Memoirs of the College of Science, Kyoto Imperial University, Series B, Vol. XIV, No. 1, Art. 3, 1938.

# On the Roseki-Deposit (Aggregation of Pyrophyllite, Kaolinite and Diaspore) at Mituisi, Bizen Province, Japan

By

# Kojirō Kimizuka

(Geological and Mineralogical Institute, Kyöto Imperial University)

With 44 Text-figures

(Received March 17, 1938)

14.

#### Contents

Introdu	ction.	•	• •		•			•	•	•	•	•	•	•		•	٠	٠	•	74
Geology	v of the Mituisi Rōseki District.																			77
1.	Liparite																			78
2.	Felso-liparite																÷			79
3.	Pyrophyllitized liparite																			81
4.	Shale																			83
5.	Pyrophyllitized shale																			85
6.	Rōseki.																			85
Compos	nent minerals and kinds of Rōseki																			86
Princip	al component minerals of Rōseki.																			89
1.	Diaspore	-																		89
2.	Kaolinite																			92
3.	Pyrophyllite	-																		93
Descrip	tion of typical kinds of Rōseki					•		•					•		•					94
1.	Tokusyu-Rōseki								•					•						95
2.	Zyō-isi					•														97
3.	Tyū-isi																	•		97
4.	Haku-Rōseki.							• •								•		•		99
5.	Ma-isi.												•					•		99
Occurr	ence of Rōsoki								•									•	•	101
Origin	of Röseki																	•		106
A. (	Original rocks																		•	106
1.	From the geological point of view	7.																		106
2.	Considerations from the point of	vi	ew	of	01	re	dej	203	sits	s.										110
3.	Considerations based on chemical	c	om	pos	siti	on														111
В. <i>А</i>	Agents of metamorphism																		-	115
1.	Earlier theories.																	•		115

a. Alteration by hydrothermal action
b. Alteration by weathering
c. Alteration by pneumatolytic action.
2. Mechanism of Röseki formation at Mituisi.
a. Diaspore, kaolinite and pyrophyllite
b. Pyrite
c. Alunite
Summary and conclusion
Literature

#### Introduction

The Japanese term "Rōseki" meaning "Wax stone" is generally given to a waxy and greasy soft and compact aggregation of minerals, and it is classified in many ways according to its constituent materials. In order of their quantities, the components found in Rōseki-deposit are pyrophyllite, kaolinite, diaspore, and unaltered portion of original rocks. In Japan proper such deposits are found at Mituisi, Bizen Prov., Syōkōzan, Bingo Prov., Kanakura, Sinano Prov., and elsewhere.



Fig. 1. The Mituisi Rōseki Mines are located 50 km east of Okayama City, and 1 km southwest of Mituisi railway station along the San-yō-line. A. Akō town. C. The Tigusa-river. G. The town of Katagami. H. The Coast of Harima. J. Yakiyama Village. K. The town of Kamigōri. M. The town of Mituisi. S. The San-yō-R.R. Line. Y. The Yosi-i-river.

and other factors; (2) the sedimentary rock, mostly shale, which was a xenolithic mass in liparite or other igneous rocks, was altered mostly into pyrophyllite but sometimes kaolinite or diaspore, and it seems that the

These deposits have been considered by geologists to be hydrothermal alteration products of igneous rocks as liparite or porphyrite.

The Mituisi Rosekideposit has been studied by the writer since 1932, and as a result of his investigation its origin differs from that assumed by earlier investigators in the following points: (1)some ore-bodies composed of pyrophyllite originated from liparite as supposed by earlier theories and it is considered by the writer that the mass was altered by pneumatolytic exhalation, judging by its occurrence, the process of alteration

74

masses were not altered by hydrothermal action, but principally by heat and gases which were exhaled from the cooling magma of the rocks.

The writer has inspected many other localities of Rōseki-deposits, such as Syōkōzan (Bingo Prov.), Syōzakai (Tazima Prov.), Simozasa and Okumura (Harima Prov.), Minamiyamagata (Bizen Prov.), and Kanakura (Sinano Prov.), and he has found that the materials there are the same in origin as those in Mituisi, in that a large portion of the deposits were produced from sedimentary rock and were altered by thermal and pneumatolytic gas which were exhaled from igneous rocks.



Fig. 2. Geological map of the district around the Mituisi Rōseki Mines. A. Alluvium. B. Talus. C. Palaeozoic formation. D. Felso-liparite. E. Liparite. F. Granite.

- D. The Mituisi Röseki Mines.
- I. The road to Katagami.
- III. The road to Akö.
- V. The San-yō-Line.

M. Mituisi.

- II. The road to Wake.
- IV. Kongō-River.



Fig. 3 Geological map of the Mituisi Roseki Mines.

A. Talus. B. The rubbish of the Poor-ores and country rocks. C. Quartz vein. D. Felso-liparite. E. Felso-liparite crumbling to small fragments. F. Liparite. G. Pyrophyllitized shale (Rōseki preserving the bedding planes of original shale). H. Xenolithic shale masses of Palæozoic formation.

Röseki-Deposit (Aggregation of Pyrophyllite etc.) at Mituisi, Japan 77



Fig. 4. Liparite from Yakiyama (comparatively fresh sample, but a small part of the phenocryst of feldspar was altered into pyrophyllite flakes), ×50, (crossed nicols).

F. Feldspar. P. Pyrophyllite. Q. Quartz.



Fig. 5. Liparite from Ikegami gallery (comparatively fresh sample, but a part of the feldspar was altered into pyrophyllite and hydrargillite flakes),  $\times$ 50, (crossed nicols).

H. Hydrargillite. P. Pyrophyllite. Q. Quartz.

# Geology of the Mituisi Rōseki District

The Mituisi Roseki Mines are located 50 km east of Okavama City. and 1 km southwest of Mituisi railway station on the San-yō-Line. On the east side of the Yakiyama-Mituisi fault the Palæozoic formation is distributed; on the other side liparite. The mining district extends over the west side of the fault, and the shattered zone along the fault; the district is mainly composed of liparite, and the highest point (240 m) in the district is the center of the radial felso-liparite dykes with maximum width, 100 m, which were injected into liparite.

The various directions of the dykes, N20°W, S20 W, N70 W, and S70 W are geologically significant. More or less metamorphosed masses of various sizes and shapes of the Palæozoic shale are observed as xenoliths in these liparites. The rock formations in this district including the following rocks were observed by the writer (Figs. 2 and 3).

1. Liparite; 2. Felsoliparite; 3. Pyrophyllitized liparite. 4. Shale; 5. Pyrophyllitized shale; 6. Rōseki.



Fig. 6. Liparite from Ikegami gallery (a large part of the feldspar and one part of the feldspar in groundmass were altered into pyrophyllite flakes),  $\times 50$ , (crossed nicols).

F. Feldspar, H. Hydrargillite, P. Pyrophyllite, Q. Quartz.



Fig. 7. Liparite from Ikegami gallery at MituisiRōseki Company's Mine, ×50, (parallel nicols).F. Feldspar. H. Hydrargillite. P. Pyrophyllite.Q. Quartz.

## 1. Liparite

As shown on the map (Fig. 3), the Roseki-region is mainly composed of liparite which extruded from the Palæozoic formation. catching up blocks and fragments of shale in various shapes and sizes. The liparite was more or less altered by pyrophyllitization caused by the pneumatolytic action of the felsoliparite dykes. A fresh sample of this could hardly be found on the surface or in the gallery of the district.

The liparite varies in colour, the prevailing colour being pale grey, which shades off into white, and various shades of yellow and brown, depending on the iron content. Many phenocrysts of feldspar and quartz are scattered through it.

The rock consists of alkali-feldspar, plagioclase, quartz, chlorite and sometimes biotite or hornblende as phenocrystic constituents cemented minute felsitic groundmass which sometimes contains some glass.

*Plagioclase* is the predominant phenocryst. It is idiomorphic and mostly colourless. Twinnings, both of Carlsbad and albite, are seen under the

78



Fig. 8. Looking up the felso-liparite dykes, West-northwest; and a view of the characteristic topographic features of the felso-liparite dyke.

crossed nicols. The feldspar is determined to be oligoclase and acidic andesine.

Alkali-feldspar is sometimes found to be smaller and more irregular in shape than the plagioclase, its optical axial angle is very small with negative optical character, and zonal structure and twin are observed in it, but the perthitic structure is rarely and faintly observed.

The pyrophyllite flakes are observed along the fissure or cleavage of the feldspar, and sometimes, the greater part of the feldspars were altered into aggregates of fine pyrophyllite flakes, preserving the original shape of the feldspar; and rarely hydrargillite from feldspar is observed in a thin section.

*Quartz* is the phenocryst ranking next to the plagioclase in amount, and occurs in irregular shapes, more or less corroded with inlets of matrix.

*Chlorite* is presented in considerable amount, and might be the altered product of biotite and hornblende. Biotite and hornblende are rarely seen as residual minerals at the core of aggregates of chlorite.

As accessory minerals, pyrite and zircon crystals are contained in liparite.

Groundmass is of felsitic structure, and sometimes is partly associated with glass. Feldspar as the constituent of the groundmass is rarely pyrophyllitized, but in this case, the pyrophyllite flakes are much smaller than those from the phenocrysts.

#### 2. Felso-liparite

The highest point (240 m) in the district is the center of the radial felsoliparite dykes with maximum width of 100 m, injected into the liparite.

Among the directions of the dykes, N20 W, N70 W, S20 W, and S70 W are significant as already described, and a dyke N10 W, separated from them, occurs at Daiyama on the east slope of the Roseki mine.

Since felso-liparite is more resistant than liparite to weathering, its topographical features are very characteristic as shown in Fig. 8.



Fig. 9. Felso-liparite from Kitagawa gallery (it is located on the east side near the top of the mine),  $\times$ 95, (crossed nicols).

Q. Phenocryst of quartz. P. Pores. No signs of pyrophyllitization were observed



Fig. 10. Felso-liparite from the Gallery no. 1 at Mituisi Rōseki Company's Mine, ×95, (crossed nicols).

Q. Phenocryst of quartz. P. Pores (the wall of the pores consists of comparatively coarse grained felsitic groundmass). It varies in colour, being dark grey, grey, pale yellowish grey, pale bluish white, and white, and is generally porous except the porcelain-like variety which has a fluidal structure; but the texture observed under the microscope is compact. The dark variety usually contains many small crystals of pyrite. It is very noticeable that some varieties of felso-liparite are very brittle, being easily broken into small pieces of almost 5 cm in size.

It is generally found in the galleries, in spite of the fact that there is no outcrop except the northeast portion of the dyke at Daiyama.

It may be that the felsoliparite dykes have an intimate connection with the genesis of the Rōseki-deposit at Mituisi, as the ores, rich in alumina, are always distributed within the dykes and in their immediate vicinities.

Through microscopic examination, it is ascertained that the rock is composed of rich felsitic groundmass which rarely contains only phenocryst of quartz of small size. The rock from Kitagawa-Honkō located on the east slope, and near the top of the mine, reveals well its characteristics in the following points:

1. The groundmass is hollocrystalline, and composed of very small grains ranging from 0.00130 mm to 0.00005 mm in diameter.

2. The phenocryst of the

in it.

quartz ranges from  $0.015 \times 0.032 \text{ mm}^2$  to  $0.01 \times 0.01 \text{ mm}^2$  in size.

3. The rock contains many small xenolithic masses of diaspore or kaolinite flakes.

4. The porous texture is very significant; the pores ranging from  $0.0061 \times 0.006$ mm<sup>2</sup> to  $0.026 \times 5.00$  mm<sup>2</sup>. The wall of the pores is generally composed of comparatively coarse-grained minerals.

The last two facts point to the fact that the felso-liparite magma contained more abundant gases than the liparite around it.

## 3. Pyrophyllitized Liparite

The liparite in this district had more or less suffered from pyrophyllitization as already described. But the pyrophyllitized liparite, here mentioned, means a rock which can be worked as ore of pyrophyllite, though all the rock of this district may be the same name from the petrological point of view.

The pyrophyllitized liparite occurs as a xenolithic mass within the felso-liparite dykes and in their immediate vicinities. Through a microscopic investigation on the liparite and pyrophyllitized liparite, the process of the pyrophyllitization in liparite is successively observed; the following being the most noticeable features.

1. Alteration of the phenocryst.... Pyrophyllitization



Fig. 11. Liparite from Ikegami gallery. A portion of the phenocryst of feldspar was altered into pyrophyllite flakes, while the pyrophyllitization is not obserbed in ground-mass,  $\times 50$ , (crossed nicols).

