MIXED-LAYER CLAY GEOTHERMOMETRY IN THE WAIRAKEI GEOTHERMAL FIELD, NEW ZEALAND

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Abstract—Mixed-layer clays of variable composition and structure occur in core samples from two drillholes (WK207 and WK210) drilled into the Te Mihi sector of the Wairakei geothermal field. These were identified by X-ray diffraction analysis of glycolated and oriented sample fractions at less than 2 μ m and less than 0.2 μ m.

Low permeability lacustrine sediments encountered by drillhole WK207 contain a well-developed sequence of mixed-layer clays. The shallowest downhole appearance of mixed-layered illite/smectite ($I_{0.6}$ /Sm) occurs at 146 m depth where temperature is only 100°C. Discrete illite is present only below 297 m (200°C) in the finer size fraction (less than 0.2 μ m). Chlorite first appears downhole, in association with illite-smectite, at 177 m depth (110°C).

Drillhole WK210 encountered predominantly ignimbrites and rhyolites, and fluid flow here is mainly in channels. Within these rocks, a sequence of interlayered clays is poorly developed. Discrete illite and chlorite are present in core from only 244 m (180°), but the measured temperatures where interlayer clays occur ranges from 140 to 209°C.

Differences in the identity of clay minerals present in the Wairakei reservoir, where conditions are otherwise the same, demonstrate the strong control that the type of fluid flow has on their formation. In poorly-permeable sediments, where diffuse fluid flow prevails, a clearly-defined sequence of mixed-layer clays occurs. These are absent where channel flow dominates, the discrete chlorite and illite deposit directly from solution.

Key Words-Geothermal system, Hydrothermal alteration, Mixed-layer clays, Reservoir permeability.

INTRODUCTION

The significance of clay minerals and mixed-layer clays as mineral geothermometers has long been recognized, both from the study of active hydrothermal systems and low-temperature metamorphic mineral assemblages. There are three ways in which clays are typically useful: (1) the presence or absence of a clay mineral may coincide with a specific temperature range, (2) changes in crystallinity or degree of organization of the structures of some mixed-layer clays takes place at a specific temperature, and (3) the transition between two end-member clays involves changes in chemical composition that may be temperature dependent.

Over the past two decades, major advances have been made in the study and understanding of mixedlayer clays. These studies have been undertaken within several independent disciplines that include soil science, petroleum geology, sedimentary geochemistry, geothermal petrology and clay mineralogy. Active geothermal systems are especially fruitful places to study the genesis of clay minerals since they are natural laboratories where fluid/rock interactions occur under measurable conditions.

Studies of the clay alteration mineralogy of the Wairakei geothermal system were first undertaken in the

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1950s by Steiner (1953, 1968, 1977) as part of his work on its subsurface geology and hydrothermal rock alteration. Subsequent investigations have been made by Zhang (1989) and Youngman (1989). The clay mineral studies of Steiner were among the earliest attempts to use clays as mineral geothermometers, and they contributed significantly to understanding the thermal stability of discrete clay minerals and some mixed-layer clays.

Previous studies of hydrothermal alteration drew attention to the usefulness of calc-silicate minerals as mineral geothermometers (Browne, 1979; Bird *et al.*, 1984). The identity of the secondary feldspars that form in a geothermal reservoir is also often a clue to the amount of fluid that has flowed through the host rocks (Browne, 1970), i.e., a clue to its permeability. Any information provided by clay minerals about temperature and permeability therefore adds to the mineralogical tools that geologists can use in interpreting the hydrology and paleohydrology of geothermal reservoirs.

The objectives of this study were: (1) to determine the distribution of mixed-layer clays in cores and cuttings obtained from the Te Mihi sector of the Wairakei geothermal system, taking into account our current understanding of mixed-layer clay structures, and (2) to evaluate their usefulness as mineral geothermometers and indicators of reservoir permeability.

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THE WAIRAKEI GEOTHERMAL SYSTEM

The Wairakei geothermal system is one of the largest active hydrothermal areas in the Taupo Volcanic Zone of the North Island of New Zealand (Figure 1). This zone is 30–50 km wide extending in a north-easterly direction for 200 km between the active andesite volcanoes of Ruapehu and White Island (Cole, 1990).

