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Salt-poor and Salt-rich Fluid Inclusions in Quartz from Two Boreholes in Northern Switzerland

by

J. Mullis and H.A. Stalder

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Vorwort der Schweizerischen Geotechnischen Kommission

Von den Tiefbohrungen Böttstein und Weiach aus dem Untersuchungsprogramm der NAGRA (Nationale Genossenschaft für die Lagerung radioaktiver Abfälle) liegen bereits umfangreiche Publikationen in geologischer, geotechnischer sowie hydrologischer Richtung vor. Die vorliegende Publikation, welche den Teilaspekt der salzhaltigen Flüssigkeitseinschlüsse in Quarz von diesen beiden Bohrungen zum Thema hat, bildet eine wertvolle Ergänzung zu den NAGRA-Arbeiten, welche hauptsächlich in den „Technischen Berichten" der NAGRA sowie zum Teil in den „Beiträgen zur Geologie der Schweiz, Geotechnische Serie" der Schweizerischen Geotechnischen Kommission veröffentlicht werden.

Die Kommission dankt den Autoren für ihren Beitrag zum Verständnis der Flüssigkeitseinschlüsse dieser beiden wichtigen Bohrungen in der Schweiz.

Für den Inhalt von Text und Figuren sind die Autoren allein verantwortlich.

Zürich, Juni 1987

Schweizerische Geotechnische Kommission
Der Präsident
C.M. Schindler

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SALT-POOR AND SALT-RICH FLUID INCLUSIONS IN QUARTZ FROM TWO BOREHOLES IN NORTHERN SWITZERLAND

J. MULLIS* and H.A. STALDER

Institut de Minéralogie et Pétrographie, Université de Fribourg, CH-1005 Fribourg (Switzerland)
Naturhistorisches Museum, CH-3005 Bern (Switzerland)

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Abstract

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Microthermometric studies on fluid inclusions in quartz from two boreholes in northern Switzerland which cored the Böttstein granite at Böttstein and the crystalline basement with the overlying Permo-Carboniferous trough sediments at Weiach enable to elucidate the evolution and the *P-T* trapping conditions of the fluid compositions.

Two fluid groups, one salt-poor and the other salt-rich, were identified within inclusions from boreholes. The salt-poor inclusions are older than the salt-rich inclusions, whose age is post-Stephanian. The salt-poor fluids consist mainly of NaCl and are trapped under relatively high temperatures (up to 350-400°C). The salt-rich inclusions reached temperatures of at least 100-130°C. Low temperatures observed during the first melting (−70° to −37°C) indicate that other chloride salts besides NaCl (i.e. $CaCl_2$, $MgCl_2$, etc.) must be present. These salt-rich fluids probably originated in the Permian and/or Triassic evaporites, and imply significant vertical fluid migration.

1. Introduction

Studies of cores from two boreholes drilled in northern Switzerland indicated that the crystalline rocks were partly hydrothermally altered (Peters et al., 1985). Fluid inclusion studies were therefore carried out to evaluate the composition and trapping conditions of the fossil hydrothermal solutions.

One of the boreholes is situated near Böttstein (Fig. 1). The borehole penetrated and cored a rather homogeneous Hercynian biotite granite with large phenocrysts of K-feldspar below a Mesozoic cover (Peters et al., 1985). Two types of hydrothermal alterations were observed comprising: (1) an earlier alteration of the granite body attributed to cooling; and (2) a later, more intense alteration, restricted to fissures and cataclastic zones (Peters and Hofman, 1984).

A thick sequence of Permian and Upper Carboniferous sediments was unexpectedly found in the other borehole, located near Weiach (Fig. 1; Matter et al., 1987). The crystalline basement cored in this borehole comprises gneisses with aplites that were altered along fault zones during the intrusion of Late Carboniferous granites (Meyer, 1987).

**Present address*: Mineralogisch-Petrographisches Institut, Universität Basel, Bernouillistr. 30, CH-4056 Basel, Switzerland.

