"HYDRATED TALC" — AN ALTERATION PRODUCT OF WOLLASTONITE BY REACTION WITH MAGNESIUM-BEARING HYDROTHERMAL SOLUTION

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ABSTRACT

A clayey material consisting predominantly of the trioctahedral 2:1 phyllosilicate which is similar to talc rather than stevensite, with a basal spacing of 9.7 Å, was found in hydrothermal-alteration products of wollastonite-rich skarn at the Akatani copper-iron mine, Niigata Prefecture, Japan, by one of the present authors (N. I.). The material occurs as a small pocket in the wollastonite-rich skarn, and is pseudomorphous after wollastonite. Field relations indicated that the small pocket is located in an intermediate subzone between inner talc and outer stevensite zones.

Chemical analysis of the clay fraction less than 2μ leads to the following structural formula for the mineral in question per unit-cell-layer on the basis of O_20(OH)_4 anion unit:

(Mg_{5.81}Mn^{2+}_{0.40}Fe^{3+}_{0.06})(Si_{7.73}Al_{0.15})O_20(OH)_4(Na_{0.02}K_{0.02})\cdot nH_2O.

X-ray diffraction, thermal and infrared absorption analyses revealed that the mineral in question is a variety less hydrated than stevensite, and may be named as "hydrated talc".

Based upon the field relations, information on the formation temperatures of minerals from the mine obtained by means of fluid-inclusion thermometry, and the synthetic data for the system MgO-SiO₂-H₂O, the mineral in question was identified as an alteration product of the pre-existing wollastonite at temperatures of 250°~300°C by the action of magnesium-bearing hot water, which journeyed through a channel-way immediately before ore-formation and simultaneously dolomitized limestone wallrock.

INTRODUCTION

During the course of an intensive study on the hydrothermal-alteration products of wollastonite-rich skarn at the Akatani copper-iron mine, Niigata Prefecture, it was confirmed that the wollastonite has been converted into

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talc (9.4 Å) near the channel-way along which magnesium-bearing hot water ascended. In the alteration halo, not only stevensite (15.4 Å) associated closely with “disordered talc” (9.5 Å) and a serpentine mineral but also a talc-like mineral with a basal spacing of 9.7 Å have been found by the first author and others (Imai, 1960; Imai, 1964; Imai and Otsuka, 1965; Imai, 1966; Imai and Yamazaki, 1967; Imai et al., 1970). The latter talc-like mineral seems to resemble closely “stevensite” from the North Tyne district, England, described by Randall (1959). In a previous paper concerning the stevensite from the Akatani mine, Imai et al. (1970) discussed the nomenclature of this talc-like mineral, and claimed that the mineral is not stevensite in a strict sense, but “hydrated talc”.

“Hydrated talc” and nickeliferous talc were already synthesized from gels with appropriate compositions by Caillere et al. (1963). Hydrothermal syntheses of the minerals in the system MgO-SiO₂-H₂O from various starting materials as well as hydrothermal treatments of natural stevensites at temperatures up to 500°C and under water-vapour pressure of 1000 kg/cm² have recently been carried out by the present authors and others. These studies revealed that there exists “hydrated talc” between stevensite and talc in their P-T stability fields (Otsuka et al., 1969; 1970; 1972).

Available information on “hydrated talc” in nature is fairly scarce. However, Makisimovic (1966) showed in his study on the clay minerals of β-kerolite-pimelite series that these minerals form a series of solid solution, where β-kerolite stands as a magnesian end member and pimelite as a nickeliferous end member. He also suggested that β-kerolite and pimelite represent hydrated and nickeliferous forms of talc, respectively. Recently, Brindley and Hang (1972) considered in their study on the so-called garnierite that the above-mentioned nickeliferous minerals have a mica-type structure with an interlayer water sheet, instead of interlayer alkali cations, and they called these minerals “talc monohydrate”.

The principal objective of the work reported here is to offer the mineralogical data on “hydrated talc.” The genesis of “hydrated talc”, based upon its occurrence in the field, its formation temperature as revealed by fluid inclusion thermometry, and synthetic data for the system MgO-SiO₂-H₂O, is also discussed briefly. The manuscript of this paper was prepared by the first author (N. I.).

