids)*

The distribution of fluid inclusions is very complex. As it was not possible to relate them to any growth patterns in the quartz grains, a clearcut distinction between primary and secondary inclusions can not be made. We can however establish a relative chronology based on the following criteria (ROEDDER, 1971, 1984; TOURET, 1981; HOLLISTER, 1981): a) inclusions isolated or dispersed in clusters are earlier than those clearly related to more or

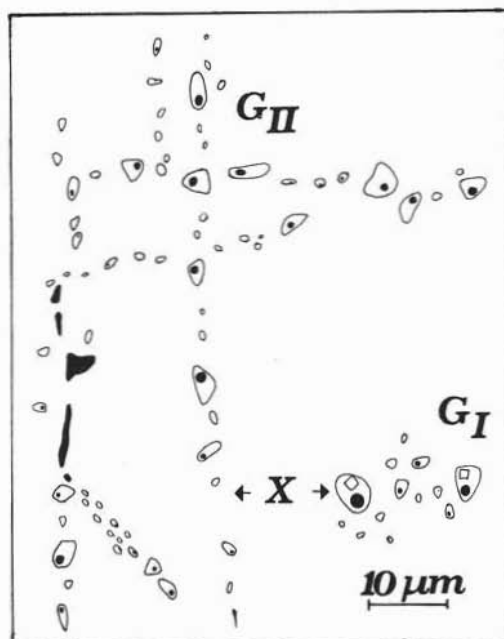


Fig. 3. — Fluid inclusion distribution in a quartz crystal from the leucogranite.  $G_I$ : early inclusions (two-phase and three-phase halite bearing);  $G_{II}$ : different trails of two-phase aqueous inclusions, intersecting each other. The illustrated example is particularly demonstrative; sometimes there is a partial overlapping of the two groups. (X zone: no inclusions in the present case but, often some inclusions more or less isolated difficult to relate to  $G_I$  or  $G_{II}$  groups).

less healed fractures. b) Same types of fractures are more or less coeval. c) Intracrystalline inclusion trails may be older than those which crosscut grain boundaries (intercrystalline).

According to these criteria, inclusions in the leucogranite may have been formed during at least two different episodes (Fig. 2g and 3):

— Type  $G_I$ : primary or early secondary inclusions in small groups with no preferential orientation; two phase (L + V) and three phase (L + S + V) halite bearing inclusions (Fig. 2e).

— Type  $G_{II}$ : clearly secondary inclusions confined in healed fractures; constantly two-phase (L + V), no daughter phase is ever present (Fig. 2d, h).

*Melting data:*  $G_I$  and  $G_{II}$ , two separate fluid generations. All  $G_{II}$  secondary inclusions show a range in temperatures of ice final melting ( $T_m$ ) between  $-10$  and  $0^\circ\text{C}$ , corresponding to salinities from 0.5 to 8 wt.% NaCl equivalent (POTTER et al., 1978). Despite the small size of the studied inclusions, some temperatures of first melting could be measured between  $-20.8$  and  $-22.8^\circ\text{C}$ . These values are close to the eutectics of the system  $\text{H}_2\text{O} + \text{NaCl}$  at  $-21.1^\circ\text{C}$  and the system  $\text{H}_2\text{O} + \text{KCl} + \text{NaCl}$  at  $-22.9^\circ\text{C}$  (ROEDDER, 1971). Together with the final melting values, they suggest that the dissolved ions were mainly  $\text{Na}^+$  with some  $\text{K}^+$  but no  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  (Te  $\text{H}_2\text{O} + \text{MgCl}_2 + \text{NaCl} = -35^\circ\text{C}$ ; Te  $\text{H}_2\text{O} + \text{CaCl}_2 + \text{NaCl} = -52^\circ\text{C}$ ) (CRAWFORD, 1981).

