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Stevensite Scale Precipitation At The Mori Geothermal Power Plant, Japan

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ABSTRACT

A large amount of smectite scale with light gray, black or light brown colors has been found in the two-phase fluid flow zone of production wells, surface instruments and re-injection wells at the Mori geothermal power plant, Hokkaido, Japan. The mineralogy of the scale has been studied by petrologic microscope, X-ray diffraction, EPMA and TG/DTA analyses. These data suggest that the scale is Mn-bearing stevensite as one of trioctahedral smectite group, and also that the structural formula of a sample from a production well is (Ca_{0.02} K_{0.03})(Mg_{2.78} Mn_{0.14} Cu_{0.04} Fe²⁺0.05)Si_{3.98}O₁₀(OH)₂. Its crystallinity is strongly related to the fluid temperature; it is highly crystalline in the production wells, whereas it is poorly crystalline in the surface instruments and the re-injection wells. The stevensite scale might be precipitated from the MgO and SiO₂ rich hydrothermal liquid (120-240°C) by boiling of reservoir fluid in the production well.

Introduction

Stevensite, a trioctahedral magnesian smectite, was first found as a pseudomorph after pectolite in the vicinity of Bergen Hill Tunnel, Hoboken, New Jersey, USA (Faust and Murata, 1953). It occurs in various ancient and modern saline lakes (e.g., Darragi and Tardy, 1987) and also skarn ore deposits (Otsu et al., 1963; Sakamoto et al., 1975). The first author (Y.M.) found that stevensite scale with a large amount of calcite has been deposited in production wells at the Mori geothermal field (Muramatsu et al., 1988b). Smectite scale has also been found at some geothermal fields such as the Fushime (Yagi and Akaku, 1989), Onikobe (Ajima et al., 1998), Takigami (Taguchi et al., 1999) in Japan, and Philippine (Reyes and Cardile, 1989) and Iceland (Kristmannsdottir et al., 1989).

In this paper, we present the mineralogy on the stevensite scale from the production wells,

surface instruments and re-injection wells at the Mori geothermal power plant. Moreover, we discuss the hydrothermal stability of the stevensite scale based on the temperature and pressure logging, and the fluid geochemical data.

Mori Geothermal Field and Power Plant

The Mori geothermal field is located in the Nigorikawa Basin, about 5 km southwest of the seacoast and about 20 km northwest of Mt.Komagatake, which is an active volcano, southwest Hokkaido, Japan (Figure 1). Geothermal development of the field started in 1977 by Dohnan Geothermal Energy Co., Ltd. with the cooperation of Japan Metals and Chemicals Co., Ltd. and Hokkaido Electric Power Co., Ltd. Twenty-six geothermal wells had been drilled to depths from 430 to 2464 m before 1982 in the field.



Figure 1. Schematic geologic section of the Mori geothermal field (modified from Kajiwara et al., 1996). B-D and F: well bases.

The geology of the field was described by Ide (1982), Ando et al. (1992) and Kurozumi and Doi (1993,1994,1996). The Nigorikawa Basin with about 3 km in diameter is a small funnel shaped Crater Lake-type caldera (Ando et al., 1992), formed by volcanic eruption about 12,000 years ago (Sato, 1969; Sumi and Takashima, 1976; Figure 1). The field consists of the pre-Tertiary Kamiiso Group, Neogene Ebiyagawa Formation and Pleistocene caldera-fill deposits in ascending order (Figure 1). The Kamiiso Group is mainly composed of limestone, slate, chert and tuff. The Ebiyagawa Formation, which is composed of tuff breccia, tuff, andesite and shale with a basal conglomerate unconformably overlies the Kamiiso Group. The caldera-fill deposits are composed of alluvial, lake, landslide and vent fill (lapilli tuff and tuff breccia) deposits. Hornblende andesite intruded the caldera-fill deposits. Clay minerals consist of montmorillonite, mixed-layer sericite/montmorillonite, sericite, chlorite and kaolinite.

The Mori geothermal field is a liquid-dominated geothermal system. The reservoir fluids ascend mainly through fractures in the Kamiiso Group near the caldera wall and horizontal fractures confined to such lithofacies as each upper part of limestone and chert in the Kamiiso Group (Akazawa et al., 1993). These productive fractures were formed at the time of caldera formation (Kurozumi and Doi, 1996). The geochemistry of fluids discharged from geothermal wells in the field was described in detail by Yoshida (1991). The predominant non-condensable gas in steam is CO_2 whose concentration ranges from 0.4 to 0.8 mol % of the total discharges and from 97.4 to 98.8 mol % of the non-condensable gases. The hot water is of weakly alkaline and Na-Cl type with low salinity.

