Geochemical Study of the Sambagawa Metamorphic System in the Besshi District, Central Shikoku, Japan (Part I) : Variation of Chemical Compositions of the Schists, Related to the Local Tectonic Phenomena

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Part I

Variation of Chemical Compositions of the Schists, Related to the Local Tectonic Phenomena

By

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Abstract

Based upon the field evidences and the chemical data on about two hundreds and fifty rock-speicmens, the following problems concerning the Sambagawa metamorphic system in the Besshi district have been systematically investigated: (1) variation of the bulk chemical composition related to such local tectonic phenomena as folding, shearing and lineation (Part I); (2) mobility and fluctuation-tendency of the chemical component, relationships of the chemical composition to the metamorphic-grade and to the mineral assemblage, with special respect to the metamorphic environment (Part II); and (3) ore genesis of the bedded cupriferous pyritic ore deposits in the district (Part III).

The followings can be said as the summary of Part I: (1) bulk chemical composition of the crystalline schists, belonging to the same stratigraphic horizon and to the same mineralogical facies, fluctuates more or less with their situation, even in the closely-spaced samples, (2) the crests of the small-scale folds being composed of basic schists are commonly higher in the contents of SiO_2, Al_2O_3, FeO, and Na_2O, and lower in Fe_2O_3, MgO and CaO, compared to the wing, (3) FeO, MgO, H_2O and CO_2 increase, and SiO_2 Fe_2O_3, CaO, Na_2O and TiO_2 decrease in the central part of the shear zones of later stage, compared to the marginal part.

Introduction

Historical review: In the Besshi mining district, Central Shikoku, Japan, the

Sambagawa crystalline schists system develops with a wide area and the typical rock association, ranging from the greenschist facies to the amphibolite facies. About a hundred studies concerning stratigraphy, petrology, structure, mineralogy, tectonics, geophysics and ore genesis of this district have been published during past half century. Nevertheless, a unanimous view covering the region has not been obtained. As the result of extensive and precise geological surveys throughout the surface and the underground, which had been recently carried out by the Geologic Section of the Besshi Mine, in co-operation with the Hiroshima and the Tokyo Universities, several doubtful points in the earlier studies were settled. On the other hand, divergence of opinions on the regional structure, the metamorphism and the oregenesis become clearer and sharper than in the earlier studies.

On the geological structure of the district, some geologists, represented by M. DOI (1961), interpret it to be essentially simple, and the others, represented by K. HIDE (1961), emphasize its complexity which is partly indicated in several large-scale recumbent and tight folds such as the Shirataki anticline.

On the metamorphism, several petrological investigations have been carried out in these ten years under the influence of modern conceptions proposed by ESKOLA, TURNER, SANDER and so on, and thus comparatively generalized metamorphic zoning have been established (HIDE 1961, MIYASHIRO & BANNO 1958). On the metamorphic condition, although clear opinions have been rarely presented, two contrasting assumptions, i. e. the deep environment and the shallow tectonic environment, are prevailing. Available chemical data of the metamorphic rocks in the district are very few, so that geochemical discussions on the metamorphic condition are rather rare. From chemical viewpoint, SUZUKI (1930) emphasized the effect of original composition on the mineral assemblage of the Sambagawa system in Shikoku, (See also READ, 1957). In his discussion, about ten chemical data of the various rocks from the Besshi district were used. Recently, BANNO (1961) denied metasomatic process during metamorphism by means of partial analyses for Na_2O, K_2O etc, with a number of samples from the Besshi district.

On the ore-genesis of the bedded pyritic ore deposits in the district, the familiar controversy between syngenetist and epigenetist has been repeated. In these days, however, syngenetic theory is rather prevailing on the basis of the continuous features and the stratigraphic conformities of ores, (DOI, 1962).

Retrospecting a number of previous studies both on metamorphism and ore genesis, paucity of the available chemical data, especially on the bulk compositions of the rocks, is keenly realized.

Purpose of the study: The author was engaged in the surface and underground geological survey of the district, from 1954 to 1961, as a geologist of the Besshi Mine. During those years, the author was interested in several subjects stated below.

Since 1961, the author has been engaged in the laboratory works including chemical analyses and synthetic experiments with regard to the rocks of the Besshi district. This series of studies is mainly concerned with the chemical studies. The final purpose of these studies is to explain mechanism of the Sambagawa metamorphism from geological and geochemical standpoints. As the first approach to this purpose, fluctuations or variations of the bulk chemical compositions of the metamorphic rocks were investigated in the following local cases:

a) Fluctuation within small limited area in the same rock,
b) Lateral variation limited within the same rock formation,
c) Variation near the contact-plane between different rocks,
d) Variation related to the folding phenomena,
e) Variation related to the shearing phenomena.

Successively, from more general and regional viewpoint, the following problems have been investigated:

f) Relationships of one component to the others in fluctuation of chemical composition.
g) Mobilities of chemical components in the regional metamorphism,
h) Relation between the chemical composition and the metamorphic-grade,
i) Original rocks of the metamorphic rocks,
j) Relation between the chemical composition and the special mineral association.

On the ore genesis, the following subjects have been complementarily investigated:

k) Variation of chemical composition in the rock related to ore,
l) Variation of chemical composition of the schists, which were caused by the later-stage hydrothermal process.

The present paper is the first of a series and concerned with the subjects a) to e). Other subjects, i. e. f) to l), will be described in the succeeding papers.

It may be desirable to treat the chemical data with mathematical statistics. The total number of the analized samples, 249, however, is not sufficient yet for the mathematical treatment, because a number of factors involved in the regional metamorphism should be taken into account in sorting or stratifying of the sample-lot. Therefore, the graphic method only is used in the present paper.

The abbreviated words in the papers are explained in Table 1.

Acknowledgement: In field survey, the author has been rendered many helpful suggestions by Prof. J. KOJIMA, Dr. K. HIDE, and Dr. G. YOSHINO, Hiroshima University, Prof. T. WATANABE and Dr. S. BANNO, Tokyo University. Throughout his laboratory works, Prof. H. YOSHIZAWA, Dr. T. UEDA[1], and Dr. N. NAKAYAMA, Kyoto University,

1 The author is greatly indebted to Prof. H. YOSHIZAWA and Dr. T. UEDA for reading the manuscript.

Table 1. Explanations of the abbreviated words.

Ab	albite	G	graphite	Pump	pumpellyite
Act	actinolitic hornblende	GL	glaucophanitic amphiboles	Py	pyrite
Amf	amphibolite	Gt	garnet	Q	quartz
band	compositional band	Hb	common hornblendes	Rod	rhodonite
bl	black	Hem	hematite, specularite	R	massive sulphide ore
Bt	biotite minerals	Hor	horizon	Se	sericite
C	chlorite	Hw	hanging-wall	SK	banded impregnated ore
calc	calcareous, carbonatious	imp	impregnated, disseminated	sp	albite spotted
Cc	calcite, carbonates	Ky	kyanite	Serp	serpentinite
cg	coarse-grained, >1.5 mm	-L	level	...S	...schist
comp	compact	Ls	schistose limestone	S_1	plane parallel to rock boundary or original bedding plane
Cp	chalcopyrite	mas	massive	S_2	axial plane of close fold or common schistosity plane
cx-cut	cross-cut	mg	meduim-grained 0.5∼1.5 mm	S_3	slip-cleavage plane or peculiar schistosity plane
dg	dark green	Mag	magnetite	SG	specific gravity
Di	diopsidic pyroxenes	Mgr	massive green rock	Zoi	zoisite
Ep	epidote	Mus	white micas		
F	formations	P	piedmontite		
fg	fine-grained, <0.5mm	Per	peridotite		
fil	phyllitic	pg	pale green		
fQ, fCc	segregated Q, Cc veins	Po	pyrrhotite		
Fw	footwall				

encouraged him. Staffs of the Besshi Mine, Sumitomo Metal Mining Co., Ltd., and Staffs of the Shirataki Mine, Nihon Kogyo Co., Ltd., gave him the constant help during sampling. To these people, the author wishes to express his sincere thanks for their kindness.