The composition of fluid  $G_I$  is more difficult to establish. The presence of NaCl daughter mineral in many  $G_I$  inclusions indicates an oversaturated brine at room temperature. From the size of the halite cube, the composition may be estimated to vary between 26.3 and 55 wt.% NaCl. Microthermometric measurements were attempted for two-phase  $G_I$  inclusions close to the halite bearing ones, but they proved to be very difficult because of the small size of the inclusions. In most cases freezing could not be detected, even at temperatures as low as  $-100^\circ\text{C}$ . For three of these inclusions, the final melting could be observed between  $-3.2$

and  $-1.2^\circ\text{C}$ . From the proximity and the obvious contemporaneity with halite bearing inclusions, we infer that these values do not correspond to the melting of the ice, but of hydrohalite ( $\text{NaCl} \cdot 2\text{H}_2\text{O}$ ). If our interpretation is correct, we would be on the NaCl rich side of the  $\text{NaCl}-\text{H}_2\text{O}$  system, and a  $T_m$  of  $-3.2^\circ\text{C}$  would correspond to a salinity of 24 wt.% NaCl eq. A lower limit at 24–25 wt.% NaCl eq. is tentatively accepted as an estimate of the order of magnitude. Unfortunately, the small size of the inclusions prevented the use of optical criteria, such as higher refractive index and the difficulty to coalesce into single crystals, (SHEPHERD et al., 1985) which could be used to positively identify hydrohalite.

In conclusion, the strong salinity contrast between  $G_I$  and  $G_{II}$  indicates that the two fluids are fundamentally different, and that they must correspond to separate episodes in the fluid/rock interaction history.

*Homogenization temperatures:* 32  $G_I$  inclusions of both two-phase (L + V) and three-phase (L + S + V) were studied at high temperatures (Fig. 4). In halite bearing inclusions two homogenization temperatures were recorded: Th (disappearance of the gas

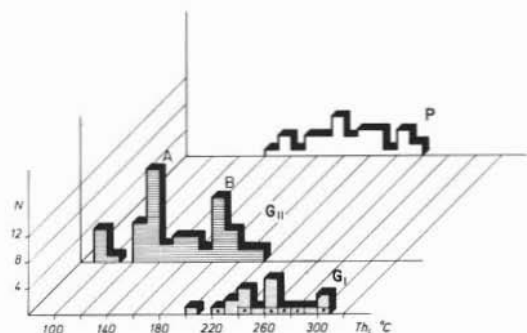


Fig. 4. — Histograms of homogenization temperatures ( $^\circ\text{C}$ ) for inclusions in the granite and in the smoky quartz in pegmatite vug (all homogenizations occurred in the liquid phase ThL). P: euhedral quartz crystal from vug (low salinity aqueous fluids);  $G_{II}$ : low salinity aqueous fluids in the granite (chemistry identical to P).  $G_{IIA}$  and  $G_{IIB}$  correspond to the two peaks ( $180^\circ$  and  $130^\circ\text{C}$  respectively as indicated by arrows).  $G_I$ : early saline fluids in the granite (ThL for both the two-phase ( $\bullet$ ) and the three-phase aqueous inclusions). Homogenization in halite bearing inclusions occurred always before NaCl dissolution ( $\text{ThL} < \text{Ts}$ ).

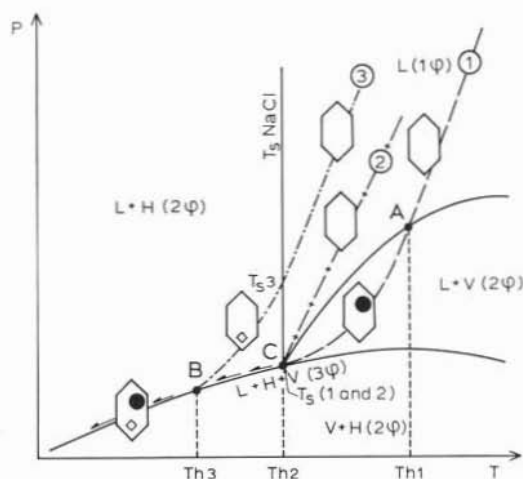


Fig. 5. — Different  $P$ ,  $T$  evolution (isochores) followed by fluid inclusions with the same NaCl content but with different  $Th$ . (1), (2) and (3) correspond to an increasing density characterized by: (1)  $T_s < Th$ ; (2)  $T_s = Th$ ; (3)  $T_s > Th$ .  $Th_1$ ,  $Th_2$ ,  $Th_3$ —homogenization temperatures observed in the different inclusions (after WEISBROD, 1984 and ROEDDER, 1984). Note that all the three correspond to the same  $T_s$ . All cases observed in the M. Pulchiana ( $G_I$  fluids) correspond to the isochore (3). H = halite; L = liquid; V = vapour.

