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FINE-GRAINED CLAY FRACTION (< 0.2 μm) : AN INTERESTING TOOL TO APPROACH THE PRESENT THERMAL AND PERMEABILITY STATE IN ACTIVE GEOTHERMAL SYSTEMS .

Patrier, P.*, Papapanagiotou, P.**, Beaufort, D.**, Traineau, H.*** and Bril, H****
* ERM - BP 25 - 86320 Civaux - France
** Lab. Pétrologie des Altérations Hydrothermales,
URA 721 du CNRS - Univ. Poitiers
40 av. du Recteur Pineau - 86022 Poitiers cédex - France
*** IMRG-BRGM - BP 6009 - 45060 Orléans cédex 2 - France
**** Lab. Géologie - Univ. Limoges - 123 Av. A. Thomas - 87060 Limoges -France

ABSTRACT

We have investigated by X-ray diffraction the very fine grained secondary minerals (< 0.2 μm) developped in geothermal systems, in relation with their present thermal and permeability state. Because the smallest particles are the most reactive part of a rock, they are the youngest mineral phases of the geothermal fields.

This study has been performed on two active geothermal fields: Milos field, Greece ($130<T<320$°C) and Chipilapa field, Salvador ($90<T<215$°C).

In the Milos field, the mineralogical composition of the $<0.2\mu m$ clay fraction observed in the reservoir strongly differs from the overlying altered metamorphic schists in the presence of abundant quantities of saponite and talc/saponite interstratified minerals at unusually high temperature. These phases are considered to be kinetically control-led "metastable" minerals which rapidly evolve towards actinolite and talc for present temperatures higher than 300 °C. Their occurrence is a good indicator of discharge in highly permeable zones.

In the geothermal field of Chipilapa, the mineralogical composition of the $<0.2\mu m$ clay fractions fairly agrees with the temperatures presently measured in the wells, whereas several discrepancies may be pointed out from the compositions of coarser clay fractions ($<5\mu$m) which contain minerals inherited from higher temperature stages. Permeable zones may be evidenced from an increase of expandable components in the interstratified minerals and a decrease of the coherent domain of the unexpandable clay particles (chlorite).

INTRODUCTION

Alteration assemblages in hydrothermal systems have received a considerable attention (Lonker et al., 1990; Reyes, 1990). Many authors have shown how temperature, fluid composition and permeability could influence the compositional and structural variations displayed by secondary minerals. However the use of alteration minerals (especially clay minerals) as condition indicators in geothermal environments seems rather complicated by the previous history of these systems. Overprinting during cooling stages or kinetics during heating stages often lead to incompatible assemblages.

An interesting tool to approach the recent thermal and permeability state of a system (and to predict its evolution) may be searched in the mineralogical composition of the very fine-grained fraction of secondary minerals because it contains the youngest cristalline phases of the altered rocks. So, in our ongoing research we have investigated by X. R. D. the mineral fraction less than 0.2 μm in several drill holes of two active geothermal systems:

- the active system of Milos, Greece (drill holes MI1, MI2, MI3).
- the active geothermal system of Chipilapa, Salvador (drill holes CH9, CH7b).

GEOLOGICAL CONSIDERATIONS

1- Milos geothermal area

Milos is one of the main volcanic centres of the Aegean volcanic arc. This volcanic arc is of Pliocene age.

The very strong thermal gradient measured in the exploratory wells indicates that at the present time, the thermal activity is in a prograde stage. The reservoir (below 900m depth) is hosted in fractured metamorphic formations. The drill hole MI1 crossed alluvial deposits (0-20 m depth), polygenic formations (calcareous and volcanic rocks) and the metamorphic basement (60-1180 m depth) which belongs to greenschist and locally blueschist facies. Measured temperatures range from 130 to 320 °C. According to Liakopoulos (1987) the hydrothermal fluid in the reservoir consists of boiling sea water near 250-350 °C.

2- Chipilapa geothermal area

The area of Chipilapa, close to the Ahuachapan geothermal field, is located in the Western part of Salvador. The stratigraphy includes pyroclastic rocks and lavas of andesitic composition, of Pliocene and Pleistocene age. Temperatures up to 215 °C (1791 m depth) have been measured in exploratory wells. However, data of fluid inclusions and the presence of relicts of epidote at shallow depth indicate that the system was subjected to higher temperatures during a previous stage. High permeable levels (total circulation loss) have been identified below approximately 1000 m depth.