F. Feldspar. Q. Quartz. P. Pyrophyllite flakes.



Fig. 12. Liparite, xenolith in the felsoliparite dyke from Motoyama gallery. The phenocryst of feldspar was altered into aggregates of pyrophyllite flakes, but the pyrophyllitization is scarecely observed in groundmass, ×50, (crossed nicols).

P. An aggregate of pyrophyllite flakes. Q. Quartz.



Fig. 13. Pyrophyllitized liparite (Ma-isi) from the Gallery no. 3 at Mituisi Rōseki Company's Mine. Whole crystals of the phenocrysts, feldspar were altered into pyrophyllite flakes, and the pyrophyllitization is partly observed in groundmass, ×50, (crossed nicols).

P. An aggregate of the pyrophyllite flakes. Q. Phenocrysts, quartz.



Fig. 14. Pyrophyllitized liparite (Ma-isi) from the Gallery no. 3 at Mituisi Rōseki Company's Mine. Phenocryst, feldspar and large portion of the groundmass were altered into pyrophyllite flakes,  $\times 50$ , (crossed nicols).

P. Pyrophyllite flakes. Q. Phenocryst, quartz.

in the liparite was begun from phenocryst of feldspar, and the pyrophyllite flakes were formed along its fissures. The pyrophyllitization gradually proceeded into the inner part of the feldspar became completely altered into pyrophyllite aggregation.

Even at the stage in which the liparite is highly altered into the ore which mainly consists of pyrophyllite, the aggregates of pyrophyllite keep the original shape of the feldspar, as shown in Figs. 11, 12, 13, and 15.

2. Alteration of groundmass.... It was noticed that the pyrophyllitization of the groundmass began in a later stage, but the alteration did not exactly proceed to the complete pyrophyllitization of the phenocryst, and it went simultaneously in certain stages as seen in Figs. 11, 12, 13, 14, and 15.

3. In comparing these pyrophyllite flakes from phenocryst with those from groundmass, the former is large in shape ranging 1.4 microns to 13.4 microns in length, but the latter, from 0.25 to 1.0 micron. There is a very remarkable difference in size between those of different origin, as illustrated in Fig. 16.

The boundaries of these pyrophyllite aggregations are so distinct that the original porphyritic structure of the liparite can be observed even in the best portion of the ore, while the ore from shale consists of rather small pyrophyllite flakes of almost equal sizes, without any porphyritic structure as seen in Figs. 31 and 28.

#### 4. Shale

It was first reported by Mr. H. Awazu<sup>7</sup> that shale-mass is distributed at the top of the mine and that small fragments of shale were caught up by liparite near the top of the mine, as xenolith, but he did not notice that the shale played an important role in forming Rōseki.

As shown on the geological map (Fig. 3), at many other places in this district, it is observed also that blocks and masses of Palæozoic shale were caught up by liparite and felso-liparite as xenolith, and these xenolith, and these xenolithic shale-masses were more or less metamorphosed into Rōseki or pyrophyllitized shale, but the aspect of the masses was changed so much from that of the original shale by metamorphism that the bedding planes of the shale even in the least altered masses, were scarcely revealed even on careful scruting.

Shale masses occur also in many places in the gallery, some having few altered into pyrophyllite, and some into kaolinite or diaspore masses. It was observed that the core of a large ore-body consists of the shale preserving an original bedding plane.



Fig. 15. Pyrophyllitized liparite (Zyō-Maisi) from the Gallery no. 3 at Mituisi Rōseki Company's Mine. Though feldspar and groundmass were completely altered into pyrophyllite flakes, the quartz grains scattered within pyrophyllite flakes, and the porphyritic structure of the liparite are clearly observed,  $\times$  50, (crossed nicols).

P. pyrophyllite flakes. Q. Quartz grains.



Fig. 16. Pyrophyllitized liparite ( $Zy\bar{o}$ -Maisi),  $\times 310$ , (crossed nicols).

The boundary dividing pyrophyllite flakes from phenocryst and from groundmass, is clearly observed on account of the difference in their sizes.

Half part, P. Aggregation of pyrophyllite from phenocryst. Half part, G. Aggregation of pyrophyllite from groundmass.



Fig. 17. An outcrop of xenolithic shale at 700 feet on the eastern slope of the mine. It is 100 m from north to south, and its thickness is about 20 m. Strike, N-S, Dip, W20°-10°, or N10°W, NE25°.



Fig. 18. An outcrop of xenolithic shale at  $\overline{O}$ giri gallery at the north-western corner of the mine, Katō Rōseki Co. . It was more or less altered into pyrophyllite, but it does not contain enough pyrophyllite to warrant exploitation.

Upper part of dotted line, Liparite. Lower part of dotted line, pyrophyllitized shale.

The xenolithic shale-masses range from 50 m to 1 cm in length, and they are black, dark grey, yellowish grey and yellowish brown in colour. The darker colour is mainly due to the formation of pyrite by contact meta-morphism.

Under the microscope, it is observed that the mass consists of shaly materials of equally minute size, and that the pyrophyllite flakes, the sizes of which are almost equal to those of the shaly substances, are scattered between them.

## 5. Pyrophyllitized shale

One might call the xenolithic shale-masses in this district "Pyrophyllitized shale," for the masses were more or less altered into pyrophyllite, but the name is used here only for those rocks that have been altered mainly into pyrophyllite aggregations, and which preserve the original bedding planes.

The important outcrops of pyrophyllitized shale are as follows: at the top of the mine, on the northwest slope of the mine, and elsewhere as indicated on the geological map (Fig. 3).

A protuberance at the top of the mine, about 15 m from northeast to southwest and 10 m from northwest to southeast, with a hight of 4 m, consists of whol1y pyrophyllitized shale called Ma-isi or Aiisi (consisting of almost pure pyrophyllite flakes). The disturbance of the bedding planes is very conspicuous as indicated in Figs. 19, 20 and 21.

The pyrophyllitized shale-mass on the northwestern slope of the mine is about 30 square meters in area, and 20 m in thickness. A large portion of it altered into Ma-isi, but the remainder into Tyūisi, which consists of pyroand kaolinite phyllite flakes, shows a thickness of 10 cm in the individual bed striking N-S, N20°W, with a dip of 50°-40' eastward.

The microscopic investigation of the pyrophyllitized shale will be



Fig. 19. A protuberance located at the top of the mine extends about 15 m from northeast to southwest, and 10 m from northwest to southeast, and is 4 m in height. It consists wholly of pyrophyllitized shale. The disturbance of the bedding planes of the original shale in the upper portion is more conspicuous than at the base.

described in the chapter on the Roseki-ores.

#### 6. Rōseki

The xenolithic shale-mass which is altered into pyrophyllite, kaolinite,

K. KIMIZUKA



Fig. 20. One portion of protuberance consisting of pyrophyllitized shale at the top of the mine. The pyrophyllitized shale contained enough pyrophyllite to warrant exploitation while the bedding planes of the original shale may be clearly observed. Strike,  $N30^{\circ}-50^{\circ}E$ ,  $NW10^{\circ}-50^{\circ}$ .



Fig. 21. A sketch map of the protuberance at the top of the mine; the disturbance of the bedding planes of the original shale is indicated in it.

and diaspore, and which does not preserve the original bedding planes of the shale, will be called by the name of "Rōseki". It is distributed on the southern slope of the mine in the vicinity of the upper part of the Eisei gallery, about 700 feet above sea level, and on the eastern side of the mine, and elsewhere some of the places being shown on the geological map.

# Component Minerals and Kinds of Rōseki

Rōseki is variously classified by miners in this mine according to its fusibility.

The principal component minarals are restricted to pyrophyllite, kaolinite, and diaspore, with occasional associations of pyrite, or unaltered portions of the original rocks, and rarely alunite.



Fig. 22. A pyrophyllitized shale mass on the northwestern slope of the mine. A large portion of the mass was altered into Ai-isi, which consists of pyrophyllite, and another portion into Tyū-isi. But the bedding planes of the original shale are clearly observed.



Fig. 23. Showing contact between felso-liparite and  $Ty\bar{u}$ -isi composed of pyrophyllite and kaolinite, which were altered from shale.

F. Felso-liparite. C.  $Ty\bar{u}$ -isi (the bedding planes of the original shale is obviously observed).

## K. Kimizuka

Though it is only rarely that Rōseki-ore consists exclusively of pyrophyllite, kaolinite, or diaspore, yet Haku-Rōseki consists of almost pure pyrophyllite, Zyō-isi of almost pure kaolinite, and Tokusyu-Rōseki no. 1 of almost pure diaspore. Rōseki-ore is usually a mixture of several kinds of minerals in various proportions. A low grade ore is mainly composed of pyrophyllite, which is poor in alumina among the component minerals, and it is generally associated with an unaltered portion of the original rocks.

A medium grade ore consists mainly of kaolinite and pyrophyllite, or pure kaolinite, and it is in some cases associated with small quantities of diaspore. A high grade ore contains principally diaspore and is sometimes associated with kaolinite in small amounts.

There may be found a gradual increase of each mineral according to the grade of the Rōseki; moreover, there is no sharply marked distinction between one kind of Rōseki and another. For that reason a purely mineralogical study can hardly reveal its character, origin, and genesis. We are thus forced to approach the subject from the standpoint of geology, mineralogy, and ore-deposits.

The kinds of  $R\bar{o}$ seki and their component minerals as determined under the microscope are shown in the following table:

Component minerals	Local names of the ores.	Fusibility.
Diaspore $(Al_2O_3 \cdot H_2O)$	Tokusyu-Röseki no. 1	41 (S. K.)
	Tokusyu-Rōseki no. 2	38
	Tokusyu-Rōseki no. 3	
	Medama-isi	
Kaolinite	Zyō-isi no. 1	36
(Al <sub>2</sub> O <sub>3</sub> .25IO <sub>2</sub> .211 <sub>2</sub> O)	Zyō-isi no. 2	35
	Ao-Rōseki	35
Burophullite	Zyō-isi no. 3	34
(Al <sub>2</sub> O <sub>3</sub> .4SiO <sub>2</sub> .H <sub>2</sub> O)	Tyū-isi	34
	Ao-Tyū-isi	34
	Kuro-Tyū-isi	34
	Haku-Rōseki	34
Unaltered	Aka-Tyū-isi	33
substances	Ai-isi	33
Pyrite	Zyō-Ma-isi	32
e f	Ma-isi	32-31
Alunite ,	Ge-Ma-isi	28
	Containing a large amount.	
and the second se	Containing sometimes a moderate amount.	
	Containing a small amount.	
	Containing sometimes a small amount.	

Kinds of Roseki-ores, their Average Components, and Fusibility.

. . .

. .

As shown in the foregoing table, each kind of Rōseki is composed of two or three or more minerals, except Tokusyu-Rōseki no. 1, Zyō-isi no. 1 and Haku-Rōseki, which are almost monomineral ores. It is difficult to get pure mineral from the Rōseki except that crystal may be isolated from it.

The highest grade of Rōseki, Tokusyu-Rōseki no. 1 represents the final stage in the alteration into Rōseki. Though it is seen to be composed of almost pure diaspore under the microscope, yet monomineral specimens are rarely found. Other ores representing an intermediate stage of alteration into Rōseki, are not monomineral ores, strictly speaking, and some of them, though differing in quantity of their component minerals, are called by the same names as various Rōseki ores.

A great many chemical analyses of the Rōseki have been already published by other investigators, but great differences will be found in the analyses of the same named ores from the same locality. These differences may be due to the fact that the Rōseki is a mixture of component minerals in various proportions, even when it was detected from the same mass. It is reasonable to believe that the chemical analyses of Rōseki would parallel the results of a microscopic investigation.

## Principal Component Minerals of Roseki

#### 1. Diaspore

*Occurrence.*.. Crystalline diaspore occurs generally as the main constituent of Tokusyu-Rōseki and Medama-isi, though sometimes, as an associated mineral within Zyō-isi and Tyū-isi, which are usually composed of kaolinite and pyrophyllite. The crystal of the diaspore occurs in coarse grained Tokusyu-Rōseki; microscopical crystals are contained within Medama-isi occurring along the marginal zones of Zyō-isi or Tyū-isi. The crystal



Fig. 24, (a). Crystal of diaspore from Mituisi Rōseki Mine.



Fig. 24, (b). Stereographic projection of crystal faces of the diaspore from Mituisi Rō-seki Mine.

K. KIMIZUKA



Fig. 25. The diaspore crystals growing on the wall of the druse in coarse grained Tokusyu-Rōseki. One half of the crystal was transformed into the grained part of the Rōseki, and no perfect crystal was isolated from it,  $\times 23/18$ , (from the 2nd gallery at Mituisi Rōseki Company's Mine).



