Grain growth and re-orientation of phyllosilicate minerals during the development of slaty cleavage in the South Kitakami Mountains, northeast Japan

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Abstract—Using X-ray diffraction analysis, the mutual relations among illite crystallinity, degree of preferred orientation of chlorite, grain size change of chlorite and illite during metamorphism and development of slaty cleavage have been investigated for argillaceous rocks in the South Kitakami Mountains, northeast Japan.

The metamorphic grade of IC (illite crystallinity index) = 0.29 is a critical one, beyond which the homogenization of chlorite composition, coarsening of chlorite and illite grains and degree of preferred orientation of chlorite are abruptly advanced. Grain coarsening is also promoted by the development of slaty cleavage, especially in the range of coarser grain size.

The oriented growth by the effects of both the anisotropy of intrinsic growth rate of mineral grains and that of the environment in which grains grow, is considered to bring about the preferred orientation of chlorite and illite.

INTRODUCTION

SLATY cleavage is a foliation due to preferred orientation of platy minerals such as chlorite and illite. Several mechanisms of re-orientation have been proposed (Siddans 1972, Wood 1974, Means 1977, Oertel 1983, for review). They can be grouped into two end members: (1) mechanical rotation of unequidimensional particles into parallelism with the *XY*-plane of the finite-strain ellipsoid and (2) syntectonic crystallization, recrystallization and grain growth, concerned with oriented nucleation or oriented growth under deviatoric stress.

The former hypothesis is based largely on the agreement between the preferred orientation of phyllosilicate minerals and that estimated from the March model, which supposes rotation of platelets in a deformation field (Oertel 1970, Tullis & Wood 1975, Tullis 1976, Wood *et al.* 1976, Wood & Oertel 1980). However, several examples of preferred orientation incompatible with the March model have been reported (Holeywell & Tullis 1975, Siddans 1976). From chemical and microstructural studies, it has been suggested that considerable chemical reaction occurs through crystallization and recrystallization during the formation of slaty cleavage (White & Knipe 1978, Stephens *et al.* 1979, Gray 1981, Knipe 1981, White & Johnston 1981). However, the details of the re-orientation process are not clear.

This study investigates the relation between metamorphism and the development of slaty cleavage in argillaceous rocks of the South Kitakami Mountains, northeast Japan. Illite crystallinity and maximum distribution density of preferred orientation of chlorite were used as indicators of the metamorphic grade and degree of development of slaty cleavage, respectively. Special attention was paid to changes in grain size of these mineral species. The processes of grain growth and re-orientation of chlorite and illite were examined with respect to metamorphic grade, degree of development of slaty cleavage and change of grain size.

GEOLOGICAL SETTING

Permian to Lower Cretaceous strata in the southern part of the South Kitakami Mountains (Fig. 1) are composed of shallow marine sedimentary rocks and Lower Cretaceous andesitic volcanics. The stratigraphic details of the Kitakami Mountains are summarized in Onuki (1969, 1981).

These strata were folded, cleaved and intruded by granitic bodies during the early Cretaceous. The axial surface of the folds and slaty cleavage trend N–S to NNE–SSW and dip steeply. Slaty cleavage is normal to the maximum finite shortening axis (Ikeda 1984, Ishii 1985). The development of slaty cleavage is thought to be closely related to a rise in temperature by magmatism (Oho 1982, Ikeda 1984, Ishii 1985).

MICROSTRUCTURE

The argillaceous rocks with slaty cleavage are made up of three microstructural components (Figs. 2a & b) (Ishii 1985). The first comprises grains larger than 20 μ (i.e. 20 μm), most of which are detrital in origin and are mainly composed of quartz, feldspar and muscovite, and minor biotite, chlorite and ore minerals. The second is mica-rich seams which are observed under the optical microscope as dark stripes parallel to the slaty cleavage, mainly composed of fine grained (generally <5 μ) illite, chlorite and dark materials (probably oxides and carbonaceous materials). The third is a matrix of uniformly dispersed fine grains (<10 μ) which are mainly composed of illite, chlorite and quartz. The development of
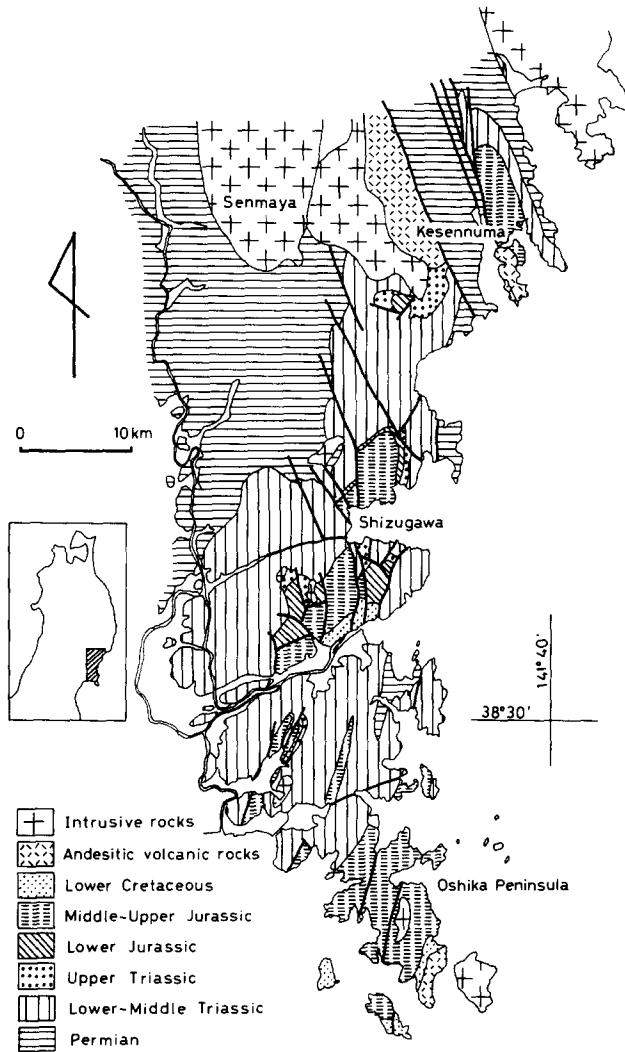
Fig. 1. Geological sketch map of study area.