METHODOLOGY

The determination of the clay fraction associated to recent hydrothermal activity needs a specific methodology which consists in extracting the fine-grained fraction of the altered rocks to minimize the "obscuring effect" of inherited minerals.

According to the available sampling, 5 to 25 g of rock were ground to obtain a powder. This fraction was dispersed in distilled water by ultrasonic treatment. The fine fraction ($<0.2\mu m$) was extracted from the $<5\mu m$ suspension by continuous ultracentrifugation (BECKMAN J2 21). The bulk suspension was introduced in the rotor with a 152 ml/mn flow rate. The rotor speed was 5000 rpm. The fine clay suspension was drawn through a millipore filter. Oriented preparations were realized by putting the filter on glass slide.

Because it strongly minimizes the influence of the non platy minerals, this method highly improves the recorded X.R.D. signals.

X-ray diffraction was carried out on air dried and ethylene glycol solvated preparations using a Philips PW1730 diffractometer (Co Kα radiation, 40 mA, 40 kV) monitored by a DACO MP numerical system. Spectra were recorded between 2 and 35 °2Θ with a step size of 0.025 °2Θ and a counting time of 6s. Distinction between dioctahedral and trioctahedral minerals has been made on the basis of the (060) reflection of randomly oriented powders.

Experimental diffractograms were smoothed to remove major statistical counting errors and complex spectra were decomposed in Lorentzian bands according to the method of Lanson (1990).

RESULTS

1- Mineralogy of the <0.2 μm clay fraction identified in both geothermal fields

Clay minerals have been identified according to the works of Brindley and Brown (1980) and Reynolds (1980, 1985).

- The trioctahedral clay sequence

Saponite shows typical diffractograms with basal spacings at about 17, 8.5 and 5.6 Å after ethylene glycol solvatation.

Irregular chlorite/smectite mixed-layers with high smectite content are characterized by a shift of basal spacings towards higher 2Θ angle. The 001 reflection ranges between 16 and 17 Å according to their percentage of smectite component (Fig. 1).

Corrensite is a regular chlorite/smectite mixed-layer (50/50). The diffraction pattern shows peaks at about 31, 15.5, 7.8, 5.19, 4.46 and 3.45 Å.

Chlorite is a non swelling mineral characterized by basal spacings about 14, 7, 4.7 and 3.5 Å (Fig. 1).

The following minerals are observed only in Milos samples.

Talc is a non swelling mineral characterized by peaks at 9.34 and 4.6 Å.

Saponite/talc mixed-layers are characterized by a second order reflection at intermediate spacing values between the (002) reflection of smectite and the (001) reflection of talc. In the studied samples, saponite component predominates over talc component. Considering the important width of the (002) reflection, several types of randomly

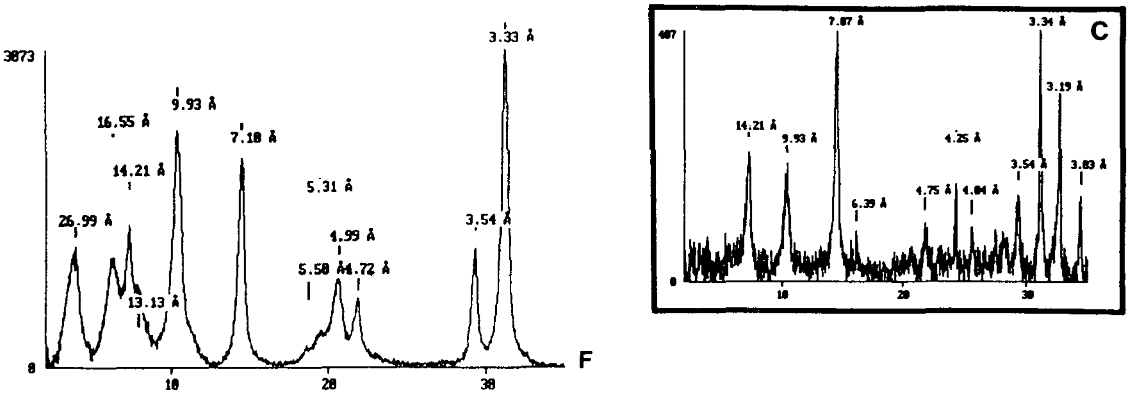

Fig. 1. X-ray diffraction patterns of the $<0.2\mu m$ clay fraction and of the coarser fraction "C" ($<5\mu m$). Sample CH7b 1066 m.

interlayered minerals probably coexist.