The double flash system was adopted for the Mori geothermal power plant (50MWe). A flasher and secondary steam supply



Figure 2. Distribution map of the steam supply system at the Mori geothermal power plant (after JMC, 1987).



Figure 3. Photograph of smectite scales from the Mori geothermal power plant.
A. Smectite scale (ST) with calcite (CA) from 80m depth of the production well ND-1 (sampling date 1988.7).
B. Smectite in white scale sample from 308-430m depths of the production well NF-1 (sampling date 1987.8). Crossed polarized light under the microscope. An airlift tube is on the left side.

| Table 1. Occurrence of smectite and its associated scales deposited before 1988 at the Mori geothermal power plant. |
|--|
| ST,smectite; CA,calcite; AR,aragonite; QZ,quartz; AS,amorphous silica; AH, anhydrite; TE,tetrahedrite; CP,chalcopyrite; PY,pyrite; HA,halite |

| | Sampling point | ST | CA | AR | QZ | AS | AH | TE | Others | Sampling date |
|-----------------------|---|-----|------|-----|----|----|-----|-----|--------|---------------|
| Production well | | | | | | | | | | |
| ND-1 | Injection tube | ++ | ++ | +++ | | | | | | 1984.4 |
| | Injection tube (0-200m) | +++ | | | | | | | | 1984.9 |
| | Injection tube (360m) | + | + | +++ | | | | | | 1984,9 |
| | Injection tube (80m) | t | +++ | | | | | | | 1988,7 |
| ND-3 | Injection tube (732m) | ++ | +++ | ++ | | | | | | 1987.8 |
| | Injection tube (1250m) | Ħ | | | | | | | | 1987.8 |
| ND-5 | Injection tube | +++ | ŧ | | ŧ | | | | | 1983.12 |
| ND-7 | Sampling pot | + | + | | | | ŧ | +++ | | 1986.5 |
| | Centralizer | +++ | | | | | | | | 1986.5 |
| | Sampling pot (1684-1688m) | +++ | ÷ | | | | | +++ | СР | 1987.8 |
| NF-1 | Sampling pot (989m) | ++ | ·### | | + | | | +++ | | 1983.3 |
| | Reaming scale (550-650,778m) | ++ | +++ | | + | | | ++ | | 1983,4 |
| | Airlift tube (white sample at 308-430m) | ++ | +++ | | | | | | | 1987.8 |
| | Airlift tube (black sample at 308-430m) | +++ | | | | | | ++ | | 1987.8 |
| | Reaming scale (450-1592m) | + | +++ | | + | | | | | 1987.8 |
| | Reaming scale (2315m) | + | +++ | | ŧ | | +++ | | | 1987.8 |
| NF-9 | Reaming scale (395m) | +++ | | | | | | | | 1983.4 |
| | Reaming scale (745m) | +++ | + | | | | | | | 1983.4 |
| | Dummy (490,1800m) | + | | | ŧ | | | | PY | 1984.6 |
| | Dummy (1503m) | +++ | | | | | | | | 1984.6 |
| | Sampling pot (594,596m) | +++ | +++ | + | | | | | | 1986.12 |
| Re-injection well | | | | | | | | | | |
| NF-6 | Sampling pot (1235 - 1244m) | +++ | | | | | | | | 1987.4 |
| | Sampling pot (<1224m) | +++ | | | + | | | | | 1987.7 |
| NF-7 | Sampling pot (<1032m) | +++ | | | + | | | | | 1987.7 |
| NF-8 | Sampling pot (1441m) | +++ | | | | | | | | 1987.4 |
| Surface Installations | | | | | | | | | | |
| | Separator (200 line) | +++ | | | + | | | | | 1984.9 |
| | Separator (300 line) | +++ | | | | | | | | 1985.1 |
| | Level header (200 line) | ++ | ++ | | + | | | | РҮ,НА | 1984.9 |
| | Level header (300 line) | +++ | | | | | | | | 1984.9 |
| | Injection water flow line (D-F) | +++ | ++ | + | | + | | | | 1984,10 |
| | Two phase flow line (200,300line) | ++ | ++ | | + | | | | PY | 1986.5 |
| | Heat exchanger | ++ | ++ | + | | ++ | | | | 1984.5 |
| | Butter valve (300 line) | ++ | | | | | | | | 1984.9 |

system are set up in addition to the usual components such as two-phase flow pipeline, separator, flasher, steam flow pipeline, hot water tank and injection water flow pipeline in the double flash system (Figure 2). Unfortunately large amounts of calcite and smectite scales have occurred in the production wells, re-injection wells and surface instruments.

