# The Mineralogy and Chemistry of Clay Minerals of the Middle Ordovician Jigunsan Formation: Implications for the Metamorphic Grade

중부오오도비스기 직운산층 점토광물의 광물학적 및 화학적 연구: 변성정도에 대한 의의

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**ABSTRACT:** Illite 'crystallinity' and chlorite chemistry are applied to the evaluation of the thermal grade of the Jigunsan Formation. Illite 'crystallinity' value of the formation has the range from 4.48 to 32.5 in Weaver index (W. I.) and from  $0.14\triangle^{\circ}$   $2\theta$  to  $0.30\triangle^{\circ}$   $2\theta$  in Kubler index (K. I.). Most of illite 'crystallinity' values in this formation belong to the epizone field (K. I.  $\langle 0.21 \triangle^{\circ} 2\theta \rangle$ ). The chemistry and calculated temperature of chlorites form this formation (Fe/(Fe + Mg)=0.45, Tet. Al/Octa. Al=0.84, the calculated temperature=250-270°C) are similar to those of epizone chlorites in the literature.

The results of this work show that the metamorphic grade of the Jigunsan Formation belongs to the epizone and the formation is believed to have been reached paleotemperatures of at least 300°C.

요약: 직운산층의 열적변성도를 밝히기위해 일라이트의 '결정도'와 녹니석의 화학성분을 이용하였다. 직운산층의 일라이트 '결정도' 값은 Weaver index로는 4.48에서 32.5에 이르며, Kubler index로는  $0.14 \triangle$ °  $2\theta$ 에서  $0.30 \triangle$ °  $2\theta$ 에 이른다. 대부분의 일라이트 '결정도' 값은 epizone의 영역  $(K.\ I.\ \langle\ 0.21 \triangle$ °  $2\theta$ )에 속한다. 직운산층 녹니석에 의해 계산된 온도와 그 화학 분석 치 $(Fe/(Fe+Mg)=0.45,\ Tet.\ Al/Octa.\ Al=0.84,\ the calculated temperature=<math>250-270\,^{\circ}\mathrm{C}$ )는 기존의 epizone 녹니석의 연구결과와 잘 일치한다. 이 연구의 결과는 직운산층의 변성정도가 epizone에 속하고 직운산층은 과거에 적어도  $300\,^{\circ}\mathrm{C}$ 의 온도에 도달했다는 것을 나타낸다.

#### INTRODUCTION

Progressive thermal evolution of sedimentary rocks with increasing depth of burial results in the gradual transformation of organic and inorganic materials. The studies of vitrinite reflectance, conodont color alteration, clay mineralogy, etc. have been used to establish the thermal grade of sedimentary rocks.

The diagenetic changes occur in mud and shale at temperature ranges from 20°C to nearly 200°C (Burst, 1959; Dunoyer de Segonzac, 1970; Foscolos and Kodama, 1974; Boles and Franks, 1979). In general, the greenschist facies, the beginning of "true" met-

amorphism starts at the temperature above 400°C (Weaver and Associates, 1984; Frey and Kisch, 1987). Low-grade metamorphism is located in the temperature ranges between 150-200°C and 350-400°C (Frey and Kisch, 1987). In the case of very low- and low- grade metaclastites, this metamorphic grade is divided into anchimetamorphism (anchizone) and epimetamorphism(epizone) based only on the illite 'crystallinity' (Kubler, 1968).

Several workers reported that the chemical composition of chlorite varies as a function of temperature (Brown, 1967; Black, 1975; McDowell and Elders, 1980; Weaver and Associates, 1984; Cathelineau and Nieva, 1985). Cathelineau

and Nieva(1985) studied correlations between chlorite composition, ranges and nature of site occupancy, and temperature in geothermal system. They concluded that variations of site occupancy (mainly Al(IV)) and the octahedral occupancy (6-Al(VI)—(Mg+Fe<sup>2+</sup>)) are considered mainly temperature dependent.

The purposes of this study are to analyze the mineralogy and chemical aspects of clay minerals of the Middle Ordovician Jigunsan Formation and to establish the thermal grade of this formation using illite 'crystallinty' and chlorite chemistry data. This report forms part of a larger study on the evaluation of the unconformity between the Lower Paleozoic Joseon and the Upper Paleozoic Pyeongan supergroups. The Jigunsan Formation was chosen for a clay mineral study because other strata in the Joseon Supergroup are mostly carbonates with subordinate sandstones. Although slates occur in the lower part of the supergroup, they were not suitable for such purpose.

## **GEOLOGICAL SETTING**

The study area is located in the northeastern part of the Ogcheon belt(Fig.1). The E-W trending syncline, Baegunsan Syncline, runs through the middle of the study area. The study area is largely covered with the Paleozoic sedimentary rock sequence consisting of the Lower Paleozoic Joseon and the Upper Paleozoic Pyeongan supergroups (Fig. 1).