Fig. 26. Tokusyu-Rōseki no. 1 from the 2nd gallery at Mituisi Rōseki Company's Mine, ×95, (parallel nicols).

#### from the wall.

*Crystal.*.. The crystal is generally prismatic in habitus, with a maximum length of 1 cm, and a vitreous lustre; the cleavage paralleling to (010)

within the coarse grained Tokusyu-Rōseki was sometimes cemented with kaolinite, and in this case, the crystal may be easily isolated from the ore as a perfect crystal, but it is generally milky white in colour, and the crystal faces are generally curved, so that it is not usually suitable for mineralogical study.

When the crystal attached to the wall of the fissure or druse of the coarse grained Tokusyu-Rōseki, it is colourless or grey in colour, and its crystal faces are perfect. It is very suitable for mineralogical study, but half of crystal usually breaks in detaching it is perfectly developed. The crystallographical constants are determined as following:

Interfacial angles measured with reflection goniometer,

## $210^{210}=50^{\circ}10'$ $110^{111}=58^{\circ}37'$

The axial ration ca	lculated from these interfacial	angles was:
Diaspore from Mituisi. (Кімідика)	Diaspore from Syōkōzan. (В. Yosнiкi)	Diaspore. (DANA: A System of Min.)
ă: b: c 0.9362 : 1 : 0.5943	0.946:1:0.593	0.93722:1:0.60387
The following faces	s are usually observed (Fig. 24)	
(111), (210),	(110), (010).	

The crystal faces (100) and (011) are rarely observed.

*Chemical Composition.* . The Tokusyu-Rōseki from the Third gallery of the Mituisi Rōseki Co., composed of almost pure grained diaspore under the microscope, was analysed by the late U. USHIJIMA, to whom the writer feels greatly indebted.

The result of the analysis is as follows:

Weight % by analysis	Recalculated weight %	Weight %, in mineral- ogical diaspore
SiO <sub>2</sub> 2.654	2.667	0.000
$Al_2O_3$ 62.167	82.570	85.000
H <sub>2</sub> O 14.691	14.763	15.000
Total 79.512	100.000	100.000

The recalculated values are very similar to those mineralogical diaspore, but if these are precisely compared, there are such differences as an excess in SiO2, and deficiencies in Al<sub>2</sub>O<sub>3</sub> and H<sub>2</sub>O in the recalculated values. These differences may be due to impurity in the Tokusyu-Roseki, for it is sometimes observed under the microscope that the kaolinite or the pyrophyllite flakes are associated with diaspore in small amounts. The normative minerals, kaolinite, pyrophyllite, and diaspore, as the component minerals of the Roseki, are indicated from the recalculated values as follows:



Fig. 27. Tokusyu-Rōseki no. 1 from the 2nd gallery at Mituisi Rōseki Company's Mine, ×95, (crossed nicols).

Diaspore		•			95.304
Kaolinite		•			2.425
Pyrophyllite					2.304
Total			•		100.033

The following table indicates the chemical composition of diaspore from many localities:

	1	2	2	4	5	6	7
$SiO_2 \ldots \ldots$	1.37	3.33	21.92	10.34	2.44	4.38	6.37
$Al_2O_3\ \ldots\ .$	84.24	81.30	65.07	74.56	81.60	80.30	73.72
$Fe_2O_3 \ldots$			<b>1</b>	0.95	0.42	) +	-
FeO $\ldots$		1.41		Revenue ou		} u.	0.28
СаО	0.10					-	
$Na_2O$				0.56			
$\mathrm{K}_2\mathrm{O}$ ,		Reserved					
Ig. loss	14.67	13.97	11.96	13.24	15.01	15.63	14.52
Total	100.38	100.01	99.47	99.65	99.47	100.31	94.89

1. Diaspore<sup>20</sup> from Syōkōzan.

 Diaspore from Kanakura. Journ. Ceramic Soc., Vol. 38, no. 450, pp. 328–336, 1930 (in Japanese).

- Diaspore from Mituisi. Journ. Ceramic Soc., Vol. 37, no. 444, pp. 545-556, 1928 (in Japanese).
- 4. Diaspore<sup>21</sup> from Mituisi.
- 5. Diaspore<sup>21</sup> from Mituisi.
- 6. Diaspore13 from Syöközan.
- 7. Diaspore from Mituisi. Loc. cit.

As shown in the foregoing table, a great many differences in chemical composition are found among these.

Therefore it may be considered that the samples are not mineralogically pure diaspore, and we may call these by the name of Tokusyu-Rōseki.

#### 2. Kaolinite

Kaolinite occurs as a constituent of  $Zy\bar{o}$ -isi and  $Ty\bar{u}$ -isi, and sometimes, it is contained in Tokusyu-Rōseki, Medama-isi, Haku-Rōseki and Ai-isi in small amount. The kaolinite in the ore is seen as fan-shaped flakes, and varies from 0.25 micron to 0.5 micron in length. It is difficult to determine the mineralogical constant on the isolated flakes, but some of its optical properties were determined under the microscope as follows:

Its optical character is negative, and its extinction angle referring to cleavage ranges from 9' to 8'.

*Chemical Composition.* . . Though pure kaolinite can not be isolated from Rōseki, it is observed that Zyō-isi no. 1 is composed almost wholly of pure kaolinite under the microscope. The sample was analysed by Mr. T. OYAMA, to whom the writer tenders his sincere thanks. The result of the analysis is as follows:

	1	2	
$SiO_2 \ldots \ldots$	51.23	46.50	
$\mathrm{TiO}_2 \ldots \ldots$	0.26		
$Al_2O_3$	38.12	39.50	
$Fe_2O_3$ FeO $f \sim \cdot \cdot$	0.14		
MgO	0.04		
СаО	0.08		
$Na_2O$	0.58		
$\mathrm{K}_{2}\mathrm{O}$	0.63		
$MnO \ \ldots \ .$	nil.		
$P_2O_5$ . ·	0.11		
$H_2O(-)$	0.50		
$H_2O(+)$	8.55	14.00	
Total	100.24	100.00	

1. Zyō-isi no. 1 from Mituisi Rōseki Mine.

2. Kaolinite (mineralogical).

In column 1 of the foregoing table, the weight percentages of the three components  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  in the principal components of Roseki were recalculated as follows:

 $SiO_2 \dots 52.063$   $Al_2O_3 \dots 38.739$   $H_2O \dots 9.198$ 

Comparing the weight percentages of the components shown by the recalculated value with those in column 2, it is found that the excess in

SiO<sub>2</sub>, and deficiencies in Al<sub>2</sub>O<sub>3</sub> and H<sub>2</sub>O in the recalculated values are indicated to a certain extent, while there may be found similarities between these. This condition may be caused by the impurity, pyrophyllite, for it is sometimes observed that the pyrophyllite flakes occurring as the impurity of  $Zy\bar{o}$ -isi no. 1 are associated with kaolinite, in small amount.

## 3. Pyrophyllite

Occurrence. . . Pyrophyllite is the principal component mineral of Tyū-isi and other inferior ores of Rōseki. Haku-Rōseki and Ai-isi are composed almost wholly of pyrophyllite, while Zyō-Ma-isi, Ma-isi, and Ge-Ma-isi are composed of pyrophyllite and an unaltered portion of the original rocks.



Fig. 28. Kaolinite in Zyō-isi,  $\times$  310, (crossed nicols). The kaolinite within Zyō-isi appears as very minute flakes, ranging from 0.25 micron to 0.50 micron in length. The sizes of the kaolinite flakes within Zyō-isi are almost equal; the texture of the Zyō-isi is similar to that of shale, and porphyritic structure is never found.

Lath-shaped, scaly, or fan-shaped pyrophyllite ranges from 50 microns to 0.25 micron in length. The large sized pyrophyllite occurs as a cementing material of the coarse grained diaspore-mass, which is called by the name of Tokusyu-Röseki. Medium sized pyrophyllite with a length of from 1.4 to 13.4 microns, occurs as alteration product from phenocryst, feldspar within pyrophyllitized liparite. Small sized pyrophyllite varying from 0.25 to 1.0 micron, occurs as pyrophyllite from groundmass within pyrophyllitized liparite from shale.

The pyrophyllite is colourless, its optical character is negative, and it has straight extintion.

*Chemical Composition*. . The pyrophyllite cementing the interstices of the coarse grained diaspore-mass, which is called by the name of Tokusyu-Rōseki, and which came from the 2nd gallery at the Mituisi Rōseki Company's Mine was analysed by Mr. T. OYAMA, with the following results:

	1	2
$SiO_2 \ldots \ldots$	62.20	66.66
$Al_2O_3$	29.93	28.34
$Fe_2O_3$	0.11	
FeO	0.04	
${\rm TiO}_2$	nil.	
CaO	0.14	
MgO	0.06	4
$MnO \ . \ . \ .$	tr.	
$\mathrm{P}_2\mathrm{O}_5$	0.06	
$\mathrm{K}_2\mathrm{O}$	0.18	
$Na_2O$	1.74	
${\rm H_2O}(-)$	0.32	
$H_2O(+)$	6.20	5.00
Total	100.98	100.00

1. Pyrophyllite from 2nd gallery at Mituisi Röseki Company's Mine.

2. Chemical composition of mineralogical pyrophyllite.

Comparing the chemical composition of the foregoing table, small excess of  $Al_2O_3$  and  $H_2O$ , and deficiencies in  $SiO_2$  are observed in analysis. The weight percentages of the three components,  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  in (1) have been recalculated and the component minerals of this sample may be expressed as follows:

Diaspore . . . . 4.360 Pyrophyllite . . . . . 94.392

## Description of Typical Kinds of Roseki

As shown in the foregoing table, Rōseki is classified into various kinds corresponding to the degree of metamorphism. When their fusibility and component minerals are gradually changed, it is difficult to differentiate the ores very strictly. However, Tokusyu-Rōseki, Zyō-isi, Tyū-isi, Haku-Rōseki, and Ma-isi may be here selected as the most typical and their occurrence, distribution, component minerals and general characters will be described in convenience of the explanation of the original rocks and mechanism of alteration in Rōseki formation.

## 1. Tokusyu-Rōseki

The Tokusyu-Roseki is composed mainly of diaspore, and sometimes, is associated with kaolinite or pyrophyllite in small amounts. It occurs as the mass within felso-liparite dykes or in the immediate vicinity of the dykes, but if a large mass of Roseki was altered from xenolithic shale mass caught up by the dyke, the Roseki mass generally consists of various kinds of Roseki. The outer part contact with the dykes is usually composed of Tokusvu-Roseki no. 1 (consisting almost wholly of diaspore); and the next zone of Tokusyu-Roseki no. 2, which is associated with kaolinite in small amounts. The diaspore gradually decreases towards the inner part; kaolinite, on the contrary increases, and the Tokusvu-Roseki zone gradually changes into Zyō-isi, which is mainly composed of kaolinite. It is difficult to determine exact boundary of each zone.

Its occurrence and relation with the other kinds of Rōseki are shown on a specimen from Naka-gallery, Ōhira Company's Mine at Mituisi. In Fig. 32, *a*-zone (the zone, in contact with felso-liparite) is composed of Tokusyu-Rōseki no. 1 (consisting almost wholly of pure diaspore); *b*-zone, of Tokusyu-Rōseki no. 3, which is composed of diaspore, associated with kaolinite in small amounts; and *c*-zone, chiefly of Zyō-isi, which is



Fig. 29. Large shaped pyrophyllite flakes associated with diaspore, and from 50 to 13.4 microns in length,  $\times 310$ , (crossed nicols).



Fig. 30. Medium sized pyrophyllite flakes in pyrophyllitized liparite (Ma-isi). Porphyritic structure caused by the differences between pyrophyllite from phenocryst, and from groundmass, is observed,  $\times$  310, (crossed nicols). The pyrophyllite ranges from 13.4 microns to 0.25 micron in length.



Fig. 31. Small sized pyrophyllite in  $Zy\bar{o}$ -Ma-isi, ranging from 1.0 to 0.25 micron in length. The flakes are almost equisized, so porphyritic structure is not observed. The structure is similar to that of shale,  $\times$ 310, (crossed nicols).

composed almost wholly of kaolinite.

The Tokusyu-Rōseki varies in colour, being pale bluish white, pale brownish white, grey and dark grey. The various shades of brown and grey probably depend mainly upon the content of minute crystals of pyrite which are present as impurity.

Hardness ranges from 6.5 to 7.0; fractured surface is uneven, and it glistens in sunlight by perfect cleavage, parallel to (010) of the diaspore; the Tokusyu-Rōseki is easily recognized by miner because of its characteristic hardness and glistening.