The Wairakei geothermal system is contained within a sequence of mainly Quaternary calc-alkaline silicic volcanic rocks interbedded with lacustrine sediments (Grindley, 1965). Hydrothermal fluids, at temperatures up to 270°C, ascend along a series of steep normal faults and hydraulic fractures. Lateral flow also occurs within several horizontally-bedded permeable formations.

Wairakei, the first water-dominated geothermal system in the world to be exploited for electric power generation, was commissioned in 1958 and has been producing almost continuously since then. Drilling for this development was undertaken between 1950 and 1987, with most drilling taking place prior to 1966. Over one hundred deep (to 2400 m) and shallow drillholes have been drilled. The majority of these holes were cored at intervals of 6-30 m, including ten drilled into the Te Mihi sector of the field. Tests made include downhole temperature and permeability measurements, and some of these data have been used in this study. The Te Mihi sector, in the western part of the Wairakei field, is a zone of major upflow. Wells discharging here have fluids with some of the highest gas



Figure 1. Location of drillholes in the Te Mihi sector of the Wairakei geothermal field in the Taupo Volcanic Zone, New Zealand. Line A-A' is the position of the geological cross-section (Fig. 2).

contents, temperatures and salinities present in the field. There is a trend of lower temperature and pressure towards the east of Te Mihi, which is consistent with their being a subsurface outflow path in this direction.

TEMPERATURE DEPENDENCE OF MIXED-LAYER CLAYS

The temperature dependence of smectites, illitesmectites, chlorite-smectites and chlorites has been studied both in hydrothermal systems and in buried diagenetic sequences, such as those that occur in the Gulf Coast sediments of the southern United States. A summary of the temperature stability ranges of several clay mineral structures is given in Table 1. Transformation from smectite to either illite or chlorite depends upon both temperature and the structure of the smectite. Dioctahedral smectites transform through illite-smectite to illite (Burst, 1958), while the less common trioctahedral smectites (e.g., saponite) transform through chlorite-smectite structures to chlorite (Iijima and Utada, 1971; Chang et al., 1986). The illite-chlorite mixed-layer clays progress towards discrete illite and chlorite with increasing temperature.

The smectite-to-illite transformation

Smectite transforms to illite with increasing temperature or depth of burial through a series of progressively more ordered illite-smectite interlayer structures. This process has been studied by workers who include Weaver (1956), Burst (1958, 1969), Steiner (1968), Perry and Hower (1970), Hower *et al.* (1976), and Nadeau and Reynolds (1981). The calculated X-ray diffraction data of Reynolds (1980) have been used in this study to determine the proportion of illite interlayering present in the mixed-layer structures.

In the Gulf Coast sediments of mid-Tertiary age, Hower et al. (1976) noted 20% illite layers present in randomly interstratified illite-smectite (I/Sm) clays at shallow depths. The illite contents increased monotonically with increasing depth of burial. At 3-4 km depth, an ordered, interstratified I/Sm phase, allevardite, occurred. This phase has about 60% illite layers and an inferred formation temperature of 100°C. With the increasing proportion of illite layers, there is an increasing degree of ordering in the structure, classified by Nadeau and Reynolds (1981) as R = 0 random interstratification, R = 1 allevardite or ISISIS type ordering (60–85% illite layers), and R > 3 Kalkberg or IIIS type ordering (>85% illite layers). Nadeau and Reynolds recognized, in the Mancos Shale of the western United States, a sequence of progressive illitization with increasing depth, with the threshold temperature for the formation of R = 1 type being close to 100-150°C.

Jennings and Thompson (1986) studied the diagenesis of Pliocene-Pleistocene sediments in the Salton Trough (southern California), where heat flow is very

Permeability and lithology Low permeability sediments	Transformation temperatures								
	70°C			90°C			190–220°C		
	Dioctahedral smectite Trioctahedral smec	\rightarrow ration rates related to rates related to rates relation rates ra	andom I/Sm chlorite/s	 smect	ordered $I_{0.5}/Sm$ + chlorite bi- product ite \rightarrow progressive c	→ orderi	progressive illitization + chlorite ng towards → c	→ hlorit	illite + chlorite (+? I/Chl) e
Permeable zones in sediments and volcanics									Direct precipita- tion of illite + chlorite

Table 1. General transformation temperatures of mixed-layer clay minerals.

high. These workers demonstrated that the younger the system, the higher the temperature that was necessary for a particular transition to occur. They recognized a sequence of increasing illite proportions present in I/Sm structures from 13% at 68°C to 63% at 129°C to 90% at 176°C, and complete transformation to illite at 208°C. The degree of ordering of the I/Sm structure was considered to be R = 0 for up to 130°C, R = 1 from 130–160°C, and R = 3 from 150–190°C, based on the ordering classification of Nadeau and Reynolds (1981).