MATERIAL INVESTIGATED

The material in question was found from wollastonite-rich skarn developed in the South-East Tunnel on the Bawari-zawa 410 m adit level of the Akatani mine. It occurs as a small pocket attaining to 70 cm in a longer dimension at a marginal portion of the wollastonite-rich skarn which is in direct contact with crystalline limestone on the northeast adit-wall of the tunnel (Fig. 1).

The geologic feature in the vicinity of ore deposits on the adit level, especially with respect to the hydrothermal alteration of the wollastonite-
rich skarn, was already described in a previous paper (Imai et al., 1970).
From a viewpoint of zonal distribution of the altered minerals in the alteration halo, the material in question occurs in an intermediate subzone between inner talc and outer stevensite zones, although the above-mentioned spatial relationship becomes obscure in some places on account of a sporadic distribution of the skarn.

A hand specimen containing the material in question is pale brown in colour, and consists largely of fibrous or radiating aggregates of a talc-like mineral with soft feel. In thin section, the material is seen to consist almost entirely of this talc-like mineral with small amounts of diopside and wollastonite relics and new calcite; namely, wollastonite is replaced by the talc-like mineral, that is pseudomorphous after wollastonite. It is worthy to note that the material contains dusty inclusions of extremely fine grain-size and assumes a brownish-red colour in transmitted light (Fig. 2).

The material was easily crumbled and was readily dispersed in distilled water.

CHEMICAL ANALYSIS

The sample used for chemical analysis was a clay fraction less than 2μ, separated from the specimen (No. A6414A) by the combined methods of sedimentation and centrifugation in distilled water. The sample thus prepared was examined under the polarizing and electron microscopes, and with an aid of the electron-probe X-ray microanalyzer. As a result, it was confirmed that the sample contains minute inclusions which were identified as manganese oxide. Because of their extremely fine grain-size, it was impossible to obtain a pure sample free from impurities. Therefore, the sample
contaminated to some extent with the impurities was analyzed chemically using a normal wet method by the third author (T. N.).

The result of chemical analysis is listed in Table 1, together with that of the unaltered wollastonite-rich skarn, and with those of the talc, "stevensite" and β-kerolite in previous papers for comparison. No deduction of manganese oxide in the impurities from the analysis was made, because it was difficult to estimate their exact amount. Calculation on the basis of an anion unit of \( O_{20}(OH) \), in the 2:1 layer structure leads to the following structural formula per unit-cell-layer:

\[
(Mg_{5.81}Mn_{0.40}Fe_{0.06})_{(Si_{7.73}Al_{0.15})}O_{20}(OH)_{4}(Na_{0.02}K_{0.02})\cdot n \text{H}_2\text{O}.
\]

The formula indicates that there are deficiency and excess in number of cations per formula unit in the tetrahedral and octahedral sites, respectively.

**X-RAY DIFFRACTION ANALYSIS**

The X-ray diffraction patterns of the crude sample (specimen No. A6414A) and its clay fraction less than 2μ both untreated and treated, have been recorded with a "Geigerflex" X-ray diffractometer, using Ni-filtered CuKα-radiation. On the X-ray pattern of the crude sample, the reflections from the talc-like mineral in question appeared together with those from diopside, calcite and wollastonite. The X-ray patterns of the clay fraction are reproduced in Fig. 3, and the powder data for the untreated sample are tabulated in Table 2, together with those for the well-crystallized Manchurian talc.

As is well known, interlayer binding forces in talc are so weak that the mechanical process has a possibility to produce disorder in its structure,
TABLE 1. Chemical analyses of "hydrated talc," unaltered wollastonite-rich skarn, "stevensite" and β-kerolite

