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The Pegmatites of the Oku-Tango District of Kyoto Prefecture researched chiefly from the View-point of Trace Elements

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Abstract

The writer statistically made a comparative study of trace elements contained in the granite and pegmatite of the Oku-Tango district, Kyoto Prefecture, and made researches into the mode of the distribution of trace elements during the magmatic differentiation from granite to pegmatites and also into the relation between the characteristic points of the mineral paragenesis of each pegmatite and those of concentrations of trace elements contained in the essential minerals of each pegmatite.

As a result of this study it was found that a tendency in the distribution of trace elements was predominant between granite and pegmatite and among pegmatites themselves, and that even in the same pegmatite a tendency prevailed in the mode of the distribution of trace elements in the same mineral according as a zone varies.

In regard to the above result a few attempts of discussion are made.

Introduction

In the granite of the Oku-Tango district of Kyoto Prefecture, there exist several pegmatites of irregular lenticular shapes about 10 m wide and about 20 m long as shown in Fig. 1.

In Table 1 are shown the mineral parageneses of these pegmatites and from that it is found that these pegmatites are remarkable in the parageneses of their accessory minerals.

By statistically making a comparative study of the trace elements contained in the essential minerals—feldspar, quartz and biotite—of these pegmatites and the granite, in which these pegmatites are distributed, the writer has the follwing two things in veiw—one, the mode of the distribution of trace elements that took place during the magmatic differentiation of granitic magma from granite to pegmatite, and the other—the relation between the characteristics of each pegmatite in the mineral paragenesis and those of the distribution of the trace elements contained in the essential minerals of each pegmatite.

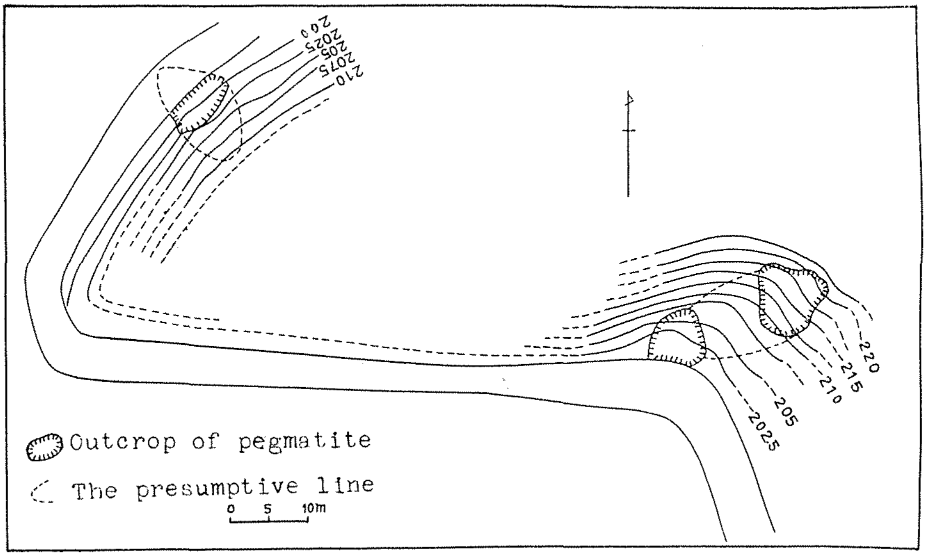

Fig. 1. Map of pegmatite, Oro, Goka village, showing irregular lenticular shape.

Samplings

At first it was planned to collect granites at random throughout the district, but it was impossible to collect fresh samples at some places, therefore these were finally collected at the spots marked with solid circles in Fig. 2.

As regards the feldspar, biotite and quartz of pegmatite they were collected from every clearly distinguishable zone in each pegmatite (see Fig. 3 & Fig. 4).

Feldspars, biotites and quartzs were picked out from granite and prepared other samples according to the methods announced in the previous paper (pp. 301–302)[1].

Spectrographic method for determining trace elements

The analytical method of Ba and Sr in biotite and potash feldspar was modeled after the method announced in the previous paper (pp. 302–305)[1].