bubble, homogenization temperature, always to the liquid) and  $T_s$  (dissolution of the salt cube).  $Th_L$  is always lower than  $T_s$ , a characteristic of relatively high density fluids (Fig. 5). Only four NaCl dissolution temperatures could be measured from 380°C to 463°C, indicating salinities from 45 to 53 wt.% NaCl equivalent. These results fit with the visual estimates from the size of the salt cube, which suggest salinities from 35 to 55 wt.% NaCl. Homogenization temperatures ( $Th_L$ ) for  $G_I$  (both two-phase and three-phase) range from 200 to 300°C. The absence of vapour rich inclusions suggests that no boiling phenomena could have occurred.

Homogenization temperatures for  $G_{II}$  inclusions are significantly lower than those for  $G_I$ .  $Th$  histograms (Fig. 4) show a loosely defined distribution with two maxima at 130 and 180°C (respectively  $G_{IIA}$  and  $G_{IIB}$ ). There is a slight overlap between the highest  $Th$  in  $G_{II}$  (Fig. 4) and the lowest  $Th$  in  $G_I$ . This is significant in that there is always the possibility that some biphasic  $G_I$  for which no melting temperatures have been recorded,

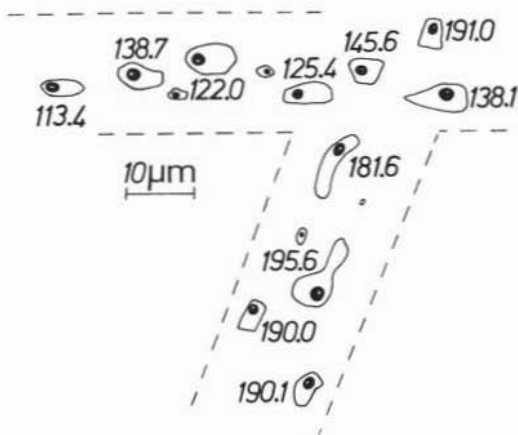


Fig. 6. — Homogenization temperatures in two intersecting trails (fluid  $G_{II}$ , quartz in the leucogranite). All homogenizations in the liquid ( $Th_L$ ).  $Th_L$  = numbers indicated near each inclusions (°C).

could infact belong to  $G_{II}$ .

Regarding the relative chronology, it is fundamental to know precisely the evolution of  $Th$  within  $G_{II}$  inclusions. Generally, temperature dispersion within a single trail is in the order of tenths of degrees; much lower than the overall variation in the histogram. This observation, common in most deep seated rocks (TOURET, 1981), leads to a picture of successive fracture openings, trapping relatively homogeneous fluids of changing density (Fig. 6). The example described in Figure 6, shows that the relative age of the different trails is very difficult to establish. However, there is a general trend, which can be observed in most comparable rocks, indicating a decrease in density for older inclusions (lower  $Th = G_{IIB}$  in figure 8 for relatively younger inclusions).

#### *Fluids in the smoky quartz of the pegmatite («P fluid»)*

In thick section the crystal consists of a smoky rim of about 3–5 mm and an internal colourless part of about 4 cm (Fig. 7). The presence of well defined growth zones makes it possible to recognize primary, pseudosecondary and secondary inclusions (ROEDDER, 1984). Microthermometric investigations have been performed on primary inclusions related to successive steps



of crystal growth and they follow, in a more or less precise pattern, some growth zones. Hence, the relative chronology is essentially based on the distance from the core of the crystal toward its border. We have already reported that some inclusions contain a solid phase (muscovite Fig. 2 f, i). These are only a few, in the most internal part, among widely dominant two-phase (L + V) inclusions (Fig. 2c). All inclusions in the quartz crystal are considered to represent a fluid phase that we will call «P fluid».

**Melting and homogenization data:** no salt cube has ever been observed. A number of final melting temperatures could be determined between  $-4$  and  $0^{\circ}\text{C}$ . Therefore, the P fluid is assumed to be low salinity water (between 6 and 0 wt. % NaCl), very similar to the  $G_{II}$  fluid in the granite. The results of Th determinations are plotted in Figure 4. They are scattered with no preferential distribution between 140 and  $260^{\circ}\text{C}$ . Figure 7 shows the distribution of individual Th values labeled according to the successive growth zones of the quartz crystal.