Legend:
- Intrusive rocks
- Andesitic volcanic rocks
- Lower Cretaceous
- Middle–Upper Jurassic
- Lower Jurassic
- Upper Triassic
- Lower–Middle Triassic
- Permian

Fig. 3. Localities of samples and their IC value (in parentheses). Preferred orientations of chlorite and illite are measured only for samples indicated by closed circles.

mica-rich seams leads to a heterogeneous distribution of mineral species and grain sizes, resulting in a domainal microstructure, which characterizes slaty cleavage. Moreover, Oho (1981) showed that the chemical composition of the mica-rich seams is poorer in SiO_2, CaO and richer in Al_2O_3 and K_2O compared with whole-rock chemistry.

Illites and chlorites in both matrix and mica-rich seams are aligned with their basal planes parallel to the slaty cleavage and are not deformed (Fig. 2b). On the other hand, large detrital muscovite, biotite and chlorite grains are aligned with their basal planes parallel to the bedding plane and are generally deformed to a variable extent (Ishii 1985). The deformed detrital phyllosilicate minerals show bending, kinking or splitting along their basal plane (Figs. 2c–e); spaces formed by splitting are filled by illite or minor chlorite, which are rarely deformed. These texural features of the composite grains resemble 'chlorite–mica aggregates' widely reported from slaty rocks (Roy 1978, van der Pluijm & Kaars-Sijpesteijn 1984, Woodland 1985, Gregg 1986). They are thought to be formed by deformation of detrital phyllosilicate minerals synchronous with crystallization of illite and chlorite. Around some rigid grains, such as framboidal
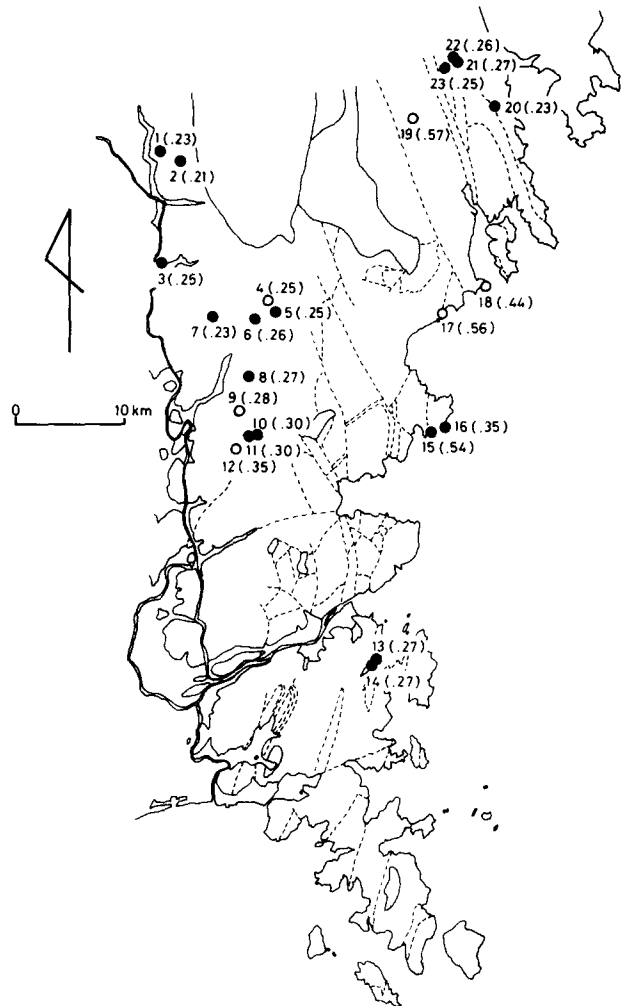
pyrite, there develop pressure fringes composed of illite or chlorite whose basal planes are aligned parallel to the slaty cleavage (Ishii 1985).

The microstructures described above indicate crystallization and/or recrystallization of illite and chlorite synchronous with deformation associated with the formation of slaty cleavage. Slaty cleavage is not developed in the argillaceous rocks distributed between Senmaya and Shizugawa (Oho 1982). These rocks have no mica-rich seams and phyllosilicate minerals are weakly aligned with their basal planes parallel to the bedding plane.