General Geology—Geological Background.

Outlines of geology of the Besshi district are summarized in Table 2 and Figures 1 to 4. For the convenience of the succeeding discussion, brief explanations are made as follows.

Stratigraphy: Besshi mining district is composed of basic schists, pelitic schists and amphibolites, with minor siliceous schists, ultrabasic rocks, eclogite and bedded pyritic ore, which belong to the Yoshinogawa Series, Sambagawa System. The Sambagawa System develops along the outer zone of the southwest Japan and shows the widest distribution (about 25 km in N-S direction) in the Central Shikoku.

In the northern part of the district, the system contacts unconformably with the Izumi Sandstone Group of Cretaceous age, and in the southern margin it is distinguished from the Mikabu System of lower metamorphic grade by the Kiyomizu

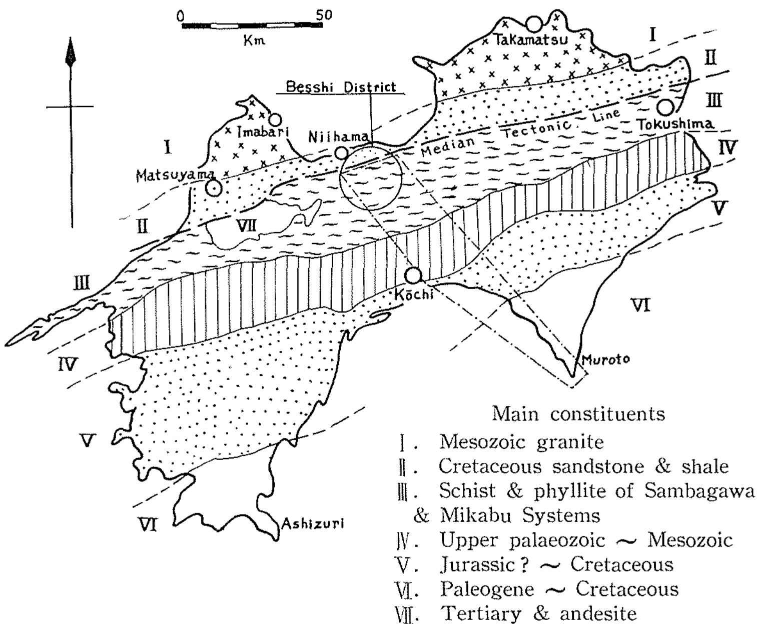


Fig. 1. Outline of geology, roughly classified, of shikoku, Japan. A chain-line encloses the area in which the sampling has been carried out.

tectonic zone which is a steeply-dipping shear-zone along the phyllitic black schists.

Stratigraphic classifications of the system in the district have been made by several authors (OGAWA 1902, OZAWA 1926, SATO 1938, KOJIMA 1951). Above all, HIDE's classification (HIDE 1961) seems to be the most precise and acceptable as a standard. For convenience of the present study, however, a little modification is added to HIDE's classification and new nomenclatures which are named according to type-localities near the Besshi Mine are given to a few formations.

Absolute age of the Sambagawa metamorphism is estimated to be 82×10^6 years from biotite in the district (MILLER, J. and others 1961).

Petrography: Metamorphic rocks in the district are so completely recrystallized that no original texture is found. Detailed petrography has been described by several authors, (SUZUKI 1930, HORIKOSHI 1938, SATO 1938, YOSHINO 1961, etc.). To avoid duplication, petrographic features of the analized samples are briefly expressed together with chemical data and sketches.

Geological structure: Many of divergent opinions have been presented on the regional structure (OGAWA 1902, OZAWA 1926, SATO 1938, DOI 1961, HIDE 1961). Refering to Figure 3, however, the regional structure can be briefly summarized as follows. As a regional scale, elongated domes, e. g. Nakahichiban dome, Akaboshi

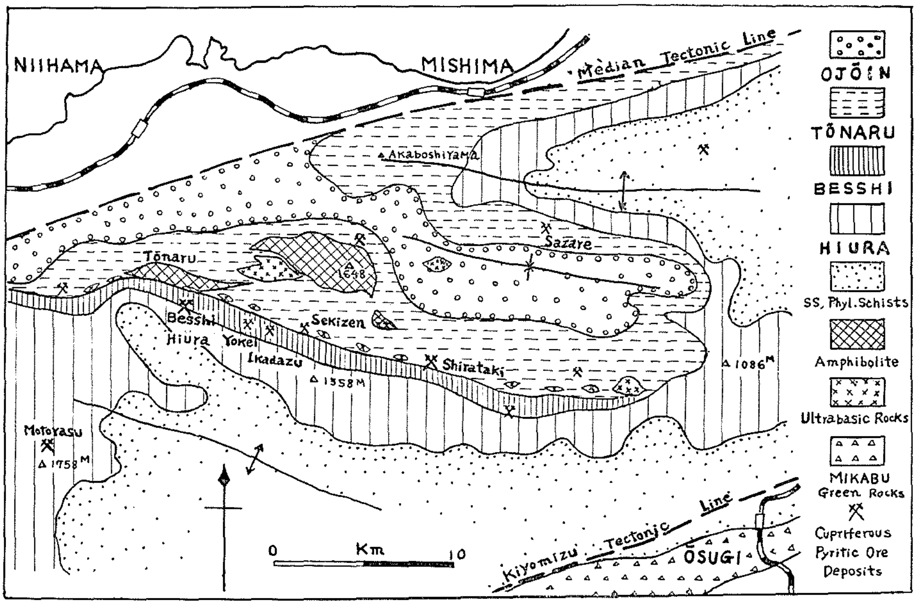


Fig. 2. Simplified geologic map of the Besshi district, Central Shikoku.

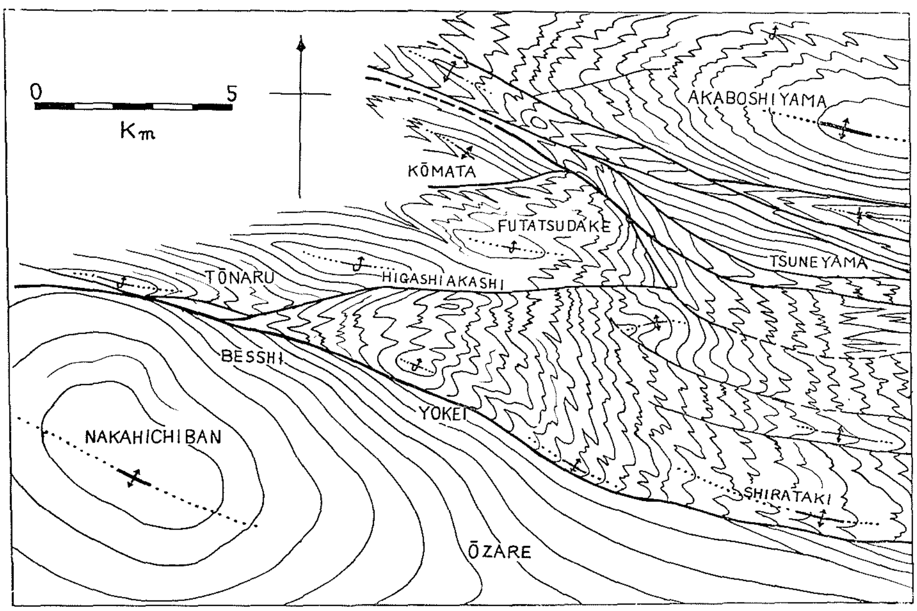


Fig. 3. Simplified structural map of the Besshi district, showing the general trend of principal schistosity. Projection of the schistosity was made on the 700—1400 M plans, according to lineation map.

Table 2. Summary of general geology, structure, tectonics and metamorphic zoning in the Besshi district.