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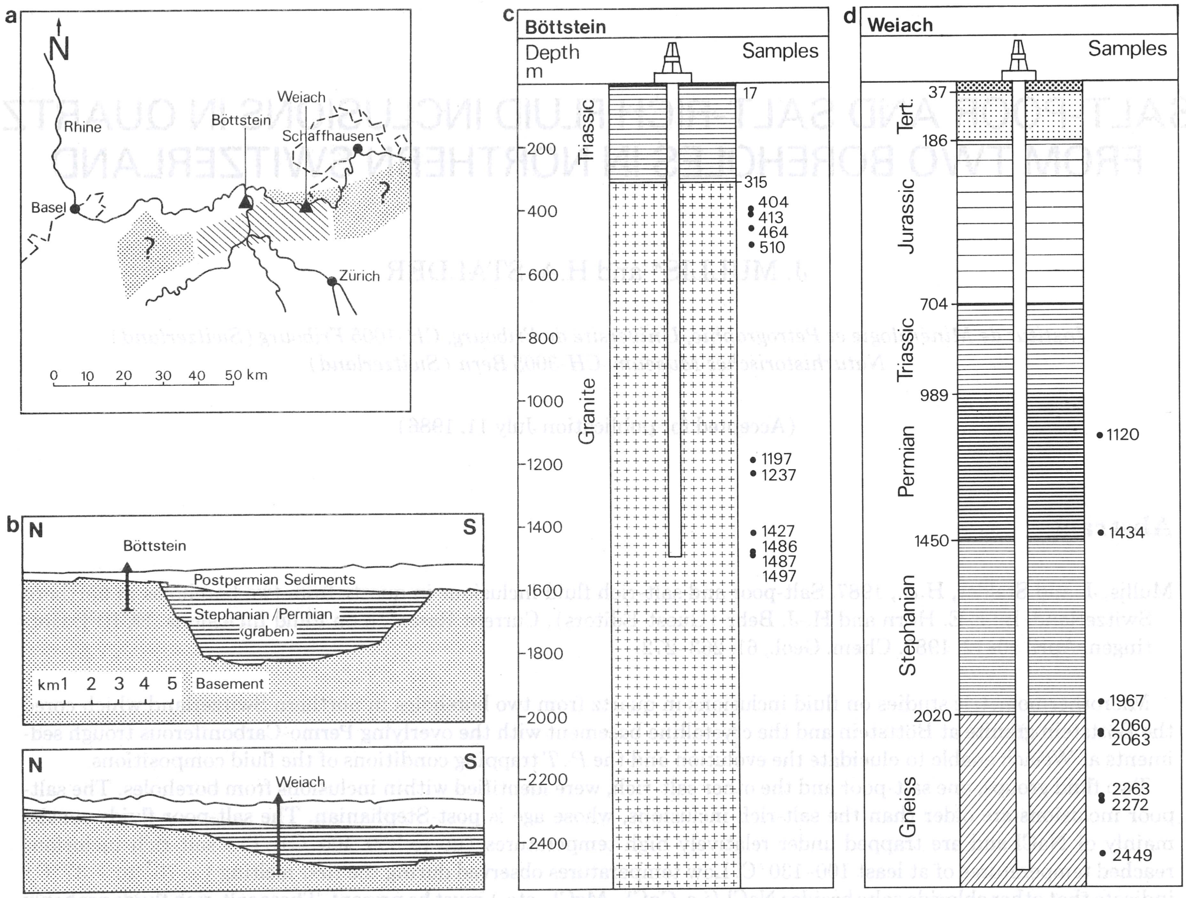

Fig. 1. a. Northern Switzerland showing the two drilling sites and the Permo-Carboniferous trough (proved and supposed).
b. Profile through the Permo-Carboniferous trough at Böttstein and Weiach.
c. Geological profile of the Böttstein core.
d. Geological profile of the Weiach core.
(According to information published in *Nagra Aktuell*, 1983–1984.)

2. Analytical techniques

Fluid inclusions were studied in rock-forming and fissure quartz from crystalline rocks in both the Böttstein and Weiach boreholes, as well as in detrital, fissure and authigenic quartz from sedimentary rocks in the Weiach borehole.

