MINERALOGICAL STUDIES ON DEWEYLITE AND POORLY-CRYSTALLIZED SERPENTINE FROM WAKAMIYA-CHO, FUKUOKA PREFECTURE

MASATOMO MUCHI, Mitsunori Ido and Yoshie Shibata

Abstract: The clayey or garnular specimens of deweylite and poorly-crystallized serpentine found from Wakamiya-cho, Fukuoka Prefecture, occurring as fine veins, have indicated the following chemical formulae respectively:

$$\text{3.6 MgO} \cdot 3 \text{SiO}_2 \cdot 5.6 \text{H}_2\text{O}..........(Y \ 43),$$

$$\text{3.9 MgO} \cdot 3 \text{SiO}_2 \cdot 5.7 \text{H}_2\text{O}..........(Y \ 50_b),$$

$$\text{3.7 MgO} \cdot 3 \text{SiO}_2 \cdot 5.3 \text{H}_2\text{O}..........(Y \ 51),$$

$$\text{2.9 MgO} \cdot 2 \text{SiO}_2 \cdot 1.9 \text{H}_2\text{O}..........(Y \ 40).$$

The chemical formulae of the first three specimens are close to the standard formula of deweylite and the last one to that of serpentine. These specimens have given the very similar X-ray powder patterns one another, but the slight differences have been recognized in intensities of the first three diffraction peaks. And no marked structural differences were indicated on the heat treatment between the two mineral species. On the differential thermal analysis curves, some features were recognized: deweylite showed a large, dual endothermic reaction with a remarkable peak at 100 to 200°C and being immediately followed by a small shoulder at about 250 to 300°C, and it showed no plain endothermic reaction at about 700 to 800°C, however, poorly crystallized serpentine did a sharp reaction in the same temperature region. The particle shapes of these minerals are also close, showing generally rounded shapes with polygonal or irregular outlines. Deweylite would readily appear to be fluffy or take rounded forms with fine projections, however, poorly-crystallized serpentine would rather take angular forms.

INTRODUCTION

Deweylite is generally found in small fissures in serpentine rocks, as a white, gray, brownish or yellowish earthy mass with a gum-like appearance as described by Dana (1915), Selfridge (1936) and Sudo and Minato (1949). This mineral is closely related to serpentine minerals chemically and structural-
Deweylite and poorly-crystallized serpentine

The chemical formula of it is considered to be $4 \text{MgO} \cdot 3 \text{SiO}_2 \cdot 6 \text{H}_2\text{O}$ and that of serpentine mineral is $3\text{MgO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$. Selfridge (1936) described that the powder lines of deweylite have much the same interplanar spacings as serpentine minerals and he pointed out that the variation of the intensities of the first three lines in X-ray patterns can differentiate deweylite from serpentine mineral species. In the case that serpentine minerals always have high degrees of crystallization, the differentiation will be probable. But if serpentine minerals are low in degrees of crystallization, it may be difficult.

The name serpentine is commonly applied to two varieties (platy and fibrous one) through the discussions by Nagy and Faust (1956), Fleischer (1957) and Zussman, Brindley and Comer (1957). The analogy between serpentine and kaolin groups is remarkable morphologically and also structurally. The morphological habits of these minerals, tubular or platy, are much related to the crystal chemistry as pointed out by Kalouseck and Muttart (1957) and Bates and Comer (1959). This study, in a part, was undertaken to compare the particle shapes of deweylite and poorly-crystallized serpentine with those of hydrated-halloysites with low crystallinity.

MATERIALS USED FOR THIS STUDY

The specimens used for this study occur as fine veins or lenticular forms in serpentine or peridotite bodies widely developed near Rikimaru Dam, Wakamiya-cho, Fukuoka Prefecture, where is a well known locality for serpentine minerals, being shown in Fig. 1. These minerals are generally white, gray, light yellow or light brown and appear to be granular or clayey in hand specimens, and often associate with leuchtenbergite. These specimens display the appearance similar to one another, so it is very difficult to differentiate macroscopically deweylite from such a poorly-crystallized serpentine.

Deweylite and serpentine minerals in the specimens have been identified particularly by chemical analysis and X-ray powder diffraction patterns in addition to differential thermal analysis curves and electron micrographs.