A mixed-layer phase characterized by a broad reflection at about 12 Å, that does not shift after ethylene glycol solvatation is locally identified. It may be an interstratification of non swelling minerals such as talc/chlorite (9.3, 14 Å).

- The dioctahedral clay sequence

Montmorillonite: diffractograms show basal spacings similar to saponite ones. The distinction of these smectites is possible by studying the position of their (060) reflection: this reflection ranges between 1.49 and 1.50 Å instead of 1.52-1.54 Å for saponite.

Regular illite/smectite mixed-layers (R1), observed in Chipilapa, show diffraction peaks at about 27-28, 13, 9-9.3, 6.7, 5.3, 4.5 and 3.3 Å (Fig. 1).

Illite and illite rich I/S mixed-layers (R3) show strong reflections near 10, 5 and 3.33 Å (Fig. 1). Minor quantities of mixed-layers generate the enlargement of the (001) reflection toward lower 2θ angle after ethylene glycol treatment.

2- Clay minerals distribution

In Milos, the following trioctahedral clay sequence has been established (Fig. 2):

Saponite (100-250m ; 130<T<170°C)
Saponite + corrensite (250-350m ; 180<T<220°C)
"12 Å interlayered mineral" + chlorite + saponite (400-600m ; 220<T<280°C)
Chlorite (650-850m ; 280<T<300°C)
Saponite ± talc + saponite/talc mixed layers are associated with actinolite below 903m depth (300<T<320°C).

The dioctahedral clay sequence is poorly developped in this field.

In Chipilapa

The alteration sequence recorded in CH9 well can be considered as a conventional pattern. The trioctahedral phyllosilicate sequence includes the successive appearance of smectite, chlorite/smectite mixed-layers and chlorite. In the same way, the dioctahedral sequence shows the development of montmorillonite, R1 and R3 mixed-layers and finally illite. However overprinting of different events is demonstrated by the crystallization of epidote and chlorite in shallow levels for lower temperatures compared to litterature data.

In CH9 well, the clay phases appear successively in a narrow range of temperature, from 185 to 214 °C.

In CH7bis well, the dioctahedral clay sequence is characterized by the successive occurrence of montmorillonite at shallow depth, R1 and R3 mixed-layers for depths which respectively exceed 306 and 748 m (T>155 °C and T>195 °C). The concentration of R1 and the disappearance of R3 mixed-layers at 898 m depth do not agree with a classical scheme of montmorillonite to illite conversion. The trioctahedral clay sequence comprises smectite in the whole well (110<T<204°C), corrensite between 408 and 556 m depth (174<T<197°C), chlorite from 652 m depth (T>195°C).

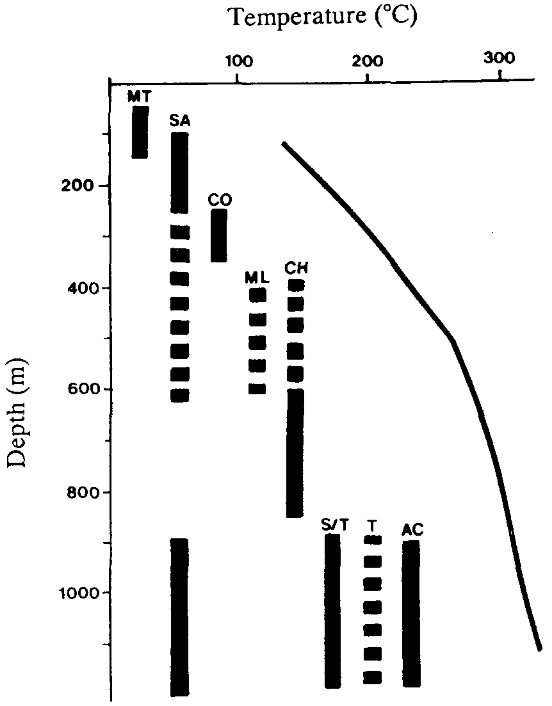

Fig. 2. Fine-grained clay minerals (<0.2μm) and present temperatures versus depth in Milos drill hole MI1. MT: montmorillonite, SA: saponite, CO: corrensite, ML: "12 Å phase", CH: chlorite, S/T: saponite/talc, T: talc, AC: actinolite.