It is classified into fine,



Fig. 32. The mode of occurrence is shown in this picture, representing one part of the Röseki-ore mass from Naka gallery at Öhira Röseki Co., ×<sup>4</sup>/<sub>10</sub>. *a*-zone. Tokusyu-Röseki no. 1 (composed almost wholly of grained diaspore). *b*-zone. Tokusyu-Röseki no. 3 (composed of diaspore and kaolinite). *c*-zone. Zyö-isi (composed of almost pure kaolinite).
M. Medama-isi (composed of diaspore).

medium, and coarse grained types depending on the size of the constituent mineral. The coarse grained Tokusyu-Rōseki is generally composed of crystals of diaspore and diaspore grains, while fine and medium grained ones consist of crystalline diaspore, of almost the same size, and of which the structure is almost the same as that of sandstone, when seen under the microscope.

#### 2. Zyō-isi

The Zyō-isi is composed of kaolinite, and is sometimes associated with diaspore or pyrophyllite in small amounts. It occurs within felsoliparite dykes or in the immediate vicinity of the dykes, as a small mass consisting almost wholly of Zyō-isi, and if it constitutes one part of a large Rōseki-mass, the Zyō-isi zone is distributed at the next inner zone of the Tokusyu-Rōseki, as seen in Fig. 32.

Hardness ranges from 2 to 2.5; and it has a dull lustre. It is classified into several groups, no. 1, no. 2 and no. 3, according to the amount of associated minerals and impurity. It varies in colour, showing pure white, pale bluish white and pale brownish white, the various shades of blue and brown probably depending upon the content of minute crystals of pyrite or iron hydroxyde.

Through microscopic observation, it is ascertained that the Zyō-isi is composed of minute and almost equisized flakes of kaolinite, ranging from 0.25 to 0.5 micron in length. Porphyritic structure is never found as shown in Fig. 28.

The Zyō-isi, in some cases, associated with Medama-isi ("Eye-Ball Stone" or "Ball Stone," the Japanese term "Medama<sup>6</sup>-isi" or "Tama-isi" being given to the globular body composed of the same component minerals as the Rōseki, and besides being associated with Zyō-isi and Tyū-isi in some cases, is easily detached from the Rōseki.). The Medama-isi is usually composed of minute grains or acicular crystals of diaspore, and then it may be called Tokusyu-Rōseki.

Fig. 33 shows that the Medama-isi is contained within Zyō-isi. The Medama-isi will be discussed by the writer in another paper.

# 3. Tyū-isi

The Tyū-isi is composed mainly of kaolinite and pyrophyllite flakes, and occurs as a mass within the felso-liparite dykes or in the immediate vicinity of the dykes. If it is associated with other better ores of the Rōseki as a large mass, the Tyū-isi is always distributed at the next inner zone of the Zyō-isi and is gradually changed into Zyō-isi on the outer side, and into Ai-isi or Ma-isi on the other side.

It varies in colour, pale yellowish white, pale brown, brown, pale red, pale bluish grey, grey and dark grey having been observed. The various shades of the colours apparently depend upon the content of ferric matter or minute crystals of pyrite. It is classified into several groups by miners according to its colour and the proportional amounts of the principal component minerals, as shown in the foregoing table.

Through microscopic observation, it is ascertained that the Tyū-isi is composed of minute pyrophyllite and kaolinite flakes, irregularly mingled. The pyrophyllite flake ranges from 0.25 to 1 micron; the kaolinite, from 0.25



Fig. 33. Zyō-isi containing Medama-isi. From Naka-gallery at  $\overline{O}hira$  Rōseki Co.,  $\times 2/3.$ 

A. Medama-isi composed of diaspore. B.  $Zy\bar{o}$ -isi composed of kaolinite. If we crush the sample into pieces, the Medama-isi is easily isolated from  $Zy\bar{o}$ -isi on account of differences in hardness.

to 0.5 micron. There is no great difference in their sizes, and the porphyritic structure was not observed, as in the case of the  $Zy\bar{o}$ -isi. It is sometimes observed that the  $Ty\bar{u}$ -isi is associated with the Medama-isi having a smooth surface.

#### 4. Haku-Röseki

The Haku-Rōseki occurs as a xenolithic mass enveloped by pyrophyllitized liparite within felso-liparite dykes as shown in Figs. 34, 35 and 36, and sometimes, it occurs as a small xenolithic mass within liparite. It is usually small and platy or irregular fragments in shape.

As shown in Figs. 34 and 35, the Haku-Rōseki (A-zone) is compact in texture, and is never associated with quartz grains. Pyrophyllitized liparite (B-zone) is associated in a scattered manner with a great many quartz grains. The boundary between two parts is clearly detected, a gradual change never being observed. Fig. 36 shows the occurrence of the irregular fragmentary Haku-Rōseki mass; A-part is the Haku-Rōseki, having a compact texture, and B-part is the pyrophyllitized liparite, having a rough surface. The distinct difference between two parts can be observed macroscopically.

Through microscopic observation, it may be seen that the Haku-Rōseki is composed of almost pure pyrophyllite flakes, ranging from 0.25 to 1 micron in length, the texture being the same as that of Zyō-isi and Tyū-isi.

#### 5. Ma-isi

The Ma-isi is composed partly of pyrophyllite and partly of an unaltered portion of the original rocks; the greater part of the Rōseki from Mituisi belongs to this class. The Ma-isi is classified into two groups, the one, from shale, and the other, from liparite, according to its original rocks.

The Ma-isi from shale... This originated from the xenolithic shale-mass which was caught up by intruded liparite; the shaly substance of the xenolithic shale-mass was recrystallized into pyrophyllite by pneumatolytic action which produced the liparite. Tokusyu-Rōseki, Zyō-isi, Tyū-isi, and Haku-Rōseki are the highly altered products of shale, but Ma-isi is a slightly altered product. It is observed that some xenolithic shale-masses altered into pyrophyllite aggregations preserve the original bedding planes. So the gradual alteration from shale into Ma-isi may be seen not only microscopically but macroscopically.

Through microscopic examination, it is ascertained that the Ma-isi is composed of minute equisized pyrophyllite flakes, some being composed of pyrophyllite and shaly substances. Porphyritic structure is never detected as in the case of products highly altered from shale.

It occurs not in liparite as the xenolithic mass, but also at the core of the large Rōseki mass, originating from the xenolithic mass of the shale within felso-liparite dykes or as a xenolithic mass, enveloped with pyrophyllitized liparite within the dykes.

*Ma-isi from liparite*... It is composed of pyrophyllite flakes and partly

K. KIMIZUKA



Fig. 34. Haku-Rōseki is associated with pyrophyllitized liparite,  $\times 9/6$ . A. Haku-Rōseki composed of pure pyrophyllite, having a greasy feeling, and not associated with quartz grains. B. Pyrophyllitized liparite having a rough surface, and a great many grains of quartz scattered through it. Q. Quartz grains.



Fig. 35. Haku-Rōseki inserted into pyrophyllitized liparite from 2nd gallery at Mituisi Rōseki Co.,  $\times 3/2$ .

A. Haku-Rōseki. B. Pyrophyllitized liparite (Ma-isi). Many corroded quartz grains are observed in it. Q. Quartz grains.



Fig. 36. Irregular shaped Haku-Röseki-mass within pyrophyllitized liparite from Turuta Röseki Mine, near Öhira Röseki Co., ×2/3.
A. Haku-Röseki enveloped by pyrophyllitized liparite (Ma-isi).
B. Pyrophyllitized liparite.

of an unaltered portion of liparite. The pyrophyllitization process from liparite into Rōseki is gradually observed, and in this case, the porphyritic structure is clearly observed through the difference in size of pyrophyllite on the one hand and phenocryst and groundmass on the other, and by the scattered quartz grains, even in the best ores.

It occurs as the xenolitic mass within felso-liparite dykes or along the contact zone of liparite with the dykes. The contact part is highly altered, degree of the alteration decreasing to go far the dykes, and the ore gradually changing into normal liparite. The degree of alteration is proportional to its shape, size, and situation.

#### Occurrence of Roseki

The Roseki-mass originating from liparite, occurs simply as the xenolithic mass within felso-liparite dykes or as the contact zone of liparite within the dyke, and it is generally composed of pyrophyllite or pyrophyllite and an unaltered portion of liparite. The mass originating from shale varies in its component minerals according to its size, shape, distribution and the degree of metamorphism. Some are composed of pyrophyllite and unaltered portion of shale, and some almost exclusively of pyrophyllite, of pyrophyllite and kaolinite, of pure kaolinite, of kaolinite and diaspore, or of diaspore. It occurs in various ways. The mode of occurrence for each typical kind was described in the preceding pages, but the mode of occurrence of all kinds will be summarised in this chapter. The mass being slightly altered, and being composed of pure pyrophyllite or pyrophyllite and an unaltered portion of shale occurs as the xenolithic mass within liparite, or as the core of the large Roseki-mass within felso-liparite dykes, or as the mass enveloped by pyrophyllitized liparite was caught up by the dyke.



Fig. 37. Zyō-Ma-isi originating from shale within pyrophyllitized liparite (Ma-isi). From 1st gallery at Mituisi Rōseki Co.,  $\times 1$ .

A. Zyō-Ma-isi having compact texture; quartz grains are not found in the area. B. Pyrophyllitized liparite (Ma-isi); a great many grains of quartz are found in the area. Q. Quartz grains. When the mass has been highly altered, and is composed of minerals, rich in alumina, it occurs as the xenolithic mass within the felso-liparite dyke, or as the crustal part of the large Rōsekimass within the dyke. Some ore-bodies at Mituisi Rōseki Mines have been selected to explain the mode of occurrence of the Rōseki originating from shale as follows:

Fig. 39 is a sketch of the outcrop along one part of the Motoyama gallery. As shown in the figure, the right side of the Rōseki-mass was bounded by felso-liparite, while the upper, lower and left sides were not excavated to the dyke. The xenolithic mass within the dyke may easily be surmised from the distribution.

The Rōseki-masses having two origins are alternately distributed

from right side (East) to left side (West) as follows:

- 1. Ma-isi originating from liparite, composed of pyrophyllite and an unaltered portion of liparite.
- 2. Tyū-isi originating from shale, composed of kaolinite and pyrophyllite. It is associated with Medama-isi composed almost wholly of diaspore; the Medama-isi is densely contained at the contact part with pyrophyllitized liparite (Ma-isi), and it decreases the farther it goes from the contact; the mass is gradually changed into Tyū-isi.



Fig. 38. Zyō-Ma-isi associated with pyrophyllitized liparite. From Moto-yama gallery at Mituisi Rōseki Co.,  $\times 1.$ 

A. Zyō-Ma-isi originating from shale, and preserving the bedding planes of original rock.

B. Pyrophyllitized liparite. Bedding planes are not observed.

3. Tyū-isi originating from shale.

4. Tyū-isi originating from shale, and containing Medama-isi.

5. Ma-isi from liparite.

6. Tyū-isi originating from shale, and associated with Medama-isi.

7. Ma-isi originating from liparite.

That the Roseki-mass have alternate origins may be geologically explained as follows:

The xenolithic mass of shale being altered into Roseki was first caught up by liparite which had erupted through the Palæozoic formation, and was



Fig. 39. Sketch of outcrop, the Rōseki from shale and from liparite being distributed in alternating manner. At motoyama gallery, Mituisi Rōseki Co. .

A. Tyū-isi (principal components, kaolinite and pyrophyllite) with Medama-isi (principal component, diaspore). B. Tyū-isi. C. Ma-isi (principal component, pyrophyllite, associated with an unaltered portion of liparite) from liparite. D. Felso-liparite.



Fig. 40. The profile through the center of the spherulitic mass of Rōseki at the First gallery, Mituisi Rōseki Co. Mine. It is perfectly enveloped by felso-liparite. The best ore is distributed at the crustal part, and the low grade ore at the core, while the whole mass is composed of minerals rich in alumina. The zonal arrangement of the ore is observed.

A. Tokusyu-Rōseki (consists almost wholly of diaspore). B. Zyō-isi no. 1 (almost pure kaolinite) with Medama-isi (of diaspore). C. Zyō-isi no. 2 (kaolinite and pyrophyllite in small amounts). D. Tyū-isi (kaolinite and pyrophyllite). E. Felso-liparite. F. Fragments of felso-liparite. A cross-section of small tongues of felso-liparite intruded into fissures of the shale-mass. more or less altered into Röseki by contact metamorphism. It was later caught up with blocks of liparite by the felso-liparite and dvke, they were altered into Rōseki by pneumatolytic action, due in turn to the cooling magma of the dyke. Thus the alteration of the xenolithic shalemasses was influenced by double metamorphism, and they were composed of minerals rich in alumina.