CHEMICAL ANALYSIS

The chemical analyses of the specimens were done by K. Nuruyu, Kyushu University, and are listed in Table 1 together with those of deweylites found in the literature. The data of the specimens (Y 43, Y 50b, Y 51 and Y 40) show that all of them are composed of hydrous magnesium silicates, but the reasonably distinct differences have been recognized among the data. Namely, the magnesium content is considerably lower in Y 43, Y 50b and Y 51 than in Y 40, and also the iron content is slightly lower in the first three specimens than in the fourth. The difference of the adsorbed water content is more noticeable.
Fig. 1 (A) Showing fine veins of poorly-crystallized serpentine near Riki-
maru Dam, Fukuoka Prefecture, (Y 40)

(B) Fine veinlets of deweylite in serpentine rock near Rikimaru
Dam, Fukuoka Prefecture, showing somewhat network appear-
ance.

Table 1 Chemical analyses of deweylites and poorly-crystallized
serpentine

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>42.22</td>
<td>42.60</td>
<td>40.16</td>
<td>41.05</td>
<td>41.1</td>
<td>40.16</td>
</tr>
<tr>
<td>TiO₂</td>
<td>—</td>
<td>—</td>
<td>tr.</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>1.12</td>
<td>0.15</td>
<td>1.15</td>
<td>2.41</td>
<td>2.9</td>
<td>tr.</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>1.67</td>
<td>0.39</td>
<td>1.25</td>
<td>1.74</td>
<td>tr.</td>
<td>1.16</td>
</tr>
<tr>
<td>FeO</td>
<td>2.18</td>
<td>0.17</td>
<td>0.21</td>
<td>0.16</td>
<td>3.2</td>
<td>—</td>
</tr>
<tr>
<td>MnO</td>
<td>0.04</td>
<td>—</td>
<td>tr.</td>
<td>0.03</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>MgO</td>
<td>40.80</td>
<td>34.00</td>
<td>35.05</td>
<td>32.68</td>
<td>30.6</td>
<td>36.00</td>
</tr>
<tr>
<td>CaO</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>1.2</td>
<td>0.80</td>
</tr>
<tr>
<td>Na₂O</td>
<td>tr.</td>
<td>tr.</td>
<td>tr.</td>
<td>tr.</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>K₂O</td>
<td>tr.</td>
<td>tr.</td>
<td>tr.</td>
<td>tr.</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>H₂O(⁺⁺)</td>
<td>12.26</td>
<td>12.33</td>
<td>12.21</td>
<td>12.28</td>
<td>15.7</td>
<td>21.60</td>
</tr>
<tr>
<td>H₂O(⁻⁻)</td>
<td>tr.</td>
<td>10.63</td>
<td>10.52</td>
<td>9.46</td>
<td>7.1</td>
<td>—</td>
</tr>
<tr>
<td>Total</td>
<td>100.29</td>
<td>100.27</td>
<td>100.55</td>
<td>99.81</td>
<td>101.8</td>
<td>99.72</td>
</tr>
</tbody>
</table>

1. Y 40 (Poorly-crystallized serpentine) from Wakamiya-
cho, Fukuoka Prefecture.
2. Y43 (Deweylite), from Wakamiya-cho.
3. Y50b (Deweylite), from Wakamiya-cho.
4. Y51 (Deweylie-like mineral), from Wakamiya-cho.
5. Deweylite from Yagiyama, Fukuoka Prefecture, desc-
6. Deweylite from Dana’s “System of Mineralogy”.
The specimens from Wakamiya-cho were analysed by K.
Nuruyu, Kyushu University.
Deweylite and poorly-crystallized serpentine

than that of magnesium content. The chemical formulae of Y 43, Y 50, and Y 51 are 3.6 MgO · 3 SiO₂ · 5.6 H₂O, 3.9 MgO · 3SiO₂ · 5.7 H₂O and 3.7 MgO · 3 SiO₂ · 5.3 H₂O, respectively, and these formulae correspond closely with 4 MgO · 3 SiO₂ · 6 H₂O, being the standard formula of deweylite. On the other hand, the formula of Y 40 is 2.9 MgO · 2 SiO₂ · 1.9 H₂O, which is entirely identical to 3 MgO · 2 SiO₂ · 2 H₂O, the formula of serpentine.

X-RAY POWDER DIFFRACTION PATTERNS

The X-ray powder diffraction patterns of natural specimens, prepared as purely as possible by the repetition of elutriation and ultra-centrifuge, were recorded with a Shimazu geiger counter X-ray diffractometer. Ni-filtered copper radiation (Cu Ka: 1.5418 Å) was used for every specimen under the following experimental conditions: 30 KV, 20 mA, time constant 2.5 seconds, scanning speed 2° per minute, chart speed 2 cm per minute, full scale 500 counts and slit system 1°-2°-0.8 mm. These patterns are shown in Fig. 2 and the observed intensities and spacings measured from the diffractometer charts are listed in Table 2.