The Jigunsan Formation forms the upper strata of the Duwibong sequence of the Lower Paleozoic Joseon Supergroup(Fig. 2). This formation overlies the Maggol Formation conformably, and is characterized by the dark grey to black shales. The shales are often calcareous and nodular in the upper part, whereas limestone beds are well developed in the lower part of the formation. This formation is fossiliferous and 2 species of graptolites, 1 of the Plumulites, 3 of brachiopods, 13 of pelycypods, 6 of ostracods have been reported

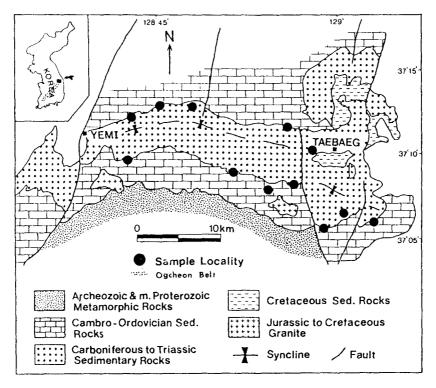


Fig. 1. Simplified geological map of the study area. Sampling localities are shown as dots. The shaded area in the inset represents the Ogcheon belt.

by Kobayashi(1966). The Jigunsan Formation is of the Llanvirnian to Llandeilian in age (Cheong et al, 1979; Lee, 1977). This formation is estimated 50–100m in thickness.

The Jigunsan Formation is conformably overlain by the Duwibong Formation which is the uppermost strata of the Lower Paleozoic Joseon Supergroup in the study area. The Joseon Supergroup is unconformably overlain by the Pyeongan Supergroup. The unconformity between the two supergroups represents a great hiatus ranging from Late Ordovician to Late Carboniferous (more than 100 million years).

## **METHODS**

For a clay mineral study, argillaceous samples were collected at 12 localities throughout the Baegunsan Syncline area(Fig. 1). At each locality, more than 10 fresh samples were collected from outcrops of the Jigunsan Formation with intervals of 1 to 2 meters.

Clay fractions ((2,4m) were separated in order to carry out the X-ray diffraction (XRD) analysis. The oriented samples were prepared by

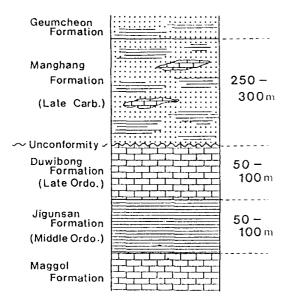


Fig. 2. Simplified columnar section of the upper sequence of the Joseon Supergroup and lower sequence of the Pyeongan Supergroup.

sedimentation of mixture of clay fraction and distilled water on a glass slide, which were dried under room conditions overnight. For the illite polytype analysis, the dried and sized material was ground slightly in an agate mortar to break any reaggregated material (Velde and Hower, 1963). The clay mineral analysis was carried out by using Rigaku Model RAD-3C diffractometer with Ni-filtered CuK  $\alpha$  radiation.

In order to determine the illite 'crystallinity', the oriented samples were run on a X-ray diffractometer through 3° to 10° (2 $\theta$ ) at operation settings of 30kV, 20mA, scanning rate  $0.5^{\circ}$   $(2\theta)$ / min, divergence slit 1°, scatter slit 1° and receiving slit 0.15mm. The illite 'crystallinity' values were represented by Weaver index(Weaver, 1960) and Kubler index(Kubler, 1967 cited in Kubler 1968). For the Kubler index, illite 'crystallinity' was determined by measuring the half height width of the 10 Å peak and this measurement, in millimeters, was converted to  $\triangle^{\circ}$  2 $\theta$ . For measuring the Kubler index the peak widths are dependent upon the experimental conditions of X-ray diffraction, such as the widths of the divergence, scatter and receiving slits used, the time constants, the kind of radiation used, and the scanning rate (Kisch, 1980, 1990; Blenkinsop, 1988; Robinson et al., 1990). For this study, instrumental settings for determinations of illite 'crystallinity' were based on Kisch(1990)'s setting: time constant 2 and scan rate  $0.5^{\circ} (2\theta)/\text{min}$ .

Clay mineral polytype analysis was done by scanning from 3° to 50°  $(2\theta)$  at the setting of 35kV, 20mA, scanning rate 1°  $(2\theta)$ /min.. Also, the expandability of clay minerals is checked with both heated and glycolated samples. The oriented samples were heated at 300°C for 2 hours, and the oriented samples were ethylene glycol solvated by the vapor method. The heated samples and glycolated samples were scanned form 3° to 50°  $(2\theta)$  at a scanning rate of 4°  $(2\theta)$ /min..