Since the reservoir fluids are saturated with respect to calcite, plugging of the production wells by precipitation of calcite scale occurred directly after operating the plant. At the early stage of the scaling trouble, reaming and acid treatment were done to remove the scale in the related well. Afterwards, a calcite scale inhibitor (polyacrylic acid) was injected below the flash point through injection or airlift tubes inserted in the related well. Subsequently, calcite scaling has been prevented, thus solving the trouble.



production well ND-1 (sampling date 1984.9). 1,tuffaceous mud; 2,altered andesite: 3,slate; 4,basal conglomerate; 5,limestone; 6,chert; 7,no cuttings; 8,complete lost circulation. ST,smectite; CA,calcite; AR,aragonite; PL,plagioclase; KF,K-feldspar; AM,amphibole; DO,dolomite; MS,magnesite; QZ,quartz; TR,tridymite; MO,montmorillonite; S/M, mixed-layer sericite/montmorillonite; SE, sericite; CH,chlorite; LA,laumontite; GY,gypsum; AH,anhydrite; PY,pyrite; Ebi.F., Ebiyagawa Formation; Dep.,caldera fall deposits.

Description of Smectite Scale

The smectite scale occurs in the production wells, re-injection wells and various kinds of surface instruments such as separator, level header, hot water and two-phase flow pipelines, heat exchanger and butter valve. It has various colors such as light gray, black and light brown. The occurrence and mineral assemblage of the scales deposited before 1988, which were determined by petrologic microscope and X-ray diffractometer, are presented in Table 1. The smectite scale is associated with various proportions of calcite (Figure 3A), quartz, amorphous silica, tetrahedrite (Muramatsu et al., 1988a), anhydrite, aragonite and pyrite. Figure 3B shows the photomicrograph of smectite from production well NF-1. It occurs at the airlift tube side as aggregates of minute fibrous crystals (≤0.2mm in size) elongated in the vertical direction of the tube. Distribution of the scale products with depth in the production well ND-1 is shown in Figure 4. Surface of a injection tube inserted in the well has been clogged by smectite and minor amounts of calcite and aragonite above about 400 m depth, where it is under two-phase flow conditions according to the dynamic temperature and pressure logging data. The smectite scale occurs limitedly at the two-phase flow zone shallower than the flashing point in each production well.

X-Ray Diffraction Study

X-ray powder diffraction for the smectite scale was performed using a Rigaku diffractometer. X-ray diffraction patterns under untreatment and treatment for the scales from three different locations are shown in Figure 5. The basal spacings are variable among the production well NF-9, two-phase flow line (300 line) and re-injection well NF-6. The smectite scales from NF-6 and two-phase flow line appear to be trioctahedral group because the (06, 33) peak is 1.523-1.525Å and the (001) peak of 16.1-17.0Å shifts to 17.7-18.0Å on treatment with ethylene glycol. The scale from NF-9 has a short (001) peak of 10.3Å, probably resulting from its dehydration. A comparison of the shapes of (001) peak between samples suggests that the smectite scale from the production well is highly crystalline, whereas those from the two-phase flow line and re-injection well are poorly crystalline.

Chemical Analysis

Chemical analyses of the smectite scale associated with tetrahedrite from 1684-88m depth of ND-7 were performed by



- Figure 5. X-ray diffraction patterns of smectite scales from the Mori geothermal power plant.
 - U.T.,untreated; E.G., ethylene glycol treated; CA,calcite; QZ,quartz.
- A. Smectite scale from 594m depth of the production well NF-9 (sampling date 1986.12).
- B. Smectite scale from 1235-1244m depths of the re-injection well NF-6 (sampling date 1987.4).
 - C. Smectite scale from the two-phase flow line (300line) (sampling date 1986.5).

 Table 2.
 Chemical compositions of smectite scale from the Mori geothermal power plant and stevensite from the two other localities.

I Smectite scale from 1684-1688m depths of the production well ND-7 of the Mori geothermal power plant (sampling date 1987.8).