The smectite-to-chlorite transformation

Iijima and Utada (1971) recorded the diagenetic transformation of trioctahedral smectite (saponite) to chlorite via a chlorite-smectite (corrensite) mixed-layer structure present in tuffaceous sediments of the Niigata oilfields in Japan. Smectite is absent where temperatures exceed 85-95°C, the lowest temperature at which corrensite occurs. Hoffman and Hower (1979) subsequently confirmed this as the smectite-corrensite transformation temperature in evaluating the use of corrensite as a mineral geothermometer. Tómasson and Kristmannsdóttir (1972) reported that the transformation of iron-rich saponite, via mixed-layer chloritesmectite phases, to chlorite occurs in some geothermal fields of Iceland. The change of saponite into mixedlayer clays takes place at about 160°C. Subsequently, Kristmannsdóttir (1976) reported the gradual transformation of trioctohedral smectites to discrete chlorite. This change is completed above 270°C. More recently, Chang et al. (1986) reported corrensite forming at 70°C in the Cassipore Basin shales of Brazil. Using chemical analyses they showed that the transformation of trioctahedral smectite to chlorite involves the loss of interlayer cations, an increase in the extent of tetrahedral substitution of aluminum for silicon, and the fixing of hydroxide sheets into interlayer positions to balance the charge deficiency created by the tetrahedral substitution.

Illite-chlorite mixed-layer clays

There are few data describing the occurrence of these mixed-layer clays in hydrothermal systems. Lee *et al.*

(1984), using high-resolution transmission electron microscopy (HRTEM) in sedimentary basin studies, report mixed-layer illite-chlorites that are randomly ordered at the individual layer level, but where burial metamorphism was more intense, separate into discrete chlorite and illite. In this study, the identification of illite-chlorites was based on the calculated X-ray diffraction data of Reynolds (1980).

Chlorite

Chlorite forms over a wide range of temperatures. From sedimentary basin studies, Cathelineau (1988) quotes work on the Colorado and North Sea basins where chlorite first formed at between 100 and 140°C. In the Wairakei geothermal system, Steiner (1968) recorded chlorite as present at temperatures as low as 110°C, but stable up to the highest temperature encountered at Wairakei (270°C). Minor chlorite occurred at much lower temperatures in the $<2-\mu m$ size fraction, but some of this may be of detrital origin.

Jennings and Thompson (1986) identified the first appearance of chlorite in the finest clay fraction (<0.1 μ m) at 194°C in the Salton Sea sediments. It was stable up to 270°C. They considered that chlorite present in the coarser size fractions was detrital. Yau *et al.* (1988), also working on the Salton Sea sediments, identified euhedral crystals of chlorite present in pores at temperatures above 220°C. In Iceland, Kristmannsdóttir and Tómasson (1975) recorded chlorite present in association with epidote forming above 230°C in basalts.

Cathelineau and Izquierdo (1988), working on samples from the Los Azufres geothermal field in Mexico, recognized that the amount of tetrahedrally coordinated aluminum and octahedrally coordinated iron present in the chlorite structure correlated positively with drillhole temperatures, which permitted chlorite compositions to be used as a mineral geothermometer.