Measurements on diffraction spectra of the fine fraction can be realized considering their good resolution. The thickness of chlorite particles (coherent domain) and the variation in percentage of expandable layers in chlorite/smectite have been investigated.

3- Coherent domain of chlorite particles

The coherent domain of a particle is the thickness (number of layers) which diffracts coherently. (001) peak width at half height is inversely proportional to the average thickness of the phyllosilicate population. In order to obtain the chlorite size all along the well, we have decomposed the (002) reflection in one or two lorentzian bands, considering or not the existence of associated mixed-layers chlorite/smectite. Such an investigation has been realized on samples from CH9 well, because of the great occurrence of chlorite in this well. Peak width at half height deduced from the whole reflection ranges from 0.42 to 0.71 °2θ and from 0.31 to 0.49 °2θ for the chlorite component of the decomposed reflection (Fig. 3). For comparison, the same values deduced from the coarser fraction (<5μm) have also been calculated: they range from approximately 0.33 to 0.16 °2ϴ.

4- Distribution of smectite and chlorite/smectite with low chlorite content

These phyllosilicates are of great interest because they are sensitive to thermal conditions (Bettison and Schiffman, 1988). According to Reynolds (1980), the (001) reflection of these minerals shifts towards high 2ϴ angles with increasing chlorite content. These minerals have been investigated in samples collected from CH7b well.

The position of their (001) reflection ranges between 16.9 and 16.5 Å (Fig. 4). It decreases with depth up to 1006 m, if we except the level 808m. For depth higher than 1006 m, an opposite trend is evidenced.

INTERPRETATION

1- Mineralogy of Milos geothermal area

The mineralogy of the <0.2μm clay fraction points out several interesting results. The trioctahedral sequence recognized in the formations which overly the reservoir (weak fracture permeability) obeys a classical alteration scheme largely described in geothermal systems and in diagenetic environment (Bettison and Schiffman, 1988; Inoue, 1987). However, the paragenesis observed in the reservoir (high permeable zone) consists of non common minerals for such thermal conditions (T>300°C). Saponite, talc/saponite mixed-layers, actinolite and talc are incompatible phases according to litterature and therefore cannot be interpreted in term of thermodynamical equilibrium.

Saponite has been hydrothermally synthetized by Iyama and Roy (1963) for temperature as high as 850 °C. However the reactivity of this mineral is high for such an elevated temperature and it evolves towards more stable phases such as anthophyllite + talc. According to Whitney (1983), for temperature close to 400 °C, after 200 days, saponite is transformed into saponite/talc mixed-layers.

The association described in Milos includes therefore all the minerals produced in synthesis experiments. They probably belong to the same sequence of mineral transformation, in which saponite and talc/saponite mixed-layers are metastable and so the youngest phases. Indeed the precipitation of smectite must be very recent with