X-RAY ANALYSES

Samples of Permian argillaceous rocks were collected from localities indicated in Fig. 3. The samples were crushed and three size fractions ($<2\ \mu$, 2–6 μ, 6–20 μ) were separated for X-ray diffraction studies: illite crystallinity, chlorite composition and X-ray diffraction intensities of chlorite and illite. These were carried out using an X-ray diffractometer XD-3 (Shimadzu Seisakusho Ltd), with Ni-filtered Cu–Kα radiation, 35 kV × 20 mA. Each sample was analysed three times and a mean value determined.
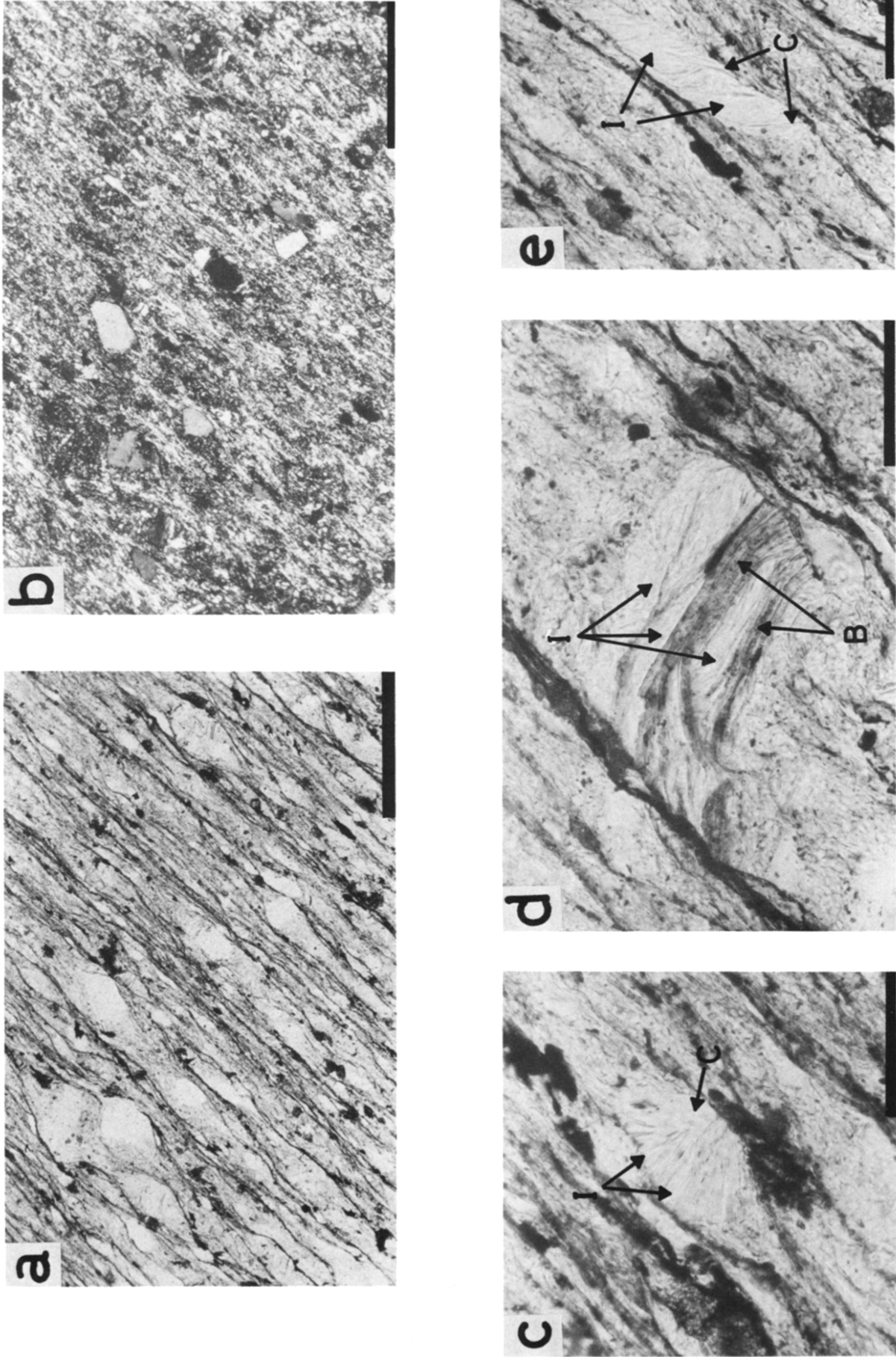
Fig. 2. (a) and (b) Photomicrographs of argillaceous rock with slaty cleavage: (a) open nicol; (b) crossed nicols. Scale bars are 200 μ. (c)–(e) Photomicrographs of composite grains: C, chlorite; B, biotite; I, illite. Scale bars are 50 μ.
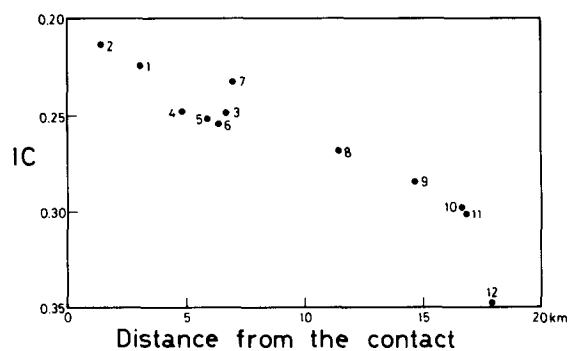
Fig. 4. Relation of IC to the distance from the Senmaya granitic body. The numbers show their localities (Fig. 3).