Fig. 40 is the sketch of the profile through the core of an almost spherical mass of Rōseki originating from shale formed in the 1st gallery at Mituisi Rōseki Co. Mine. It is completely enveloped by felso-liparite, and is about 8 m in

diameter. It is composed of the best ores originating from shale. The zonal arrangement of the ores is clearly observed in the mass; the crustal part is composed of minerals comparatively rich in alumina, and the core of minerals poor in alumina, and the zones gradually change, so that the boundary of each zone was bounded with dotted lines.

- 1. Crystal part composed of Zyō-isi no. 1 (it consists of almost pure kaolinite, and is associated with Medama-isi), but the bottom part is composed of Tokusyu-Rōseki (consisting of almost pure diaspore).
- 2. The next inner zone composed of Zyō-isi no. 2. It is not associated with Medama-isi.



Röseki-Deposit (Aggregation of Pyrophyllite etc.) at Mituisi, Japan 105

Fig. 41. A map of the distribution of Rōseki at Mituisi Rōseki Mines, grouped high above sea level.

1-column, Tokusyu-Rōseki, Zyō-isi, and Tyū-isi. 2-column, Ai-isi and Ma-isi. Mituisi Rōseki Co. Mine.

A. Motoyama gallery. B. 1st-gallery. C. 6th-gallery. D. Taisei gallery. E. Kisitake gallery. F. Kitagawa-Sin-gallery. G. Kitagawa-Hon-gallery. H. Eisei gallery. I. Yokozaka-Sin-gallery. J. Yokozaka-Hon-gallery. K. 2nd gallery. L. Takahasiyama-gallery. M. 3rd gallery. N. 5th gallery.

Öhira Rōseki Co. Mine.

O. Ögiri gallery. P. Sinkuti gallery. Q. Naka gallery. R. Tatu-gallery. S. 1st gallery. T. 3rd gallery. U. 2nd gallery. V. Hon-gallery.

3. The core composed of Tyū-isi (consisting of kaolinite and pyrophyllite).

As the mass is comparatively small, it is not associated with pyrophyllitized liparite, and is perfectly enveloped by felso-liparite. The alteration into Rōseki was strongly influenced by felso-liparite, while differential alteration is observed at the crustal part and the core.

The forms of the Rōseki-masses which were inspected by the writer are various in shape, being spheroidal, lenticular, platy, reticular (caused by intrusion of liparite or felso-liparite) and irregular. The forms doubtless originated from blocks and fragments of the original rocks which were caught up by the intruded rocks.

The distribution of the Roseki-masses of the best quality was surveyed in all the galleries and on the surface of the mine, and was mapped by the writer as indicated in Fig. 41. It will be seen that the ore-masses are distributed within or along the dykes in scattered manner, and that ore-masses are separated from one another.

## Origin of Roseki

## A. Original Rocks

Though all Rōseki-deposits have been considered by geologists to be the hydrothermally<sup>3, 4,7, 14, 15, 17, 18, 19, 22, 25, 41, 45, 49</sup> altered product of igneous<sup>40</sup> rocks, usually liparite<sup>4, 7, 15, 33, 41, 45, 40</sup> or porphyrite,<sup>3, 4, 12, 7</sup> yet the greater part of them did not originate from igneous rocks, but mainly from sedimentary rock, shale usually being caught up by igneous rocks, which intruded through the sedimentary rock, while one part of the deposit composed of minerals poor in alumina originated from igneous rocks caught up by other igneous rock which intruded through various other surrounding rocks.

The xenolithic masses in both cases were changed into Rōseki by heat and gases exhaled from the intruded rocks. The difference in texture and component minerals between the ore originating from igneous rocks, and shale is so conspicuous that the difference may be macroscopically observed as described in preceding pages.

The original rocks of Roseki will be discussed here from the geological, petrological, ore-deposit and chemical points of views.

#### 1. From the Geological Point of View

It will be clear that the Rōseki-deposit at Mituisi originated mainly from sedimentary rock as mentioned in the preceding pages. The same conditions were observed at other Rōseki mines inspected by the writer.

## Syōzakai Rōseki Mine, Tazima Prov.

The Syōzakai Rōseki Mine is located 5 km northwest of Ebara railway station on the San-in-line, and mining district is composed of liparite which erupted through the base composed of alternating shale, sandstone and conglomerate, which may be considered to belong to the Tertiary. It is observed here and there that the blocks and fragments of the Tertiary formation in various shapes and sizes were caught up by liparite, and that the xenolithic masses were altered into Ma-isi composed of pyrophyllite. The best ore and the ore produced from liparite are not distributed. It may be assumed that the metamorphism in this district was not strongly influenced, because the felso-liparite was not distributed.

The mode of occurrence of the Rōseki-mass is like that at Mituisi; the ore-body is distributed in a scattered manner, and the masses are separated from one another as indicated in Figs. 42 and 43.

Okumura and Simozasa Rōseki Mines, Harima Prov.

Okumura Rōseki Mine is located about 20 km north of Himezi railway station on the San-yō-line: it stands on the contact zone of liparite and the Palæozoic formation. The



Fig. 42. The photograph of specimen of Rōseki indicating the mode of occurrence at Syōzakai Rōseki Mine.

M. Ma-isi from shale. Its fractured surface is comparatively smooth; compact in texture.

L. Liparite. It is not pyrophyllitized, and its fractured surface is rough, so the boundary between liparite and Rōseki is easily observed. Each Rōseki-mass is separated from the others.

Rōseki mine is called by the name of Oiti-dani by the local people, the outcrop standing on the eastern slope of the mountain, which is about 280 m in height above sea level. The mountain is composed of liparite containing a large xenolithic mass of Palæozoic formation which was caught up by intruded liparite.

The xenolithic massis composed of alternate strata of shale and sandy shale.

It is to be observed that the shale and sandy shale were altered into an aggregation of pyrophyllite, and preserving the bedding plane of the original rocks. The pyrophyllitized shale was mined as Ma-isi ore in the former times, but it is not mined at present, because it is composed of a mineral poor in alumina.

K. KIMIZUKA



Fig. 43. Photograph of the contact part of Ma-isi mass with liparite at Myōto-iwa gallery, Syōzakai Rōseki Mine.
L. Liparite. M. Ma-isi (pyrophyllitized shale).
Strike and dip of the contact plane are, N10°W, NE50°.
Strike and dip of the pyropoyllitized shale are, N10°W, SW30°.
The boundary between pyrophyllitized shale and liparite is clearly observed, and is never changed gradually.

Simozasa Rōseki Mine is located about 16 km north of Tatuno railway station on the San-yō-line, and stands on the contact zone of liparite with Palæozoic formation like the Okumura Mine. The mining district is composed of liparite, and it is here and there observed that the liparite caught up many xenolithic masses of shale and sandy shale in various shapes and sizes. The xenolithic masses, especially shale-mass, were altered into Haku-Rōseki or Ma-isi, mainly composed of pyrophyllite. These ores were actively mined in former<sup>10</sup> times.

The best ore scarcely reaches the quality of Haku-Rōseki, since it is composed of almost pure pyrophyllite, poor in alumina; so it is not mined at present.

Though the ores from these mines are generally poor in alumina, and are not famous as Mituisi and Syōkōzan in ceramics, yet it is very interesting that the process of alteration from original rocks to the ore is clearly revealed.

#### Minami-yamagata Roseki Mine, Bizen Prov.

The Minami-yamagata Rōseki Mine is located about 10 km northwest of Wake railway station on the San-yō-line, and stands within the district of the Palæozoic formation which is widely distributed around the mine. The mining district is composed of alternate strata of shale and sandy shale. It is found that a felso-liparite dyke, 20 m in length and 6 m in thickness, intruded through the formation from north to south, and that the Rōseki is distributed along both sides and within the dyke, the best ore being composed of kaolinite and associated with diaspore and sometimes andalusite in small amounts. It is found distributed just at the line of contact, while the bad ore composed mainly of pyrophyllite is found to go far the dyke, and to change gradually into the Palæozoic formation.

The ore being exploitated as ceramic material extends about 2 m along the strike of the dyke; it is mined as material for fire-brick at present. Though the mine may be poor in quantity, it is very interesting that the process of alteration from original rocks to the ore is clearly revealed, and that the best ore composed of minerals rich in alumina was altered by the action of intruded felso-liparite dyke.

## Syōkōzan Rōseki Mine, Bingo Prov.

The Syōkōzan Rōseki Mine is located about 5 km north of Syōbara railway station, the end of the Geibi-line, and includes Syōkōzan (947 m in height from sea level), Imagusi (946 m) and Kayagoyama (726 m).

The Rōseki-mass is found here and there in the district, but the main orebody is distributed at the foot of Syōkōzan. Syōkōzan shows mainly porphyrite and liparite, which intruded through the sandstone believed to belong to the Palæozoic formation; more or less metamorphosed masses of Palæozoic formation are found as xenolith within the igneous rocks. The igneous rocks were intruded by the dykes of quartzose rock which had close connection with the formation of the Rōseki deposit. It is very interesting to the writer that the geological relation and Rōseki formation at this mine are like those at Mituisi Rōseki Mines.

The Rōseki is mined by two Rōseki Companies, one the East Syōkōzan Co., the other the West Syōkōzan Co. The ores vary in aluminous content, and if they are named according to the ores used at Mituisi, they are Tokusyu-Rōseki, Zyō-isi, Tyū-isi, Ma-isi and Ge-Ma-isi. It can be macroscopically and microscopically distinguished that the ore composed mainly of pyrophyllite originated from the xenolithic mass of shale within liparite or porphyrite, and the xenolithic mass of liparite or porphyrite within quartzose rock; also that the best ore composed of minerals rich in alumina originated from the xenolithic mass of shale within the dykes of the quartzose rock.

The best ore-body having various shapes and sizes occurs within or along the dykes of quartzose rock, and one mass is never connected with another.

Besides many Rōseki-masses, the pyrophyllitized shale is found here and there, and pyrophyllitized conglomerate (the matrix of the conglomerate was altered into pyrophyllite, but the pebbles are not altered, so the conglomeratic structure was preserved) shows a certain distribution.

Judging from geological relation of these Rōseki mines as described in foregoing pages, it may be concluded that the greater part of the Rōseki being especially rich in alumina, originated from the xenolithic masses of sedimentary rock within felso-liparite or quartzose rock.

Foreign Deposits relating to the Mituisi Roseki Deposit

A great many reports on the deposits relating to the Mituisi Rōseki Deposit were published by various investigators, including E. S. LARSEN,<sup>37</sup> C. H. CLAPP,<sup>40</sup> A. F. BUDDINGTON,<sup>41</sup> J. L. STUCKEY,<sup>45</sup> F. K. PAUL<sup>51</sup> and others,<sup>33,34</sup> and they considered that the deposits were alteration products of rhyorite, Triassic and Jurassic volcanics, volcanic tuff or lava, and feldspathic quartz diorite or andesite porphyry. But it may be considered from their papers that the deposits, especially at Conception<sup>41</sup> Bay, Newfoundland, the Deep River Region<sup>45</sup> of North Carolina and White Mountain<sup>51</sup> in California, originated from sedimentary rocks and that the greater part of the deposits originated from the xenolithic masses of the sedimentary rocks which were caught up by igneous rocks, the xenolithic masses having changed so much that the bedding planes of the rocks were scarcely noticed even by careful observers; the ore originating from the sedimentary rocks was consequently neglected by them.

## 2. Considerations from the Point of View of Ore Deposits

The precise study of the mode of occurrence of a deposit is very important in finding the genesis of it, especially of the Roseki deposit, and the original rocks of the Roseki deposit and the process of the alteration into Roseki can not be known except by actual, precise observation on the mode of occurrence of the deposit. A great many reports on the mode of occurrence of the Roseki deposit at various localities in Japan, have been published by different investigators, including Dr. T. KOCHIBE,<sup>2,5</sup> Mr. H. AWAZU,<sup>7</sup> Dr. T. Ogura,<sup>13, 15</sup> Dr. B. Yoshiki,<sup>17, 23, 24</sup>, Mr. K. Ihara,<sup>11</sup> Mr. Y. Kinosaki<sup>18, 22</sup> and Mr. K. TAMURA, and others, but they have also said that the deposit occurs as veins except that Mr. K. IHARA<sup>11</sup> has observed that the deposit was rarely contained within porphyrite as an amygdaloidal mass. Although many authors have declared that the Roseki deposits occur as vein, they may have perceived that the deposits generally differ from normal veins of metallic deposits, for they added in their reports that the Roseki deposits are the chain of many masses, and are irregular, amygdaloidal and lens-like in form.