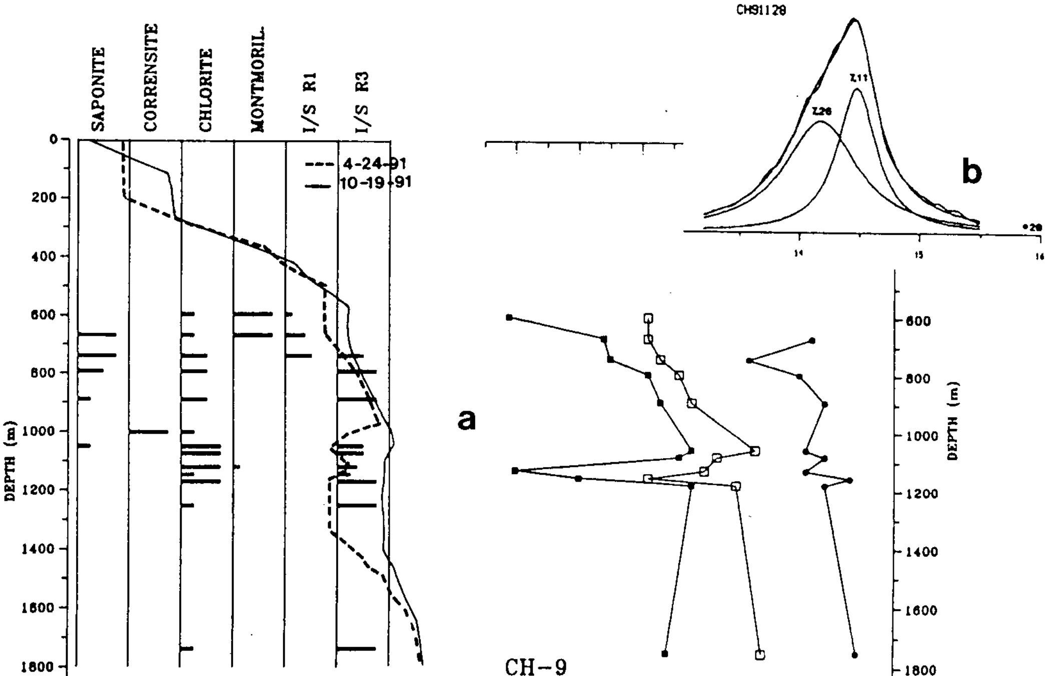

Fig. 3a. Distribution of temperatures and of <0.2μm clay minerals in CH9 well. Width at half height of (002) peak of chlorite (°2ϴ) is a plotted as a function of depth and measured temperatures.
■: (002) peak attributed entirely to chlorite
□: chloritic component of the decomposed signal (example Fig. 3b.)
●: " " for a <5μm clay fraction.

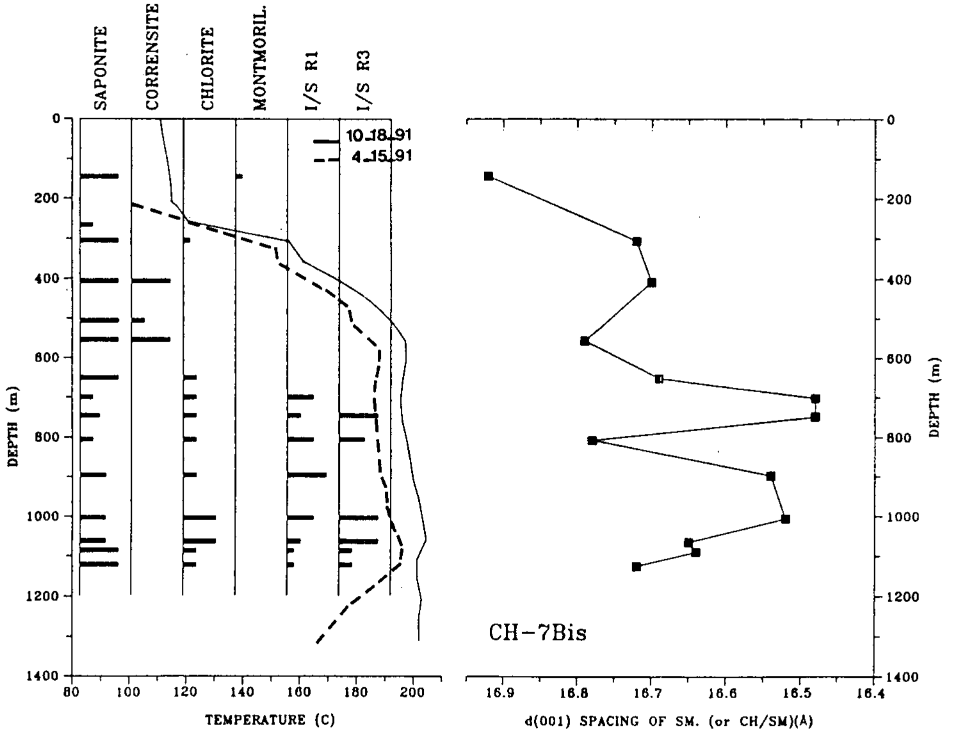

Fig. 4. Distribution of temperatures and of <0.2μm clay minerals in CH7b well. d(001) spacing of smectite or chlorite/smectite mixed-layers with high smectite content is plotted as a function of depth and temperature.

regard to its instability for such conditions.

