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CLAY MINERALOGY OF THE 0.2µm ROCK FRACTION IN THE MI-1 DRILL HOLE OF THE GEOTHERMAL FIELD OF MILOS (GREECE)

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ABSTRACT

Very fine grained mineral fractions ($<0.2\mu$ m) of cuttings coming from three drill-holes of the active geothermal field of Milos (Greece) have been investigated by X-ray diffraction in order to determine the vertical distribution of both dioctahedral and trioctahedral hydrothermal phyllosilicates. Among the clay minerals so identified, the sequence of trioctahedral phyllosilicates dominates and shows a vertical distribution which distinguishs two major zones: (1) the altered formations overlying the reservoir (with low permeability) with a classical saponite to chlorite conversion sequence as the function of increasing temperatures; (2) the reservoir (with high permeability) in which coexist expandable clays (saponite and saponite/tale random mixed layers), actinolite and tale. The coexistance of these inferred incompatible minerals at temperatures higher than 300°C is interpreted as the result of the kinetic control on the mineral reactions which presently occur in the geothermal reservoir. Saponite is considered as a discharge product of oversatured sea water in boiling zones which rapidly reacts to be transformed in more stable phases at the present temperature (tale, actinolite) via various metastable saponite/tale mixed layers. At Milos, the presence of these mixed layers saponite/tale appears as a good indicator of the high fracture permeability zones which constitute the geothermal reservoir.

INTRODUCTION.

Alteration assemblages received a considerable attention during the last twenty years as much in diagenetic environments as in geothermal systems. From this great number of studies, it appeared that both mineralogy and chemistry of alteration phases, especially clay minerals, represented a powerfull potential for the determination of petrogenetic guides and for a better knowledge of water-rock interactions at the field scale. In this way, the sequences of clay assemblage observed in geothermal fields have been used in term of geothermometry and several mineral geothermometers based on chemistry at thermodynamical equilibrium have been proposed (Walshe, 1986, Giggenbach, 1981).

In fact the apply of clay minerals in geothermal field exploration is often dramatically complicated by several factors:

-The extraction of the clay matrix and the identification of its different clay components is no so easy because of the common presence of inherited phyllosilicates (micas, chlorite) in cuttings from drill holes.

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**Laboratoire de Pétrologie des Altérations Hydrothermales URA 721 du CNRS - Université de Poitiers 40, Av. du Recteur Pineau - 86022 Poitiers cedex -France. Ψηφιακή Βιβλιοθήκη Θεόφραστος - Τμήμα Γεωλογίας. Α.Π.Θ. -Clay minerals are highly reactive in hydrothermal conditions and their transformation are controlled by kinetic factors: in other word, at given P, T, X conditions the nature of the clay phase in a conversion sequence is still time depending.

In this study, we present a detailed characterization of the clay mineral sequences in the MI-1 drill hole of the active geothermal field of Milos (Greece). We have focused our investigation on the $<0.2\mu$ m clay fraction collected in metamorphic host rocks presently altered at temperatures ranging from 130° to 320°C. We have verified the reproductivity of the results obtained in the reservoir by additional studies of samples from two other drill holes (MI-2, MI-3). The vertical distribution of these clay minerals is discussed in order to point out the influence of host rock permeability and reaction kinetics on the present crystallization of clays in high enthalpy geothermal systems.

GEOLOGICAL SETTING.

The geothermal field of Milos in Greece is located in the central part of the so-called South Aegean Active A_{n+1} (fig.1). This arc, presently extended from the Saronic golf in the west to Nisyros island in the east, was formed during the Pliocene as a consequence of the northward subduction of the African plate beneath the Aegean one (Smith, 1971).

In 1975-76 two exploratory wells were drilled in the island of Milos; they proved the existence of a high temperature geothermal reservoir.

In 1980-81 three other productive wells were drilled in Zephyria plain, on the central part of Milos. Their stratigraphic columns and the temperatures measured during the exploration are given in fig. 2.

The well MI-1 crossed alluvial deposits (0-20m), polygenic formation (20-60m) and the metamorphic basement down to a depth of 1180m.