The latest report on the Rōseki deposit at Syōkōzan was published by Dr. B. YOSHIKI,<sup>17, 20, 23</sup> and he stated that the deposit at Hiroseguti, West Syōkōzan, is a platy deposit, but that it is a chain of masses globular, lenslike, and irregular in shape, and generally known as "Komori." He also said that the deposit differs from the normal vein of metallic deposit.

The writer inspected the deposit referred to in YOSHIKI's<sup>17,20,23</sup> report, Hiroseguti, West Syōkōzan, and found that each mass of Rōseki was wholly enveloped by quartzose rock, there being no connection with one another.

Many Rōseki deposits forming xenolithic masses are distributed within or along the dyke of the quartzose rock; the direction of distribution therefore coincides with the strike and dip of the dyke. The deposit may have been wrongly observed as a vein by Dr. B. YOSHIKI. On the mode of occurrence of the pyrophyllite deposit, J. L. STUCKEY<sup>45</sup> wrote in his report " On the Pyrophyllite Deposit of the Deep River Region of North Carolina" as follows:

"A prominent feature of the pyrophyllite bodies is their irregular, oval, or lens-like form. They lie in a narrow band or zone aligned with the strike." It may be considered that the occurrence of the deposit is like that of the Rōseki deposit in Japan.

F. K. PAUL<sup>51</sup> stated in his report "On the occurrence of Andalusite and related Minerals at White Mountain, California" that the andalusite occurs in irregular aggregation in a lenticular quartz mass, which forms the prominent show, and that a large portion of the quartz contains andalusite, only a limited area containing enough mineral to warrant exploitation. The andalusite from North stope, the Big stope, and the South stope are referred to as C. P. andalusite; the material, however, is not chemically pure, pyrophyllite and diaspore being commonly mixed with the andalusite. From his description, it may be considered that each mass of the ore was independently distributed, though he did not say so specifically, and that each mass of the ore originated from a xenolithic mass within the quartzose rock.

The Rōseki mass is platy, oval, globular, lens-like, reticular, and irregular in form, and it is observed not only at Mituisi, but at many other Rōseki mines in Japan which were inspected by the writer. It is observed that each mass of Rōseki is independently contained within igneous rocks, and there is no connection with one another. From its mode of occurrence, it may be considered that the Rōseki originated from the xenolithic mass within igneous rocks, and that the xenolithic mass originated from the blocks and fragments of sedimentary rocks which existed at the base, or occasionally from igneous rocks into which felso-liparite or quartzose rocks intruded.

## 3. Consideration based on Chemical Composition

It may be considered that the alteration into Rōseki from shale is more probable than from liparite or porphyrite, judging from the chemical composition of the ores and their original rocks.

	1	2	3	4	5	6	7
$SiO_2 \dots \dots$	81.67	75.58	75.06	62.20	47.37	51.23	2.654
$TiO_2 \dots$	0.08	0.35	0.19	nil.	0.24	0.26	0.068
$Al_2O_3 \ldots$	10.41	17.66	15.41	29.93	36.47	38.12	82.167
$Fe_2O_3 \ldots$	0.29	0.15	0.81	0.11	0.12)	0.14	0.007
FeO	0.29	0.10	0.33	9.04	0.08∫	0.14	0.007
MnO	0.03	nil.	0.01	tr.	nil.	nil.	tr.
MgO	0.01	0.04	0.04	0.06	0.05	0.04	0.016
CaO	nil.	0.08	0.03	0.14	0.51	0.08	tr.
Na <sub>2</sub> O	0.74	0.86	0.27	1.74	0.59	0.58	tr.
К2О	4.64	0.05	3.24	0.18	5.44	0.63	tr.
$F_2O_5 \ldots \ldots$	0.01	0.07	0.03	0.06	0.16	0.11	nil.
$H_2O(+)$	1.30	3.57	3.16	6.20	6.20	8.55	14.691
$H_2O(-)$	0.54	0.69	1.09	0.32	2.69	0.50	0.398
F	0.01						
Total	100.02	100.40	99.67	100.93	99.92	100.24	100.001

- 1. Country rock (Liparite).
- 2. Ma-isi from liparite (from 6th gallery at Mituisi Roseki Co. Mine).
- 3. Xenolithic mass of shale (near the Eisei gallery, Mituisi Roseki Co. Mine).
- 4. Pyrophyllite, matrix of coarse grained Tokusyu-Rōseki from 2nd gallery at Mituisi Rōseki Co. Mine.
- 5. Zyö-isi from 5th gallery at Mituisi Röseki Co. Mine.
- 6. Zyō-isi no. 1.
- 7. Tokusyu-Rōseki no. 1 (composed of crystalline diaspore) from 3rd gallery at Mituisi Rōseki Co. Mine.

In the foregoing table, column 1 indicates the chemical composition of liparite composing the Mituisi Rōseki Mine; no. 2 the Ma-isi preserving the porphyritic structure as liparite; no. 3 the shale which was caught up by liparite; and nos. 4, 5, 6, and 7 the pyrophyllite, Zyō-isi, Zyō-isi no. 1, and Tokusyu-Rōseki no. I which are thought to have been altered from xeno-lithic masses of shale within felso-liparite. Porphyritic structure is never observed.

In comparing the chemical composition of the ores with their original rocks, a significant decrease in  $SiO_2$  and alkalies and an increase in  $Al_2O_3$  and  $H_2O$  in the composition of the ores are detected, but it may be assumed that the addition of  $Al_2O_3$  in the ores was not caused by a supply of  $Al_2O_3$  from another source, and that it increased proportionally according to the subtraction of  $SiO_2$  and other components from the chemical compositions of the original rocks.

F. K. PAUL<sup>51</sup> seems to have suggested the same consideration in his report "On the Occurrence of Andalusite and Related Minerals at White Mountain, California"; thus he writes: "Judging from minerals formed, water vapour, boron, fluorine and sulphur must have been present in a more or less gaseous condition in the quartzose rock (mother rock). The corundum-andalusite-diaspore series appears, however, to have been formed largely from recrystallization of aluminous matter already at hand and not introduced alumina. The conditions under which the minerals have been formed do not appear to be those under which they would be derived directly from an igneous source.

The great excess of quartz forming the host for the andalusite mass would be expected to counterbalance an excess of alumina if produced by direct crystallization from magma."

Though he has not clearly declared in what manner "Aluminous matter already at hand" existed, the xenolithic shale mass as seen at Mituisi Rōseki Mine, and elsewhere in Japan.

It may be perceived that the concentration of  $Al_2O_3$  in the ores were originated from proportional concentration with the subtraction of  $SiO_2$ from the compositions of their original rocks, when we compare those amount in analyses of the ores with the recalculated values of  $SiO_2$ ,  $Al_2O_3$ and  $H_2O$  which were introduced from the original rocks in following manner:

(1) Comparing the chemical composition of each ore with its original

112

rock, subtracted amounts of the components, except  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  which were related to the compositions of Rōseki will be firstly distributed proportionally to the amounts of  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  in the composition of the original rock.

(2) Comparing the recalculated amount of SiO<sub>2</sub> of the original rock with that of each ore, a differential amount of SiO<sub>2</sub> will be distributed proportionally to the amounts of Al<sub>2</sub>O<sub>3</sub> and H<sub>2</sub>O in the recalculated composition of the original rock. It will therefore be found that the recalculated amounts of Al<sub>2</sub>O<sub>3</sub> and H<sub>2</sub>O are very similar to the amounts in the ore, especially the amount of Al<sub>2</sub>O<sub>3</sub>.

The recalculated amounts of  $Al_2O_3$  and  $H_2O$  which were obtained in the foregoing table are as follows:

(A) Comparing column 1 (liparite) with 2 (Ma-isi altered from liparite), weight percentages of the three components,  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  are:

Column	1	Column 2
$SiO_2 \dots \dots 81.67$		75.58
$Al_2O_3$ 10.41		17.66
H <sub>2</sub> O 1.30		3,57

The subtracted amount of the other components, excepting the upper three in column 2, compared with column 1 is 4.27. This amount (4.27) was distributed proportionally to the three components in column 1, and the recalculated values are.

SiO <sub>2</sub> .					85.40
$Al_2O_3$		•			10.99
$H_2O$ .					1.36

Comparing the recalculated value of SiO<sub>2</sub>, 85.40 with 75.58 in column 2, the differential amount 9.82 was introduced, and this amount was distributed proportionally to the amounts of  $Al_2O_3$  and  $H_2O$  in column 1.

The result are:

 $Al_2O_3$  , , , , 19.14 (17.66 in analysis of column 2)

 $H_2O\ldots\ldots$  2.39 (3.57 in analysis of column 2)

It will be found that the recalculated values of  $Al_2O_3$  and  $H_2O$  are similarly situated to those in the analysis, especially the amount of  $Al_2O_3$ .

(B) The following table indicates the recalculated volues compared with amount in the analysis:

			4	5	6	7
a.	Weight % of three com- ponents in analysis.	$(Al_2O_3 \ldots 2$	9.93	36.47	38.12	82.167
		{H₂O	6.20	6.20	8,55	14.691
		$(SiO_2 \dots G)$	2.20)	(47.37)	(51.23)	(2.650)
b.	Recalculated values	$(Al_2O_3 \ldots 2)$	8.66	38.75	37.97	80.27
		$H_2O$	6.18	8.14	8.15	16.32

As seen in the table, the amounts of the three components will be found about the same, when we compare the recalculated values with the analyses, especially alumina.

Although the content of alumina varies in each ore, it may be clearly found that the amount of alumina in the ore originated from its proportional concentration after the subtraction of  $SiO_2$  in various amounts from

the composition of the original rock, and that the alumina in the ore was not introduced from magma or other sources.

The next table shows the chemical composition of shale from many localities. (ROSENBUSCH: Elemente der Gesteinslehre, p. 577, 1922)

•						
Chemical comp.	1	2	.3	4	5	6
$SiO_2\ldots\ldots\ldots$	55.02	60.40	67.35	57.82	47.40	61.91
$TiO_2\ldots\ldots\ldots$	0.65		August #	2.20	0.96	
$Al_2O_3$	21.02	18.58	20.66	25.80	36,10	21.73
$Fe_2O_3$	5.00	7.16	2.55	0.45	0.40	
FeO	1.54	******			-	4.73
$MnO \ \ldots \ldots \ldots$	sp.			+	terest.	*****
MgO	2.32	2.89	0.31	0.55	0.64	0.59
CaO	1.60	0.43	sp.	0.05	0.36	0.09
$Na_2O$	0.81	0.21	not good		0.15	0.25
$K_2O$	3.19	4.70	not good	L	0.17	3.16
$H_2O$	5.65	5.04	6.70	13.21	13.68	6.73
$P_2O_5\ldots\ldots\ldots$	0.06					
Total	100.54	99.41	97.57	100.08	99.86	99.89

- Mittelcambrischer Schieferton. Coosa Valley, Alabama. Mit 0.04 BaO, 0.03 Li<sub>2</sub>O, 2.44 hygroskop. Wasser, 0.83 CO<sub>2</sub>, 0.02 SO<sub>3</sub>, 0.32 kohliger Substanz.
- 2. Schieferton, graublau mit roten Streifen, cambrisch. Am Kundaschen Bach. östlich Revel, Estland.
- 3. Schieferton aus dem Liegenden der Saarbrückener Kohle.
- 4. Schieferton karbonisch, hellgelblichgrau mit grauen Adern. Rothellschacht, Blatt Zweibrücken, Rheinbayern.
- 5. Schieferton karbonisch. Grube Nordfeld, ebenda, mit Spur Li<sub>2</sub>O.
- 6. Schieferton karbonisch, England. Mit 0.70 organischer Substanz.

From the foregoing table, the three components in each column  $SiO_2$ ,  $Al_2O_3$  and  $H_2O$  which were related to the principal component minerals of Rōseki are selected and the recalculated percentages between them are as follows:

Chem. comp.	1	2	3	4	5	6	
$SiO_2 \dots \dots$	67.26	71.89	71.11	59.47	48.78	63.48	
$Al_2O_3$	25.73	22.11	21.81	26.54	37.15	24.04	
$H_2O$	7.01	6.00	7.08	13.09	14.07	7,48	
Mineralogical	compo	nents of	pyropy	llite and	kaolin	ite are a	us follows:
Chem. comp.		Pyroph	yllite		Kaol	inite	
SiO2		66.70	)		46.	50	
$Al_2O_3$		26.30	)		39.	50	
$H_2O$		5.00	)		14.	00	

In comparing these recalculated values with the components of pyrophyllite or kaolinite, columns 1, 4 and 6 are almost similar to that of pyrophyllite, and column 5, to kaolinite; it may therefore be considered that some shale having the same amounts to the chemical composition of pyrophyllite and kaolinite, will be easily recrystallized into these minerals without the subtraction or addition of varying amounts of the three components from the shale, and that most shales contain more  $AI_2O_3$  than the liparite or porphyrite. It may be considered that if there were agents to subtract SiO<sub>2</sub> in any degree from the composition of the shale, the  $Al_2O_3$  would be easily concentrated, and they would be recrystallized into aluminous minerals; also, the alteration into Rōseki, especially rich in alumina, from shales is more probable than from liparite or porphyrite.