Microthermometry as described in Roedder (1984) was used to measure the temperatures at the first melting ($=T_{m1}$; Fig. 2), the melting ($=T_m$), the dissociation of hydrate (T_{dHy}) and the bulk homogenization ($=T_h$).

No gas could be found through microthermometry except for limited amounts of CO_2 in some of the early salt-poor inclusions from Weiach. Selected fluid inclusions, from Böttstein (1486 m) and Weiach (2063 m), where microthermometric measurements had not shown any dissolved gas, were analysed with the Raman microprobe (CREGU, Vandoeuvre-lès-Nancy), but neither CH_4 nor CO_2 were detected.

3. Results

Microthermometric measurements were performed on liquid- and vapour-rich fluid inclu-

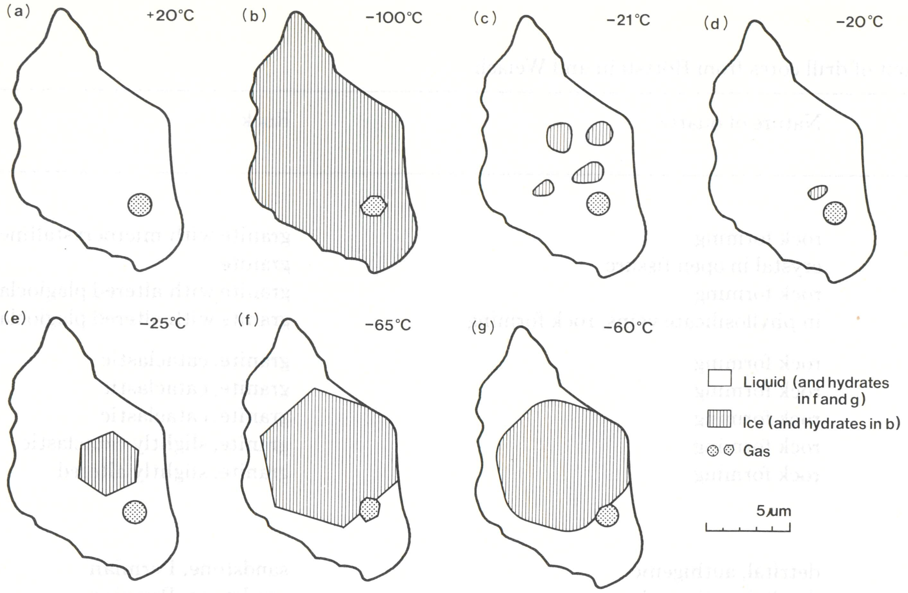

Fig. 2. Microthermometric determination of the first melting by sequential freezing and reheating for $CaCl_2$-bearing fluid inclusions. Roundig of the ice boundary in (g) indicates the first melting (T_{m1}).

sions of trails with constant gas/liquid ratio. Clusters of fluid inclusions showing different degrees of filling were also found, indicating heterogeneous trapping and necking down (Roedder, 1984).

3.1. The Böttstein core (Tables I and II; Fig.3)

Two groups of fluid inclusions were detected: a group of salt-poor inclusions in rock-forming and early fissure quartz; and a group of salt-rich inclusions in rock-forming quartz.

3.1.1. Salt-poor inclusions. These inclusions show a T_m between $-4.5°$ and 0°C and T_h between 350° and 90°C. Certain systematic differences exist between inclusions from shallower and those from deeper borehole levels. The early inclusions derived from the upper level exhibit a more or less continuous decrease in T_m down to $-3°C$ and an increase of T_h of up to 310°C with increasing depth. Below the 1190-m level, T_m decreases down to $-4.5°C$, whereas T_h increases up to $+350°C$.

3.1.2. Salt-rich inclusions. These inclusions have only been observed in deeper levels, where T_h is between 118° and 43°C: at the 1237-m level T_m ranges from $-27°$ to $-14°C$, with T_{m1} at around $-65°C$; at the 1497-m level T_m ranges from $-11.5°$ to $-6.5°C$, with T_{m1} at around $-39°C$.