As for other minerals and other elements, the spectrographic photographs were taken on the condition mentioned later on, and the relative concentration of every element was detected, based on the number of the spectral lines that could be detected and on their visual relative intensity of every spectral line, and it is shown with the following arbitrary scales: the numeral 10 indicates the highest relative concentration of all the samples of the same mineral kind

Table 1. Mineral parageses of pegmatites

Locarity for pegmatite	Essential Mineral				Accessory Mineral													
					Rare element bearing Mineral									Common Mineral				
	Quartz	Perthite	Plagioclas	Biotite	Zircon	Monazite	Xenotime	Fergusonite	Tscheffkinite	Kōbelite	Allanite	Ilmenite	Sphene	Garnet	Tourmaline	Pyrite	Cupper Pyrite	*** Muscovite
Ōro	+	+	(+)	(+)	×	—	—	×	—	—	—	(×)	(×)	—	—	×	—	×
Kōbe	+	+	—	(+)	(×)	×	(×)	—	×	×	—	×	—	(×)	(×)	×	—	×
Morimoto	+	+	—	(+)	(×)	—	—	—	—	—	×	—	—	—	—	—	—	×
Taniuchi	+	+	—	(+)	(×)	—	—	—	—	—	×	—	—	—	—	—	—	×
Mié	+	+	—	(+)	(×)	—	—	—	—	—	?	—	—	—	—	—	—	×
Nagaoka	+	* +	—	** (+)	—	—	—	—	—	—	—	—	—	—	—	×	×	×

+ : Abundant as an essential mineral.
(+): Considerably abundant as an essential mineral but less than "+".
× : Accessory minerals, and smaller in quantity than essensials
(×): Accessory minerals, and much smaller in quantity than "×".
? : Occurrence is uncertain.
—: Not found.

* The kaolinized mineral.
** The chloritized mineral.
*** The muscovite of a small scale shape which occurred at the latest stage of the occurrence of the pegmatite.

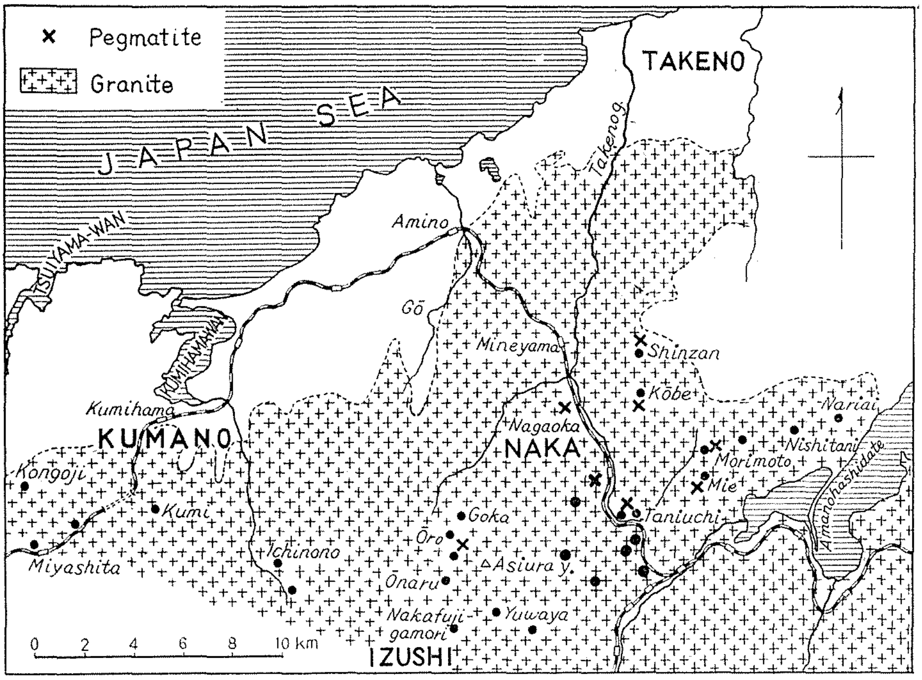

Fig. 2. Locations for pegmatites and samples

in every element and the numeral 1 indicates the lowest, and the symbol "—" indicates the amount of the element contained below the limits of detection.

The scale 1–10 of the concentrations are significant only when these are used for comparison of the concentrations of the same element contained in the same mineral kind except that scales exceedingly vary, such a comparison between different mineral kinds does not convey any meaning, much less between different elements.

To make surer of this experiment, the following experiment was carried out: The samples were divided into groups, each group consists of six samples which are selected at random from samples of the same mineral kind, and these samples were analysed spectrographically according to the above procedure three times respectively. From this result it was ascertained that the scale obtained for every sample was almost the same and the order of the scales was never reversed. Therefore the writer firmly believes that this method holds true with such a statistical comparative study.

Conditions of spectrographic analysis

(1) Respecting the wave length range of 2335Å–4000Å, the Adam Hilger E_2 spectrograph with the width of slit of about 15/1000 mm was used according to

the "Kollimatorabblidung" method. A pure carbon 5 mm across was used as a supplementary electrode and a hole 2.5 mm across and 5 mm deep was made in the lower carbon electrode and the powdered sample of the same weight was put in the hole each time, and the distance between the upper and lower electrodes was held about 5 mm, and the sample was arced at 9–10A.* 220 V., A. C. . Then it was exposed on the Fuji process hard spectral plate for 40–50** seconds, and after it was developed with F. D.–31 (18°C) for 4 minutes according to the brushing method, it was fixed with F. F.–1 (18°C) for 10 minutes.