Fig. 5. I(14 Å)–I(7 Å)–I(4.7 Å) triangular diagram.

Illite crystallinity

Illite crystallinity (Weaver 1960, Kubler 1968) is defined as the sharpness of the illite 10 Å peak on the X-ray diffractogram. This sharpness reflects the grade of metamorphism and has been used widely in studies of low-grade metamorphism (e.g. Frey et al. 1980, Kisch 1980a,b, Duba & Williams-Jones 1983, Weaver 1984, Merriman & Roberts 1984). In this paper, peak width at half-height expressed in degrees 2θ is used as an indicator of sharpness to define the illite crystallinity index (IC); higher metamorphic grade is expressed as a smaller value of IC.

The measurement was carried out on oriented samples of $<2\,\mu$ size fractions (prepared by smearing onto a glass slide and allowing to air-dry) at the following X-ray diffractometer settings: slit system $0.5 \times 0.25 \times 0.5$ mm; time constant 5 sec; scan speed $0.5°\,2\theta$/min.

Figure 4 shows the relationship of IC to distance from the Senmaya granitic body; metamorphic grade increases toward the granitic body. This strongly suggests that IC in this area reflects the thermal effects of the granitic body rather than diagenesis. However, this correlation is not clear over all the study area (Fig. 3). For example, samples from localities 13 and 14 show relatively low IC values in spite of being far from exposed granites, possibly due to the influence of buried granites.

Argillaceous rocks adjacent to the granites have metamorphic minerals such as biotite, cordierite, andalusite and garnet. The ICs of these rocks were not measured, because the diffraction peak of biotite overlaps that of illite. The biotite isograd is estimated to correspond to IC $\simeq 0.22$.

Chlorite composition

Chemical composition of chlorite can be determined from lattice spacing and relative X-ray diffraction intensities of basal planes (Bailey 1972, Brown & Brindley 1980). Relative X-ray diffraction intensities of chlorite (001), (002) and (003) reflect the chemical composition and their distribution in structural sites within the chlorite crystal. Oinuma et al. (1972) expressed these relations on a I(14 Å)–I(7 Å)–I(4.7 Å) triangular diagram.
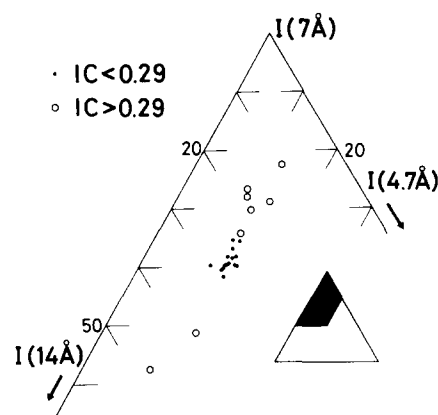
Following their method, relative X-ray diffraction intensities of basal planes of chlorites were measured on random samples (prepared by packing $<2\,\mu$ size fraction into an aluminium holder) at the following X-ray diffractometer settings: slit system $0.3 \times 0.2 \times 0.5$ mm; time constant 2 sec; scan speed $1°\,2\theta$/min. The peak height of the diffractogram was regarded as the diffraction intensity.

The results (Fig. 5) are widely scattered, but those of IC < 0.29 are distributed in a limited area on the diagram. No correlation can be recognized between IC and the relative X-ray diffraction intensities among the samples of IC > 0.29. This indicates that chlorites with a variable chemical composition were homogenized by the metamorphism and this homogenization occurred abruptly across the metamorphic grade of IC $= 0.29$.

Grain size change of chlorite and illite through metamorphism

From the relation between IC and grain size of chlorite and illite, their change of size as a result of the metamorphism can be investigated. To this purpose, X-ray diffraction intensities (peak areas) of chlorite and illite (002) were measured on oriented samples of three size fractions; $<2\,\mu$, 2–6 μ, 6–20 μ. The diffraction intensity is proportional to the volume in the sample, but it is also affected by the chemical composition of the mineral and the absorption coefficient of the sample. Therefore, the difference in diffraction intensities for each size fraction only roughly reflects the change of grain size.

Figure 6 shows variation of the X-ray diffraction intensities of chlorite and illite in relation to IC for each size fraction. The diffraction intensity of chlorite for the $<2\,\mu$ size fractions is independent of IC, but that for the $>2\,\mu$ size fractions increases abruptly in the field of IC $<$ 0.29. In the case of illite, the intensity for the $<2\,\mu$ size fractions decreases abruptly in the field of IC < 0.29, but that for $>2\,\mu$ size fractions is independent of IC.