		Izumi Sandstone	Median Dislocation Line	(Upper) Yoshinogawa Group, Sambagawa Metamorphic System				(Lower)	Kiyomizu Tectonic Zone	Mikabu
Formation				Ōjōin	Tōnaru	Besshi	Hiura			
Thickness				apparent 4000 M+ estimated 1000 M+	apparent ? estimated several hundreds M	500～600 M	400～500 M	1000 M+		
Rocks				G-S, Q-S, Talc-S, Amf, Serp.	Hb-S, Amf, G-S, Q-S, P.Q-S, LS, Serp.	Act. Ep. C-S, Gl-S, G-S, Q-S, P.Q-S, LS.	Act. Ep. C-S, Gl-S. Q-S, G-S, C-S.	G-S, Q-S, C-S phyllitic S.		C-S, LS, phyllites
Ore Deposits				none	Shirataki, Sekizen, Ehime, Iyo etc.	Besshi, Yokei, Ikadazu, etc.	Hiura, Kannabe, Shimokawa etc.	none		none
General	Strike			N30°-130°W, average EW	N20°-120°W average EW	N40°～80°W, general EW	N0°～180°W, general N70°W	general N70°W		
	Dip			30°-70°N, occasionally S-dip	30°-75°N, occasionally S-dip or ⊥ al	30°～75°N, general 45°N	0°～75°N	irregular general N-dip		
Tectonics	Folding			complicated strong folds	complicated strong folds	warping & interformational tight folds	warping & interformational tight folds	complicated open folds		?
	Faulting			large-scale thrusts & normal faults	large-scale thrust faults; normal: rare	parallel thrust faults & oblique normal faults	thrust faults: rare normal faults: common	small-scale thrust & normal		
	Schistosity			S_1, S_2, S_3, etc.	S_1, S_2	$S_1 \doteq S_2 \doteq S_3$ (S_3: rare)	S_1 : S_2 : S_3	$S_1 \neq S_2 \neq S_3$		$S_1 \neq S_2 \neq S_3$
	Lineation			various directions	N60°～110°E (0°～40°). N50°～80°W (0°～35°)	S60°～90°E(5°～35°), locally N80°E(10°), N35°W(55°) etc.	S70°～85°E (10°～20°)	S80°～90°E (0°～10°)		indistinct
Metamorphic Facies				Amphibolite Facies	Epidote Amphibolite Facies	Greenschist Facies	Greenschist Facies, locally Glaucophane Facies	Greenschist F. Glaucophane F.		Greenschist Facies
Spot-, Non-spot-				Ab.-spotted zone (coarse-grained texture)		Ab.-non-spotted zone (fine-grained texture)				
Index Minerals in Basic Schists	Chlorite			- - - - -	- - - - -	———	———	———		
	Actinolitic Hornblende			- - - - -	- - - - -	———	———	———		- - -
	Glaucophanitic Amphiboles					- - -	———	———		-
	Common Hornblende			- - - - -	———	- -				
	Epidote			- - - - -	- - - - -	———	———	———		
	Zoisite			- - - - -	———	- - - -				
	Garnet			- - - - -	———	- - - - -				
	Pyroxenes				- - ———	- -				
	Kyanite				— —					
	Biotite			- - - - -	- - - - -	- -				
	Serpentine			- - - - -	- - - - -	- - -				

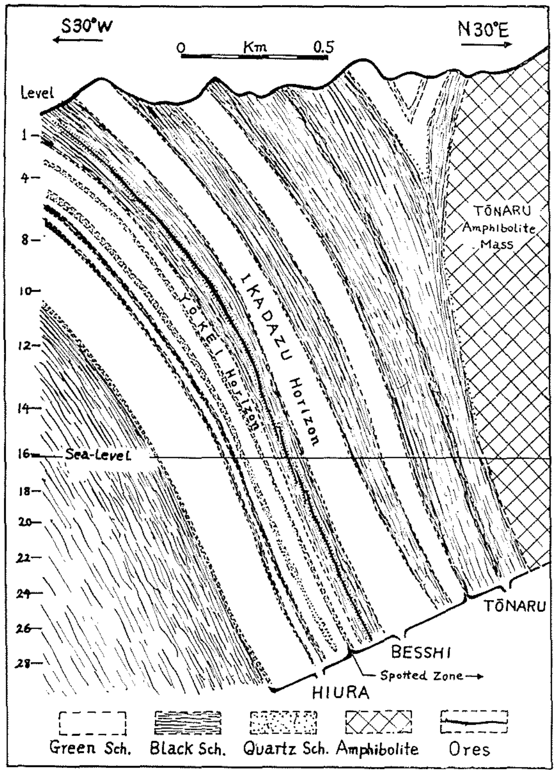


Fig. 4. Vertical geologic section through the Besshi Mine. The grade of metamorphism increases towards the north, i. e. towards the shallower horizon. To the depth, any of conspicuous petrological changes, except for the decreases of thickness and grain-size of the schists, is not observed.

anticline, Nishinokawa anticline, develop sporadically or rather *en echelon*, and synclinal parts or basins between these domes show complicated folded and sheared structures, in which repeating and branching of rock-layers are very common.

Metamorphic zoning and facies classification: The grade of metamorphism increases northwards, i. e. towards apparently shallower depth, from the greenshist facies up to the amphibolite facies[2].

Lines or planes of equal metamorphic grade are in general nearly parallel to the regional geological trend. For example, the lower limit of albite-spotted zone can be traced along the Yokei horizon for a long distance, (Fig. 2, 4).

Sampling and Chemical Analyses

Sampling: Rock sampling, including the reference samples from other district, were limited within the area illustrated in Figure 1. In most cases, exposed rocks on the ground surface were more or less weathered[3], and therefore underground sampling in several mines were mainly adopted. In the surface samplng, localities were restricted only to the fresh exposures along several mountain-routes which had been recently cut.

The following sampling methods were used according to place and purpose:

2 On this subject, several detailed examinations from geochemical standpoint will be described in the succeeding papers. For the present purpose, these rough classifications will be enough.

3 Even in a fresh sample from the ground-surface, leachings of carbonates are occasionally observed under the microscope.

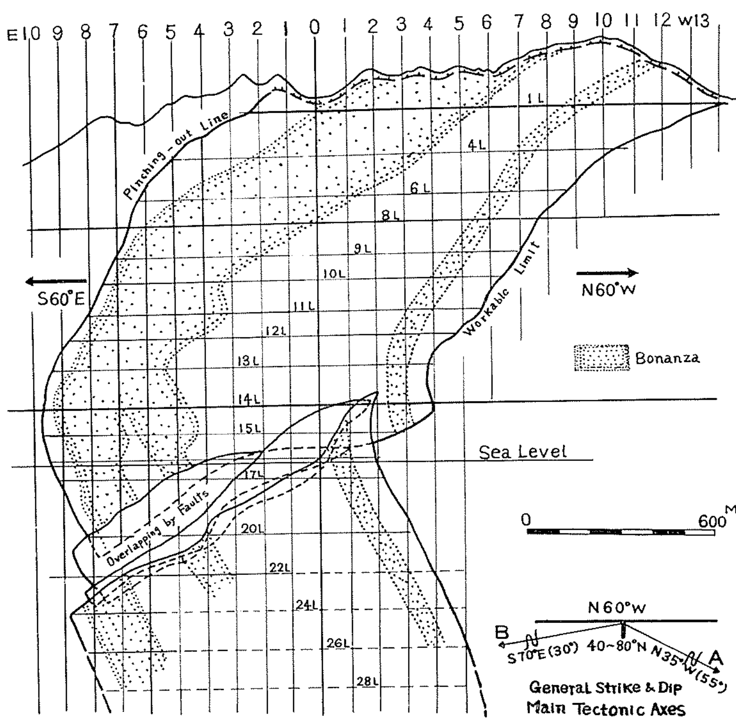


Fig. 5. Longitudinal projection of the Besshi ore deposit. Average dip is 40° at the outcrop, steepening to 70° in depth. Ore was assured down to −900 M level in 1962.