More than 30 polished-thin sections were made to analyze the chemical composition of authigenic chlorites. The analyzed chlorites are replacement minerals of fossils (usually trilobite). The chemical composition was analyzed by JEOL JXA-733 Electron Microprobe.

#### RESULTS

The clay mineral assemblage of the Jigunsan Formation consists of illite and chlorite (Fig. 3). The discrete mixed-layer clay mineral peaks are absent in XRD analysis.

#### Illite

Illite 'Crystallinity': The illite 'crystallinity' values of the Jigunsan Formation measured by Weaver and Kubler indices are shown in Table 1.

The illite 'crystallinity' values range from 4.48 to 32.5 in Weaver index and from  $0.14 \triangle^{\circ} 2\theta$  to  $0.30 \triangle^{\circ} 2\theta$  in Kubler index. Fig. 4 shows a crossplot of illite 'crystallinity' and incipient metamorphic grade. Most of illite 'crystallinity' values belong to the epizone field(K. I.  $\langle 0.21^{\circ} \triangle 2\theta \rangle$ ). Some illite samples are plotted in anchizone field. Comparing the illite 'crystallinity' between the southern and northern regions of Baegunsan syncline, both

regions equally represent mean Kubler index value of  $0.19\triangle^{\circ} 2\theta$ .

Illite Expandability: The mixed-layer illite/smectite (I/S) peaks are not shown in the XRD patterns of the Jigunsan clay. In order to investigate the possible existence of the mixed-

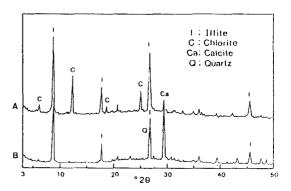


Fig. 3. Typical XRD patterns of the Jigunsan Formation. A: illite-chlorite assemblage; B: illite.

Table 1. Illite crystallinity values and I(002)/I(001) intensity ratio of the Jigunsan illites.

Sample number	K. I.	W. I.	1002/1001	Sample number	K. I.	W. I.	1002/1001
J01-1	0.18	18.86	0.406	J05-4	0.18	6.57	0.344
J01 <i>-</i> 2	0.16	12.82	0.304	J05-5	0.26	6.83	0.399
J01 <del>-4</del>	0.16	17.30	0.400	J05-7	0.21	5.50	0.314
J01-5	0.18	6.93	0.308	J06-2	0.16	11.00	0.389
<b>J02-</b> 1	0.14	18.30	0.400	J06-3	0.18	9.30	0.336
J02 <del>-6</del>	0.15	12.33		J06-5	0.20	8.50	0.476
J02-7	0.18	11.00	0.382	J06-9	0.20	6.39	0.385
J02-11	0.20	11.62	0.235	J06-11	0.24	4.55	0.289
J02-14	0.15	12.64	0.393	J06-14	0.21	9.07	0.419
J02-18	0.25	4.48	0.339	J07-5	0.21	7.29	0.333
J02-20	0.27	4.87		J07 <del>-6</del>	0.18	8.80	0.303
J03-3	0.16	19.50	0.425	J08-3	0.16	9.10	0.315
J03-5	0.18	9.44		J08-4	0.20	7.00	0.283
J03-7	0.14	22.00	0.397	J08-6	0.22	7.10	0.245
J03-9	0.14	32.50	0.424	J09-1	0.18	11.07	0.275
J04-1	0.14	24.50	0.261	J09-3	0.14	7.86	0.273
J04-3	0.23	7.30	0.455	J09-5	0.15	8.40	0.235
J04-4	0.18	11.00	0.308	J010-1	0.18	11.93	0.310
J04-5	0.18	10.29	0.380	J010-2	0.14	20.40	0.329
J04-6	0.22	10.00	0.400	J010-4	0.20	5.60	0.333
J04-7	0.30	4.93	0.304	JO11-1	0.22	5.71	0.404
<b>J05-</b> 1	0.18	9.25	0.314	J011-2	0.23	7.76	0.284
J05-2	0.16	10.50	0.371	J011-3	0.18	11.53	0.313

K. I.: Kubler index; W. I.: Weaver index; I(002)/I(001): intensity ratio of the second- and first-order diffraction peaks of illites.

layer I/S in illite, ethylene glycol-treated, heattreated and air-dried XRD patterns for all illite samples were examined and the results were compared with one another.

The first step in the identification procedure is to plot the positions of the 002 and 003 reflections from glycolated preparations onto Srodon (1984)'s diagram(Fig. 5). Samples are plotted in the illite field containing illite or mixtures dominated by illite. The second step was to check the intensity ratio(Ir) of Srodon and Eberl (1984). The Jigunsan illites are thought to be pure illite because the intensity ratio is just about 1 (Fig. 6).

Another method for checking illite expandability is heat treatment. Fig. 7 shows the difference in K. I. values of the 10 Å peak before and after heating at 300°C for 2 hours. There are no difference between two values which implies that no expandable layers exist.