The well MI-2 crossed alluvia deposits (0-15m), volcanosedimentary deposits (15-25m), a polygenic formation (25-145m), neogene limestones (145-150m), and finally the metamorphic basement down to a depth of 1381m.

The well MI-3 crossed a polygenic formation (0-20m), altered pyroclastites (20-125m), neogene limestones (125-173m) and the metamorphic basement down to a depth of 1017m (Fytikas, 1989).

In these wells the reservoir is hosted in the fractured metamorphic basement, which is essentially composed of schisteous rocks containing abundant micas (muscovite, biotite) chlorite and locally talc.

The thermal gradient is high (near 4.5-5.5°C/10m) in the volcanosedimentary cover of MI-1 and MI-2, whereas it is lower 0.77-0.79°C/10m in the deeper part.

In the well MI-1 the reservoir was identified from 897m to the bottom hole. The measured temperature ranged from 307°C to 323°C the depths of between 900 and 1100m.

In MI-2 the reservoir is located from 930m to the well-bottom. The temperature of 282°C was measured at 1300m.

In MI-3 the reservoir was identified from the depth of 900m to the bottom of the drill hole.

The fluid collected in the reservoir consists of near boiling water and a steam phase (Report PPC).

According to Liakopoulos (1987) the reservoir is fed by sea-water which has undergone a distillation near 250-330°C, which are the temperatures presently measured in the reservoir. The formed steams can released of the system along the fractured zones.

METHODOLOGY.

Considering the fact that must of the host rocks of the geothermal field of Milos are schists which contain micas, chlorites and sometimes talc, the determination of the clay minerals of hydrothermal origin needed a specific methodology which consisted in the extraction of the very fine grained fraction of the rocks in order to minimize the influence of the inherited metamorphic phyllosilicates.



Figure 1: Milos island: geographic location and drill sites.



Figure 2: Litholo Ψηφιακή Βιβλιοθήκη Θεόφραστος - Τμήμα Γεωλογίας. Α.Π.Θ.

MI-3.

1. Sampling.

48 samples were studied in three borcholes. All consisted of cuttings collected at different depths. Their location and the associated temperatures are listed at table 1.

2. Samples preparation.

Approximately 25g of each sample (cuttings) were grinded to $< 50\mu$ m powder fraction in automatic scatterbox. A suspension of $< 5\mu$ m clay fraction (so called coarse clay fraction) was collected after ultrasonic treatment of the powder in distilled water and sedimentation during 4 hours. The $< 0.2\mu$ m clay fraction was extracted from the coarse clay fraction by continuous ultracentrifugation. The bulk suspension was introduced within the rotor at 152ml/min flow rate and the rotor speed was 5000 rpm. In such conditions particles larger than 0.2μ m were sedimented in the rotor whereas smaller particles remained in suspension leaving the rotor.

Clay oriented preparations were realized on glass slides following the method outlined by Bish and Reynolds (1989), which produces preparations having fair crystallite orientation and giving accurate intensity.

3. X-Ray Diffraction.

X-Ray diffraction was carried on air-dried and ethylene glycol saturated oriented preparations using a Philips PW 1730 diffractometer with Fe-filtered Co-Ka(1+2) radiation. Motor and intensity acquisition were effected using a DACO MP computer system.

The diffractograms of air-dried oriented clay preparations have been acquired with a step size of $0.025^{\circ}2\Theta$ between 2° and $35^{\circ}2\Theta$, whereas the preparations saturated with ethylen glycol were analysed between 2° and $15^{\circ}2\Theta$. Counting time per step was varying from 4 to 10 sec in order to obtain XRD patterns of high quality.

The distinction between the dioctahedral and the trioctahedral clay minerals has been made on the basis of the position of their (060) reflection of randomly oriented powders. Diffraction patterns obtained for ethylen-glycol saturated clay preparations were decomposed using the methods outlined by Lanson (1990) (fig. 3c). This method allows the substraction of the background and the decomposition of the superimposed peaks into elementary ones of gaussian or lorentzian shape.

RESULTS.

All the XRD patterns obtained from the coarse clay fraction the mineralogy is strongly obliterated by the presence of great amounts of inherited chlorite and micas.