<table>
<thead>
<tr>
<th></th>
<th>(1)</th>
<th>(2)</th>
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<th>(4)</th>
<th>(5)</th>
<th>(6)</th>
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<tr>
<td>SiO₂</td>
<td>57.01</td>
<td>52.89</td>
<td>54.5</td>
<td>53.86</td>
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<td>0.00</td>
<td>0.02</td>
<td>0.01</td>
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<tr>
<td>Al₂O₃</td>
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<td>0.68</td>
<td>&gt; 0.1</td>
<td>0.01</td>
<td>0.26</td>
<td>0.15</td>
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<td>Fe₂O₃</td>
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<td>0.52</td>
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<td>0.14</td>
<td>0.10</td>
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<td>FeO</td>
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<td>0.24</td>
<td>0.45</td>
<td>0.14</td>
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<td>MnO</td>
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<td>0.19</td>
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<td>27.52</td>
<td>18.43</td>
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<td>CaO</td>
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<td>K₂O</td>
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<td>P₂O₅</td>
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<td>n.d.</td>
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<td>n.d.</td>
<td>n.d.</td>
<td>n.d.</td>
<td>n.d.</td>
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<td>H₂O (+)</td>
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<td>0.80</td>
<td>9.9</td>
<td>8.14</td>
<td>6.49</td>
<td>4.80</td>
<td>4.75</td>
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<td>H₂O (−)</td>
<td>2.27</td>
<td>0.19</td>
<td>6.6</td>
<td>7.60</td>
<td>8.41</td>
<td>n.d.</td>
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<tr>
<td>Total</td>
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<td>99.66</td>
<td>99.9</td>
<td>99.92</td>
<td>99.90</td>
<td>100.00</td>
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</table>

(1) "Hydrated talc", the mineral in question (specimen No. A6414A), Akatani mine; (2) Unaltered wollastonite-rich skarn (specimen No. A6305), Akatani mine; (3) "Stevensite", North Tyne, England (Randall, 1959); (4) "Stevensite II", Ohori mine (Otsu et al., 1963); (5) β-kerolite, Goles Mountain, Yugoslavia (Maksimovic, 1966); (6) Talc (Pask and Warner, 1954); (7) Calculated from ideal talc of chemical composition Mg₃Si₄O₁₂(OH)₂. (1) and (2), analyst: T. Nakamura.

Fig. 3. X-ray powder diffraction patterns of "hydrated talc", the mineral in question, specimen No. A6414A.
1 Treated with H₂O, 2 Treated with ethylene glycol, 3 Untreated (room temperature), 4 Heated to 225°C, 5 Heated to 700°C.
TABLE 2. X-ray powder diffraction data for "hydrated talc" and talc

<table>
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<th>hkl*</th>
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<td></td>
<td>d (Å)</td>
<td>I</td>
<td>d (Å)</td>
<td>I</td>
</tr>
<tr>
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<td>9.7</td>
<td>100</td>
<td>9.3</td>
<td>100</td>
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<td>004</td>
<td>4.73</td>
<td>20</td>
<td>4.68</td>
<td>85</td>
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<td>020</td>
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<td>202</td>
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<td>132, 204</td>
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<td>2.109</td>
<td>5</td>
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<td>0.0.10</td>
<td>1.888</td>
<td>10 (br)</td>
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<td>5</td>
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<td>060, 352</td>
<td>1.526</td>
<td>15</td>
<td>1.527</td>
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<td>330</td>
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<td>2</td>
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<td>1.3.10</td>
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<td>2 (br)</td>
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<td>2.0,10, 1.3,12</td>
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<td>5</td>
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<td>0.0.14</td>
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<td>0.0.16</td>
<td></td>
<td></td>
<td>1.168</td>
<td>5</td>
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</table>

* Indices on the basis of the two-layer monoclinic cell with a=5.287 Å, b=9.158 Å, c=18.95 Å, β=99°30' given by Stemple and Brindley (1960).

(1) "Hydrated talc," the mineral in question (specimen No. A6414A), Akatani mine; (2) Well-crystallized Manchurian talc (specimen No. 723).

especially that due to b-axis displacements, and grinding in dry state for a long time changes it into an amorphous substance (Takahashi, 1959). Therefore in the present X-ray study, pulverizing the samples in an agate mortar for a long time was avoided.