A first important separation boundary is located between the colourless internal zone (x) and the smoky rim (y). In the internal part at least five different fluid pulses have occurred (Fig. 7, a-e) with a general trend of decreasing temperature during the growth of the crystal core. Nevertheless, several small reversals are superimposed; in any given zone, the temperature jump is variable (up to hundred degrees). The zoning of the crystal thus reflects deposition by discrete fluid pulses and a general decrease in temperature with time, but at the smoky border (y), this general evolution changes abruptly and the last fluid pulse (f) reaches temperatures similar to those found in the core.

The variation between the x and y zones is consistent with the hypothesis of some physico-chemical discontinuity during the last stages of growth. However, the two fluids most likely have the same overall origin, as Tm variation is very small and the only real difference is indicated by the absence of muscovite in inclusions at the smoky border. Both types represent some variation in a fluid which otherwise is very similar to  $G_{II}$  in the

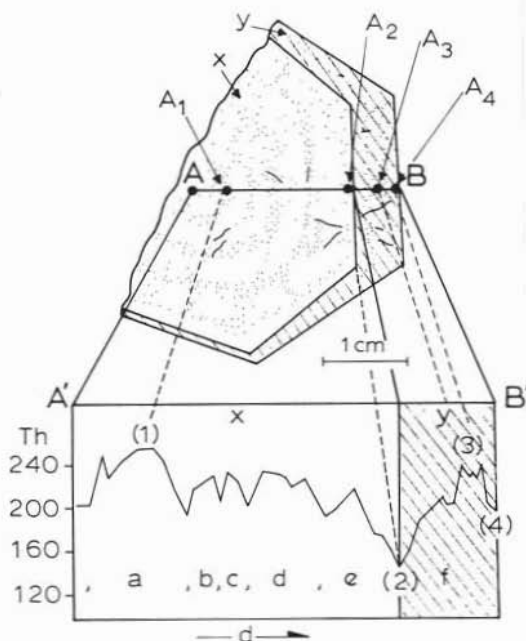


Fig. 7. — Above: schematic drawing of fluid inclusion distribution in a thick section ( $\perp$  to «c» axis) of the smoky quartz crystal in the pegmatite vug. Only primary and pseudosecondary inclusions are reported. (Hatched area corresponds to the last small smoky episode of quartz growth). Below: topological distribution of Th values versus distance from the core zone. Five different preferential distribution zones (a-b-c-d-e) of fluid inclusions are present in x internal zone and at least one (f) in y smoky rim. (1), (2), (3) and (4): representative ThL values selected for isochore interpretation ( $A_1$  to  $A_4$ , Fig. 10).

granite. The overall pattern therefore reflects the local oscillations and variations in a complex system of hydrothermal circulation during the late cooling stages.

#### Comparison of fluids in the granite ( $G_I$ and $G_{II}$ ) and in the pegmatite (P)

The graph of homogenization temperatures (Th) versus salinity (Fig. 8) shows distinct trends:

$G_I$  (saline fluids in the granite) corresponds to a relatively high Th which partially overlaps the Th values of the «P fluid» in the pegmatite with a distinct gap in salinities. The wide range of salinities indicates an heterogeneity of the brines and

a local NaCl oversaturation at the time of trapping. The absence of vapour rich inclusions points out that this oversaturation cannot be due to a boiling effect. From this it follows that the dissolution temperatures of halite ( $T_s$ ) are not to be considered minimum trapping values of the fluid, as is generally assumed for homogeneous fluid inclusions homogenizing «by halite disappearance» ( $T_s > T_h$ ) (BODNAR, 1982; ROEDDER, 1984). The trapping temperature must be below the maximum  $T_s$  NaCl measured ( $T_s = 480^\circ\text{C}$ ), which in any case, is well below any magmatic temperature. Thus, it is very difficult to know the original composition of the circulating fluids. However, considering the salinity range from

24 to 55 wt.% NaCl eq., we can propose an average value of about 30 wt.%.