Intensity Ratio (I(002)/I(001): The intensity ratio of the second- and first-order diffraction peaks of illite, I(002)/(001), is thought to be related to illite composition (Weaver, 1965; Esquevin, 1969).

Fig. 8 shows the relationship between I(002)/I (001) ratio and illite 'crystallinity' of the Jigunsan Formation. The I(002)/I(001) values of this formation ranged from 0.235 to 0.476. According to Fig. 8, these values are plotted on biotites+muscovites and phengites regions.

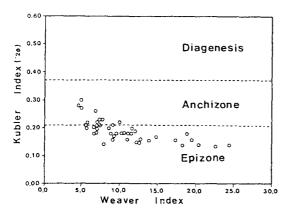


Fig. 4. Diagram of the distribution of illite 'crystallinity' values of the Jigunsan Formation based on Kubler index vs. Weaver index. Most Jigunsan illites belong to epizone field.

Illite Polytype: The experimental curve of Maxwell and Hower(1967) has been used in order to determine the Jigunsan illite polytype. In this mehtods, 2.80 Å and 2.58 Å peaks are used. As a result of this analysis, the proportions of 2M polytypes of the illites is more than 85%. The amount of 2M polytype gradually increase with increasing illite crytallinity(Fig. 9). In the epizone, the samples consist of almost 100% 2M.

#### Chlorite

Chlorites are also the abundant clay mineral in the Jigunsan Formaion. The chlorite polytype is II b type.

The chemical composition of chlorites in the formation is shown in Table 2. For chlorite classification, chlorites were plotted in the diagram of Foster (1962) (Fig. 10). Chlorites of the Jigunsan Formation are classified as brunsvigite. The area outlined by dashed lines shows the ranges of compostion of chlorites from the metasedimentary lower greenschist facies of the Tennant Creek area, Central Australia (Ramamohana, 1977). Chlorites in Jigunsan Formation are included in this area.

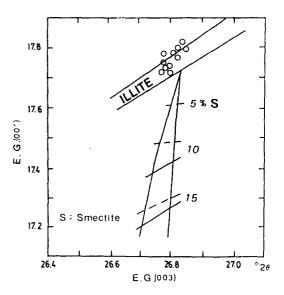


Fig. 5. Diagram for determining smectite:illite ratio from (002) and (003) reflections of etylene glycolated specimens(after Srodon, 1984). The Jigunsan illites are plotted in the illite filed.

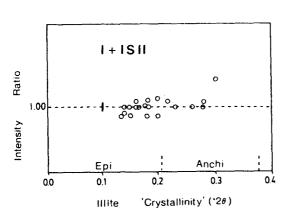


Fig. 6. Relationship between illite 'crystallinity' (K.I.) and intensity ratio(Ir). I:illite; ISII:kalkberg-type illite/smectite. The Ir values of the Jigunsan illites are just about 1, indicative of pure illite.

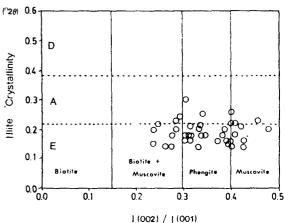


Fig. 8. Relationship between I(002)/I(001) ratio and illite 'crystallinity' of the Jigunsan Formation. The distribution pattern corresponds to Dunoyer de Segonzac (1970)'s) Type 5 field. The chemical composition of the illites belong to the region of biotite+muscovite and phengite(Esquevin, 1969). D: diagenesis; A: anchizone; E: epizone.

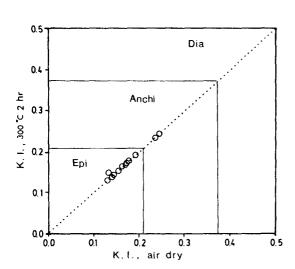


Fig. 7. Diagram showing the changes in Kubler index (K. I.) values when samples are heated at 300°C for 2 hours. The peak widths of illites are not changed after heating.

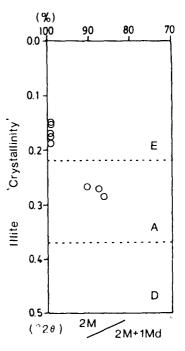


Fig. 9. Realationship between illite 'crystallinity' and 2M%. The proportion of 2M polytype increases with increasing illite 'crystallinity'. Samples in the epizone consist of almost 100% 2M illite.

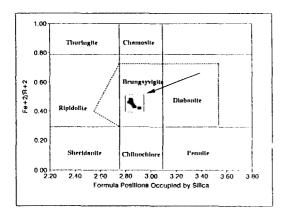


Fig. 10. Diagram showing chlorite classification (after Foste, 1962). Dotted lines enclose compositions of chlorite in low-grade metamorphic phyllites (Ramamohana, 1977). Arrow shows direction of compositional change with increasing metamorphic grade in southern Appalachian(after Weaver and Associates, 1984).