1. Mineralogy of the fine fraction.

Clay minerals have been identified according to the recognition works of Brindley and Brown (1980), and Reynolds (1980, 1985). They are determined by the position of their diffraction bands after ethylen glycol saturation.

a. Trioctahedral phyllosilicates.

Saponite (trioctahedral smectite): X-Ray diffraction patterns of oriented preparations show basal spacings close to 17, 8.5 and 5.5Å. In this study, saponite is used as a general term for trioctahedral smectites (fig. 3a,d).

Corrensite is a regular chlorite/smectite mixed layer (50/50). This mineral is characterized by diffraction peaks at approximatively 31, 15.5, 7.6, 4.77 and 3.56Å (fig. 3a).

Chlorite is a non swelling mineral, characterized by basal spacings about 14, 7, 4.7 and 3.53Å (fig. 3a,b,d).

Talc is a non swelling mineral with basal spacings at 9.34 and 4.6Å (fig. 3a).

Saponite/Talc Mixed Layers are characterized by XRD reflections which differ from those of saponite by a second order reflection at intermediate spacing values between the second order reflections of saponite (8.5Å) and the first order reflection of talc (9.34Å) (fig. 3b,c). Considering the average position of

MI-1

Depth	Temperature	Depth	Temperature
(m)	(°C)	(m)	(°C)
50		650	285
100	130	700	290
150	150	745	295
200	170	804	300
250	185	843	300
300	210	903	300
350	220	955	310
400	235	1002	312
450	245	1049	318
500	265	1095	320
550	270	1152	
600	280	1180	

MI-2

Depth	Temperature		
(m)	(°C)		
800	298		
850	300		
900	302		
950	304		
1000	306		
1045	308		
1100	312		
1150	312		
1200	302		
1255	300		
1300			
1350			
1380			

MI-3

Depth	Temperature		
(m)	(°c)		
650			
700			
755			
800	204		
850	236		
900	230		
950	210		
1000	200		
1015			

Table 1. Estimated temperatures corresponding to the depths of selected samples according to the temperature-depth diagram (Fig. 2)



Figure 3: X-Ray diffraction patterns after ethylen glycol saturation, Co-Ka radiation:

- 3a: corrensite (corr), measured in the range 2°-15°20, 350m depth.
- 3b: saponite/talc randomly interstratified minerals (sap/talc), measured in the range 2°-15°2θ, 1180m depth.
- 3c: decomposition of the pattern 3b in the range 9.7°-12.7°20.
- 3d: mixel μαριακή Βιβλιδθήκη Θεύφραστος 2 Τμήμα Μεωλογίας. Α.Π.Θ.

their reflection and its very large width, it seems reasonable to consider that it represent the sum of several types of randomly interstratified saponite/tale. From a general point of view, the saponite component predominates in these interstratified minerals.

A Mixed Layer phase with a broad XRD reflection near 12Å (fig. 3d) have been observed in 5 consecutive samples, this reflection did not shift after ethylon-glycol saturation. This fact suggests the interstratification of two unexpandable layers as mica/chlorite $(10\text{\AA}/14\text{\AA})$ or tale/chlorite $(9.3\text{\AA}/14\text{\AA})$.

b. Dioctahedral phyllosilicates.

Montmorillonite (dioctahedral smeetite) gives spacings very similar to saponite. Its identification is possible on the basis of the (060) reflection which ranges from 1.49 to 1.50Å instead of 1.52Å to 1.54Å for saponite.

Micas, Illite and Illite rich 1/S mixed layers are characterized by strong reflections near 10, 5 and 3.33Å (fig. 3a,b,c,d. The presence of minor quantities of illite/smectite mixed layers containing more than 90% of illite may be suspected in a few shallow samples. They generate an enlargement of the near 10Å reflection towards the small angular values in the ethylen glycol saturated samples.

c. Other minerals.

Actinolite is characterized by a peak near 8.45Å (fig.3c). Calcite is characterized by a peak about 3.86Å and another one more intense about 3.04Å. Anhydrite is characterized by a peak about 3.49Å.