The widely different formation temperatures for chlorite, close to 100°C in some settings and to 220°C in others, is explained by Yau *et al.* (1988) as being due to two different permeability regimes. In sedimentary basins with rocks of low permeability, the transformation from smectite to illite is a progressive reaction whereby the smectite structure is incrementally



Figure 2. Cross-section through the Te Mihi section of the Wairakei geothermal system. Reproduced with kind permission of C. P. Wood (1990).

illitized from a threshold temperature of about 100°C. Ahn and Peacor (1985) have shown, by HRTEM studies, that diagenetic chlorite is a direct by-product of this smectite-to-illite transformation in argillaceous sediments undergoing burial. This is commonly intergrown with packets of layers of smectite and/or illite, or else present as mixed-layer phases together with illite. Yau *et al.* (1988) carried out similar HRTEM studies on sediments from the Salton Sea geothermal field and also showed that authigenic chlorite occurs as grains of less than 2 μ m that are associated with patches of illite within smectite crystals.

In summary, therefore, in open systems above 220°C, where channel flow prevails, a dissolution-precipitation mechanism is likely, and chlorite forms by direct precipitation from fluids. By contrast, in systems of low permeability, where fluids move via interconnected pores, a gradual transformation of smectite to illite occurs. In this environment, chlorite forms at temperatures as low as 100°C as a by-product of this transformation (Table 1).

MATERIALS AND METHODS

Sample selection and preparation

Drillhole WK207 is situated in the center of the Te Mihi sector of the Wairakei field (Figure 1). Over the temperature interval of most interest with regard to clay minerals (i.e., 100–230°C), WK207 is drilled almost entirely within sediments of the Huka Falls Formation and Rautehuia Breccia (Wood, 1990).

Drillhole WK210 is located 1 km south of drillhole WK207 and penetrated only about 50 m of Huka Falls Formation and Rautehuia Breccia at shallow depth. Over the 100–230°C temperature interval WK210 penetrated tuffs, ignimbrites and rhyolites. A section

through this part of the Wairakei field is shown in Figure 2. Core samples were recovered from drillholes WK207 and WK210 at 6 to 10 m intervals. In places, however, the coring interval was approximately 20 m.

X-ray diffraction analysis

The samples were fractionated at 2 and 0.2 μ m and each size fraction studied separately by X-ray diffraction.

The two size fractions were prepared as oriented mounts on glass slides and allowed to dry below 40°C. X-ray diffraction analysis was carried out on these and on glycolated samples. X-ray diffraction charts were run on a Philips PW2243/20 diffractometer at 20 mA and 40 kV between 2 and 45°2 θ using CuK α radiation. Background correction and smoothing were applied to the data. Interpretation of the X-ray diffraction data was based on Brindley and Brown (1980) and Reynolds (1980).

RESULTS

The distributions of the clay minerals and zeolites identified in samples from drillholes WK207 and WK210 are given in Tables 2, 3 and 4, together with data on the sample lithology and formation temperatures considered to be closest to the pre-exploitation conditions. The sequence of mixed-layer clays present in drillhole WK207 is also shown graphically in Figure 3.

Drillhole WK207

Mordenite. This zeolite occurs in altered tuffs at depths of 104 m and 128 m where the temperature is below 100°C. No clay minerals were encountered.

Illite-smectite. At 146-152 m drilled depth, regular

Drilled depth (m)	Elevation R.S.L. (m)	Lithology	Formation temp. (°C)	2-0.2-µm fraction	<0.2-µm fraction
104	393	Tuff	<100	Mordenite	Mordenite
128	369	Tuff	<100	Mordenite	Mordenite
146-152	345-351	Sandstone	100	$I_{0.6}/Sm$	$I_{0.7}/Sm$
177-183	314-320	Mudstone	110	$Chl + I_{0.8}/Sm$	$Chl + I_{0.7}/Sm$
183-198	299-314	Mud siltstone	130	$Ch1 + I_{0.8}/Sm$	$Chl + I_{0.7}/Sm$
198-213	286-299	Tuffaceous silt	140	$Chl + I_{0.6}/Sm$	$Chl + I_{0.6}/Sm$
213-244	253-286	Breccia	155	$Chl + I_{0.9}/Sm$	$Chl + I_{0.8}/Sm$
244-250	247-253	Breccia	170	$Chl + I_{0.9}/Sm$	$Chl + I_{0.8}/Sm$
250-256	241-247	Muddy breccia	180	$Chl + I_{0.9}/Sm$	$Chl + I_{0.9}/Sm$
268-274	223-229	Soft breccia	190	Illite $+$ Chl	$Chl + I_{0.9}/Sm$
297-305	195-200	Firm breccia	200	Illite + Chl	Illite + Chl

Table 2. Clay and zeolite mineralogy or cores from drillhole WK207.

mixed-layered illite-smectite with 60% illite layers occurs. This is represented as $I_{0.6}$ /Sm in Table 2. The X-ray diffraction trace for the <0.2- μ m fraction after glycolation of this sample (Figure 4) shows the presence of a well-defined superstructure peak at 29–32 Å (2.8– 3.1°2 θ), and rational peaks at 13.81 (6.4°2 θ) and 9.45 Å (9.36°2 θ), indicating it has an ordered I/Sm structure.