Figure 7 shows relationships between the ratio of the diffraction intensities of each size fraction and IC. For both chlorite and illite, the ratio of the diffraction intensity of $>2\,\mu$ size fraction to that of $<2\,\mu$ size fraction
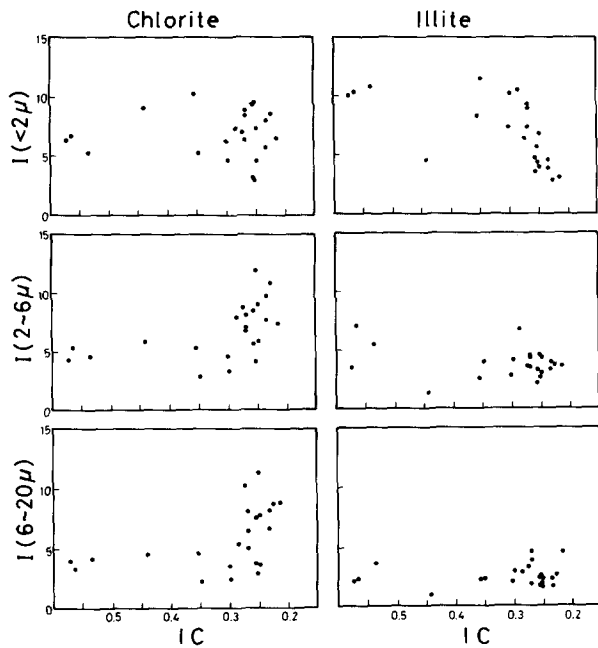
Chlorite   Illite

Fig. 6. Relation of the X-ray diffraction intensities of chlorite and illite to IC.

increases in the field of IC < 0.29. Here we should remember that the diffraction intensity reflects the volumetric proportion of chlorite or illite in each size fraction and not in the sample as a whole. The studied samples tend to show that the lower values of IC are associated with smaller contents of <2 μ size fraction. Taking this into account, for the bulk rock sample, the increase in the proportion of chlorite and illite of >2 μ size in the field of IC < 0.29 would be more intense than that shown in Figs. 6 and 7.

Consequently, it is concluded that both chlorite and illite grains are coarsened in the field of IC < 0.29,
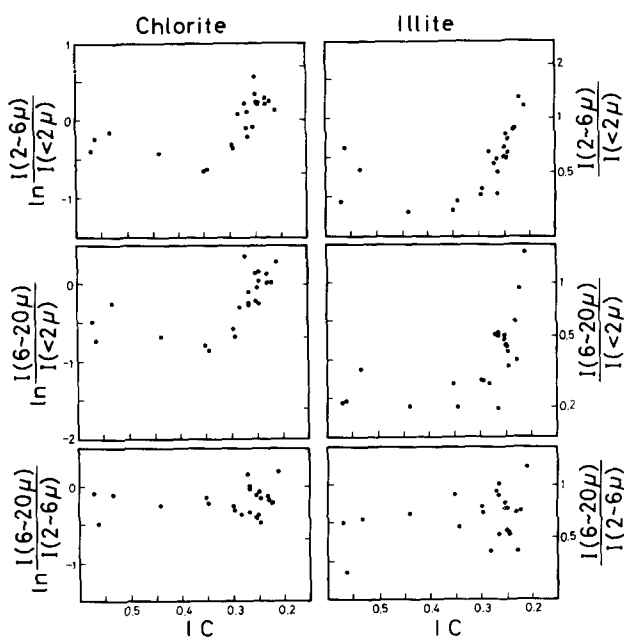
Chlorite   Illite

Fig. 7. Relation of ratio of the X-ray diffraction intensities of chlorite and illite of different size fractions to IC.

compensated to some extent by the decrease of smaller grains. But the coarsening mode is somewhat different between chlorite and illite; that is, a minor decrease of smaller grains and major increase of larger ones for chlorite, compared with illite. Such differences may be attributed to the dominant grain growth of illite at the expense of smaller grains and the dominant crystallization of chlorite grains larger than 2 μ. In order to examine the change in amount of chlorite and illite in the bulk rock, their diffraction intensities were measured for powdered rock samples. No correlation was found between the diffraction intensity of illite and IC, but the diffraction intensity of chlorite shows a tendency to increase with lowering IC. This result supports the interpretation mentioned above.

It is to be emphasized that IC = 0.29 is a critical value not only for the homogenization of chlorite composition, but also for the coarsening of chlorite and illite grains.

Preferred orientation of chlorite and illite

The preferred orientation of chlorite and illite (002), indicated by closed circles in Fig. 3, were measured in transmission mode (Decker et al. 1948) using a diffractometer fitted with a universal sample holder and pinhole slit (the divergent and receiving slit were 1 mm and 3 mm in diameter, respectively). The beam used was Ni-filtered Cu–Kα radiation, 40 kV × 40 mA.

The measured samples were thin sections with μt = 1.1–1.5 (μ: linear absorption coefficient, t: thickness of thin section). No absorption correction was done, because it is smaller than 8% for the ranges of μt = 1.1–1.5 and tilting angles up to 35° (Decker et al. 1948). Since only one thin section was measured for a sample, the whole area of a pole figure cannot be covered. To calculate the mean intensity, intensities in the unmeasured area were estimated by extrapolation, but the calculated mean intensity is precise enough, due to low intensities in the unmeasured area.