2. Vertical distribution of very fine grained minerals.

The vertical distribution of the $<0.2\mu m$ minerals identified by XRD in the well MI-1 is illustrated fig. 4. Two main clay sequences may be differentiated.

<u>The dioctahedral sequence</u>. In MI-1 drill hole the dioctahedral sequence is not clearly distinguishable because of its small proportion in the clay matrix. The main variation is characterized by the presence of illite in the upper part of the drill hole (down to 100-150m), illite rich I/S mixed layers minerals are observable between 150-200m and illite is clearly identified down to 400m. At greater depth the small peak observed near 10Å in almost all samples may be attributed to relics of inherited micas.

The trioctahedral sequence. A very complete sequence of trioctahedral clay minerals is observable in the MI-1 drill hole.

SAPONITE is observed between of depths 100 and 600m, however its proportion in the clay matrix decreases below 250m.

CORRENSITE appears in association with saponite near 250m and predominate down to 350m.

INTERSTRATIFIED PHASE at 12Å appears with saponite near 400m and is clearly identifiable down to 600m.

CHLORITE strongly predominates between 550 and 850m. The traces of chlorites observed as much at lower depths as at greater depth probably represent a background of metamorphic chlorites inherited in the $<0.2\mu$ m clay fraction.

SAPONITE/TALC RANDOMLY INTERSTRATIFIED MINERALS have been identified at depth between 745m and the bottom of the well (1180m) however these minerals are strongly represented in the reservoir (at depth below 903m). This particular occurence of such clay phases has been confirmed in samples from wells MI-2 and MI-3. Saponite/talc randomly interstratified minerals may be associated to discrete saponite and discrete talc in some samples. They constitute the major clay phase and are always associated to abundant and very fine grained actinolite crystals in the reservoir.

TALC is essentially observed above the reservoir. Considering the presence of tale schists in the metamorphic host rocks it seems probable that this mineral is often an inherited phase in the clay fraction. Hydrothermal tale has been detected in several samples of the reservoir. The hydrothermal origin of this mineral is suggested by the fact that this mineral is only present in the $<0.2\mu$ m clay matrix of samples which contains abundant muscovite and chlorite of metamorphic origin in their coarse grained clay fraction.

ACTINOLITE is restricted to the reservoir, in which it coexists with the saponite/tale mixed layers. ANHYDRITE is essentially present in the reservoir.



Figure 4.

Figure 4: Diagram showing the vertical distribution of the clay mineral assemblages along the well MI-1.

CALCITE is abundant in the main part of the MI-1 well, however its proportions strongly decreases in the reservoir and it seems to desappears below 1100m.

3. Interpretation.

The mineralogical study of the $<0.2\mu$ m fraction of the MI-1 drill hole of the geothermal field of Milos characterizes a very interesting sequence of Fe-Mg minerals in which clays are present in great amounts. The following sequence have been established:

-Saponite at depths between 100 and 250m and temperatures between 130° and 185°C.

-Corrensite + saponite between 250 and 400m and temperatures between 185° and 230°C.

-12Å mixed layer + chlorite ± saponite between 400 and 600m and temperatures between 230° and 280°C.

-Chlorite ± Saponite/talc mixed layers between 600 and 900m and temperatures between 280 °and 300°C.

-Actinolite + saponite/talc mixed layer \pm saponite \pm talc between 900 and 1180m and temperatures between 300 and 323°C.

The vertical distribution of the assemblages presented above clearly distinguished two contrasted zones:

-The altered formations overlying the reservoir (weak fracture permeability zone),

-the reservoir (high permeability zone)

The sequence of Mg-Fe clay minerals observed in the formations which overlies the reservoir has been reported in many geothermal fields. The transition saponite-corrensite-chlorite which appears with increasing depth and temperature has been interpreted by several authors as a conversion sequence of saponite to chlorite in natural systems (Inoue, 1987, Bettison and Schiffman, 1988 among others). In this respect, the interstratified mineral with a basal spacing at 12Å identified at Milos as a transitional phase between corrensite and chlorite.