The arrow indicates the changes in chemical composition with increasing metamorphic grade of the southern Appalachians (Weaver and Associates, 1984). The composition of the Jigunsan chlorite corresponds to that of epizone samples of Weaver and Associates(1984), which belong to the highest metamorphic grade and show 350°C in temperature.

Fig. 11 shows a distribution of tetrahedral and octahedral Al in the Jigunsan chlorites. The chlorites in the formation have more octahedral Al than tetrahedral Al, and the average value of tetrahedral Al/octahedral Al ratios is 0.84. A value of 1.2 tetrahedral Al generally appears to distinguish between high-(metamorphic) and low-temperature(low-grade metamorphic) chlorites(Weaver and Associates, 1984). The Jigunsan chlorites are plotted on low-grade metamorphic chlorites range. The compositional range of the chlorites corresponds to that of the Weaver and Associates (1984)' chlorites which are associated in epizone

The chlorite formation temperature can be inferred by the following equations proposed by Cathelineau and Nieva(1985).

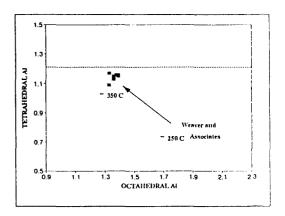


Fig. 11. Distribution of tetrahedral and octahedral Al in chlorites. The horizontal line at 1.2 tetrahedral Al divides the high-temperature chlorites in the upper field from low-temperature chlorites in the lower field. Arrow shows trend of low-grade metamorphic chlorites(after Weaver and Associaltes, 1984) with increasing temperatures.

Al(
$$\mathbb{N}$$
)=4.71×10<sup>-3</sup> T -8.26×10<sup>-2</sup> (1)  
6- $\Sigma$  VI=-8.57×10<sup>-3</sup>T+2.41 (2)

The average tetrahedral Al of Jigunsan chlorites is 1.13 and the inferred temperature estimated by Equation(1) is 257.45°C. The  $6-\Sigma$  VI values for the Jigunsan Formation average 0.13 and chlorite formation-temperature calculated by Equation (2) is 266.04°C.

## DISCUSSION

In general, as metamorpohic grade increases the illite 'crystallinity' increases also. Between two illite 'crystallinity' indices, the Weaver index is restrictedly used in unmetamorphosed sediments, due to the increasing error in determining the sharphness ratio on very narrow, high 'crystallinity' peaks(Kubler, 1968; Blenkinsop, 1988), whereas the Kubler index has been more widely used to determine the metamorphic grade. There are a few problems in determining the boundaries of metamorphism by means of the Kubler index; the difference among instrumental settings in experiments brings about the discrepancy. The lower boundary values for anchizone and epizone used

sample	1	5	6	7	26	27	28	29
SiO <sub>2</sub>	26.83	26.57	27.66	27.12	26.60	26.55	26.88	26.92
TiO <sub>2</sub>	0.01	0.08	0.04	0.03	0.05	0.03	0.05	0.04
Al <sub>2</sub> O <sub>3</sub>	20.38	19.92	19.49	19.99	20.17	20.27	20.14	19.95
Cr <sub>2</sub> O <sub>3</sub>	0.01	0.00	0.04	0.00	0.00	0.00	0.03	0.02
FeO	24.03	23.40	21.33	22.10	23.53	23.77	23.00	22.11
MgO	15.02	15.58	16.85	16.19	14.84	14.76	15.58	16.02
MnO	0.03	0.03	0.09	0.02	0.07	0.07	0.04	0.0
CaO	0.04	0.06	0.05	0.05	0.07	0.03	0.06	0.03
Na <sub>2</sub> O	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00
Total	86.34	85.64	85.56	85.49	85.32	85.49	85.79	85.17
Si(IV)	2.84	2.83	2.91	2.87	2.85	2.84	2.85	2.86
Al(IV)	1.16	1.17	1.09	1.13	1.15	1.16	1.15	1.14
Al(VI)	1.38	1.33	1.33	1.36	1.39	1.39	1.36	1.30
Fe	2.13	2.09	1.88	1.96	2.11	2.13	2.04	1.97
Mg	2.37	2.48	2.65	2.55	2.37	2.35	2.46	2.54
Mn	0.00	0.00	0.01	0.00	0.01	0.01	0.00	0.01
acancy	0.12	0.10	0.13	0.13	0.13	0.13	0.14	0.13
/Fe+Mg	0.47	0.46	0.42	0.43	0.47	0.48	0.45	0.44

Table 2. The chemical composition of the Jigunsan chlorites.

in this study are Kubler index 0.37  $\triangle^{\circ}$  2 $\theta$  and 0.21  $\triangle^{\circ}$  2 $\theta$ , respectively (Kisch, 1990).