As seen from curve 3 in Fig. 3, the X-ray diffraction pattern of the talclike mineral untreated is not identical with those of stevensite, hectorite, β-korolite and pimelite, which are characterized by few hk bands in addition to 00l reflections (Faust and Murata, 1953; Brindley, 1955; Faust et al., 1959; DeRudder and Beck, 1963; Otsu et al., 1963; Maksimovic, 1966; Imai et al., 1970), but shows a similarity to those of talcs, especially to that of dark green massive talc from Chester County, Pennsylvania (Stemple and Brindley, 1960). Indexing of the resolved peaks in Table 2 was made on the basis of a two-layer monoclinic cell, and it is noticed that all observed reflections have l indices which are even, indicating that there is no necessity
"Hydrated Talc"

for assuming the two-layer cell.

In comparison with the X-ray pattern of well-crystallized talc, however, the talc-like mineral in question shows the following differences: 1) The 002 reflection appears at a d-spacing of 9.7 Å. This shifts considerably towards the low-2θ side from that of talc, which usually appears at 9.3~9.4 Å. Also, the basal reflections are broadened and do not form a rational series. 2) Including the basal reflections, only few reflections appear. 3) The 020 reflection appears as a diffuse band with a “tail” extending to the high-2θ side, showing a two-dimensional reflection effect. 4) The 132 and 132 reflections are not well resolved and spread, the 004 reflection is weaker and the 0.0.10, 0.0.12 and 0.0.14 reflections, which usually appear as relatively strong peaks on the X-ray patterns of talcs, are scarcely seen.

The fact that almost all the resolved peaks correspond to the \(k=3n\) reflections, as shown in Table 2 (1), is in accordance with random or nearly random layer displacements of \(nb/3\). The 060 reflection appears at 1.526 Å, which indicates the trioctahedral nature of the mineral in question. The treatments of the mineral with ethylene glycol and water do not give any noticeable change in the X-ray pattern, as shown in Fig. 2. After heating to 225°C, the 9.7 Å reflection shifts to 9.6 Å and subsequently to 9.5 Å when heated to 700°C.

It is well recognized that crystalline materials consisting of small particles which are equal in their size to “coherent” small domains, give anomalous X-ray diffraction effects such as broadening and shift of reflections; and these phenomena were well explained theoretically by Ross (1968). In order to ascertain the cause of non-rational series of the basal reflections of the talc-like mineral in question, Nakajima performed a Fourier analysis of its X-ray diffraction profiles using Warren and Averbach's method (1950, 1952). From this analysis he concluded that the non-rational series may mainly be due to the particle-size effect and that the spacings of 00l reflections are “apparent” (Nakajima, 1970).

**THERMAL ANALYSIS**

The thermal analysis of the material in question has been performed in the following two different ways; 1) simultaneous recording of the thermogravimetric (TG), derivative thermogravimetric (DTG), differential thermal (DTA) analyses and temperature (T) curves with a “Simultaneous Thermobalance Analyzer”, and 2) quasi-static thermogravimetry with an ordinary thermobalance by a stepwise method.

Simultaneous TG, DTG, DTA and T curves of the clay fraction separated from the crude sample (specimen No. A6414A; sample weight 397.9 mg) are shown in Fig. 4. The TG and DTG curves show three steps of weight loss, indicating that dehydration of the talc-like mineral in question proceeds in three steps, provided that the weight loss takes place only upon dehydration. The first step of dehydration due to expulsion of the majority of interlayer water is abrupt, starting at about 60°C and terminating at 150°C.
The maximum rate of weight loss reaches to 0.75 mg (0.19 per cent) per minute at 125°C. The second step of weight loss due to elimination of the remainders of interlayer water is very gradual, ranging from 150° to 780°C, as indicated by the gentle slope of the TG curve. The rate of weight loss is nearly constant, being 0.15 mg (0.03 per cent) per minute throughout this temperature range. The third step of the weight loss due to release of constitutional water (OH-group) starts at 780°C and terminates at 860°C.

The DTA curve shows a small and broad endotherm at about 125°C and a large endotherm at 825°C, and is very similar in shape to the DTG curve. The DTA curve of the talc-like mineral is comparable with those of stevensite, β-kerolite and pimelite.

The quasi-static thermogravimetry has been performed for the mineral in question and the well-crystallized Manchurian talc, using a thermobalance made by Moriya Co., Japan. It was equipped with a temperature regulator made by Chino Co., Japan, which controls the temperature within an error of ±1°C. In this procedure, the sample was heated at a constant temperature until an equilibrium was reached, and then raised by every 5°C.