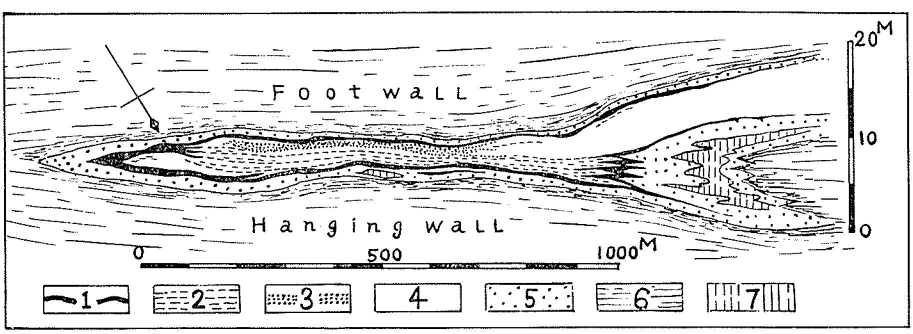


Fig. 6. Horizontal section through the Besshi ore deposit, showing a generalized feature on the levels from 14-L to 22-L. On the upper levels the feature is to be modified in some degree. 1–Massive sulphide ore. 2–Banded ore of sulphides impregnated in greenschist. 3–Massive green rock being composed mainly of chlorite and magnetite. 4–Greenschist. 5–Quartz schist. 6–Black schist. 7–Calcareous schist.

1) Channel sampling—Samples were obtained by cutting a channel (5 cm in width, 3 to 5 cm in depth) perpendicularly to bedding schistosity along the length of underground exposure. This method was applied mainly to phyllitic schists for the purpose of investigating the problems (b) (c) and (k) stated in Page 51.
2) Chip sampling—To prevent as much as possible the influence of heterogeneous character of a schistose rock, minimum size of sample at one spot was fixed to 5 cm × 5 cm in dimension parallel to schistosity and to 20 to 25-times of the maximum thickness of compositional band in thickness. In equigranular rocks, minimum chip-size was fixed to about twenty times of the diameter of maximum crystal. Chip sampling was applied for the subjects (a) (c) and (d) in Page 51.
3) Core Sampling—Whole bore-core samples were obtained from the places lacking fault and fold. The core-recoveries in the sampled boreholes were 95 to 100 per cent and the core-angles were 60° to 90° to the schistosity plane.
4) Composite sampling—With interval of a twentieth of the entire length of exposure, blocks or chips defined above were collected and thus about twenty blocks were mixed into one sample.

Preparation of powder-samples: Core- and several block-samples were splitted by a diamond saw. All samples were washed, dried in the sun, crushed by jaw crusher under 1 cm across in size and quartered into 200 to 400 grams. These crude samples were crushed by a steel mortar and quartered in two steps, i. e. under 50 mesh, into 40 to 50 grams and under 65 mesh, into 10 to 15 grams and then were ground into the size under 200 mesh and desicated at 110°C for several hours. In firm rocks such as quartz schist and eclogite, contaminations of impurities from the grinding apparatus were so conspicuous that corrections of the chemical data were made[4]. Most rocks, however, were easily crushed and these contaminations were imperceptibly small.

Chemical analyses: Contents of SiO_2, Al_2O_3, Fe_2O_3, FeO, MgO, CaO, Na_2O, K_2O, TiO_2, MnO, P_2O_5, H_2O^+, CO_2, S, and Cu of all samples were determined by following methods[5]: (1) gravimetric—SiO_2, Al_2O_3, MgO, CaO, S, Cu; (2) volumetric—Fe, FeO, P_2O_5; (3) colourimetric—MnO, TiO_2, (4) flame-photometric—Na_2O, K_2O. Special care was taken to determine the H_2O-content. It was determined by BRUSH and PENFIELD's methods as shown in Figure 7, at heating conditions defined as 5 min. at 25°—300° C, 10 min. at dull red of the ignited powder colour and 10 min. at white or yellow glow, corresponding the tempereture 1000°—1100°C. H_2O^+ contents of the samples of every other number were checked off by the same method, but

4 For example, total amount of impurities from crusher and mortars were 0.7% in Fe and 1 to 2% in SiO_2 in case of eclogite.

5 Specific gravities were determined by suspension of rock blocks in water.

the differences were in general negligible small. CO_2 were qualitatively tested by pouring of the hot 6N-HCl solution on the rock powders and then calculated from the following formula:

$$CO_2\% = \text{ignition loss}\% + 0.111 \cdot FeO\% - 0.625 \cdot S\% - H_2O\%$$

Value for TiO_2 contains some amounts of V_2O_5 which is, as a rule, higher in the hematite bearing rocks. P_2O_5 in most rocks, except in the hematite bearing rocks, are less than 0.5%, so that P_2O_5 are not taken into account in this paper. Total weight percentages are within the limit 100±1% in most samples except for those containing much sulphur[6].

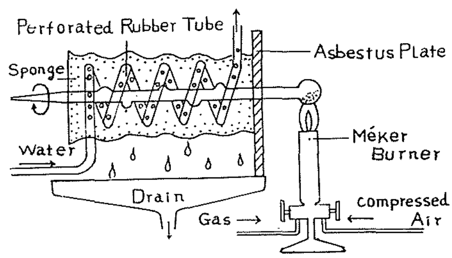


Fig. 7. Schematic illustration of the modified apparatus for H_2O-determination.

Owing to limited space, the whole data can not be numerically expressed. In the present paper graphic representation only is adopted.

Fluctuation within Narrow Limited Area

In cases of igneous rocks, it is often practised to regard a single chemical datum obtained from a typical handspecimen as the average chemical composition of the massive rock with a certain dimension. For the study on metamorphism from geochemical standpoint, it seems to be first necessary to ascertain dimension of the sample-lot in which an analytical value can properly represent the mean value. In addition, if chemical equilibrium in the rock of a certain dimension is attained differentially according to time and space, chemical heterogeneity of the rock will be intensified by the metamorphic processes. With respect to these matters, following three cases were examined by means of bulk chemical analyses.

1) Non-spotted, comparatively homogeneous greenschists, belonging to the Hiura formations near the Besshi Mine, (Sketch: Fig. 8-A, Variation diagram: Fig. 9-A).
2) Spotted hornblende schists belonging to the Tōnaru formations near the Shirataki Mine, (Sketches: Fig. 8-B, Fig. 8-C, Variation diagrams: Fig. 9-B, Fig. 9-C).
3) Spotted black schists belonging to the Besshi formations near the Besshi Mine, (Sketch: Fig. 8-D, Variation diagram: Fig. 9-D).

In each case, the rock was cut into several blocks as shown in Figure 8 by a diamond saw.

The results obtained from these data are summarized as follows:

1) Bulk chemical compositions of the closely-spaced rocks fluctuate in some

6 In most cases, errors arose from impurities from glass ware during pecipitation of hydroxides.

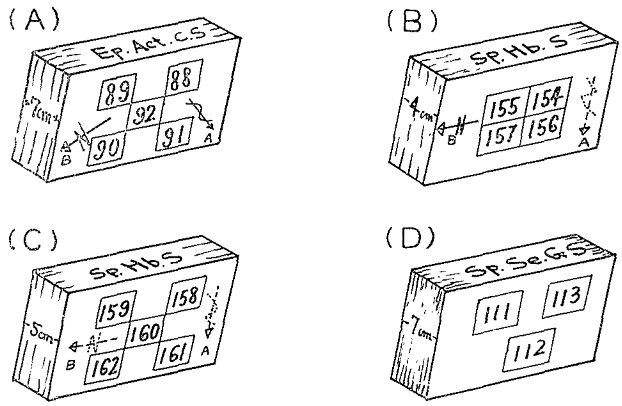


Fig. 8. Diagrams showing the location of closely-spaced samples. A broken arrow indicates indistinct or estimated tectonic axis. (See Fig. 9)

degree, or in other words, even in the rocks having a similar petrographic features, chemical values are not equal in strict sense. Therefore, it may be said that crystalline schists are chemically heterogeneous, irrespectively of dimension of the rocks. This result should be taken into account in sampling and also in geochemical discussion on metamorphism.