The metamorphic grade of the Jigunsan Formation belongs to the epizone by means of illite 'crystallinity'. The clay mineral assemblage of the Jigunsan Formation is correlated well with that of the epizone(Hoffman and Hower, 1979; Hunziker et al., 1986). Generally mixed-layer illite/smectite(I/S) mineral is converted into pure illite at the boundary between anchizone and epizone (Perry and Hower, 1970; Reynolds and Hower, 1970; Hoffman and Hower, 1979; Reynolds, 1980; Hower, 1981 a, b). The Jigunsan illites contains, if any, few mixed-layer illite/smectite minerals, which implies that the formation had experienced high thermal history. The 2M polytype of illite is stable at above 200-350°C (Yoder and Eugster, 1955). From the results of polytype analysis more than 85% 2M polytypes occur in Jigunsan illites, which corresponds well to their metamorphic grade.

Some illites plotted in the anchizone field are attributed to the lithology of the host rock. Those samples were collected form the upper part of the Jigunsan Formation, where it is more calcareous than other parts of the Jigunsan sequence due to its position being gradational to the overlying

Duwibong Formation. In calcareous lithology, the aggradation of illite may be retarded compared with noncalcareous clastics due to a deficiency in potassium(Frey, 1987; Kisch, 1987).

Many workers argued the intensity ratio of the second and first-order diffraction peaks of illite, I(002)/I(001), is related to illite composition (Weaver, 1965; Esquevin, 1969; Dunoyer Segonzac, 1970). Dunoyer de Segonzac(1970) distinguished five types of the relationships between peak-width and I(002)/I(001) ratio in an illite population. The distribution of I(002)/I(001) values of the Jigunsan illites is in good agreement with the Type 5 field, the epizone illites. According to Esquevin(1969), the ratio is an indicator of the chemical composition of the octahedral layer. Namely, smaller value of I(002)/I(001) ratio reflects existence of biotite and phengite with a high Fe content than mica. As shown in Fig. 8, the recrystallization of some biotites and phengites seems to have begun already in the Jigunsan Formation, which implies that the formation had experienced high thermal history. In the southern Appalachian (Weaver and Broekstra, 1984), phengite is the highest grade micaceous material in the epizone.

As anchizone and epizone are only deter-

mined by illite 'crystallinity', the temperature range of each zone does not represent the specific values, and varies with the locality. Based on the mineral conversion temperatures and stability ranges, anchizone begins at about 200°C and extends to 300°C (Dunoyer de Segonzac, 1970; Winkler, 1979; Hunziker, 1986). In the case of the southern Appalachian the beginning temperature of the anchizone is at about 250-280°C (Weaver and Associates, 1984), which is higher than the general beginning temperature of the anchizone. From the illite 'crystallinity' study, the Jigunsan Formation is believed to have been reached paleotemperatures of at least 300°C.

The recrystallization of chlorite occurs during late diagenesis. Amon the chlorite polytypes, the II b polytype is more stable in the higher temperature range (Brindley and Gillery, 1956; Hayes, 1970; Walker, 1989). The II b polytype chlorites of the Jigunsan formation imply that the formation had experienced high temperature.

The chemical composition of the Jigunsan chlorites is similar to that of epizone chlorites by McDowell and Elders (1980) and Weaver and Associates (1984). The chlorite formation-temperatures calculated by Cathelineau and Nieva(1985)' equation are about 250-270°C. This value represents the temperature at the time of chlorite formation. The maximum temperatures at which the Jigunsan Formation had undergone might be higher than the chlorite formation temperatures, which indicate that this formation had experienced the temperatures of at least 250-270°C. This result is in good agreement with the result of illite 'crystallinity'.

Illite 'cystallinity' is the one of the most suitable and generally applicable monitors in very low-grade metamorphism of clastic sedimentary rocks. But at the present state of knowledge it cannot be used for geothermometer purposes (Frey, 1987), because the illite 'crystallinity' is dependent on many variables including temperatures as well as other physical, chemical and experimental factors. Also chemical composition of chlorite is dependent on not only temperature but also host rock chemistry.

Therefore, the study of incipient metamorphism requires oxygen isotope and fluid inclusion

studies, whereby tmeperautre can be inferred independently and more accurately. The study for the fluid inclusion from the Duwibong Formation (the strata above the Jigunsan Formation) indicate that the maximum homogenization temperature is 290°C which is not pressure-corrected (Ko, 1991). This temperature well corresponds to the results of this study.

## **CONCLUSIONS**

The results of the clay mineral study on the Jigunsan Formation of the Lower Paleozoic Joseon Supergroup are summarized as the followings.