#### B. Agents of Metamorphism

## 1. Earlier Theories

Before discussing the agents of metamorphism, the views already expressed by many investigators on the agents of alteration will be referred to.

The agents of the metamorphism mentioned by these authors may be grouped as follows:

- a. Alteration by hydrothermal action which may be attributed to the post volcanism.
- b. Alteration by weathering.
- c. Alteration by pneumatolytic action which may have been introduced by hot gases caused by exhalation from cooling magma.

## a. Alteration by hydrothermal action

A great many investigators believe that the Rōseki-deposits or the deposits associated with the Rōseki were formed by ascending hydrothermal replacement of the country rocks. Among such, we mention Mr. S. OTSUKA<sup>5</sup> (1896), Dr. B SUZUKI<sup>4</sup> (1897), Mr. N. KIYONO<sup>10</sup> and Mr. K. MATSUNO (1919), Mr. T. SAKAMOTO<sup>14</sup> (1922), Dr. B. YOSHIKI<sup>17, 20, 23, 24</sup> (1923, 1933, 1934, 1935), Mr. Y. KINOSAKI<sup>18</sup> and Mr. K. TAMURA (1928), Mr. Y. KINOSAKI<sup>22</sup> (1934) and others<sup>5</sup> in Japan, Mr. L. F. YIH,<sup>40</sup> Mr. F. LEE and Mr. K. CHANG (1931) in China, C. H. CLAPP<sup>40</sup> (1915), A. F. BUDDINGTON<sup>41</sup> (1916), J. L. STUCKEY<sup>45</sup> (1925), F. K. PAUL<sup>51</sup> (1932) and others<sup>33, 57, 39, 46</sup> in foreign countries.

But F. K. PAUL<sup>51</sup> expressed the opinion that the andalusite-deposit, associated with corundum, diaspore, pyrophyllite, muscovite, and alunite as a contact metamorphic deposit, was partly formed by pneumatolytic, and partly by hydrothermal action from his precise investigation of thin sections of the ores, and that the aluminous minerals, andalusite, corundum, and diaspore were formed in the pneumatolytic stage, while pyrophyllite, muscovite, alunite and lazurite were formed in the hydrothermal stage.

If these deposits were formed by ascending hydrothermal solution along the fissure, as these writers supposed, it might be supposed that each mass of the deposit would have close connection with another, and that the passage of the solution within the mother rocks would be more or less traceable.

In the occurrence of the Roseki-deposits at Mituisi, Syokozan and elsewhere as stated by the writer in the preceding pages, each mass was perfectly enveloped by mother rocks, there being never any connection between them; moreover the passage of the solution can not only be traced in the surrounding rock, but the rock has never suffered of feature compared with the other part of the rock. The significant features were reported by Mr. F. YAMANARI<sup>12</sup> in his report "On the Report of Rosekideposit at Indo," and F. K. PAUL,<sup>51</sup> "The Occurrence of Andalusite and related Minerals at White Mountain," PAUL has discussed the agents of metamorphism as follows:

"All the features of occurrence, mineralization, and surroundings suggest aqueo-igneous and pneumatolytic influence. The physical characters of the deposit and its immediate surroundings support this condition."

Though he accepts the pneumatolytic influence in the formation of the deposit, he has expressed the opinion that pyrophyllite, muscovite, alunite, and lazurite were formed in the hydrothermal stage. But the writer, judging from the geological, petrological and ore-depositary points of view, is forced to the view that all the minerals composing the Röseki-deposit at Mituisi, Syöközan and elsewhere were formed by pneumatolytic action.

## b. Alteration by Weathering

T. SAKAMOTO<sup>14</sup> (1922) and H. AWAZU<sup>7</sup> (1919) in Japan, and C. H. CLAPP<sup>40</sup> abroad have expressed the belief that the Rōseki, kaolinite, pyrophyllite and alunite deposits were formed by the influence of descending water, but it may be considered that the Rōseki deposit was not altered by weathering, judging from its occurrence, distribution and other characters.

R. J. LEONARD<sup>47</sup> concluded in his report, "Hydrothermal Alteration of Certain Silicate Minerals," that the silicate minerals were not altered into kaolinite to any noticeable degree by the action of many acidic or alkali solutions. It is to be assumed that the influence of the descending water did not caused the formation of the Rōseki-deposit at Mituisi and other Rōseki mines, judging from various features of the deposit.

#### c. Alteration by Pneumatolytic Action

The pneumatolytic influence on the formation of China-clay and kaolinite deposits has already been noticed by many investigators. R. H. RASTALL<sup>45</sup> (1925) suggested the formation of Cornish China-clay by the action of steam on the feldspar without fluorine or boron; A. STAHL<sup>46</sup> (1912) reported that the China-clay of the Naab Valley in Bavaria, is due to pneumatohydrolysis, caused by ascending carbonic acid solution connected with basalt-magma; H. RIES gave his opinion that the kaolinite of Cornwall, and possibly those of Zettliz in Bohemia, were of deep seated origin and due to fluoric exhalations.

Though the pneumatolytic influence in the formation of the kaolinite deposit has been accepted by many authors, yet such influence in the Rōsekideposit associated with many aluminous minerals, such as diaspore, kaolinite, and pyrophyllite, was not mentioned except that a report on andalusite and related mineral deposits resembling Rōseki at Mituisi was published by F. K. PAUL,<sup>51</sup> and he expressed the opinion that only diaspore was formed by pneumatolytic, and pyrophyllite, by hydrothermal influence. It is possible to consider that all the principal component minerals of the Rōseki-deposit were formed by pneumatolytic action exhalated from the cooling magma of liparite or felso-liparite or other igneous rocks.

## 2. Mechanism of Roseki Formation at Mituisi

The geological, petrological, ore-depositary and chemical features of the Rōseki-deposit at Mituisi may indicate that the deposit was altered from xenolithic masses of shale and liparite by pneumatolytic action, which was due to the cooling magma of liparite and felso-liparite being intruded through the Palæozoic formation.

Judging from the minerals formed, vapour, fluorine, and sulphur must have been presented in gaseous condition within the cooling magma. On the other hand, it has been reported that a fluorite-vein occurs within liparite in the vicinity of Tuyama City near Mituisi. It was found by the writer that the fluorite associated with aluminous minerals at Sammeizi Mine, which is situated along the contact zone of liparite with Palæozoic formation. So it may be considered that the liparite, especially the felsoliparite dykes in this district, contained fluorine in magma stage, and that the fluorine and vapour acted to diminish SiO<sub>2</sub> in the original rocks.

## a. Diaspore, Kaolinite and Pyrophyllite

Though the principal component minerals. diaspore  $(Al_2O_3,$ 85.00, H<sub>2</sub>O, 15.00), kaolinite (Al<sub>2</sub>O<sub>3</sub>, 39.50, SiO<sub>2</sub>, 46.50, H<sub>2</sub>O, 14.0), and pyrophyllite (Al<sub>2</sub>O<sub>3</sub>, 28.34, SiO<sub>2</sub>, 66.66, H<sub>2</sub>O, 5.00) differ in their mineralogical characters, it will be found that there is intimate relation between their chemical compositions, the difference in them being essentially due to the proportionate amounts of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, and H<sub>2</sub>O.

2

The kaolinite must have been chemically introduced from the composition of the pyro-



Fig. 44. Relation of the components of pyrophyllite, kaolinite, and diaspore and their original rocks at Mituisi Röseki Mine.

- 1. Liparite and Roseki from liparite.
- 2. Shale and Roseki from shale.

Black part is the amount of  $SiO_2$  exhaled to alter into each mineral from the original rock.

phyllite by the diminution of  $SiO_2$  and the addition of  $H_2O$  in some amount;  $Al_2O_3$  may be relatively concentrated in amount in kaolinite; diaspore will be introduced from haolinite the same as in the preceding relation.

The same relation may be found between pyrophyllite and xenolithic masses of liparite or shale. If the metamorphic agent was fluorine exhalation, then alkali components in the rocks would be diminished to parallel the loss of SiO<sub>2</sub> from the rocks. Vapour was contained in the magma in all cases, and the H<sub>2</sub>O in the mineral compositions might easily facilitate the recrystallization into pyrophyllite and other aluminous minerals. So we need only discuss the relation of SiO<sub>2</sub> with Al<sub>2</sub>O<sub>3</sub> in the compositions of the minerals and their original rocks.

In Fig. 44, group 1, is diagrammatically indicated the comparison of the components  $SiO_2$  and  $Al_2O_3$  in the aluminous minerals with liparite, and in group 2, with shale. Each column represents the recalculated percentages of  $SiO_2$  and  $Al_2O_3$  which were derived from analysis of the original rock and composition of the minerals. The black part in each column represents the diminished amount that was altered into minerals from the original rocks. It is clearly indicated that the kaolinite and pyrophyllite are intermediate product altered into diaspore, and that the diaspore is the end product of the Röseki formation.

From this, it may be supposed that all the ores except Tokusyu-Rōseki which is composed of pure diaspore, coincide with intermediate steps in the process of alteration into diaspore, that the Rōseki formed from shale needed a weaker agent than that from liparite to produce ores of the same grade, and that the alteration from shale is generally more probable than liparite.

#### b. Pyrite

Pyrite crystal is associated with many Rōseki ores being found especially at the contact part of the xenolithic shale-mass and liparite, or felso-liparite.

It occurs in various ways, sometimes, along the fissure or bedding planes of the shale-masses, and sometimes, at the core of  $R\bar{o}$ seki-mass, or as impregnation.

From its occurrence, it may be allowed to believe that the component, Fe in pyrite did not come from magma or other sources, but originated from the ferric matter in the shale, the ferric matter being recrystallized into pyrite by the action of sulphur dioxyde, vapour, and hydrogen sulphide, which were generally conceived to be present in magma.

It may be generally considered that ferric matter was contained in shale as Fe(OH)<sub>3</sub>, and that it was recrystallized into pyrite by pneumatolytic action somewhat as indicated by the following process:

$$\begin{split} &2SO_2 + O_2 = 2SO_3, \quad SO_3 + H_2O = H_2SO_4, \\ &2Fe(OH)_3 + 3H_2SO_4 = Fe_2(SO_4)_3 + 6H_2O, \\ &2Fe_2(SO_4)_3 + 2H_2S = 4FeSO_4 + 2H_2SO_4 + 5S, \\ &FeSO_4 + H_2S + S = FeS_2 + H_2SO_4, \end{split}$$

Though the ferric matter was contained in shale as  $Fe_2O_3$  or  $Fe_2O_3$ .  $nH_2O$ , it may be supposed that it was easily altered into  $Fe(OH)_3$  by the presence of vapour, and the pyrite being easily produced as indicated in the preceding equation.

# c. Alunite

Alunite was rarely associated with Tokusyu-Rōseki no. 3 composed of diaspore, kaolinite. The well known alunite deposits are distributed in the southern parts of Tyōsen Peninsula, such as Seizan, Kuhiganzan, Gyokumaisan Mines in Kainan District, the Kasatori Mine in Tintō District, and the Tyōzantō Mine in Muan District. The writer visited the Gyokumaisan and Seizan Mines in 1935, and found that the alunite-deposit was like the Rōseki-deposit from the stand-point of all the features.

From the geological, petrological, and ore-depositary features of the alunite-deposit, it may be supposed that the deposit also originated from the xenolithic shale or tuff or porphyrite masses within liparite, and that the masses were altered into alunite by pneumatolytic action due to the cooling magma of the igneous rocks. Geologists considered it to be hydrothermally altered product of liparite or liparitic tuff or porphyrite. The alunite-deposit will be precisely discussed by the writer in another paper.

## Summary and Conclusion

1. The Mituisi Röseki Mine stands on the contact zone of liparite with Palæozoic formation, and on the shattered zone along the fault, from Yakiyama to Mituisi. The geology of this district may be classified into liparite, felso-liparite, pyrophyllitized liparite, shale, pyrophyllitized shale, and Röseki. The shale and pyrophyllitized shale masses originated from the Palæozoic formation which was intruded by liparite and felso-liparite dykes, and they were the xenolithic masses within these igneous rocks.