The situation is more complicated for  $G_{II}$  and P. In the Th-salinity diagram both fluids are well separated, but with a partial overlap between low salinity, low density fluids in the granite ( $G_{IIA}$ ) and the highest density fluid in the pegmatite (P), (Fig. 8). These P fluids, being in an euhedral quartz from a vug, are obviously the latest ones in the pegmatite.

Considering their identical salinity, it can be tentatively assumed that  $G_{II}$  and P belong to the same generation, but that fluids in the pegmatite present lower density (higher homogenization temperature than in the granite).

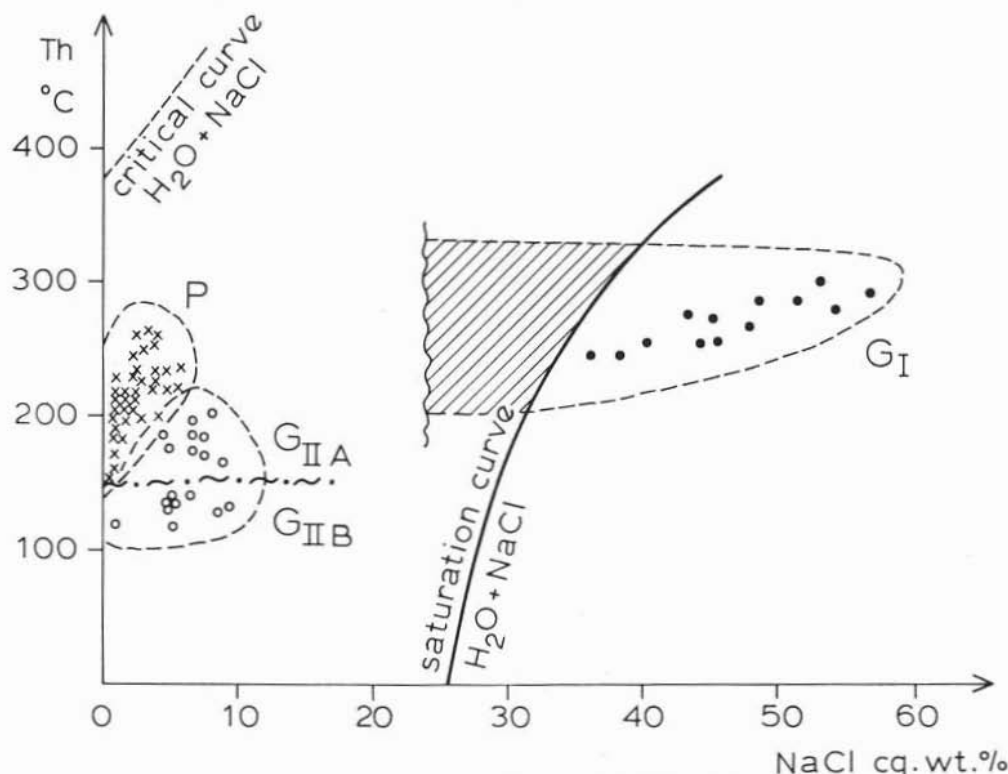


Fig. 8. — Homogenization temperatures versus composition (wt.% NaCl eq.) in the system  $\text{H}_2\text{O} + \text{NaCl}$ :  $G_I$  (•): three-phase halite bearing inclusions;  $G_{II}$  (o): two-phase aqueous inclusions; P (x): two-phase aqueous inclusions.  $G_I$  and  $G_{II}$  + P inclusions are situated in different areas (dotted lines);  $G_I$  distribution gives evidence of a large salinity range with values from 24 to 55 wt.% NaCl eq. which can suggest oversaturation at time of trapping. As for  $G_I$  biphasic inclusions the salinity could not be determined precisely,  $T_h$  measurements can only be represented as a field bound between NaCl saturation curve and the lowest qualitative estimate of  $G_I$  salinity (hatched area). P and  $G_{II}$  (in the low salinity field) are distributed in such a way that at least the  $G_{II}$  inclusions homogenizing at highest temperatures ( $G_{IIA}$ ) can be considered to belong to the same fluid episode than P. Both  $G_{IIA}$  and P represent the same fluid/rock interaction episode, the former in the leucogranite, the latter in the pegmatite euhedral quartz crystal.  $\text{H}_2\text{O} + \text{NaCl}$  saturation and critical curves from POTTER and BROWN data (1977).