3.2. The Weiach core (Tables I and II; Fig. 4)

The same two groups of fluid inclusions can be observed in the Weiach borehole. Salt-poor inclusions are found almost exclusively in rock-forming quartz. Salt-rich inclusions, by contrast, can occur in all the observed forms of quartz, but are the only inclusions found in the younger fissure quartz of the crystalline basement, the Permo-Carboniferous sediments and in the authigenic quartz overgrowths of the Permian sandstones.

3.2.1. Salt-poor inclusions. Special attention has been given to the evolution of T_m, T_h and the liquid/gas ratios of these inclusions. T_m increases from at least $-4.5°$ to 0°C, whereas

TABLE I

Quartz samples out of drill cores from Böttstein and Weiach

Depth (m)	Nature of quartz	Rock
Böttstein:		
404	rock forming	granite with microcrystalline quartz veins
413	crystal in open fissure	granite
464	rock forming	granite with altered plagioclase and ore veins
510	in phyllosilicate veins, rock forming	granite with altered plagioclase
1,197	rock forming	granite, cataclastic
1,237	rock forming	granite, cataclastic
1,486	rock forming	granite, cataclastic
1,487	rock forming	granite, slightly cataclastic
1,497	rock forming	granite, slightly altered
Weiach:		
1,120	detrital, authigenic	sandstone, Permian
1,434	detrital, authigenic	sandstone, Permian
1,967	detrital, fissure quartz	greywacke, Stephanian
2,063	rock forming, fissure quartz	aplite in gneiss
2,064	rock forming, fissure quartz	aplite in gneiss
2,272	rock forming, fissure quartz	gneiss, cataclastic
2,450	rock forming, fissure quartz	aplite in gneiss

TABLE II

Salt-rich inclusions

Depth (m)	T_{m1} (°C)	T_m (°C)	T_h (°C)	Gas bubble (vol.%)	n
Böttstein:					
1,237	−65, −70 to −64	−21, −27 to −14	97, 65–118	3, 2–5	30
1,497	−39, −40 to −37	−9.1, −11.5 to −6.5	72, 43–103	3, 2–4	5
Weiach:					
1,120	−58, −65 to −50	−18.7, −20 to −18	25, 19–32	1	17
1,434	−55	−20	67, 55–73	2	3
1,967	−60	−17.8, −18.4 to −17	99, 91–125	3	18
1,967	−58, −60 to −50	−12, −14 to −9.5	100, 91–117	3	39
2,060	−60	−17.8, −18.3 to −16.8	97, 91–109	3	26
2,060	−60	−9.8, −11 to −9	102, 97–110	3	20
2,063	−52	−17.5, −18.8 to −16	97, 90–107	3	50
2,263	−60	−9.0, −10.2 to −6.3	106, 94–121	3	19
2,272	−60	−17.7, −17.8 to −17.6	83	3	2
2,449	−53, −60 to −50	−19.8, −20.6 to −18.3	94, 83–107	3	20

n = number of studied inclusions. Values before the comma are median temperatures.