The X-ray powder patterns of these specimens are closely similar to one another, and the diffraction peaks are few and very broad, giving the two dimensional features with long tails towards high angle side (2θ). From the

![X-ray Powder Diffraction Patterns](image)

**Fig. 2** X-ray powder diffraction patterns of the natural specimens
patterns, it will be estimated that these specimens are composed of the serpentine type minerals. Selfridge (1936) pointed out that, although the X-ray lines of deweylite indicated the same interplanar spacings as serpentine, the intensities of the first three lines were distinctly inverse in each powder pattern of deweylite and serpentine. And also he differentiated deweylite from serpentine according to the variation of the intensities of the three lines.

The diffraction peaks of Y43, Y50b and Y51, as compared with the X-ray data of deweylites described by Selfridge (1936) and Sudo and Minato (1949), are also considerably near to those of the deweylites in the spacings, intensities and sharpness. For Y40, on the other hand, the 7.5 and 3.6Å peaks are slightly lower than the 4.5Å peak in intensities. From this point, this specimen

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>d(Å)</td>
<td>1</td>
<td>d(Å)</td>
<td>I</td>
<td>d(Å)</td>
<td>I</td>
<td>d(Å)</td>
</tr>
<tr>
<td>7.4</td>
<td>6</td>
<td>7.5</td>
<td>6</td>
<td>7.4</td>
<td>5</td>
<td>7.5</td>
</tr>
<tr>
<td>4.59</td>
<td>10</td>
<td>4.59</td>
<td>10</td>
<td>4.58</td>
<td>10</td>
<td>4.55</td>
</tr>
<tr>
<td>3.71</td>
<td>8</td>
<td>3.67</td>
<td>7</td>
<td>3.68</td>
<td>6</td>
<td>3.65</td>
</tr>
<tr>
<td>2.61</td>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.56</td>
<td>8</td>
<td>2.57</td>
<td>8</td>
<td>2.55</td>
<td>8</td>
<td>2.55</td>
</tr>
<tr>
<td>2.50</td>
<td>9</td>
<td>2.50</td>
<td>9</td>
<td>2.50</td>
<td>8</td>
<td>2.49</td>
</tr>
<tr>
<td>2.48</td>
<td>8</td>
<td>2.48</td>
<td>8</td>
<td>2.48</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>1.540</td>
<td>8</td>
<td>1.537</td>
<td>8</td>
<td>1.535</td>
<td>7</td>
<td>1.537</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1: Deweylite from Wakamiya-cho, Fukuoka Prefecture (Y 43).
2: Deweylite from Wakamiya-cho, Fukuoka Prefecture (50b).
3: Deweylite from Wakamiya-cho, Fukuoka Prefecture (Y 51).
4: Poorly-crystallized serpentine from Wakamicho, Fukuoka Prefecture (Y 40).
may be considered in a way to be deweylite, however, it should be comprised in the serpentine group by a combination of chemical analysis and differential thermal curve.

**THERMAL CHANGES**

The transformation of serpentine minerals to forsterite by heating in air has been studied by many workers. Brindley and Zussman (1957) pointed out that the transition for highly-crystallized serpentines appears to be related to the structural and chemical characteristics of the initial material. This investigation was undertaken in part to check the differences of thermal behaviours on deweylite, poorly-crystallized serpentine and highly-crystallized one.

The specimens for this experiment were respectively heated in an electric furnace in air under the heating rate of about 10°C per minute and kept at successively higher temperatures of 500, 600, 700 and 800°C for one hour respectively. After quenching those specimens in a desicater, the X-ray powder patterns were obtained under the same experimental conditions described above. These patterns are shown in Fig. 3. All the patterns, in general, indicate almost similar appearance respectively. The other specimens from

![Fig. 3 (A) X-ray powder diffraction patterns of the specimens heated at 500°C for 1 hr.](image-url)
different localities, giving somewhat higher crystallinity, were used to compare with these ones on the effect of heating.

On heating up to 400°C, all these specimens preserve well their original structures. When heated at 500°C, the basal peaks disappeared but the other peaks remained distinctly, as shown in Fig. 3 (A). After heating at 600°C, the very broad peak showing the dome-like appearance was faintly seen on the chart of Y 40 in the region of 5 and 8 degrees (2θ), newly, but on both of Y 43 and Y 50b, no peak appeared (Fig. 3 (B)). On Y 51, as seen in the same figure, the peak in this region was very indistinct for a certain content of alumina. It is considered that such a peak will exhibit the transformation to 10-14Å mineral (chloritic material), its transformation being very imperfect. And such imper-
fect transition may be due to the low degree of crystallization and the want of heating hours. On the other hand, the comparative X-ray patterns for common serpentines found from other localities gave the sharp and strong peaks in the same region.