- 1. The clay mineral assemblage of the Jigunsan Formation consists of illite and chlorite. The Jigunsan illitic material is pure illite and the prportion of 2M polytype is more than 85%.
- 2. The illite 'crystallinity' of this formation shows a range from 4.48 to 32.5 in Weaver index and from  $0.14\triangle^{\circ}$   $2\theta$  to  $0.30\triangle^{\circ}$   $2\theta$  in Kubler index. Most of illite 'crystallinity' values in this formation belong to the epizone.
- 3. The I(002)/I(001) ratio of the Jigunsan illite ranged from 0.235 to 0.476 suggesting that the recrystallization of some biotites and phengites seems to have bugun already.
- 4. The chemistry and calculated temperature of chlorites from the Jigunsan Formation (Fe/(Fe+ (Mg) ratio = 0.45, Tet. Al/Octa. Al = 0.84; the calculated temperature = 250-270 $^{\circ}$ C) are similar to those of the epizone chlorites in the literature.
- 5. According to the results of clay mineral studies, the upper atrata of the Joseon Supergroup had been reached paleotemperatures of at least 300°C.

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# REFERENCES

Black, P. M.(1974) Oxygen isotope study of metamorphic rocks from the Ouegoa District, New Caledonia. Contrib. Mineral. Petrlo, 47, 197-

- 206.
- Blenkinsop, T. G.(1988) Definition of low-grade metamorphic zones using illite crystallinity. Jour. Meta. Geol., 6, 623-636.
- Boles, J. R., and Franks, S. G.(1979) Clay diagenesis in Wilcox sandstones of southwest Texas: Implication of smectite diagenesis on sandstone cementation. Jour. Sed. Petrol., 47, 55-77.
- Brindley, G. W., and Gillery, F. H(1956) X-ray identification of chlorite species. Am. Miner., 41, 169-186.
- Brown, E. H.(1967) The greenschist facies in part of Eastern Otago, New Zealand. Contrib. Mineral. Petrol., 14, 259-292.
- Burst, J. F.(1959) Post-diagenetic clay mineralenvironmental relationships in the Gulf Coast, Eocene. Clays Clay Miner, 6, 327-341.
- Cathelineau, M., and Nieva, D.(1985) A chlorite solid solution geothermometer; The Los Azufres (Mexico) geothermal system. Contrib. Mineral. Petrol., 91, 235-244.
- Cheong, C. H., Lee, H. Y., Ko, I. S., and Lee, J. D. (1979) A study on stratigraphy and sedimentological environments of the Lower Paleozoic sequences in south Korea (chiefly in Jeongseon area). Jour. Nat. Acad. Sci. R. O. K., Natural Science series, 18, 123-159.
- Dunoyer de Segonzac, G.(1970) The transformation of clay minerals during diagensis and lowgrade metamorphism. Sedimentology, 15, 282-344.
- Esquevin, J.(1969) Influence de la composition chimique des illites sur leur cristallinite. Bull. Centre. Rech. Pan-SNPA, 3, 147-154.
- Foscolos, A. E., and Kodama, H.(1974) Diagenesis of clay mineral from Lower Cretaceous shales of northeastern British Columbia. Clays Clay Miner, 22, 319-335.
- Foster, M. D.(1962) Interpretation of the composition and a classification of the chlorites. U. S. Geol. Surv. Prof. Pap., 414-A, 33p.
- Frey, M.(1987) Very low-grade metamorphism of clastic sedimentary rocks. In: Frey M.(ed.) Low Temperature Metamorphism, New York, Blackie and Sons, 9-58.
- Frey, M., and Kisch, H. J.(1987) Scope of subject. In: Frey M.(ed.) Low Temperature Metamor-

- phism, New York, Blackie and Sons, 1-8.
- Hayes, J. B.(1970) Polytypism of chlorite in sedimetary rocks. Clays Clay Miner, 18, 285-306.
- Hower, J.(1981a) X-ray diffraction identification of mixed-layer clay mineral. In: Longstaffe, F. J. (ed.) Clays and the Resource Geologists. Mineral. Assoc. Canada Short Course, Calgary, 7, 39-59.
- Hower, J.(1981b) Shale diagenesis. In: Longstaffe, F. J. (ed.) Clays and the Resource Geologists. Mineral. Assoc. Canada Short Course, Calgary, 7, 60-80.
- Hoffman, J. and Hower, J. (1979) Clay mineral assemblages as low grade metamorphic geothermometers; Application to the thrust-faulted disturbed belt of Montana, U. S. A. In: Scholle, R. A. and Schluger, P. R. (eds.) Aspects of Diagenesis. Soc. Econ. Paleontol. Mineral, Spec. Publ., 26, 55-80.
- Hunziker, J. C.(1986) The evolution of illite to muscovite: An example of the behavior of isotopes in low-grade metamorphic terrains. Chemical Geol., 57, 31-40.
- Hunziker, J. C., Frey, M., Clauer, N., Dallmeyer, R. D., Friedrichsen, H., Flehmig, W., Hochstrasser, K., Roggwiler, P., and Schwander, H.(1986) The evolution of illite to muscovite: mineralogical and isostopic data from the Glarus Alps, Switzerland. Contrib. Mineral. Petrol., 92, 157-180.
- Kisch, H. J.(1980) Incipient metamorphism of Cambro-Silurian clastic rocks from the Ja mtlan Supergroup, Gentral Scandinavian Caledonites, Western Sweden: illite crystallinity and 'vitrinite' reflectance. Jour. Geol. Soc. London, 137, 271-288.
- Kisch, H. J.(1987) Correlation between indicators of very low-grade metamorphism. In: Frey, M. (ed.) Low Temperature Metamorphism. New York, Blackie and Sons, 227-300.
- Kisch, H. J.(1990) Calibration of the anchizone: a critical composition of illite 'crystallinity' scales used for definition. Jour. Meta. Geol., 8, 31-46.
- Kobayashi, T.(1966) The Cambro-Ordovician formations and faunas of South Korea. Part X; Stratigraphy of Chosen group in Korea and south Manchuria and its relation to the Cambro-Ordovician formation of other areas,