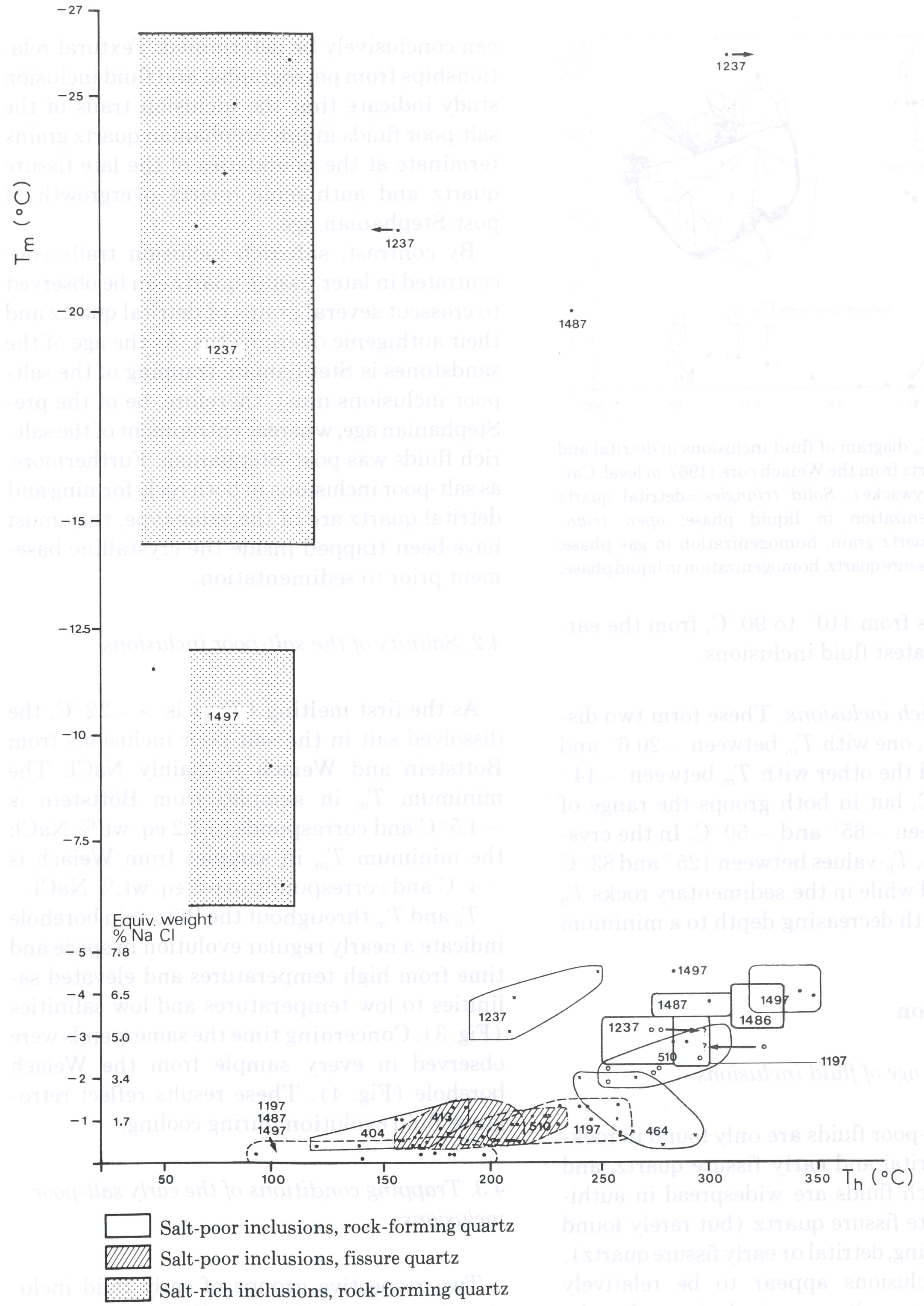

Fig. 3. T_m vs. T_h diagram of fluid inclusions in quartz from the Böttstein core. See text for discussion.

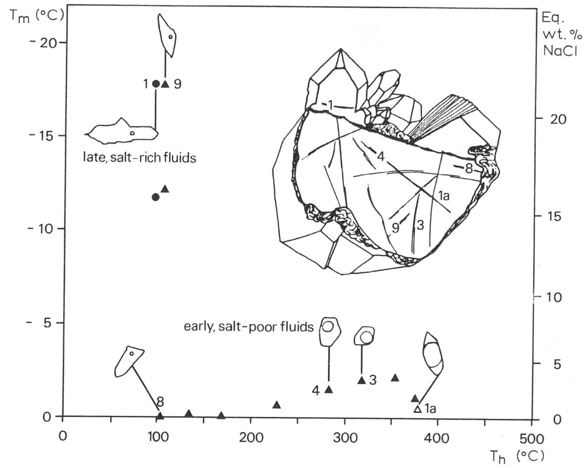

Fig. 4. T_h vs. T_m diagram of fluid inclusions in detrital and authigenic quartz from the Weiach core (1967-m level, Carboniferous greywacke). *Solid triangles* = detrital quartz grain, homogenization in liquid phase; *open triangle* = detrital quartz grain, homogenization in gas phase; *solid circles* = fissure quartz, homogenization in liquid phase.