On heating at 700°C, the every specimen (Y40–Y51) showed prominent structural change, it being nearly amorphous material in each X-ray pattern, as given in Fig. 3 (B). Namely, some peaks diffracted from the original structure, being remained up to 600°C, almost disappeared and especially for Y40, the 10–10Å peak also completely disappeared besides them. But for the specimens of Y43, Y50b and Y51, an indication of transition to forsterite may be seen by small peaks appeared in the range of 20 to 35 degrees (2θ), respectively. The transition for those specimens will be considered to begin before or after the decomposition of original structure. As indicated in Fig. 3 (C), the every transition to forsterite behaved sensitively at about 800°C.

DIFFERENTIAL THERMAL ANALYSIS CURVES

The differential thermal analyses for the specimens were carried out for about 0.2 grams and the specimens were heated from room temperature to 1000°C at the mean heating rate of 10°C per minute, respectively. The curves are given in Fig. 4.

The curves of the specimens are generally comparable to one another, but some differences are seen in the endothermic reactions. The curves of Y43, Y50b and Y51 are composed of two principal endothermic peaks and a sharp exothermic one. The low temperature endothermic peaks at approximately 150°C are considerably large and immediately followed by small shoulders at about 250 to 300°C, suggesting a characteristic for deweylite (on Y50b and Y51). The high temperature endothermic peaks at about 650°C, being attributed to the dehydroxylation, are very broad and not so large as compared to those of well-crystallized serpentines. And also no marked endothermic reaction is shown between 700 and 800°C, however, it is distinctly seen for serpentine. The exothermic peaks being very sharp and strong appear around 810–820°C and are related to the formation of forsterite.

The curve of Y40 has two endothermic peaks which differ in peak temperature from those of Y43, Y50b and Y51. The first endothermic peak appears at about 640°C and is more remarkable than those of Y43, Y50b and Y51. The high temperature endothermic peak at about 750°C is immediately followed by a very sharp and strong exothermic peak, and is more distinctly recognized than in others. The high temperature endothermic peak may be due to the break down of intermediate production during the transition process. The exothermic peak at about 810°C is resulted by the formation of forsterite as
well in Y 43, Y50b, and Y 51. The thermal reaction of Y 40 corresponds well to those characteristic for serpentine minerals, as shown by Caillere and Henin (1957).

**ELECTRON MICROGRAPHS**

In recent years, many investigations on the morphology of serpentine minerals have been reported. Generally it is considered that fibrous serpentine (chrysotile) exhibits in the range of hollow tubes of very high length/width ratio to elongate sprintery laths under an electron microscope, as shown usually in photomicrographs of halloysites (2 H₂O and 4 H₂O forms). And also antigorite or lizardite detectable morphologically appears to be in plates or broader curved laths, as seen in kaolinite photomicrographs. However, as described by Veniale and Marrel (1963), Ball (1964), Muchi, Oji and Ogawa (1967), and Muchi and Ogawa (1967), lizardite often appears to be of elogate flakes or tubes. Thus it is very difficult to differentiate lizardite from antigorite only by electron micrographs.

The pictures for the specimens obtained by using an electron microscope,
Fig. 5, Y 40: Electron micrograph of poorly-crystallized serpentine
(A) The general view of a part where essentially consist of aggregates of rounded particles having angular outlines.
(B) The detailed view of a small portion in Fig. 5 (A), indicating aggregates of relatively rounded particles with polygonal outlines.

Fig. 6, Y 43: Electron micrograph of deweylite
The general view of a portion where chiefly composed of aggregates of rounded and fibrous particles. The rounded particles indicate mostly irregular shapes and project fibrous material as seen well at the lower left side of the picture.
Fig. 7, Y50A: Electron micrograph of deweylite
(A) The general view of aggregates of rounded particles giving polygonal outlines. These particles mostly have very fine fibrous projections and fuzzy appearance.
(B) The highly magnified view of rounded particles composed of aggregates of hair-like materials, showing well fuzzy appearance.