- Sect. A, The Chosen Group of South Korea. Jour. Fac. Sci. Univ. Tokyo, Sect II, 16(1), 1-84.
- Ko, H. K.(1991) The significance of the unconformity between Early Palezoic Joseon Supergroup and Late Paleozoic Pyeongan Supergroup. M. S. Thesis, Seoul National University, Seoul, 125p.
- Kubler, B.(1968) Evolution quantitative de me tamorphisme par la cristallinite de l'illite. Bull. du Centre de Recherches de Pan-SNPA, 2, 385-397.
- Lee, H. Y.(1977) Conodonten aus den Jigunsan und den Duwibong-Schichten (Mittelordovizium) von Kangweondo. SudKorea. Jour. Geol. Soc. Korea, 13, 121-150.
- Maxwell, D. T., and Hower, J.(1967) High-grade diagenesis and low-grade metamorphism of illite in the Precambrian Belt Series. Am. Miner, 52, 843-857.
- McDowell, S. D., and Elders, W. D.(1980) Authigenic layer silicate minerals in borehole Elmore 1, Salton Sea Geothermal Field, California, U. S. A. Contrib. Mineral. Petrol., 74, 293-310.
- Perry, E. A., Jr. and Hower, J.(1970) Burial diagenesis in Gulf coast pelitic sediments. Clays Clay Miner, 18, 165-177.
- Ramamohana R. T.(1977) Distribution of elements between co-existing phengite and chlorite from the greenschist facies of the Tennant Creek area, Central Austalia. Lithos, 10, 103-112.
- Reynolds, R. C.(1980) Interstratified clay minerals. In: Brindley, G. W. and Brown, G. (Eds.) Crystal Structures of Clay Minerals and Their X-ray Identification. Mineral. Soc. London, 289-304.
- Reynolds, R. C., Jr., and Hower, J.(1970) The nature of interlayering in mixed-layer illite-

- montmorillonite. Clays Clay Miner, 18, 25-36.
- Robinson, D., Warr, L. N., and Bovins, R. E.(1990)
  The illite crystallinity technique: a critical appraisal of its preicision. Jour. Meta. Geol., 8, 333-344.
- Srodon, J.(1984) X-ray powder diffraction identification of illitic materials, Clays Clay Miner, 32, 337-349.
- Srodon, J., and Eberl, D. D.(1984) Illite. In: Baily, S. W. (ed.) Micas. Reviews in Mineralogy. Mineral. Soc. Am., 13, 495-544.
- Velde, B., and Hower, J.(1963) Petrological significance of illite polymorphism in Paleozoic sedimentary rocks. Am. Miner., 48, 1239-1254.
- Walker, J. R.(1989) Polytype of chlorite in very low grade metamorphic rocks. Am. Miner., 74, 738-743.
- Weaver, C. E.(1960) Possible uses of clay minerals in search for oil. Bull. Am. Assoc. Pet. Geol., 44, 1505-1518.
- Weaver, C. E. (1965) Potassium content of illite. Science, 147, 603-605.
- Weaver, C.E, and Associates(1984) Shale-Slate Metamorphism in southern Appalachians. Developments in Petrology, 10, Elsevier, Amsterdam, 189p.
- Weaver, C. E., and Broekstra, B. R.(1984) Illitemica. In: Weaver C. E. and Associates (Eds.) Shale-Slate metamorphism in soutern Appalachians. Development in Petrology 10, Elsevier, Amsterdam, 67-97.
- Winkler, H. G. F.(1979) Petrogenesis of Metamorphic Rocks. 5th Ed., New York, Springer-Verlag, 348p.
- Yoder, H. S., and Eugster, H. P.(1955) Synthetic and natural muscovites. Geochim. Comochim. Acta, 8, 225-280.