The quasi-static TG curves thus obtained are shown in Fig. 5. Curve 1 for the present mineral may be divided into three parts; namely, low-, intermediate- and high-temperature ranges. The weight loss and the corresponding number of water molecules released in the respective temperature ranges are shown in Table 3. Curve 2 for the well-crystallized Manchurian talc indicates that dehydroxylation proceeds in a narrow temperature range from 730° to 850°C.

TABLE 3. Results of quasi-static thermogravimetry for "hydrated talc" and talc

<table>
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<tr>
<th>Stage of dehydration</th>
<th>Temperature range</th>
<th>&quot;Hydrated talc&quot;</th>
<th>Talc</th>
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</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Weight loss (%)</td>
<td>Water released (%)</td>
</tr>
<tr>
<td>1</td>
<td>~ 120 (°C)</td>
<td>1.87</td>
<td>0.81</td>
</tr>
<tr>
<td>2</td>
<td>120-700</td>
<td>1.50</td>
<td>0.65</td>
</tr>
<tr>
<td>3</td>
<td>700-1000</td>
<td>4.21</td>
<td>1.83</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td>7.58</td>
<td>3.29</td>
</tr>
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</table>

(1) The mineral in question, Akatani mine (specimen No. A6414A); (2) Well-crystallized Manchurian talc (specimen No. 723); (3) Number of water molecules released per O$_6$(OH)$_4$ unit.

From the results noted above as well as from the simultaneous TG, DTG and DTA curves in Fig. 4, it may be noticed that there is a difference in the thermal behaviour between the present talc-like mineral and talc; the present mineral closely resembles stevensite (Otsu et al., 1963; Otsu and Yasuda, 1964; Imai et al., 1970). Especially, it is worthy of note that the temperature at which the dehydroxylation initiates is lower for the mineral in question than the well-crystallized Manchurian talc.

INFRARED ABSORPTION ANALYSIS

The infrared absorption spectra of the mineral in question, both untreated and heated at various temperatures, together with the well-crystallized Manchurian talc, have been obtained with a Hitachi infrared spectrometer of grating type (EPI-G2). One to three mg of the powder of each material was mixed with 300 mg KBr and pressed by a hydraulic press in vacuum;
the disk thus prepared was examined in the region from 400 to 1300 cm\(^{-1}\). The spectra in the OH-stretching region from 3000 to 4000 cm\(^{-1}\) and in the H-O-H deformation region of 1500-1800 cm\(^{-1}\) were obtained by a paste method using Nujol as the base in order to avoid the adsorption of water.

As shown in Fig. 6, the spectrum in the OH stretching region of the unheated mineral in question gives a strong and sharp absorption band at 3675 cm\(^{-1}\) with a shoulder at 3660 cm\(^{-1}\), and weak and broad bands at 3620 and 3440 cm\(^{-1}\). By heating to 225°C, the spectrum did not show any noticeable change; but when heated to 700°C, the band at 3675 cm\(^{-1}\) shifed to 3670 cm\(^{-1}\) and the bands at 3660, 3620 and 3440 cm\(^{-1}\) almost disappeared. In the 1500-1800 cm\(^{-1}\) region, an absorption band caused by the H\(_2\)O deformation vibration appears at 1630 cm\(^{-1}\) as a single band. It decreased in intensity and shifted to 1625 cm\(^{-1}\) when heated to 225°C. After heating to 700°C, this absorption band almost disappeared.

As shown in Fig. 7, the Si-O stretching absorption appears around 1005 cm\(^{-1}\) as a strong and very broad band. In the 400-800 cm\(^{-1}\) region, there is a strong doublet absorption band at 450 and 465 cm\(^{-1}\) with shoulders at 534 and 450 cm\(^{-1}\). This doublet is characteristic to talc, and was assigned to a Mg-O vibration by Farmer (1958) and later to a Si-O deformation vibration by Wilkins and Ito (1967). In addition, there are a strong and sharp band at 670 cm\(^{-1}\) with a shoulder at 690 cm\(^{-1}\) and a weak band at 790 cm\(^{-1}\).