T_h decreases from 410° to 90°C, from the earliest to the latest fluid inclusions.

3.2.2. Salt-rich inclusions. These form two distinct groups, one with T_m between −20.6° and −16°C, and the other with T_m between −14° and −6.3°C, but in both groups the range of T_{m1} is between −65° and −50°C. In the crystalline rocks, T_h-values between 125° and 83°C are observed while in the sedimentary rocks T_h decreases with decreasing depth to a minimum of 19°C.

4. Discussion

4.1. Relative age of fluid inclusions

Since salt-poor fluids are only found in rock-forming detrital and early fissure quartz, and since salt-rich fluids are widespread in authigenic and late fissure quartz (but rarely found in rock-forming, detrital or early fissure quartz), salt-rich inclusions appear to be relatively younger than salt-poor inclusions. In the Weiach borehole (e.g., 1967-m sample; Fig. 4, Table III) the relative age of the two fluid groups can conclusively be determined. Textural relationships from petrographic and fluid inclusion study indicate that the inclusion trails of the salt-poor fluids in pre-Stephanian quartz grains terminate at the boundaries of the late fissure quartz and authigenic quartz overgrowth of post-Stephanian age.

By contrast, salt-rich inclusion trails concentrated in later fissure quartz can be observed to crosscut several grains of detrital quartz and their authigenic overgrowths. As the age of the sandstones is Stephanian, trapping of the salt-poor inclusions must, therefore, be of the pre-Stephanian age, whereas entrapment of the salt-rich fluids was post-Stephanian. Furthermore, as salt-poor inclusions in both rock-forming and detrital quartz are of the same type, they must have been trapped inside the crystalline basement prior to sedimentation.

4.2. Salinity of the salt-poor inclusions

As the first melting (T_{m1}) is > −22°C, the dissolved salt in the salt-poor inclusions from Böttstein and Weiach is mainly NaCl. The minimum T_m in samples from Böttstein is −4.5°C and corresponds to 7.2 eq. wt.% NaCl; the minimum T_m in samples from Weiach is −4°C and corresponds to 6.5 eq. wt.% NaCl.

T_h and T_m throughout the Böttstein borehole indicate a nearly regular evolution in space and time from high temperatures and elevated salinities to low temperatures and low salinities (Fig. 3). Concerning time the same trends were observed in every sample from the Weiach borehole (Fig. 4). These results reflect retrograde fluid evolution during cooling.

4.3. Trapping conditions of the early salt-poor inclusions

Two respective groups of early fluid inclusions in some samples from Weiach were found to homogenize at approximately (±10°C) the same temperature (in the 300–380°C range).

TABLE III

Microthermometric results from a representative sample of Weiach (cf. Fig. 4) - Borehole Weiach 5: 1967. 25-m depth, Carboniferous greywacke (9 quartz samples, 120 inclusions)