 $(Ca_{0.02}K_{0.03})(Mg_{2.78}Mn_{0.14}Cu_{0.04}Fe^{2+}_{0.05})Si_{3.98}O_{10}(OH)_2.$ II Stevensite from the Whin Sill, Northumberland (Randall, 1959).

III Stevensite from the Ohori mine, Yarnagata (Otsu et al., 1963).

 $Ca_{0.08}(Mg_{2.98}Mn_{0.08}Fe^{2+}_{0.01}Fe^{3+}_{0.01})(Si_{3.92}Al_{0.01})O_{10}(OH)_2.$

| Sample No. | I | п | Ш | IV |
|--------------------------------|--------|--------|--------|---------------|
| | (wt.%) | (wt.%) | (wt.%) | (wt.%) |
| SiO ₂ | 54.12 | 52.14 | 54.5 | 53.86 |
| TiO2 | none | 0.01 | 0.02 | 0.01 |
| Al ₂ O ₃ | none | 1.34 | <0.1 | 0.01 |
| Fe ₂ O ₃ | - | 0.28 | 0.34 | 0.14 |
| FeO | 0.85 | • | 0.45 | 0.14 |
| MnO | 2.19 | 3.18 | 0.41 | 1.35 |
| CuO | 0.79 | • | - | • |
| MgO | 25.40 | 25.14 | 26.8 | 27.52 |
| CaO | 0.18 | 1.44 | 0.6 | 0.96 |
| Na2O | none | 1.19 | 0.04 | 0.09 |
| K₂O | 0.32 | 0.51 | 0.09 | 0.10 |
| P_2O_5 | - | 0.03 | 0.01 | • |
| H ₂ O(-) | | 8.91 | 6.6 | 7.60 |
| <u>H₂O(+)</u> | | 5.85 | 9.9 | 8.14 |
| Total | 83.85 | 100.02 | 99.9 | <u>99.9</u> 2 |

Hitachi electron microscope (X-650S) located at Institute for Advanced Material Processing, Tohoku University. The standards employed were: Mn and Cu metal (Mn, Cu), hematite (Fe), pyroxene (Mg, Si, Ca) and synthetic glass (K). Correction factors for analysis are those of Bence and Albee (1968). The chemical compositions of smectite from the Mori field and stevensite from the Northumberland, Whin Sill (Randall, 1959) and Ohori mine (Otsu et al., 1963) are given in Table 2. The smectite from the Mori field is composed mainly of SiO₂ and MgO, and does not contain Al₂O₃. The composition of the smectite is in good agreement with stevensite from the two other localities. Its structural formula on the basis of anion unit of $O_{10}(OH)_2$ is computed in the following form: $(Ca_{0.02}K_{0.03})(Mg_{2.78}Mn_{0.14}Cu_{0.04}Fe^{2+}_{0.05})Si_{3.98}O_{10}(OH)_2$. The formula suggests that it is a trioctahedral magnesian smectite, which doesn't contain aluminum in either the octahedral or tetrahedral sheet, and also has a total octahedral occupancy of 3.01.

Differential Thermal Analysis

TG and DTA curves of two smectite scale samples were simultaneously recorded with Rigaku Thermoflex. Instrument settings in the runs for the samples from the re-injection well NF-6 and injection water flow line (F-D line) were as follows; 200 and 10 mg on full scale in TG sensitivity, ± 100 and 50μ V



Figure 6. TG and DTA curves of smectite scales from the Mori geothermal power plant.
A. Smectite scale from the re-injection well NF-6 (sampling date 1987.7).
B. Smectite scale from the injection water line (D-F line).

on full scale in DTA sensitivity; 3 and 2.5 mm per minute in chart speed, respectively, and 10°C per minute in heating rate; Pt/Pt-Rh (13%) in thermocouple; α -Al₂O₃ in reference material; air in atmosphere, commonly.

The results are shown in Figure 6. The TG curve of the smectite scale from NF-6 shows three step weight losses as follows: (1)The first step weight loss of 14% (room temperature to 180°C) results from expulsion of the majority of interlayer water (2)The second step weight loss of 6% (180-760°C) results from elimination of the remaining interlayer water, and (3)The third step weight loss of 3% (760-790°C) results from dehydroxylation of constitution water (OH-group). The TG curve of the scale from the injection water flow line is similar to that from NF-6 except for a small variation of the third step weight loss. The DTA curve of the scale from NF-6 shows three important endothermic peaks at about 140°C, 740°C and 780°C due to dehydration and dehydroxylation, and a small endothermic peak about 470°C. The curve is very closely related to those of stevensite from Springfield, New Jersey (Faust and Murata,

1953) and the North Tyne (Randall, 1959). After heating to1000°C, enstatite is formed.