2) Since changes of mineral associations are not recognized in each case, fluctuations of chemical compositions seem to correspond to those of quantities of the constituent minerals.

3) Relation between lineation and chemical composition varies according to the sampling locality. In case of the Hiura greenschist, fluctuation is small in direction of tectonic B-axis which is indicated by lineation L_{12}[7]. On the contrary, in the Shirataki hornblende schists[8], the detectable chemical trend related to lineation is found not in the case that L_{12} strongly develops[9], but in the case that linear structure is indistinct[10]. These chemical variations related to the oriented character of the schists suggest that geochemical tendencies as well as the oriented mineral arrangement may offer some informations about the tectonic career of the schists.

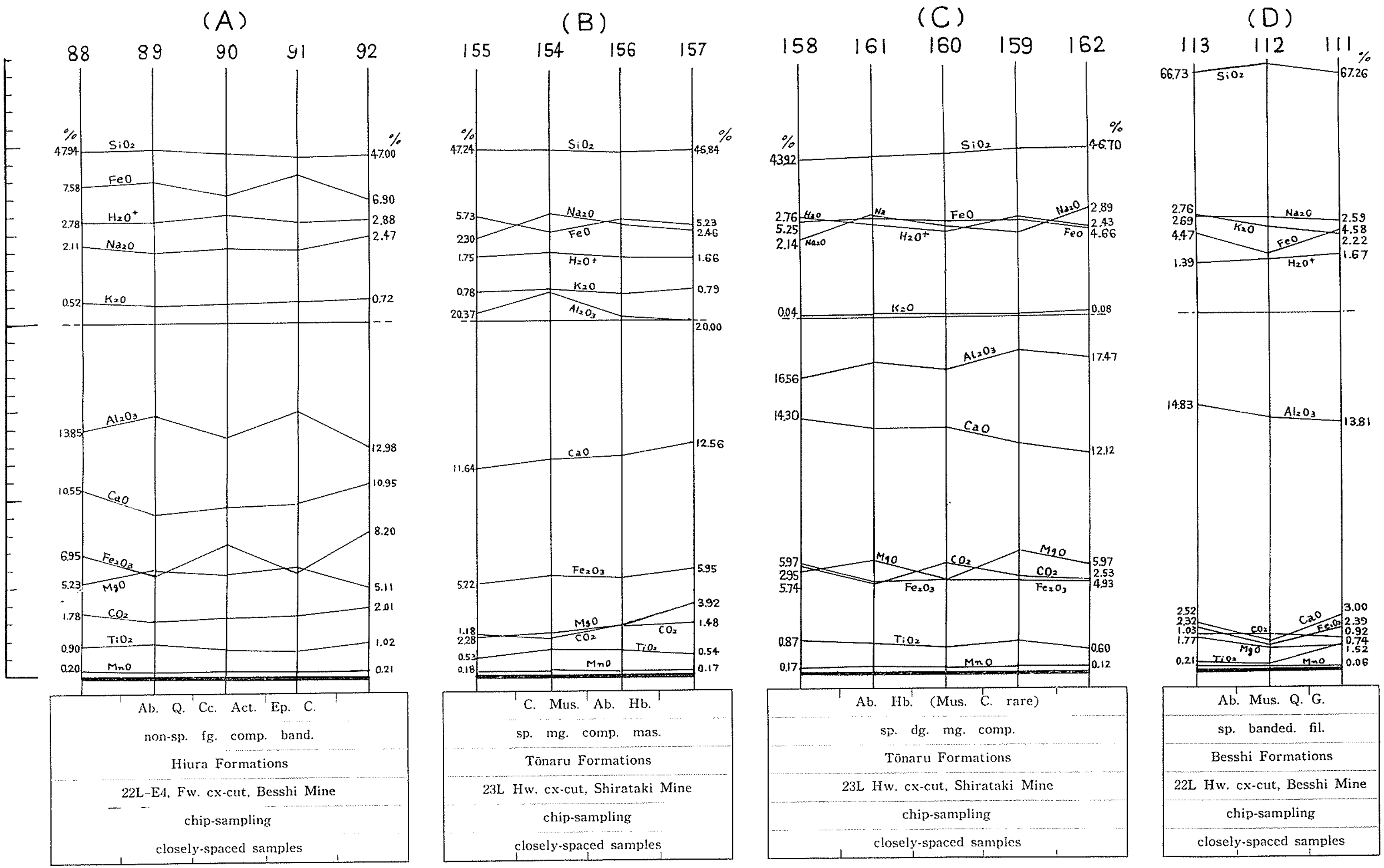
7 That is, No. 88, 92, and 90 show similar chemical compositions.

8 Secondary or later-stage folding is generally notable in the Shirataki hornblende schists. This folding is comparatively gently arcuated with the axis being parallel or oblique to the lineation L_{12} and characterized by retrogressive change of hornblende into filmy chlorite.

9 That is, differences between No. 154 and No. 155, and between No. 156 and No. 157, are rather remarkable.

10 That is, No. 158 is rather similar to No. 161 than to No. 159.

Fig. 9. Diagrams showing variation of chemical composition within narrowly limited area. The scale on the left side corresponds to 5% for SiO_2, to 1% for FeO, Al_2O_3 CaO, Fe_2O_3 MgO, and to 0.5% for H_2O, Na_2O K_2O, CO_2 TiO_2 MnO. (See Fig. 8)



Lateral Veriation of Chemical Composition, Limited within a Single Rock Formation

It is not futile, especially in prospecting for new ore-bodies, to examine whether the bulk chemical composition can be used or not as an indicator for correlation of a stratigraphic horizon or an isograd in the structurally complicated district. Following two cases were selected for the investigation.

1) Vein-forming greenschists of the Besshi ore deposit. Schematic features of the Besshi ore deposit are illustrated in Figure 4 to 6. The greenschists are sandwitched between massive sulfide ores and pinched out towards the east, together with ore. Towards the west, with diminishing of ore, they stretch laterally for several kilometres with thickness of 10 to 20 metres. Core- and channel-samples which show the same mineral association—i. e. Ab. Ep. Cc. Q. C. Act.—, and the equal structural situation, were obtained at different levels, (Localities: Fig. 5, Fig. 6, Variation diagram: Fig. 11-A)

2) Greenschists of the Yokei horizon, which is adjacent to the lower limit of the spotted zone, extend laterally for 25 kilometres or more with thickness of 20 to 50 metres[11]. Characteristic features within this horizon are the development of interformational tight folding and laminated structure, intermingling of greenshists of various kinds and so on. Several greenschist blocks having the same mineral association were collected at random from different localities, (Localities: Fig. 10, Variation diagram: Fig. 11-B)