### Fluid composition and P, T evolution

The interpretation of fluid data will be based on the following premises:

— in the granite, two fluids of very contrasting salinity:

1)  $G_I$  (24 to 55 eq. wt.% NaCl) with an average representative composition of 30 eq. wt.% NaCl; Th between 200 and 310°C with a mean value around 260°C.

2)  $G_{II}$  low salinity, (5 wt.% NaCl); Th between 100 and 220°C with two maxima around 130°C ( $G_{IIB}$ ) and 180°C ( $G_{IIA}$ ).

— In the pegmatite, one low salinity fluid (P) very similar to  $G_{II}$ , with Th between 140 and 260°C and no definite maxima at any temperature.

In Figure 9, the isochores for  $G_I$  and  $G_{II}$  in the granite and the area, limited by the isochores for the lowest and highest Th, for

«P fluid» in the pegmatite, are proposed, as well as a possible P, T evolution of the plutonite. In an earlier work on the leucogranite plutons GHEZZO and ORSINI, (1982) have reported the low water content and emplacement at shallow depths. RAMBOZ et al., (1982) have shown that any inclusion fluid which has a  $T_s$  significantly higher than Th cannot have been trapped at the liquid + vapour boundary (Fig. 5), and therefore cannot represent the liquid phase of an unmixed fluid. These statements support our findings (e.g. absence of vapour rich inclusions) and confirm that the fluid evolution did occur in the miscibility field of the  $H_2O + NaCl$  system without any boiling phenomena. This separation between magmatic aqueous fluids and melt in the fluid miscibility field (for the system  $H_2O + NaCl$  above 1.2 kb at 700°C) gives an upper limit