Figure 8 shows the pole figures which represent the preferred orientation of basal planes of chlorite and illite. Chlorite and illite in most of the samples exhibit preferred orientation with orthorhombic symmetry and their distribution densities are a maximum in orientations parallel to the pole of slaty cleavage (Fig. 8a). There is little difference between chlorite and illite in both the pattern and degree of preferred orientation. In several samples, illite exhibits preferred orientation with monoclinic symmetry, skewing toward the bedding plane (Figs. 8b & c). Even in such samples, however, chlorite exhibits preferred orientation with orthorhombic symmetry. In samples without slaty cleavage, distribution densities are a maximum in orientations parallel to the pole of bedding (Fig. 8d).

In the present paper, ϱ_{max} is the ratio of the maximum density of chlorite to mean density, and is used as an indicator of the degree of development of slaty cleavage.

Figure 9 shows variation of ϱ_{max} in relation to IC. Open circles denote samples without slaty cleavage, and hence, their ϱ_{max} reflects the preferred orientation paral-

Chlorite   Illite
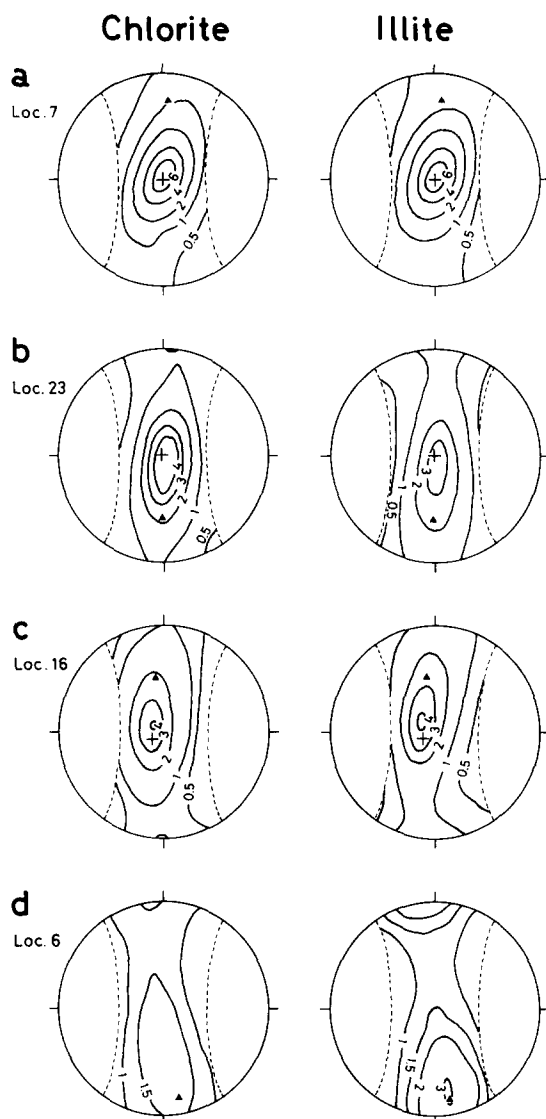

a
Loc.7

b
Loc.23

c
Loc.16

d
Loc.6

Fig. 8. Pole figures of basal planes of chlorite and illite. Contour is ratio of distribution density to mean density. Cross and triangle indicate pole to slaty cleavage and bedding plane, respectively.

Fig. 9. Relation of ϱ_{max} to IC. Open circles indicate samples with maximum distribution density oriented to bedding plane. The numbers show their localities (Fig. 3).

between ϱ_{max} and the ratio of the diffraction intensities of different size fractions is shown in Fig. 10. Four samples of low metamorphic grade in Fig. 9 were excluded to minimize the effect of metamorphic grade. For both chlorite and illite, the ratio of the diffraction intensity for 6–20 μ size fractions to that for 2–6 μ size fractions increases with ϱ_{max}. This indicates that, in the range of grain sizes larger than 2 μ, the proportion of larger grains increases with the development of slaty cleavage.

DISCUSSION

The results described above represent two distinct features of the behavior of phyllosilicate minerals during metamorphism and development of slaty cleavage. The first is that the homogenization of chlorite composition, coarsening of chlorite and illite grains and development of a high degree of preferred orientation occurred abruptly beyond the same critical metamorphic grade

lel to the bedding plane. In this figure, high ϱ_{max} values are limited to the field of IC < 0.29, suggesting that the metamorphism is a prerequisite for a high degree of the preferred orientation. The same critical value is recognized for the homogenization of chlorite composition and for the coarsening of chlorite and illite grains (Figs. 5–7). In other words, some metamorphic processes such as crystallization, recrystallization and grain growth of chlorite and illite are indispensable for a high degree of the preferred orientation.

On the other hand, in the field of IC < 0.29, the degree of the development of slaty cleavage varies widely, including no development of slaty cleavage. This indicates that other factors such as strain and stress state are also important to the development of slaty cleavage.