The kinetic control of this association explains the discrepancy which appears between the <0.2μm clay mineralogy and the phases predicted by thermodynamic modelizations, based on equilibrium statement (Liakopoulos, 1987). The abundance of smectite is explained by discharge processes of oversaturated liquid in response to the boiling of the incoming sea water (Papapanagiotou et al., 1992).

In conclusion, the mineralogy of the fine-grained fraction clearly evidences the limitation of geothermometers based on chemical equilibrium and reveals close relations between metastable clays and highly permeable levels. The mineralogy of the fine fraction could therefore be a useful tool to evidence zones of high permeability and so productive zones in high enthalpy geothermal systems.

2- Mineralogy of Chipilapa geothermal field

The <0.2μm clay fraction minimizes the contribution of paleohydrothermal minerals. The alteration sequence so determined in CH9 well is a typical clay sequence. Informations deduced from it are in good agreement with the measured present temperatures. They do not evidence a strong physico-chemical disequilibrium like in Milos.

Below 1050 m depth, chlorite is the only trioctahedral phase, for temperature in agreement with those reported in litterature (about 200°C). The "coherent domain" of chlorite particles (X.R.D. peak width at half height) has been reported as a function of depth and temperature (CH9 well)(Fig. 3). The evolution of X.R.D. peak width and the temperature profile are very similar. The increase of peak width (lower particles thickness) recognized around 1100 m depth corresponds to highly permeable level as it is suggested by the response of the temperature profile to the absorption of drilling muds. This permeable level could give way to cold water circulations. The parallel between the temperature and chlorite grain size profile recorded by the < 0.2 μm clay fraction is not perceptible for chlorites of coarser clay fractions (<5μm). Such chlorites which appeared during previous higher temperature stages totally obscur the contribution of the most recent chloritic material.

The alteration zoning recognized in CH7b differs from the previous well in the sense that smectite and chlorite/smectite mixed-layers with high smectite content crystallize up to the bottom of the well. Because lithology and temperature in CH7b are the same as in the previous well, the crystallization of smectite could be attributed to different physico-chemical characteristics of hydrothermal fluids (colder meteoric water contamination). The d(001) of smectite and mixed-layers has been reported as a function of depth and temperature (Fig. 4). The variation of the percentage of smectite interstratified with chlorite is parallel to the temperature profile. The thermal gradient, directly dependent on rock permeability, could expressed the degree of contamination by colder fluids. It seems particularly interesting to note that the occurrence of nearly pure smectite (saponite) at about 800 m depth coincides with an inversion of the vertical thermal gradient.

CONCLUSION

X-ray diffraction on $<0.2\mu m$ mineralogical fraction extracted from cuttings (or cores) of two distinctive geothermal fields provides new informations on the very recent alteration processes and hence allows to access to their present physical state. Among these informations, the most interesting one concerns the variation of the expandable components in the interstratified clay minerals (illite/smectite or chlorite/smectite) and the crystal size (coherent domain) of the unexpandable clays (chlorite) as a function of temperature and permeability data recorded in exploration wells.

In systems subjected to a prograde alteration stage, as in Milos, the presence of high enthalpy reservoir (highly permeable levels) is clearly evidenced by metastable mixed-layered minerals whose present mineralogical state is inferred to be essentially controlled by kinetic factors (t*T).

In systems subjected to a retrograde alteration stage, as in Chipilapa, the $<0.2\mu m$ clay fraction strongly minimizes the "obscuring" effect of the paleohydrothermal clay phases (chlorite). The mineralogical composition of this clay fraction fairly agrees with the present thermal conditions in most of the investigated wells. However the presence of high permeable levels is identifiable through the lower coherent domain of chlorite particles in the reservoir.

Now, it would be interesting to test the response of the $<0.2\mu m$ clay fraction to variations of hydrothermal conditions (discharge and recharge zones).

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