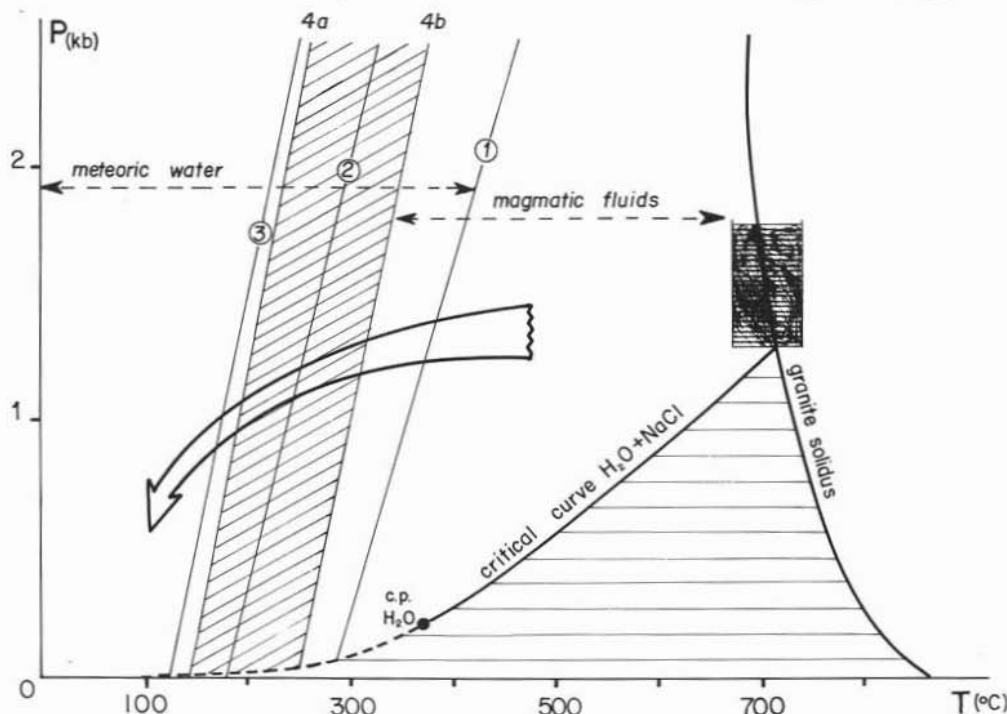


Fig. 9. — P, T interpretation of fluids  $G_I$ ,  $G_{II}$  and P. (1), (2), (3), (in circles): representative selected isochores from Th histograms (Fig. 4), corresponding respectively to  $G_I$  (magmatic, high salinity fluids),  $G_{IIA}$  (early, low salinity fluids),  $G_{IIB}$  (late low salinity fluids) in the leucogranite. (4a) and (4b): Minimum and maximum Th values for P (low salinity fluids) in the euhedral smoky quartz crystal in pegmatitic vugs. Respectively: (1): 30 wt.% NaCl after WALTHER (1981); (2) and (3): 5 wt.% NaCl after ROEDDER (1984); (4a) and (4b): 3 wt.% NaCl after ROEDDER (1984). Arrow: model of post-magmatic P, T evolution. Critical curve for  $H_2O + NaCl$  system after POTTER et al., (1977). Granite solidus after TUTTLE and BOWEN (1958). P, T conditions of granite emplacement (gray area) discussed in text; area below the critical curve: P, T conditions of immiscibility for the  $H_2O + NaCl$  system.

of emplacement for the plutonite at a depth of about 4 km.

### *Probable magmatic origin for $G_I$*

It is well known that the first fluids which evolved after the solidification of a granite magma are generally highly saline (fluid immiscibility between silicate and saline melt, e.g. Ascension Island, ROEDDER and COOMBS, 1967; CATHELINÉAU et al., 1987). For this reason, we propose that  $G_I$  fluids are initially of magmatic origin, but microthermometric data indicate a late evolution and clear postmagmatic conditions at the time of trapping;  $T_h$  in fact correspond to isochores that would reach magmatic conditions (about 700°C) at 7 kb (about 23 km depth considering lithostatic pressure regime), which is a contradiction for this kind of shallow intrusives. Magmatic immiscibility between brines and silicate melt took place when the melt became water-saturated. Due to the low initial  $H_2O$  content of the magma, this could only occur at a high degree of crystallization, in which all of the primary minerals, including quartz, had already crystallized (these leucogranites are close to the minimum eutectic composition, GHEZZO and ORSINI, 1982). No evidence of the early magmatic stages has been preserved in the rock, and this may be due to continuous quartz recrystallization at high temperatures. Final trapping has occurred at much lower temperatures (< 480°C) well after the end of crystallization.

### *Late fluids: interaction with meteoric water ( $G_{II}$ and P)*

At a late stage ( $T$  200–300°C), the pluton is invaded by homogeneous low salinity fluids ( $G_{II}$ ) that are also involved in the formation of the euhedral quartz crystals in the pegmatite vugs (P). The small differences in  $T_h$  and salinity between  $G_{II}$  and P can be related to different time relationships and mechanisms of trapping. The structural relations of the inclusions, their low salinities (about 5 wt.% NaCl) and low  $T_h$  are

inconsistent with a pure magmatic origin. In the present state of our research, a meteoric origin is much more probable, but it is not possible to state whether only meteoric waters are involved or if  $G_{II}$  result from a dilution of magmatic with meteoric fluids. Further studies on stable oxygen isotopes in fluid inclusions and in their host minerals are required in order to give a more definitive answer.

The comprehensive evolution of the hydrothermal circulation at these relatively «low» temperatures ( $T_h$  from 100 up to 260°C) must be linked to:

- the presence of a widespread system of joints and microfractures, related to brittle deformation.

- the cooling pattern of the pluton and country rocks which controlled the whole convective circulation system.

These factors allowed for the invasion of the pluton by meteoric low salinity water. The joint system, crosscutting also the vugs in the pegmatite pods, is likely to have connected them with hydrothermal circulating fluids. In any case the local conditions were very complicated; this is revealed in the granite by the presence of different generations of microfractures, characterized by different  $T_h$  values in fluid inclusions, and in the euhedral quartz crystal by the complex distribution pattern of fluid inclusions reflecting a sequence of discrete fluid pulses.

The euhedral crystals that line the vugs in some pegmatites record a considerable part of this fluid evolution during the late stages of cooling.