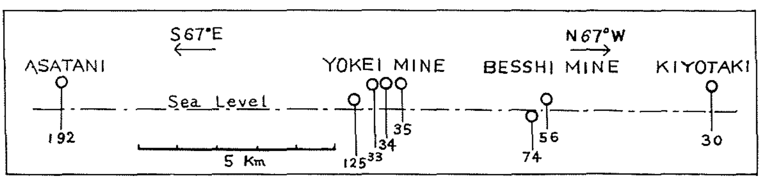


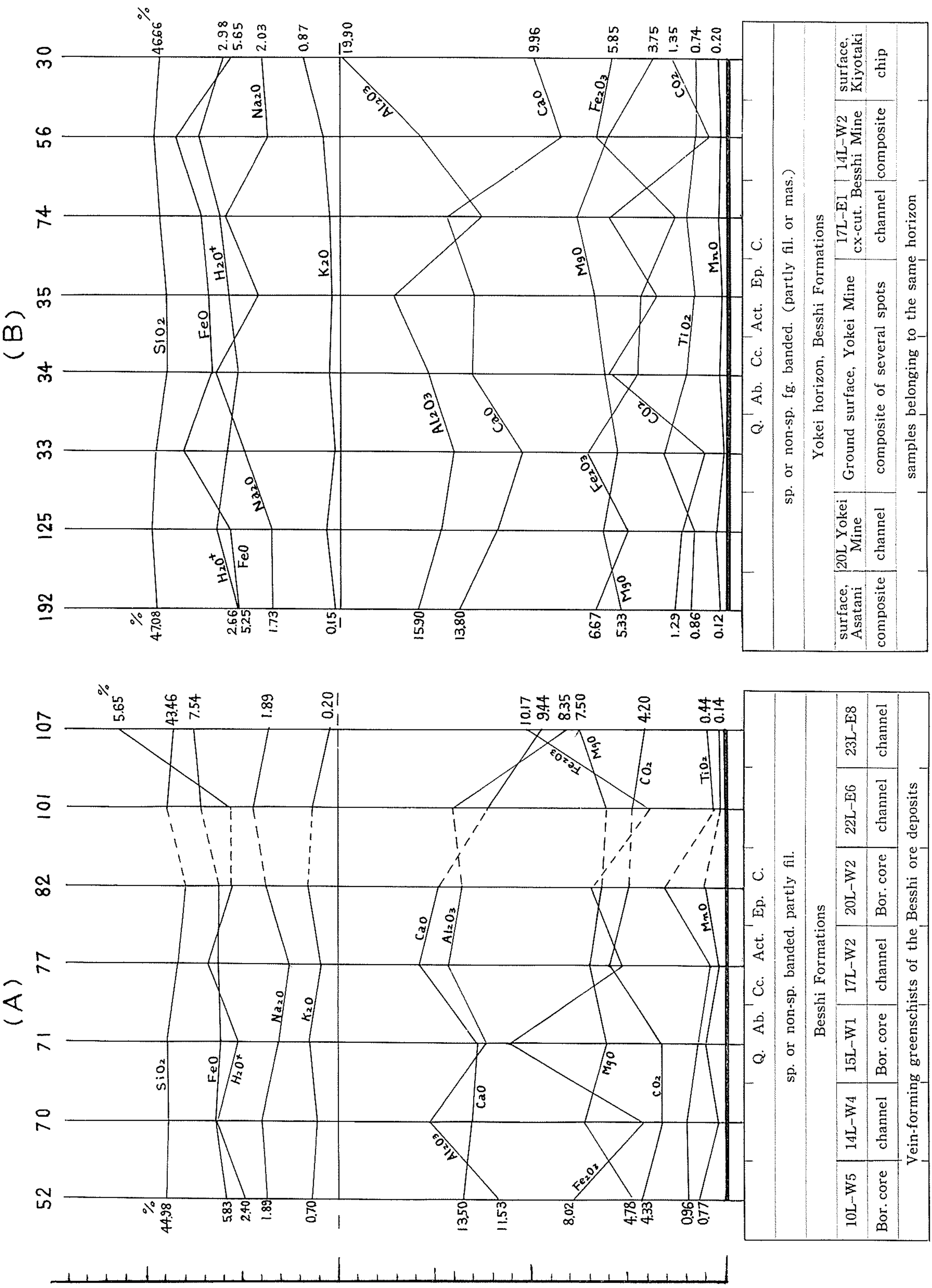
Fig. 10. Vertical projection showing location of the samples from the Yokei greenschists.

Summarizing the chemical tendencies which are recognized in the variation diagrams, it is understood that fluctuations of chemical compositions of the rocks, which belong to the same stratigraphic horizon and, at the same time, to the same mineralogical facies, are so large that bulk chemical composition itself can not be used as an indicator for correlation of a horizon and probably an isograd[12].

11 As a general trend in the district, thickness of a formation increases towards a crest of anticline.

12 Although fluctuation of each component is large, relative ratios of contents of several special components are nearly constant throughout a horizon, if mineral associations are constant. This interesting tendency is most clearly expressed in $H_2O \cdot CO_2$—$FeO \cdot CaO$.—Al_2O_3 diagram which is reserved for another occasion. (See Part II)

Fig. 11. Diagrams showing variation of chemical composition of the greenschists belonging to the same stratigraphic horizon. (A) Vein-forming greenschist of the Besshi Mine. (B) Greenschist of the Yokei horizon. The scale is equal to that of Figure 9, except for CO_2 in diagram (A) in which it corresponds to 1% for CO_2.



Variation near the Contact Plane between Different Rocks.

Many studies have been carried out to find the regularities in chemical variations related to magmatic differentiation and also to contact metamorphism during igneous process. The original chemical regularities in the parent rocks, which have been found in these studies, may be reserved during regional metamorphism, or may be destroyed to form new tendencies which is different from original one, through redistribution of chemical components.

As for field and microscopic evidences, contact planes between various rocks are as a rule very sharp, and gradual or transitional petrographic changes are hardly recognized, even in such an ordinary case that the boundary is parallel to the bedding schistosity.

Concerning this problem, chemical investigation was carried out in those cases of (1) amphibolite-mass of the Tōnaru formations, (2) hornblende schists at the Shirataki Mine, (3) so-called metagabbro of the Mikabu system, and (4) Muroto gabbro. In former two cases which are concerned with the Sambagawa metamorphism, regular variations from center to margin of the rock-bodies are not observed, and fluctuation of every component is remarkable and irrespective of situation of the specimen[13,14,15,16].

Chemical Variation Related to the Folding Phenomena

If there are any difference in plastic properties of the constituent minerals and in mobilities of the chemical components of rock, pressure gradient or stress in

13 Owing to limited space, data will be expressed in the succeeding papers. (Part IV).

14 In special cases related to mineralization, transitional variations of chemical composition near the contact planes are clearly observed. (Part III).

15 Occurence and genesis of biotite in the Besshi district: Biotite locally concentrates in the pelitic schist adjacent to amphibolites and ultrabasic rocks in the Ojoin- and the Tōnaru-formations. Biotite bearing schists from the area are higher in MgO-, FeO-, H_2O-contents than normal pelitic schists (Part II). Genesis of biotite may be attributed to metasomatic process during intrusion of the parent rock which is considered to be gabbro by some petrologists. The author, however, regards the biolite minerals as the products being connected with serpentinite intrusion at the latest stage of the regional metamorphism. Biotite minerals occur in close association with ultrabasic rock rather than with amphibolite itself. Chlorite, talc and white micas abruptly increase along the biotite zone. These evidences will supports author's assumption. Biotite, obviously post-metamorphic, occurs in the hydrothermally altered greenschist of the Besshi formations. Therefore, the author considers that absolute age (82×10^6 years) obtained from the biotite schists from the district corresponds not to the main stage of the Sambagawa metamorphism, but to the latest stage.

16 There is no strong evidence, both in field and laboratory, to demonstrate whether the siliceous and calcareous schists, developing along the boundary between basic- and pelitic-schists, are sedimentary origin or metamorphic one.

folding process will promote the tectonic differentiation, and thus divergence of chemical composition between the crest and the wing of a fold will be intensified.

The following phenomena being accompanied with the folded structures in the district are occasionally observed within the same single layer of rock, irrespectively of scale of the structures:

i) Porphyloblast of albite is enriched and much coarser in the crest part. For example, the porphyloblasts of albite, with the size of 1 to 2 cm across are found near the crest of the Shirataki anticline.

ii) Magnetite, Mn-garnet and alkali-hornblendes increase near a crest or a trough of the tight fold being composed of calcareous quartz schist.

iii) In the synclinal part of strongly folded area within the Tōnaru formation, ordinary greenshist-layers grade into lenticular masses of amphibolite and albite-rich rock, (Fig. 12).

Differences of chemical compositions between crest and wing of the small-scale folds being composed of the basic schists were investigated in the following three cases:

1) Gently warped fold in the Shirataki hornblende schists: Difference between the crest and the wing is not rocognized both in thickness and mineral association, (Sketch: Fig. 13-A Chemical data: Fig. 16-A).