Hydrothermal Stability of Stevensite Scale

X-ray diffraction data reveals that the smectite scale from the Mori geothermal power plant is a member of trioctahedral group. The structural formula of the scale from ND-7 suggests that it should be regarded as stevensite based on the definition given by Faust and Murata (1953). According to Faust and Murata (1953) and Faust et al. (1959), stevensite has some deficiency of layer charge due to the defect of cations in the octahedral sheets. However, the stevensite scale from the Mori geothermal power plant has no-deficiency of the charge because its structural formula indicates a total octahedral occupancy of 3.01.

Large amounts of bentonite have been commonly introduced into the geothermal reservoir during mud drilling. The bentonite in the mud is composed of Na-montmorillonite or sepiolite. According to Güven and Carney (1979), sepiolite was easily converted to stevensite above 204°C in the system sepiolite- H_2O -(NaCl). Therefore, if sepiolite was used for the drilling mud, an important possible mechanism for formation of the stevensite is the conversion from the mineral. Unfortu-

 Table 3. Temperature, pressure and phase condition of fluid at the stevensite scale zone in the production wells.

| Well | Stevensite scale zone (m) | Temperature (°C) | Pressure (kg/cm ² G) | Flow pattern | Measuring date |
|-------|------------------------------|---------------------|------------------------------------|--------------|----------------|
| ND-1 | <260 | <214 | 15-46 | two phase | 1984.4 |
| ND-1 | <400 | <218 | | two phase | 1984.9 |
| ND-5 | <390 | 231 | 40 | two phase | 1984.3 |
| ND-7 | <1664 | 240 | 130 | two phase | 1987.8 |
| NF- 9 | 594 -596 | 205 | 47 | two phase | 1986.12 |

Table 4. Chemical compositions of hot waters and reservoir fluids
from the production wells in the Mori geothermal field.
Original data for hot waters discharged from production
wells are from Yoshida (1991). Chemical composition of
the reservoir fluid was calculated using chemical
compositions of hot waters and quartz geothermometer
with adiabatic cooling (Truesdell, 1975).

| Component | Hot water | Reservoir fluid |
|------------------------|------------|-----------------|
| sampling date | 1985.10 | |
| Na,mg/l | 2930-6410 | 2203-4516 |
| K,mg/l | 416-923 | 313-645 |
| Ca,mg/l | 26-116 | 18-90 |
| Mg,mg/l | 0.12-2.40 | 0.09-1.86 |
| Al,mg/l | 0.06-0.19 | 0.05-0.13 |
| Fe,mg/l | 0.37-0.70 | 0.26-0.54 |
| SiO ₂ ,mg/l | 387-704 | 305-492 |
| Cl,mg/l | 4300-10100 | 3234-7171 |
| SO ₄ ,mg/l | 255-393 | 178-307 |
| CO ₂ ,mg/l | 116-343 | 2149-10992 |
| H ₂ S,mg/l | 0.1-0.4 | 9-104 |
| pН | 7.78-8.49 | |

nately, not sepiolite but Na-montmorillonite has been used for the mud at the Mori geothermal field.

The stevensite scale occurs at depth shallower than the flash point in each production well, suggesting that the scale was formed from the SiO₂ and MgO rich hydrothermal liquid, which was condensed by separation due to boiling of the reservoir fluid. Mg in the stevensite scale appears to be derived from dolomite and magnesite in the Kamiiso Group. Based on the temperature and pressure dynamic logging data at the stevensite scale zone in the production well (Table 3), the stevensite scale occurs in the two phase fluid low zone, where has temperatures of ≤240°C and pressures of ≤130kg/cm²G. Stevensite also occurs in the various kinds of the surface instruments and the re-injection wells. These reveal that the stevensite scale has been formed at temperature range of 120 to 240°C. The chemical compositions of hot waters and reservoir fluids from the production wells are shown in Table 4. Mg contents in the hot waters and the reservoir fluids range from 4300-10100 and 3234-7171mg/l, respectively. Unfortunately, it is difficult to discuss the fluidstevensite equilibria in this report because there is no thermodynamic data for stevensite. However, Kasai et al. (1997) found that the stevensite scale in the pipelines has been prevented by decreasing pH in the fluid on the pilot plant experiment. So, the stevensite scale problem will be solved in the near future.

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* in Japanese

**in Japanese with English abstract