As outlined above, the infrared absorption spectra of the mineral in question do not show any noticeable difference from those of the well-crystallized Manchurian talc examined here as well as natural and synthetic talcs studied.
Hydrated Talc

FIG. 7. Infrared absorption spectra in the region of 1300-400 cm$^{-1}$.
1 "Hydrated talc", the mineral in question (specimen No. A6414A),
2 Well-crystallized Manchurian talc (specimen No. 723), both at room temperature.

previously except for the appearance of the band due to the H$_2$O deformation vibration present in the interlayer space.

ELECTRON MICROSCOPY

The electron micrographs of the mineral in question indicated that individual crystals are remarkably variable in their particle-size; some particles are 1-5 $\mu$m in diameters, but others are extremely fine-grained, usually less than 0.1 $\mu$m. Also, they exhibit a variable morphology. Many appear as fluffy masses with ill-defined outlines, and some as long, lath-like fibers, and others as plates with well-defined outlines. A representative electron micrograph is shown in Fig. 8.

DISCUSSION AND CONCLUSIONS

Summarizing the data outlined so far, it is noted that the trioctahedral 9.7 Å layer mineral in question has mineralogical properties resembling those of stevensite in some respects and talc in other respects.

The mineral in question is very similar to stevensite in its thermal behaviours, particularly to "stevensite" from North Tyne described by Randall (1959), and to $\beta$-kerolite and pimelite by Maksimovic (1966). Randall did not describe the result of the swelling test for the North Tyne material. The mineral in question was not swollen after the treatments with ethylene glycol and water, and the same was reported for "stevensite II" from the Ohori mine (Otsu et al., 1963); and also for $\beta$-kerolite and pimelite (Maksimovic, 1966).

As pointed out by previous workers, stevensite dissolves readily in hydrochloric acid, but talc does not. Maksimovic (1966) stated that $\beta$-kerolite and
pimelite easily decompose when treated with hydrochloric acid, so they resemble stevensite in this respect. However, the mineral in question did not dissolve in hydrochloric acid, and this was the case with talc.

The X-ray diffraction patterns of “stevensite II”, β-kerolite and pimelite are characterized by few \( hkl \) bands in addition to basal reflections. These X-ray patterns are similar to those of stevensite and hectorite belonging to the montmorillonite group, except for the difference in their basal spacings (Otsu et al., 1963; Maksimovic, 1966). The X-ray patterns of the mineral in question as well as of the North Tyne material are not similar to those of the montmorillonite minerals, but closely related to those of talcs, and are similar to those of stevensites heated to 225°C (Otsu et al., 1963; Imai et al., 1970). The 9.7 Å reflection of the latter is, however, replaced by a weak and diffuse peak ranging from 11 to 12 Å in the mineral in question, when it is rehydrated (Otsu and Yasuda, 1964).

The 9.7-10 Å spacings of synthetic hydrated talc, β-kerolite and pimelite contract considerably, when treated with ethylene glycol (Maksimovic, 1966; Otsuka et al., 1972), but the 9.7 Å spacing of the mineral in question does not.

In their study on the thermal behaviours of interlayer water in stevensite, Otsu and Yasuda (1964) pointed out the appearance of various kinds of mixed-layer phases having both regular and irregular layer sequences during its dehydration process. In the case of the mineral in question, the broadening of its lower order 00\( I \) reflections, the virtual absence of 0.0.10, 0.0.12 and 0.0.14 reflections and the non-rational series of its basal reflections suggest the possibility of an irregular mixed-layer sequence. However, in the present
study, there is no other evidence to substantiate this irregular mixed-layering. The above feature of the basal reflections was also explained by the particle-size effect (Nakajima, 1970). Probably, the broadening of the lower order 00l reflections and the absence of the higher order ones may partly be due to a slight fluctuation in the basal spacings resulting from the b-axis disordering in addition to the particle-size effect.

There is a difference in the thermal behaviour between the mineral in question and normal talcs. The gradual release of interlayer water in the intermediate-temperature range from the former mineral may be due to a non-uniformity of each silicate layer, accommodation of water molecules in the interlayer space and the above-mentioned particle-size effect. To clarify these problems, however, further investigation seems desirable.