Quartz(*1)	G(*2)	n_I(*3)	T_m(*4)	eq. wt.% NaCl(*5)	ρ_{H_2O}(*6)	T_{dHy}(*7)	Vol.%$_{GB}$(*8)	T_h(*9)	H_2O(*10)	NaCl(*11)	CO_2(*12)
D	1a	2	– 1.1 to – 1.0, – 1.1	1.9	1.011	7	60	375 L	99.4	0.6	0.0
D	1b	2	– 0.8 to – 0.2, – 0.5	0.8	1.003	8	70	378 V	98.5	0.3	1.2
D	2	5	– 2.5 to – 2.0, – 2.2	3.7	1.023	-	40	340-368, 353 L	98.8	1.2	0.0
D	3	20	– 3.0 to – 1.4, – 2.0	3.4	1.021	-	30	305-328, 317 L	98.8	1.2	0.0
D	4	20	– 2.6 to – 0.5, – 1.5	2.5	1.015	-	20	256-303, 282 L	99.2	0.8	0.0
D	5	2	– 0.8 to – 0.6, – 0.7	1.2	1.006	-	10	222-235, 228 L	99.6	0.4	0.0
D	6	4	– 0.5 to – 0.0, – 0.2	0.3	1.001	-	5	163-173, 169 L	99.9	0.1	0.0
D	7	1	– 0.3	0.5	1.002	-	4	134 L	99.8	0.2	0.0
D	8	7	0.0	0.0	1.000	-	3	99-107, 103 L	100.0	0.0	0.0
D	9	14	– 18.4 to – 17.0, – 17.8	21.3	1.155	-	3	91-125, 103 L	92.3	7.7	0.0
D	10	31	– 14.0 to – 9.5, – 12.2	16.4	1.116	-	3	94-117, 105 L	94.3	5.7	0.0
F	1	4	– 18.3 to – 17.5, – 17.9	21.3	1.155	-	3	91-99, 95 L	92.3	7.7	0.0
F	2	8	– 13.0 to – 11.1, – 11.8	16.0	1.113	-	3	91-101, 95 L	94.5	5.5	0.0

*1Quartz = detrital (D) and fissure (F) quartz; *2G = generation of fluid inclusions; *3n_I = number of studied inclusions; *4T_m = melting temperature of ice (°C), values after the comma are median values; *5eq. wt.% NaCl = equivalents weight percent NaCl, determined after Schäfer and Lax (1960); *6ρ_{H_2O} = density of the aqueous solution, determined after Potter and Brown (1977), at 25°C; *7T_{dHy} = dissociation temperature of hydrate (°C); *8vol.%$_{GB}$ = volume percent of gas bubble, estimated at 25°C; *9T_h = homogenization temperature of inclusion (°C), values after the comma are median values (V = homogenization in vapour phase, L = homogenization in liquid phase); *10H_2O = approximate content of H_2O in mole%; *11NaCl = approximate salt content, in eq. mole% NaCl; *12CO_2 = approximate gas content in eq. mole% CO_2.

These inclusions form trails; each group has a constant gas/liquid ratio, but one group is vapour-rich and homogenizes into the vapour phase, the other one is vapour-poor and homogenizes into the liquid phase. Assuming that the fluids were trapped at approximately the same time, these relationships indicate that the inclusions formed during (or closely after) boiling of the solution. T_t (=temperature of trapping) for such inclusions is $\geqslant T_h$, and therefore P_t (=pressure of trapping) is $\geqslant P_h$. As the homogenization temperatures (T_h) from these inclusions are always in the range between 300° and 380°C, the trapping temperatures must also be in the same range or only slightly higher. The respective homogenization pressures (P_h) are deduced from the boiling curve for pure water (Vargoftik, 1975) (respectively between this and the curve for an aqueous solution of 5 wt.% NaCl; Haas, 1976), which is very close to the water curve): for a T_h between 300° and 380°C, the P_h's are in the range between 86 and 220 bar. As already argued, the trapping pressures of such inclusions were essentially identical with, or marginally higher than, the homogenization pressures.

It was not possible to measure identical T_h's from vapour-rich and vapour-poor inclusions in samples from Böttstein because these inclusions were too small.

However, in many samples from both Böttstein and Weiach clusters of inclusions with highly variable gas/liquid ratios were observed. These inclusions are considered to have entrapped a heterogeneous fluid; it is suggested that the T_h of these inclusions if measured would correspond to points along the two-phase boundary in the temperature–density diagram of water. Textural observations indicate that necking down of these inclusions has not occurred and it is highly probable that the fluid inclusions were trapped during (or immediately after) boiling of the solution.