Much less classical is the association of Mg-Fe clay minerals which appears in the reservoir. Effectively, the association of actinolite with saponite, mixed layers saponite/talc and sometimes talc at temperatures higher than 300°C is difficult to interpret in term of thermodynamical equilibrium. If actinolite and talc are common minerals at such thermal conditions in natural systems, saponite and saponite/talc mixed layers are well known to be formed at much lower temperature (Reynolds, 1988). How explain the association of these "incompatible" minerals in the reservoir of the geothermal field of Milos? It seems raisonable to consider that, in active geothermal systems, the fine grained clay fraction contains the youngest crystalline phases produced during the geothermal history and can be satisfactorily used for determine their recent geothermal history (Patrier and all, 1992). Most of these minerals are still growing and their relations are probably highly influenced by kinetic factors. Several experimental studies of the hydrothermal transformation of Mg-Fe phyllosilicates may be invoked for explain the mineralogical association observed in the reservoir of Milos. Saponite has been hydrothermaly synthethised at temperatures as high as 850°C (liyama and Roy, 1963), however in this case, this mineral is a metastable phase which evolves near more stable phases (anthophyllite) after a few days of experiments. The hydrothermal reactivity of saponite has been experimentally investigated by Whitney (1979). This author demonstrated that the reactivity of saponite highly increased with increasing temperature and that at temperature of 400°C, saponite is not stable for running times than 200 days; after, it transformed into various mixed layers saponite/talc. At Milos the association of Mg-Fe minerals: saponite, mixed layers saponite/tale, tale and actinolite observed at temperatures higher than 300°C in the reservoir includes all the minerals experimentally synthethised by hydrothermal transformation of saponite. All these considerations lead us to interpret the important occurence of saponite, mixed layers saponite/tale, tale and actinolite in the reservoir as the result of a continuous mineralogical transformation still in progress. If this is true, saponite and saponite/talc represent metastable phases which are very young and are probably still forming. This "kinetic" interpretation for the presence of expandable clay phases in the reservoir of Milos also explains the strong discrepancy which appears between the mineralogy of the very fine grained fraction and the phases which have been predicted from thermodynamic calculation assuming the attainment of equilibrium (Liakopoulos and all, 1991). In the present case, the abundant precipitation of saponite, saponite/talc mixed layers, talc and actinolite in muscovite bearing host-schists is not compatible with the attainment of chemical equilibrium and the chemical control of these reactions must be search in the fluid compsition of the reservoir.

According to the work of Liakopoulos (1987) these fluids are essentially sea water which is presently strongly heated then demixed in vapor and in a highly concentrated liquid phase. The precipitation of saponite from sea water is common in hydrothermal alteration (Kristmannsdottir, 1975, 1979, Pritchard and all, 1979, Searfe, 1980, Pertsev and Rusinov, 1980) and the abundance of this smectite in the present reservoir may be explained by discharge processes of oversaturated liquid in response to the change of state of the incoming sea water.

CONCLUDING REMARKS.

The mineralogical study of the $<0.2\mu$ m fraction of the altered rocks of the geothermal field of Milos points out several very interesting results which can be summarized as follows:

It is possible, using an appropriate method, to strongly minimized the obscuring effect due to the inherited phyllosilicates of host-rocks on the alteration mineralogy.

At Milos the alteration mineralogy of the reservoir highly differs from those, more classical, of the overlying weakly permeable rocks by the presence of clay phases (saponite, saponite/talc mixed layers) which are generally incompatible with the temperatures presently measured in the studied wells (300-320°C).

The presence of these minerals can be explained as the result of the discharge of overconcentrated sea water which have been partly vaporized at the contact of the very hot host-rock of the reservoir of the geothermal field. This process is still operating and the precipitation of expandable phases represent the youngest kinetically controlled phases which rapidely evolve towards more stable actinolite or tale.

The clay mineralogy of the very grained fraction of the geothermal field of Milos emphasizes the limits of the mineral geothermometry and reveals a close relation between anomalous clay minerals (saponite, saponite/talc mixed layers) and the high fracture permeability zones of the reservoir which can be an interesting guide for the exploration of active geothermal fields in similar volcanic arc environments.

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