Hower et al. (1976) and Jennings and Thompson (1986) recognized the threshold temperature for forming illite-smectite (R = 1) ordering to be 100°C. The drillhole temperature of 100°C measured in well WK207 at 150 m depth is consistent with this and shows that the composition of this clay reflects current conditions.

Samples from greater depth in WK207 show a general trend of an increasing proportion of illite present in the I/Sm structure, as evidenced by a shift in the position of the I/Sm(glycol) position from 9.3 Å in clay from 146–152 m towards the 10 Å peak of illite at a depth of 268–274 m (190°C). The values for selected samples and size fractions are summarized in Table 3.

Illite-chlorite. Crystal spacings consistent with the presence of a random mixed-layer illite-chlorite of composition $I_{0.7}$ /Chl occurs as shallow as 244–250 m (temperature here is 170°C) in the finest fraction (<0.2 µm). This identification is based on the calculated X-ray data of Reynolds (1980). Samples from below this depth have spacings progressively closer towards illite in composition, with discrete illite and chlorite present directly below the deepest occurrence of $I_{0.9}$ /Chl. In the finer clay fraction this occurs in the sample from 297–

Table 3. Difference in basal spacings of mixed-layer clays with increasing depth in drillhole WK207.

Drilled depth (m)	Elevation R.S.L. (m)	Size fraction (µm)	I/Sm (glycol) A	I/Chl A
146-152	345-351	2-0.2	9.3	
183-198	299-314	2-0.2	9.78	
212-244	253-286	2-0.2	9.94	_
244-250	247-253	< 0.2	-	11.1
250-256	241-247	2-0.2	_	10.55
268–274	223–229	20.2		10.48

305 m (200°C) whereas, in the 2–0.2- μ m size fraction, discrete illite and chlorite are present at 268–274 m (190°C). These data (Table 3) are consistent with the sedimentary basin studies by Lee *et al.* (1984), who reported mixed-layer illite-chlorites that are randomly ordered at the individual layer level, but as the effects of burial metamorphism increased, separate into discrete packages of both chlorite and illite.

The precise identification of these peaks is uncertain. A more detailed investigation is underway using the NEWMOD computer program and calculated X-ray defraction data of Reynolds (1980).

Chlorite. Chlorite is relatively abundant below a depth of 177 m (measured temperature here is 110°C). It is present in all samples from below this depth, but is most abundant in the coarser size fraction $(2-0.2 \ \mu m)$. The thermal stability of chlorite was investigated by heating to temperatures between 450 and 600°C, followed by X-ray diffraction analysis. The chlorite present at shallow depths (~200 m) showed structural collapse upon heating to 450°C. Chlorite from deeper levels (>300 m; measured temperature over 200°C) was stable upon heating to over 600°C, although some structural reorganization was indicated by an increase in the magnitude of the 14 Å reflection and a decrease in the 7 Å peak height.

Relationship between particle size, clay composition and temperature. The data from drillhole WK207 (Table 2, Figure 3) for the two size fractions indicate a higher degree of order of the clay minerals comprising the coarser fraction. Other studies (e.g., Jennings and Thompson, 1986) show, however, that the youngest diagenetic minerals concentrate in the finer size fraction. A similar situation may prevail in the Te Mihi sector of the Wairakei geothermal system where the larger clay crystals have had a longer time to grow than the smaller ones. An alternative explanation for the difference in composition of the two size fractions is that the coarser ones may have formed when the reservoir temperatures were slightly higher than those now measured, i.e., that slight cooling has occurred.