Fig. 8, Y51: Electron micrograph of deweylite
A general view of the portion where chiefly composed of rounded particles. The fine fibers are seen in the aggregates near the center and at the right hand side.
Jem-7 are given in Fig. 5-8, and these distinctly indicate that the particles appear to be generally finely spherical, exhibiting somewhat polygonal or irregular outlines. The pictures also suggest that every specimen is likely to be in initial stage in crystallization processes.

In the picture of Y 40 (Fig. 5 (A)), the particles are generally seen to give the rounded outlines, but observed in pieces, these are somewhat angular, displaying polygonal forms, and some particles have fine acicular projections, which are frequently observed in those of hydrated-halloysite particles (called geneanarly "chestnut shell-like type"). On this specimen, it may be a characteristic feature that the particles giving the flaky forms are scarcely seen in the pictures. These polygons determinable as a single particle shown an average value of approximately 350 Å, and viewed at higher magnification, some of these are distinctly pseudohexagonal as seen in Fig. 5 (B).

In the picture of Y 43 (Fig. 6), the particles generally appear to give irregular forms with somewhat angular or ellipsoidal outlines rather than rounded ones, when separated in a single particle, as seen well at the left side of the picture, and the particles mostly have very fine fibrous projections as often recognized in the rounded particles of poorly-crystallized halloysites.

The specimen of Y 50 (Fig. 7 (A)) also chiefly consists of fine rounded particles giving rather polygonal outlines, which are relatively roundish as compared with those of Y 43, however as seen in Fig. 7 (B) (highly magnified picture), these rounded particles appear to be aggregates of fine hair-like particles and display fluffy and flattened appearance.

In the specimen of Y 51 (Fig. 8), the particles dominantly indicate fine irregular forms, being somewhat roundish than in Y 43, and the fine fibrous particles were apparently recognized in the aggregates of such rounded ones.

Considering the particle shapes in these pictures, the deweylite and serpentine specimens, being in an initial stage of crystallization process, generally appear to take the fine rounded forms with various outlines and aggregates of extremely fine fibers, and so it may be impossible to differentiate deweylite from poorly-crystallized serpentine by only these pictures. But the particles of deweylite seem to exhibit more roundish outlines and fluffy appearance than in poorly-crystallized serpentine.

ACKNOWLEDGEMENTS

We wish to express our sincere acknowledgements to Professor T. Sudo, Tokyo University of Education, for his valuable advice and suggestions. Further, we are indebted to Dr. K. Ishibashi and Mr. K. Nuruyu, Kyushu University, for the chemical analyses of deweylites and serpentine minerals and for their profitable advice on the occurrence of deweylite.
REFERENCES


摘要

福岡県若宮町産デウエー石と低結晶蛇紋石の鉱物学的研究

髙 枝 叠, 井上光徳・紫田賀江

福岡県若宮町から発見されたデウエー石と低結晶蛇紋石は灰白色ないし淡黄色で結晶状あるいは細粒状をなし、蛇紋岩やカンラン岩中に細脈。時には網状をなして胚腎し、それぞれ次のような化学式を示す。

3.6 MgO・3SiO₂・5.6 H₂O…………………(Y 43)
3.9 MgO・3SiO₂・5.7 H₂O…………………(Y 50b)
3.7 MgO・3SiO₂・5.3 H₂O…………………(Y 51)
2.9 MgO・2SiO₂・1.9 H₂O…………………(Y 40)

これらのうち最初の3つの試料はデウエー石の標準化学式によく類似し、残りの1つは
Deweylite and poorly-crystallized serpentine

これらの試料はそれぞれよく類似したX線回折図を示すゆえ、両者の区別は非常に困難であるが、最初の3つの回折ピークの強度にわずかの差異を示す傾向が認められた。なお加熱処理の結果、デュエーソ石と低結晶蛇紋石との間には、明瞭な構造変化の差異は見られなかった。示差熱分析曲線ではある程度の特性を示す。即ちデュエーソ石の曲線では100℃と200℃間の大きな吸熱ピークとこれに続く250℃と300℃間の小さな付状ピークによって示されるように明らかに二重の吸熱反応が見られた。蛇紋石ではこの種の吸熱反応は生じない。また蛇紋石では700℃と750℃間に中間転移物の破壊によるかなり鋭い吸熱反応が示されるが、デュエーソ石ではこの反応は非常に不明瞭である。これらの鉱物の粒子の形状もまたよく似ており、一般に多角形または不定形な外形を示す球状体からなるが、デュエーソ石はむしろ繊維状突出を有する球状粒子からなるものが多く、これらは絹毛状の外観を呈している。しかし低結晶蛇紋石は角張った不規則な形状を示しやすいようである。