The trapping conditions for these fluids were in Weiach and in the lower parts of the Böttstein granite presumably very similar to those for the two early groups of fluid inclusions (which homogenized in the same temperature range into the vapour and the liquid phase) as described above, i.e. with a $T_t \geqslant 300$°C and a $P_t \geqslant 86$ bar. There are on the other hand considerable evidences (e.g., down to filling degrees of <4 vol.%) to suggest that boiling of the solutions in the upper parts of the Böttstein granite occurred at temperatures as low as 240°C. If this were true, then the trapping conditions for the early inclusions here would involve a $T_t \geqslant 240$°C and a $P_t \geqslant 40$ bar (deduced from the boiling curve for pure water).

4.4. Salinity of the salt-rich inclusions (see Table II)

The salt-rich solutions in the inclusions, of both boreholes, cannot be adequately described. The very low temperatures of the first melting (T_{m1}) range between −70° and −37°C and require other dissolved salt components besides NaCl. Particularly low eutectic temperatures are known in the systems containing $CaCl_2$ and $MgCl_2$:

$CaCl_2$-H_2O	−49.8°C	Linke (1958)
NaCl-$CaCl_2$-H_2O	−52°C	Yanatieva (1946)
NaCl-$CaCl_2$-$MgCl_2$-H_2O	−57°C	Luzhnaya and Vereshtchetina (1946)
Ocean water	<−70°C	Roedder (1984)

It is so far not possible to draw direct conclusions concerning the chemical composition of the Böttstein and Weiach fluids. However, it is evident that in addition to NaCl, $CaCl_2$ and other chlorides (or bromides) must be present. The low T_m between −6° and −27°C in the inclusions of both boreholes indicate salinities between 9 and >23 eq. wt.% NaCl. Such salt-rich solutions correspond to highly saline formation waters, respectively to evolved connate water (after White, 1981). Similar fluids were for example described for post-Variscan hydrothermal vein deposits in Central Europe (Behr et al., 1985). Expression of the salinity in terms of equivalent weight percent NaCl is considered problematic.

These salt-rich fluids may have originated in the Permian and/or Triassic evaporites. If such a source is accepted, these fluids must have migrated considerable vertical distances. For example in the Weiach borehole, the Triassic evaporites occur at a depth of 900 m. The salt-rich fluids must have been transported over a vertical distance of more than 1400 m down towards the crystalline basement at a depth of 2450 m.

4.5. Trapping conditions of the salt-rich inclusions

Highly saline aqueous solutions at P–T conditions of 100°C, 40 bar and 15 wt.% NaCl attain saturation with respect to methane at very low methane concentrations (0.03 mole%; Haas, 1978). Such low concentrations are close to or at the detection limit for the Raman microprobe (0.02 mole% CH_4; J. Dubessy, pers. commun., 1985) and so could not be conclusively demonstrated in the present study samples. However, aqueous solutions from coal-bearing sedimentary rocks can be expected to contain small amounts of dissolved methane. T_h can therefore be interpreted as approximative trapping temperature. In the crystalline basement measured T_h ranges between 72° and 106°C and decreases to 25°C in the Permian sedimentary rocks. The decrease of $T_h \leqslant T_t$ in Permo-Carboniferous sedimentary rocks of Weiach may be controlled through decreasing overburden during trapping. Unfortunately, it is not possible to evaluate the corresponding pressures as the exact fluid composition is unknown.

5. Conclusions

Two similar fluid inclusion groups were found in both the Böttstein and Weiach boreholes. An early group contains salt-poor inclusions and a younger group contains salt-rich inclusions.

The salt-poor fluids were relatively hot and were probably related to Carboniferous magmatism or even partly to Permian hydrothermal systems (Böttstein; Peters et al., 1985). Trapping conditions probably did not exceed 400°C and 250 bar. Homogenization of fluid inclusions with a constant gas/liquid ratio within the same temperature range into liquid and vapour phases as well as heterogeneous trapping indicates boiling of the early fluids before trapping.

The salt-rich fluids are of post-Stephanian age and are enriched in $CaCl_2$ and other chlorides in addition to NaCl. They probably originated in the Permian and/or Triassic evaporites. These salt-rich fluids penetrated deeply into the Permo-Carboniferous sedimentary rocks and the crystalline basement of the studied boreholes in northern Switzerland.

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