Grain size change of chlorite and illite through the development of slaty cleavage

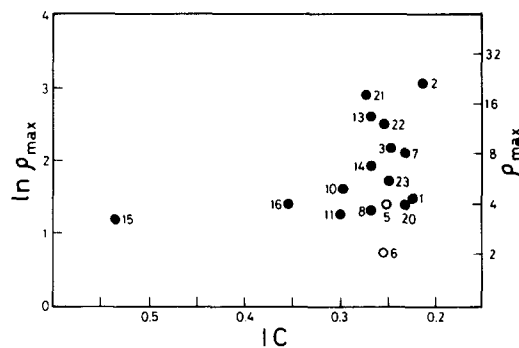To examine the grain size changes of chlorite and illite through the development of slaty cleavage, the relation
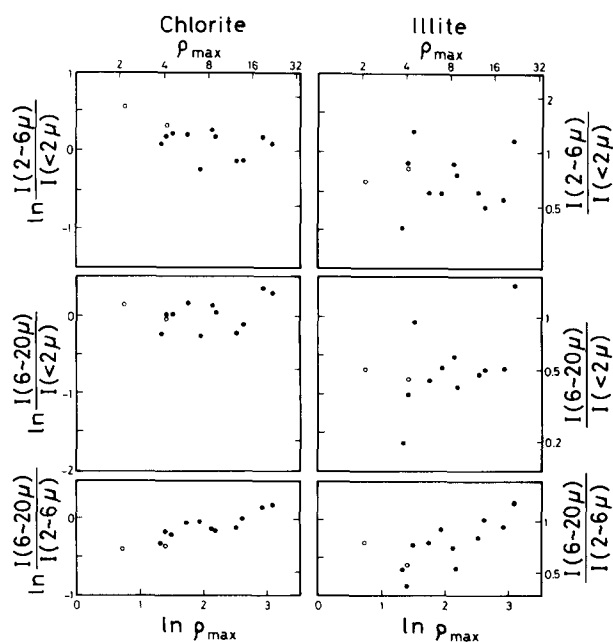

Fig. 10. Relation of ratio of the X-ray diffraction intensities of chlorite and illite of different size fractons to ϱ_{max}. Open circles indicate samples with maximum distribution density oriented to bedding plane.

corresponding to IC = 0.29. The second is that the coarsening of chlorite and illite grains correlate not only with the metamorphic grade but also with the degree of preferred orientation. These results give some suggestions on the mechanism of the re-orientation process of phyllosilicate minerals through the development of slaty cleavage, as discussed below.

Mechanical rotation

Because of their platy shape, chlorite and illite will rotate mechanically toward parallelism with the *XY*-plane of the incremental strain ellipsoid. For the studied samples, however, mechanical rotation does not explain the following observations:

(1) the distinct difference of the degree of preferred orientation across the metamorphic grade of IC = 0.29 (cf. Etheridge *et al.* 1974);

(2) the contrast in orientation and deformational features between small phyllosilicate grains in the matrix and mica-rich seams and large detrital phyllosilicate grains, suggesting that the former are not original but newly formed by syntectonic crystallization and recrystallization;

(3) the heterogeneous distribution of chemical components, mineral species and grain size in the domainal microstructure.

Process of grain coarsening

There are three different processes which affect the grain size of metamorphic minerals (Tullis & Yund 1982):

(1) dynamic or annealing recrystallization, the driving force for which is a reduction of the internal strain energy;

(2) crystallization (neomineralization), a process involving the nucleation of a new phase. The driving force for this process is a reduction of the chemical free energy;

(3) grain growth independent of the above two processes. The driving force for this process is a reduction of interfacial free energy.

In the studied samples, coarsening of chlorite and illite grains through the metamorphism was advanced at the expense of smaller grains. It is considered that grain size was mainly controlled by the third of the above processes, because grain coarsening at the expense of smaller grains reduces the total area of the grain surface and boundary. There may be some contribution by crystallization, especially to the grain size of chlorite, because chlorite was homogenized in composition and increased in total amount through the metamorphism.

Heterogeneous distribution of chemical components (Oho 1981), as well as the existence of pressure fringes and composite grains, indicates that considerable mass transfer occurred at least to distances greater than $100\,\mu$, which is similar to the spacing of mica-rich seams and the size of detrital grains. This is 10–100 times the grain size in the matrix and mica-rich seams and strongly suggests

mass transfer through the intergranular fluid. Grain growth of chlorite and illite is considered to be accelerated by increase in the diffusivity of components in the intergranular fluid. Any diffusive process is thermally activated and is reflected in the abrupt coarsening of chlorite and illite grains beyond the metamorphic grade of IC = 0.29.

The coarsening of chlorite and illite grains also correlates with the degree of preferred orientation and this is revealed in a larger grain size range than that with the metamorphic grade (Figs. 6, 7 and 10). This difference suggests that the growth of smaller grains (i.e. early growth stage of each grain) was controlled by the metamorphic grade and that the growth of larger grains (i.e. later growth stage of each grain) was also affected by the deformation which generated slaty cleavage. Namely, the further growth of larger grains was accelerated by the deformation. This can be attributed to grain growth driven by a reduction of interfacial free energy being more effective on smaller grains (Tullis & Yund 1982).

Similar relationships have been reported from the experimental study of synthetic phlogopite by Etheridge *et al.* (1974). According to them, the number and size of coarse grains increases with increasing strain, and these coarse grains, which show no obvious deformation features, are highly oriented normal to the shortening axis. These results and those of the present study suggest that deformation has a considerable effect on the grain growth.