The infrared absorption spectrum of the mineral in question does not differ significantly from those of talc and stevensite, since they are essentially the same in the structure of layer lattice. However, the spectrum in the OH stretching region is different from those of stevensites (Faust et al., 1959; DeRudder and Beck, 1963; Inouma and Hayashi, 1966, 1968; Imai et al., 1970) and is substantially the same with those of natural and synthetic talcs (Farmer, 1958; Wilkins and Ito, 1967). Maksimovic (1966) stated that the infrared spectra of β-korolite and pimelite show a similarity to talc rather than to the minerals of the montmorillonite group.

It is suggested that the hydration of talc and the presence of alkali metal cations at interlayer sites are due to some structural defects in the layer lattice, but the chemical analysis data for the present material could not demonstrate this supposition. Presumably, water molecules and alkali cations are irregularly distributed at interlayer sites, due to the lack of uniformity in the structure of each silicate layer. In this respect, the 9.7 Å mineral in question from the Akatani mine seems different from the stevensite heated to about 200°C, which also has the 9.7 Å basal spacing. The latter stevensite has a mica-type structure with a single 2H2O-sheet, and probably corresponds to “talc monohydrate” (Brindley and Hang, 1972).

As to the nomenclature of stevensite, Imai et al. (1970) stated as follows; “If one maintains that the original stevensite is a mineral of montmorillonite group (Faust and Murata, 1953), the application of the term stevensite must be restricted to “stevensite I” of the swelling type, and the naturally occurring less hydrated varieties such as “stevensite II” from the Ohori mine (Otsu et al., 1963), the 9.8 Å North Tyne material (Randall, 1959) and the 9.7 Å Akatani material belonging to the non-swelling type must be called “hydrated form of talc” (Caillere et al., 1963; Maksimovic, 1966).”

Roy and Roy (1955) studied on the synthesis and stability relations of minerals in the system MgO−Al2O3−SiO2−H2O, and noted that under a water-vapour pressure of 690 bars, magnesium montmorillonite is stable below 250°C, but is converted into talc at 275°C. Subsequently, Mumpton and Roy (1956) reported the similar result. However, the present authors and others have shown in their recent investigation on the synthesis and the stability relations of the minerals in the system MgO−SiO2−H2O under a water-vapour
pressure of 1000 kg/cm² that the natural and synthetic stevensites are not directly transformed into talc, but gradually through “dehydrated stevensite” and “hydrated talc” with increasing temperatures. Here, “dehydrated stevensite” was designated for its origin from stevensite and “hydrated talc” for its swelling properties which are different from those of stevensite with the basal spacings of 10~11 Å.

The Akatani stevensite is stable below 290°C, transforms into talc with a basal spacing of 9.4 Å at 360°C, and in the temperature range from 290°C to 360°C, the above two phases co-exist under hydrothermal conditions (Otsuka et al., 1969, 1970, 1972). Also, natural wollastonite converts into hydrated talc by treatment with 2 N and 6 N MgCl₂ solutions at 250°C under a water-vapour pressure of 250 kg/cm² (Sakamoto et al., 1972).

Strictly speaking, the Akatani 9.7 Å mineral in question is not identical with the 9.7 Å mineral in the synthetic products or from the hydrothermally treated stevensite. Also, the water-vapour pressures given above are not compatible with field relations and geologic setting of the Akatani ore deposits, because the depth at the time of ore deposition is considered to have been very small less than 500 m (Imai and Tsutsumi, 1965), so that the synthetic data noted above cannot be used to estimate the temperature at which alteration took place.

Based upon the information concerning the formation temperatures of the minerals in the Akatani mine, as revealed by fluid-inclusion thermometry (Miyazawa and Nozaki, 1959; Enjoji, 1972; Imai, unpublished data), the center of hydrothermal dolomitization, where wollastonite alters to talc, is considered to be at about 300°C. Thus, it may be concluded that the 9.7 Å mineral in question forms as an alteration product of pre-existing wollastonite at 250°C~300°C, by the action of magnesium-bearing hot water that journeyed through the channel-way immediately before ore formation and simultaneously dolomitized limestone wallrock.

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REFERENCES