2. The radial felso-liparite dykes intruded through the liparite from the centre, the highest point in the district. Felso-liparite is commonly porous and has a felsitic structure; phenocrysts are only rarely found within it. It may be considered from the structure that the felso-liparite contained a large amount of gases in its magma, and from the occurrence and distribution that the dykes had an intimate influence in the formation of the Rōseki-deposit.

3. The felso-liparite dykes have a great many xenolithic masses of shale and liparite in various shapes and sizes; these masses were almost completely altered into Rōseki. The outer portion of the xenolithic masses of liparite within felso-liparite as well as the contact zone of liparite and the dykes were altered into pyrophyllitized liparite, the alteration decreasing as it receded from the dykes. It may be clearly seen that the pyrophyllitization of liparite began from phenocryst (feldspar at first) and next from the groundmass. The size of the pyrophyllite recrystallized from phenocrysts is greater than that from groundmass. Porphyritic structure is clearly observed even in the best ore by the difference in the sizes

of the pyrophyllite flakes, and by the scattering of quartz grains within the pyrophyllite flakes as lhe residual minerals. The ore originating from liparite is easily determined from these significant features.

4. The Roseki-deposit originating from shale is contained within liparite, and felso-liparite dykes; in the former case the Roseki-body is composed of pyrophyllite, in the latter the mass is composed of diaspore, kaolinite, pyrophyllite, which were highly altered from original rock. The degrees of alteration are generally proportional to the shapes, sizes, and positions of the xenolithic masses within felso-liparite dykes. It was always observed that the margin of the ore-body was more altered than the core, and that the alteration gradually decreased as one approached the core, but distinct boundaries were not observed.

5. The Rōseki-body originating from shale is generally composed of equilateral minerals; porphyritic structure is never observed and quartz grains are not found.

6. The shape of the Rōseki-body is various, spheroidal, lenticular, platy, reticular (caused by intrusion of liparite or felso-liparite), and irregular, and varies in size from 10 cm to 50 m in length. The Rōseki-bodies are distributed in a scattered manner, each mass being perfectly enveloped by igneous rocks, and never found connected with one another. The shapes of the Pōseki-body may be due to those of the blocks and fragments of original rocks which were caught up by intruded rocks.

7. The component minerals of Rōseki are principally pyrophyllite, kaolinite and diaspore, pyrite and alunite being sometimes found in small amounts as associated minerals. Judging from the occurrence, distribution, geological relation, microscopical investigation, and chemical analysis of the ore, it may be considered that the minerals were recrystallized from original rocks by subtraction of SiO<sub>2</sub> and the proportional increase of  $Al_2O_3$ , and that the amount  $Al_2O_3$  necessary to compose these minerals did not come from other source, so it may be considered that the most important agent of alteration into Rōseki was the subtraction of SiO<sub>2</sub> from the composition of original rocks, and that the ore originating from shale is generally composed of minerals rich in  $Al_2O_3$ , originating from liparite composed of mineral poor in  $Al_2O_3$ , even with the same conditions of alteration.

8. All features of deposit support the conclusion that the metamorphic cause of alteration into Rōseki was the pneumatolytic action generated by the exhalation from cooling magma, especially the magma of felso-liparite. Fluorine, vapour, sulphur dioxyde, and hydrogen sulphide from the cooling magma of felso-liparite were the principal agents in forming the Rōsekideposit.

9. The writer hopes that his study on the Roseki-deposit may contribute to the convenience of inspecting and mining these same deposits.

In conclusion, the writer wishes to express his sincere thanks to Emeritus Prof. T. OGAWA, Prof. Sh. NAKAMURA and Assist. Prof. J. TAKUBO for valuable suggestions given him throughout his study, and he here expresses his cordial thanks to the Mituisi Rōseki Co. for the grant with which the present research has been carried out.

## Literature

- 1. J. TAKAYAMA, The Examination of Roseki. Bul. Geol. Surv. of Japan, no. 1, 1883 (in Japanese).
- 2. T. KOCHIBE, The Röseki. Bul. Geol. Surv. of Japan, pp. 227-236, 1886 (in Japanese).
- 3. S. Otsuka, Explanatory Text of the Okayama Sheet. Geol. Surv. of Japan, pp. 252–259, 1896 (in Japanese).
- 4. B. Suzuki, Explanatory Text of the Hamada Sheet. Geol. Surv. of Japan, pp. 112-115, 1897 (in Japanese).
- 5. T. KOCHIBE, Explanatory Text of the Akō Sheet. Geol. Surv. of Japan, 86-93, 1900 (in Japanese).
- K. CHITANI, Ball-stone from Mituisi. Journ. Geol. Soc. Tökyö, Vol. 22, pp. 384– 385, 1915 (in Japanese).
- H. Awazu, Localities of the Röseki in Tyūgoku. Journ. Geol. Soc. Tökyö, Vol. 26, pp. 360-367, 1919 (in Japanese).
- K. JIMBO, Diaspore from Syöközan. Journ. Geol. Soc. Tökyö, Vol. 26, pp. 283– 285, 1919 (in Japanese).
- A. TAKADA, Diaspore from Syöközan. Journ. Geol. Soc. Tökyö, Vol. 26, p. 461, 1919 (in Japanese).
- N. KIYONO, and K. MATSUNO, Röseki in Okayama and Hyögo Prefectures. Bul. Geol. Surv. of Japan, no. 79, pp. 155-159, 1920 (in Japanese).
- K. IHARA; Clay and Röseki at Gotō, Nagasaki Prefecture. Bul. Geol. Surv. of Japan, no. 79, pp. 169-171, 1920 (in Japanese).
- F. YAMANARI, The Indo Roseki Mine. Journ. Min. Soc. Tyosen, Vol. 5, no. 12, pp. 548-554, 1922 (in Japanese).
- T. Ogura, Explanatory Text of the Syōbara Sheet. Geol. Surv. of Japan, pp. 94– 99, 1922 (in Japanese).
- T. SAKAMOTO, Alunite Deposits at Gyokumaisan and its Surroundings, Kainan District, S. Zenra-Dō. Bul. Chinese Min. Industry, no. 77, pp. 15-43, 1922 (in Japanese).
- 15. T. OURA, Clay and Roseki in Sawa, Yosiki and Tuno Districts, Yamaguti Prefecture. Bul. Industrial Min. Surv., no. 18, pp. 55-59, 1923 (in Japanese).
- K. IHARA, Röseki in Abu District, Yamaguti Prefecture. Bul. Indus. Min. Surv. of Japan, no. 22, pp. 10-39, 1925 (in Japanese).
- 17. В. Yoshiki, Röseki and its Origin at Syöközan, Bingo Prov. Journ. Geol. Soc. Tökyö, Vol. 33, pp. 273-297, 1926 (in Japanese).
- Y. KINOSAKI and K. TAMURA, Diaspore Deposit in Tōei District, S. Keisyō-Dō. Bul. Min. Resource of Tyōsen, 1928 (in Japanese).
- 19. T. ISHIKAWA, On the Aluminous Ores in Työsen. Bul. of the Examination on Ore-Dressing and Smeltery, Työsen, no. 7, pp. 1-49, 1930 (in Japanese).
- 20. В. Yoshiki, Diaspore from Syöközan. Proc. Imp. Acad., IX, no. 3, pp. 109-112, 1933.
- 21. A General View of Japan Ceramics. Ceram. Soc. of Japan, pp. 100 and 108, 1933 (in Japanese).
- Y. KINOSAKI, Alunite Deposits in Kinkai District, S. Keisyö-Dö and Kainan, Tintö and Muan Districts, S. Zenra-Dö, Bul. Min. Surv. of Työsen, Vol. 8, no. 3. 1934 (in Japanese).
- B. YOSHIKI, On the Dickite from Syöközan. Journ. Jap. Assoc. Min Petro. and Econ. Geologists, Vol. 12, pp. 87-95, 1934 (in Japanese).
- B. YOSHIKI, On the Alunite and the Dickite from Seizan Mine, S. Zenra-Dö. Journ. Jap. Assoc. Min. Petro. and Econ. Geologists, Vol. 13, no. 4, pp. 1-20, 1935 (in Japanese).

- B. KAGAYA, Alunite Deposit at Ugusu Vil., Izu Prov. Journ. Min. Inst. of Japan, Vol. 51, no. 693, pp. 109-117, 1935 (in Japanese).
- EMMONS, E., Geological Report of the Middle Countries of North Carolina, Raleigh, pp. 52-54, 1856.
- COLLINS, J. H., On the Nature and Origin of Clay; the Composition of Kaolinite. Min. Mag., Vol. 7, pp. 205-214, 1887.
- 28. LEVY, M. and LACROIX, Mineraux des Roches. p. 254, 1888.
- 29. CROSS and WHITMAN, On the Alunite and Diaspore from Rosita Hills, Corolado. Amer. Journ. Sci., (3), Vol. 41. pp. 466-475, 1891.
- 30. NITZE, H. B. C. and HANNA, G. B., Gold Deposits of North Carolina. North Carolina Geol. Surv. Bul., 3, pp. 44-49, 1896.
- EMMONS, W. H., Genetic Classification of Minerals. Econ. Geol., Vol. 3, pp. 611-625, 1908.
- 32. DANA, E. S., A System of Mineralogy, p. 691, 1909.
- RANSOME, E. L., Geology and Deposits of Goldfield, Nevada. Prof. Paper, U. S. Geol. Surv., no. 66, pp. 189-195, 1909.
- LANEY, F. B., The Gold Hill Mining District. North Carolina Geol. and Econ. Surv. Bul., 21, p. 41, 1910.
- 35. ALLEN, E. T., CRENSHAW, J. L. and JOHNSTON, J., The Sulphide of Iron. Amer. Journ. Sci., Vol. 33, pp. 169-217, 1912.
- BUTLER, B. S. and GALE, H. S., Alunite, a Newly Discovered Deposits near Marysvale, Utah. U. S. Geol. Surv. Bul., no. 511, p. 37, 1912.
- LARSEN, E. S., Alunite in the San Cristobal Quardrangle, Corolado. U. S. Geol. Surv. Bul., no. 530-F, p. 7, 1912.
- 38. LINDGREN, W., Mineral Deposits. pp. 305 and 434, 1913.
- SCHRADER, F. C., Alunite in Patagonie, Arizona, Bovard and Nevada. Econ. Geol., Vol. 8, p. 756 and pp. 764-765, 1913.
- CLAPP, C. H., Alunite and Pyrophyllite in Triassic and Jurassic Volcanics at Kyuquot Sound, British Columbia. Econ. Geol., Vol. 10, pp. 70-88, 1915.
- BUDDINGTON, A. F., Pyrophyllitization, Pinitization and Silicification of Rocks around Conception Bay, Newfoundland. Journ. Geol., Vol. 24, pp. 130-152, 1916.
- BUTLER, B. S., Primary (Hypogene) Sulphuric Minerals in Ore Deposits. Econ. Geol., Vol. 14, p. 586, 1919.
- 43. RASTALL, R. H., Tr. R. G. Soc., Cornwall, 15, pp. 415-438, 1925.
- 44. WYSOR, D. C., Diaspore Clay of Arkansas and Missouri. Journ. Amer. Ceram. Soc., Vol. 6, no. 3, pp. 501-505, 1925.
- STUCKEY, J. L., Pyrophyllite Deposits of the Deep River Region of North Carolina. Econ. Geol. Vol. 20, pp. 442-463, 1925.
- 46. GREGORY, J. W., The Elements of Economic Geology. pp. 152-156 and 168-175, 1927.
- LEONARD, R. J., The Hydrothermal Alteration of Certain Silicate Minerals. Econ. Geol., Vol. 22, pp. 18-43, 1927.
- JEFFERY, and WOODHOUSE, Mining in California. California Bur. Mines, Vol. 27, no. 3, pp. 459–464, 1931.
- L. F. YIH, F. LEE and K. CHANG, The Pyrophyllite from Tsing Tien District. Contrib. from the National Research Inst. Geol. Academia Sinica, no. 1, pp. 1-31, 1931 (in Chine:e).
- BAPGER, A. E. and ALLY, A., Note on the Formation of Kaolin Minerals from Feldspar. Journ. Geol., Vol. 40. pp. 745-747, 1932.
- PAUL, F. K., The Occurrence of Andalusite and related Minerals at White Mountain, California. Econ. Geol., Vol. 27, pp. 614-643, 1932.