2) Moderately arcuated fold in the Shirataki hornblende schists: The crest is more or less thicker and more massive than the wing part, but no mineralogical difference is observed, (Sketch: Fig. 13-B, Chemical data: Fig. 16-B).

3) Tight fold at a structurally complicated part in Tōnaru formation: Difference

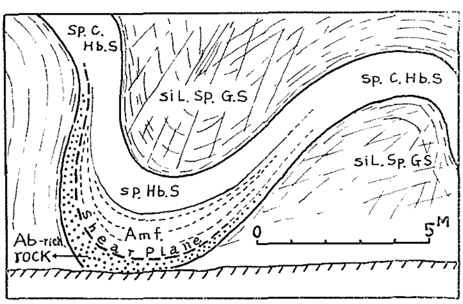


Fig. 12. Sketch showing localized amphibolitization of a greenschist at a trough of fold, near the Sekizen Mine.

Table 3. Difference of chemical composition between crest and wing of a fold being composed of basic schist. Plus sign indicates that the content in crest is higher than in wing, and minus sign is a contrary indication to a plus sign.

In Fig. 16	(A)	(B)	(C)	Average
SiO_2	+	+	+	+
Al_2O_3	+	+	+	+
Fe_2O_3	−	−	−	−
FeO	+	+	+	+
MgO	−	−	−	−
CaO	−	−	−	−
Na_2O	+	+	+	+
K_2O	−	+	−	±
H_2O^+	+	−	+	±
CO_2	+	−	−	±
TiO_2	+	−	constant	±
MnO	+	−	−	±

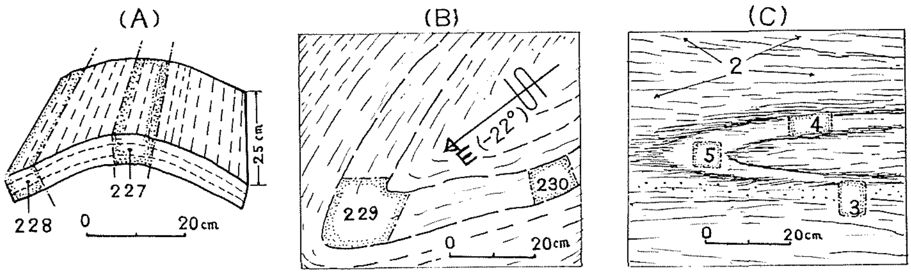


Fig. 13. Sketches of folds, showing location of the samples.

between the crest and the wing is remarkable both in thickness and mineral association, (Sketch: Fig. 13-C, Chemical data: Fig. 16-C).

Table 3 shows the summary of the chemical tendencies which are recognized in the variation diagrams. As a result, it can be said that the crest of fold being composed of basic schists is, compared to the wing, higher in contents of SiO_2, Al_2O_3, FeO and Na_2O and lower in those of Fe_2O_3, MgO and CaO.

Variation of Chemical Composition, Related to the Shearing Phenomena

Effects of shearing stress on recrystallization of minerals during regional metamorphism have been noticed by many authors. Nevertheless, actual mechanism of the effects has not been explained in quantitative sense. According to BELL, who has succeeded in synthesizing of $Al_2O_3 \cdot SiO_2$ polymorphs from unhydrous starting materials by means of shearing technique (BELL, 1963), significances of shear in the synthetic experiment are attributed to (1) bond breakage, (2) new surface exposure and (3) stored strain energy contributing to increasing the activation energy[17].

Shearing pocess in natural metamorphic system seems to play the same role as in the mineral synthesis. In the Besshi district, there are many features related to shearing process. A number of shear faults can be classified according to their scales, directions, arrangements, throws, stages, relationships to igneous activities of the later stage, and so on. However, geochemically interesting fact related to shearing is that, along a shear zone which is nearly parallel to stratigraphic trends, the center part is filled with mafic hydrous minerals and the marginal part is either more siliceous or rich in segregated quartz-lense in many cases (Fig. 14, 15).

Following two cases were selected to investigate the difference of chemical compositions between the center and the margin of shear zone:

17 Personal communication.

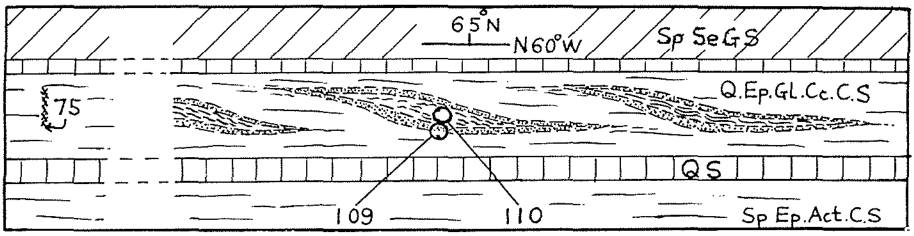


Fig. 14. Schematic illustration of the shear zone, nearly parallel to the rock-boundary, Yokei greenschist, 23-L, E 4, Footwall drift, Besshi Mine.

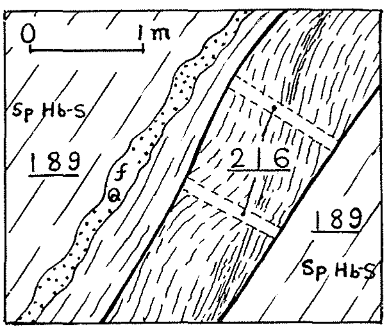


Fig. 15. Sketch of a shear zone in the hornblende schist, 23-L, Hangingwall cross-cut, Shirataki Mine.

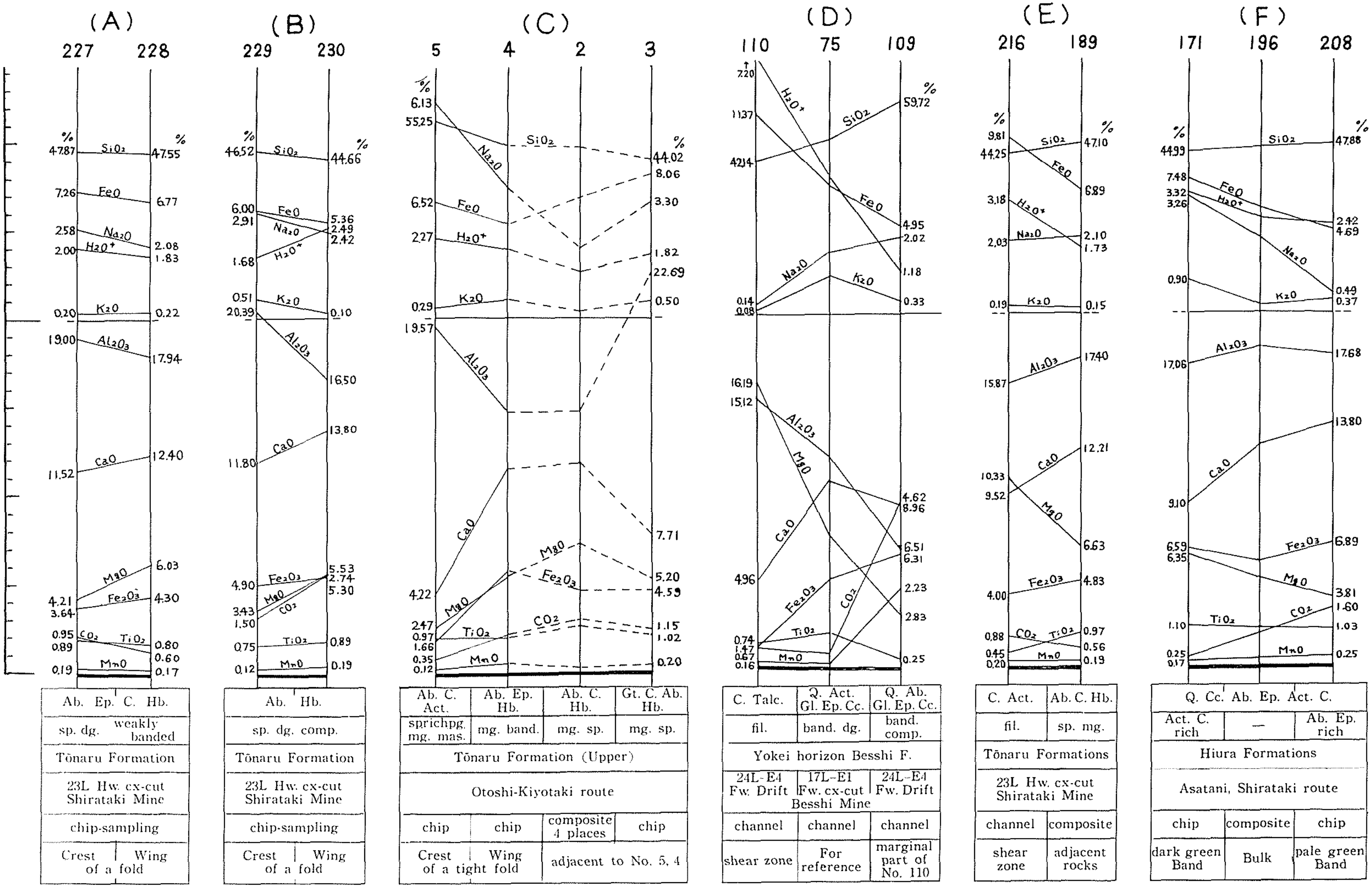
Table 4. Anomalys of chemical composition in basic schist, observed in shear phenomena. Plus and minus signs indicate "higher or increase" and "lower or decrease" respectively in shear zone, compared with marginal part.