### *Model of P, T paths*

A precise delimitation of the P, T path followed by the inclusions would require the knowledge of the pressure at the time of trapping. This is presently very poorly constrained:

- a) by the absence of boiling (P, T path above the  $H_2O + NaCl$  critical curve, Fig. 9);

- b) by a possible maximum pressure corresponding to  $P_{lithostatic}$  at the time of granite crystallization (about 1 to 1.5 kb, Fig. 9).

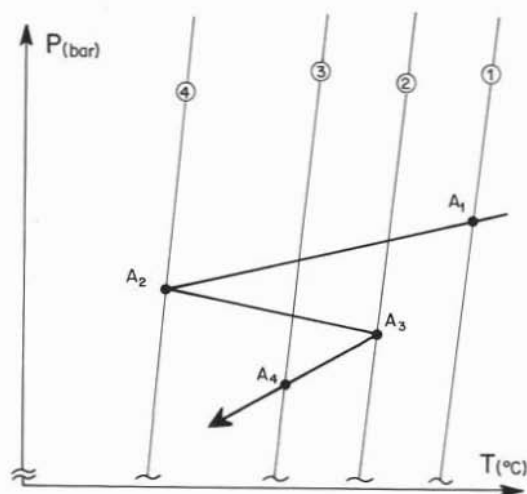


Fig. 10. — Detailed tentative interpretation of P,T conditions during the growth of the euhedral quartz in the pegmatite vug. 1, 2, 3, 4: representative isochores selected from topological distribution of ThL in the quartz (Fig. 7). Constant NaCl content (3 eq. wt. %).  $A_1$ ,  $A_2$ ,  $A_3$  and  $A_4$ : P and T conditions at the growth of respective zones in quartz crystal (Fig. 7). Unspecified pressure and temperature axes, corresponding roughly to the domain indicated in Figure 9.

The absence of discontinuity in Th values between  $G_I$ ,  $G_{II}$  and P suggests a continuous evolution and, in the present state of our research, the inferred model corresponds to the arrow indicated in Figure 9 (isobaric cooling trajectory around the pluton). In this model, the average fluid pressure has remained for a long period roughly equivalent to the lithostatic pressure at the time of granite crystallization. Locally the situation might have been much more complicated, as illustrated by successive fluid pulses during the growth of euhedral quartz (Fig. 10).

The transparent quartz (core of the crystal, Fig. 7) corresponds to the cooling of our initial fluid at a roughly constant pressure ( $A_1$ - $A_2$ , Fig. 10). This episode was not unique, but occurred several times. The last episode of the growth (smoky quartz y, Fig. 7) corresponds to the trajectory  $A_3$ - $A_4$  in Figure 10; probably at a lower pressure than  $A_1$ - $A_2$ , but by an unknown amount.

## Conclusions

In the investigated samples, two different

aqueous fluids have been characterized. The salinity gap between  $G_I$  on the one hand, and  $G_{II}$  and P on the other, suggests a different origin. A magmatic origin is proposed for  $G_I$  and a meteoric one for  $G_{II}$  and P. In this hypothesis, highly saline fluids may occur in shallow intrusive without boiling by direct magmatic immiscibility (ROEDDER and COOMBS, 1967). Similar results have been obtained by a number of other researchers, e.g. SALEMINK (1985), HARRIS (1986), CATHELINIEAU et al. (1987). Confirmation would evidently be obtained by stable isotope studies; however this is not straightforward. The meteoric signature of P (vug in the pegmatite) is in principle not difficult to establish, but the situation is entirely different for the granite. The  $G_I$  and  $G_{II}$  inclusions are so intimately intermingled that only single inclusion analysis could allow the precise determination of the isotopic signature of each group.

**Acknowledgements.** — We wish to thank C.A. Ricci for the stimulating discussions; I. Memmi and C. Baldelli for the invaluable help at the SEM and microprobe, and P. Lattanzi for extensive and constructive remarks on an earlier version of the draft of the manuscript. M.L.F. acknowledges B. De Vivo for his suggestions when working at the U.S.G.S. heating freezing stage in Rome and also the Vakgroep E.P.M. (Vrije Universiteit) for their help and support during her stay in Amsterdam. Work supported by C.N.R. grants n. 8500985/05 (to C.G.) and Art. 20/2-7 Regol. 26/1/67 Posiz. 121.05720 Prot. 067884 (to M.L.F.).

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