Drilled depth (m)	Elevation R.S.L. (m)	Lithology	Formation temp. (°C)	2-0.2-µm fraction	<0.2-µm fraction
123	388	Tuff	134	Mordenite	Mordenite + Chl
177	334	Ignimbrite	140	$I_{0.4}/Sm + Chl + I$	$I_{0a}/Sm + Chl + I$
209	302	Ignimbrite	164	$I_{0.4}/Sm + Chl$	$I_{0a}/Sm + Chl$
244	267	Ignimbrite	180	I + Chl	$\vec{I} + Chl$
276	235	Pumice tuff	190	$I_{0.9}/Sm + Chl$	I
334	177	Rhyolite	209	$I_{0.8}/Sm + Chl$	$I_n / Sm + Chl$
397	114	Rhyolite	224	Chl	Chl

Table 4. Clay and zeolite mineralogy of cores from drillhole WK210.

Drillhole WK210

Mordenite was present at shallow depth (123 m and 134°C) in samples from WK210, but the sequence of mixed-layer clays here (Table 4) is very different from that in WK207. Mixed-layer illite-smectites with variable illite proportions are present in association with minor chlorite and illite. Over much of the sampling interval, neither mixed-layer clays nor illite reflect the measured bore temperatures. A recognizable difference between these two wells is in the nature of their permeability. The lacustrine sediments encountered in drillhole 207 have few fractures, but channels in the volcanic rocks of WK210 permit conduit flow. Rapid ascent of reservoir water (at over 220°C) may result in the precipitation of illite and chlorite directly into these channels as a result of cooling. Away from the fractures, in zones of lower permeability, mixed-layer clay sequences may predominate where water-rock interactions are too slow to achieve equilibrium. Here, the alteration mineralogy does not reflect the prevailing thermal regime.

Detrital clay minerals in the lacustrine sediments and Rautehuia Breccia

To date, no systematic investigation of the detrital clay mineralogy of these sediments has been carried out. However, some are known to contain detritus from previously altered rocks, and it could be argued that some of the alteration minerals identified in this study are partly or entirely detrital. The progressive change from smectite to illite in the samples from drillhole WK207 via a mixed-layer clay type structure is in good agreement with the measured temperatures. These data are consistent with those published, confirming that the alteration clay minerals in this drillhole are in thermal equilibrium with the reservoir. Any detrital clay minerals present have clearly responded to their new thermal regime by changing their compositions.

CONCLUSIONS

The identity of the illite-smectite mixed-layer clays present within the lacustrine rocks in drillhole WK207 are consistent with the thermal stability data obtained on mixed-layer clays published elsewhere. Lacustrine sediments of the Huka Falls Formation and Rautehuia Breccia recovered from drillhole WK207 show a welldeveloped sequence of illite-smectite mixed-layer clays. The conjunction of this with the shallow appearance of chlorite, where temperatures are just over 100°C, confirms that the progressive change of smectite to illite occurs under conditions of poor permeability. This clay assemblage predominates where fluid diffuses through intergranular pores.

The host rocks in drillhole WK210 are tuffs, rhyolites and ignimbrites that show no progressive change in the identity of the illite-smectite mixed-layer clays. Mixedlayer clays are present here in association with illite and chlorite at shallow depths where fluid flows in channels. In zones or rocks where channel flow dominates, the clay minerals can form by direct precipitation. Away from these channels, poorer permeability, and perhaps slightly lower temperatures, result in the formation of mixed-layer clays because water/rock ratios are here very much lower.

Differences in the identity of clay minerals present in a geothermal reservoir, where physico-chemical conditions are otherwise the same, demonstrates the strong control that the type of fluid flow has on their formation. Under conditions of low permeability and diffuse fluid flow a well-defined sequence of mixedlayer clays commonly develops, but in zones or rock



Figure 3. Sequence of mixed-layer clays present in cores from drillhole WK207 compared with measured downhole temperatures that most closely match the pre-drilling temperatures.



Figure 4. X-ray diffraction trace of mixed-layer illite-smectite ($I_{0.6}$ /Sm glycol) sample from drillhole WK207 (146–152 m depth). Background has been subtracted.

types where channel flow is dominant, illite and chlorite precipitate directly from solution. In the host rocks away from these channels, mixed-layer illite-smectite clays may develop.

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