In Fig. 16	(D)	(E)	Average	(F)
SiO_2	−	−	−	−
Al_2O_3	+	−	±	−
Fe_2O_3	−	−	−	?
FeO	+	+	+	+
MgO	+	+	+	+
CaO	−	−	−	−
Na_2O	−	−	−	+
K_2O	−	+	±	+
H_2O^+	+	+	+	+
CO_2	+	+	+	−
TiO_2	−	−	−	+
MnO	+	+	+	−

1) Shear zone in the greenschists of the Yokei horizon.—Parallel shear zones arranged discontinuously or *en echelon* are occasionally observed in the greenschist of the Yokei horizon. They are presumed to have been caused by development of interformational tight foldings which are common along this horizon. From the evidences of secondary schistosity and linear structure, which are different from slickenside, in the shear zone, it is deduced that their stages are earlier than those of oblique faults. Mineralogical features are described in Figure 16-D. Sample No. 75 was selected for comparison as an average of the unsheared rocks in this horizon, (Sketch: Fig. 14, Chemical data: Fig. 16-D).

2) Shear zone in the hornblende schists of the Shirataki horizon.—A shear zone, sub-parallel to bedding schistosity, is filled with pale-green chlorite, and the

Fig. 16. Diagrams showing variations of chemical composition, which are related to folding and shearing phenomena in basic schists. The scale is equal to that of Figure 9.



lenticular quartz veins develop in the adjacent rocks. Sample No. 189 is a composite sample obtained from the neighbouring hornblende schists, (Sketch: Fig. 15, Chemical data: Fig. 16-E).

Table 4 shows the summary of chemical tendencies recognized in the variation diagrams. From this Table it can be said as the result that FeO, MgO, H_2O, CO_2 and MnO increase, and SiO_2, Fe_2O_3, CaO, Na_2O, and TiO_2 decrease in the central part of shear zone, compared with the unchanged part. Marginal parts show, as a rule, the reverse tendencies.

Above result will be applied to explain both the regional development of compositional band and the occurrence of serpentinite along the thrust zones in the Sambagawa system, if the differentiation due to mechanical process is adopted to occur commonly. Figure 16-F shows the chemical compositions of a dark band and a light band of the greenschist of the Hiura formations. Sample No. 196 is a composite one obtained from the vicinity. For the components of SiO_2, FeO, MgO, CaO, and H_2O, chemical tendencies are nearly equal to those in case of shear zones, (Table 4). This result may give some geochemical informations to the conceptional explanations for compositional banding, which have been made by many authors. These geochemical tendencies also support the BENNINGTON's theory that genesis of stratiform ultrabasic rock is attributed to the influence of shearing stress and pressure (BENNINGTON, 1956).

Shearing process, however, may also take place along the so-called weaker zone which is originally characterized by such the chemical peculiarities as mentioned above. If this view is valid, shearing process may have a little significance in the tectonic differentiation. Analogous question will be raised also in case of folding process. Present investigations described hitherto do not give any answer to these questions. In succeeding chapters, several answers to these questions will be given from more general viewpoint, together with another geochemical tendencies in the metamorphism. To be continued to Part II.

Literatures[18]

BANNO, S., 1961, On the so-called "Soda-metasomatism" of the Sambagawa Schists in the Besshi District, (in Japanese): Jour. Geol. Soc. Japan, Vol. 67, P. 515–525.

BELL, P. M., 1963, Aluminum Silicate System: Experimental Determination of the Triple Point: Science, Vol. 139, 1055–1056.

BENNINGTON, K.O., 1956, Role of Shearing stress and pressure in differentiation as illustrated by some mineral reactions in the system $MgO-SiO_2-H_2O$: Jour. Geol., Vol.64, P.558–577.

18 The list of literatures does not claim to be complete. The voluminous works by ESKOLA, O., KORZHINSKII, D. S., KOJIMA, G., MIYASHIRO, A., TURNER, F. J. and other authors, although these are not listed above, gave to the author many valuable suggestions throughout the studies.

Doi, M., 1962, Geology and Cupriferous Pyrite Deposits (Besshi Type) of the Sambagawa Metamorphic Zone. (I)-(III). (in Japanese): Mining Geol., Vol. 11, P. 610-626; Vol. 12, P. 1-15, P. 63-83.

Hide, K., 1961, Geologic Structure and Metamorphism of the Sambagawa Crystalline Schists of the Besshi-Shirataki Mining District in Shikoku, Southwest Japan (in Japanese): Geological Report of the Hiroshima University. No. 9.

Horikoshi, Y., 1938, On Banded Diopside-Amphibolite from Besshi, Ehime Prefecture (in Japanese): Jour. Geol. Soc. Japan, Vol. 45, P. 655-666.

Kojima, G., 1951, Stratigraphy and Geological Structure of the Crystalline Schist Region in Central Sikoku, (in Japanese): Jour. Geol. Soc. Japan, Vol. 57, P. 177-199.

Miller, J., Shido, F., Banno, S. and Uyeda, S., 1961, New Data on the Age of Orogeny and Metamorphism in Japan: Jap. Jour. Geol. and Geography, Vol. 32, P. 145-151.

Miyashiro, A. and Banno, S., 1958, Nature of Glaucophanitic Metamorphism: Am. Jour. Sci., Vol. 256, P. 97-110.

Ogawa, T., 1902, Kōchi Sheet and its Explanatory Note, (in Japanese): Geol. Surv. of Japan.

Ozawa, Y., 1926, On the Stratignaphy and Structure of the Crystalline Schists Formations in Shikoku (I), (II), (in Japanese): Jour. Geol. Soc. Japan, Vol. 33, P. 297-304, P. 309-347.

Read, H.H., 1957, The Granite Controversy: Thomas Murby & Co. London.

Sato, H., 1938, Explanatory Text of the Geological Map of Japan, "Niihama." Scale 1 : 75,000: Geol. Surv. of Japan.

Suzuki, J., 1930, Petrological Study of the Crystalline Schist System of Shikoku, Japan: Jour. Fac. Sci. Hokkaido Imp. Univ. Ser. IV, 1, P. 27-111.

Yoshino, G., 1961, Structural-Petrological Studies of Peridotite and Associated Rocks of the Higashi-akaishi-yama District, Shikoku, Japan: Jour. Sci. Hiroshima Univ. Ser. C, Vol. 3, Nos. 3-4.