The results of Etheridge *et al.* (1974) appear to indicate that coarse grains grow at the expense of strained grains to reduce the internal strain energy. In the case of the studied sample, however, reduction of the internal strain energy is considered to be a minor contribution to grain coarsening, because of the overall undeformed nature of small phyllosilicates in the matrix and mica-rich seams.

Re-orientation of chlorite and illite

The development of preferred orientation of chlorite and illite is associated with grain coarsening (Fig. 10). Further, the abrupt increase in the degree of preferred orientation of chlorite beyond the metamorphic grade of IC = 0.29 (Fig. 9) suggests that slaty cleavage also developed through a thermally activated process as well as grain growth. Thus, the development of preferred orientation can be regarded as a process of selectively oriented grain growth as follows.

The platy form of phyllosilicate minerals indicates that their intrinsic growth rate is anisotropic and is greatest in directions parallel to basal plane (Etheridge *et al.* 1974, Rosenfeld 1985). Non-hydrostatic stress will generate another anisotropy of equilibrium solubility of solid components in the enclosed fluid at grain surfaces (Paterson 1973). It will make the growth rate of grains greatest at the surface normal to the minimum compressive stress axis and least at the surface normal to the maximum compressive stress axis, even if the intrinsic

growth rate of the mineral is isotropic. In addition, foliation and layering produce an anisotropy of diffusivity, which may also cause anisotropy of growth rate of grains.

By the effect of these anisotropies, growth of phyllosilicate minerals whose basal planes are oriented normal to maximum compressive stress can be selectively accelerated. The preferred orientation produced by this oriented growth itself promotes oriented growth more effectively, resulting in the correlation between ϱ_{max} and grain coarsening of chlorite and illite.

Difference in behavior between chlorite and illite

Chlorite and illite behaved in nearly the same manner during the metamorphism and development of slaty cleavage. However, several differences are recognized. The difference in the mode of grain coarsening through metamorphism is probably due to the difference in thermodynamic character of both minerals in a metamorphic condition.

Another difference between chlorite and illite is the pattern of preferred orientation of several samples. Only illite exhibits preferred orientation with monoclinic symmetry (Figs. 8b & c). As mentioned before, large detrital grains of muscovite are aligned with their basal planes parallel to the bedding plane. The micas (10 Å minerals) in the samples can be grouped into two by their orientation; parallel to the bedding plane and parallel to the slaty cleavage. Micas of each group may exhibit preferred orientation with either axial or orthorhombic symmetry, but the whole sample exhibits monoclinic symmetry. Where this is so, the pattern of preferred orientation of whole micas depends on: (1) the relative amount of micas in each group; (2) the pattern and degree of preferred orientation of micas in each group; and (3) the angle between the slaty cleavage and the bedding plane. In general, during the development of slaty cleavage, the amount and degree of preferred orientation of micas which belong to the cleavage-parallel group increase and the angle between the slaty cleavage and the bedding plane decreases. Thus, preferred orientation with monoclinic symmetry is limited in the samples with low degree of preferred orientation ($\varrho_{max} <$ 5.7).

The studied samples rarely contain large detrital chlorite grains and thus bedding is not a controlling factor in the overall preferred orientation of chlorite. Preferred orientation of chlorite in the sample without slaty cleavage (Fig. 8d) appears to be in a transitional state to that with slaty cleavage. This also indicates the weaker effect of bedding for chlorite compared with illite.

Therefore, the preferred orientation with monoclinic symmetry is a result of superposition of subfabrics; grains in each group differ in origin, one is detrital and the other is metamorphic. Consequently, the overall preferred orientation cannot generally correlate with total strain (cf. Etheridge & Oertel 1979).

The different behavior of chlorite and illite during cleavage formation has been pointed out by several authors. The differences are revealed in the degree of the preferred orientation (Siddans 1976, 1977) or the pattern of the preferred orientation (Holeywell & Tullis 1975, Weber 1981). The data of Holeywell & Tullis (1975) and Weber (1981) indicate a stronger effect of bedding on the preferred orientation of chlorite than that of illite, contrasting to the present study. This may be related to the difference in original content of both minerals and metamorphic condition during the cleavage formation.

CONCLUSIONS

(1) Thermal metamorphism due to granitic bodies in the South Kitakami Mountains is clearly recognizable where IC < 0.29. It is characterized by the homogenization of chlorite composition and the coarsening of chlorite and illite grains in argillaceous rocks.

(2) The development of slaty cleavage was accompanied by crystallization, recrystallization and grain growth of chlorite and illite. The degree of the preferred orientation also changed abruptly beyond the metamorphic grade of IC = 0.29.

(3) The coarsening of chlorite and illite grains correlates with not only metamorphic grade but also with degree of preferred orientation.

(4) Preferred orientation of chlorite and illite is considered to develop by oriented growth due to the effect of both anisotropy of intrinsic growth rate of mineral grains and anisotropy of environment in which grains grow.

(5) Difference in behavior of chlorite and illite are revealed in the mode of their grain coarsening and in the contribution of original bedding fabrics to the pattern of overall preferred orientation.

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* In Japanese with English abstract.
† In Japanese.