(2) Respecting the wave length range of 4033.0A–6707.8A, the Adam Hilger D type spectrograph with the width of slit of about 20/1000 mm was used with a condenser lens removed. The sample was arced with the aid of the same supplementary carbon electrode as the above-mentioned, at 9–10A*, 220V, A. C. with the distance of about 5 mm between electrodes. Then after exposing it on the Fuji panchromatic plate for 20–25 seconds**, it was developed with D–76 (20°C) for 11 minutes and fixed with F. F.–1 (18°C) for 10 minutes.

In any case, plates were used out of the same case, the order of photographing the samples was determined by the random number table.

Spectral lines used in spectrographic analysis

Al ;	3082.16,	3092.713,	3944.032,	3961.527,	
Ba ;	2335.269,	4554.04,	4934.09,	5535.551,	
Mg ;	2795.53,	2802.695,	5167.343,	5172.699,	5183.618
Mn ;	2576.104,	2593.729,	4823.52,		
Na ;	2852.8,	2853.0,	3302.323,	3302.988,	
Pb ;	2833.069,	3683.471,			
Sn ;	2839.989,	3034.121,	3175.019,	3262.328,	
Sr ;	3464.57,	3474.887,	4077.714,	4215.524,	
	4305.447,	4607.331,			
Ti ;	3088.025,	3186.454,	3239.038,	3349.035,	
	3361.213,	3372.800,	3383.761,	3635.463,	
	3642.675,	3653.496,	4681.91,	4981.73,	
	4991.066,	4999.510,	5007.213,	5014.241,	
V ;	3102.299,	3110.706,	3118.383,	3183.406,	
	3183.982,	3185.396,	4379.238,	4389.974,	

Granite

It is likely that the granite of this district is a part of batholith that has come to crop out. How it is related to another sediments is plainly as follows :

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(1) * 9A with quartz and biotite ; 10A. with feldspar.
** quartz for 40 seconds ; feldspar for 45 seconds ; biotite for 50 seconds.

(2) * 9A with biotite ; 10A with feldspar.
** biotite for 20 seconds ; feldspar for 25 seconds.

the Older Tertiary sediments cover this rock unconformably and the Tertiary volcanic flows cover them. However, the relation to the Paleozoic sediments is not certain.

This rock is mainly coarse-grained biotite granite, but the granites of Nishitani and Yuwaya contain hornblende, and the granite in the vicinity of Nishitani is porphyritic granite containing large phenocrysts of potash feldspars and it is distributed only in a narrow area.

The results of the chemical analyses of the granites which occur close to the pegmatites of Ôro and Taniuchi, appear in Table 2 and this shows that these granites contain more alkali as compared with the granite in the Chûgoku district presumably of the same age as the above rock. These granites, when they are calculated in "norm" show that these contain very few colored minerals, coinciding with microscopic observation, and these granites are exceedingly acidic. As for the value of K_2O/Na_2O the granite of Taniuchi is smaller than that of Ôro.

The above two granites and the other three were analysed spectrographically, but from these results nothing peculiar was recognized, except that the concentration of V increases or decreases in parallel to the modal percentage of the biotite in granite and that the concentrations of Sr and Ba vary according as the modal percentages of alkali feldspar and plagioclase in the granite increase or decrease.

Collating all the above researches the writer thinks that the granite of this district was most probably derived from the same magma.

Table 2. Chemical analyses of granites and their calculated norms.

Locality Composition	Taniuchi, Mié village	Ôro, Goka village
	%	%
SiO_2	75.38	75.52
Al_2O_3	13.74	13.73
Fe_2O_3	0.20	0.04
FeO	0.55	0.68
MnO	0.17	0.16
MgO	tr.	tr.
CaO	0.99	0.68
Na_2O	3.80	3.78
K_2O	5.00	5.48
$H_2O+)$	0.23	0.45
$H_2O-)$	0.20	0.15
TiO_2	tr.	0.13
P_2O_5	—	—
total	100.26	100.80

Locality Norm	Taniuchi Mié villoge	Ôro Goka village
	%	%
Quartz	31.7	30.8
Orthoclase	29.5	32.3
Albite	32.0	32.0
Anorthite	5.0	3.3
Corundum	0.3	0.5
Hypersthene	0.9	0.8
Magnetite	0.2	–
Ilmenite	—	0.3
Sal./Fem.	85.0	87.3
Q / F	0.5	0.5
K_2O+Na_2O/CaO	8.9	13.6
K_2O/Na_2O	1.3	1.5

Pegmatites

All the pegmatites of this district being embedded in the above-mentioned granite, their chief veins are found in the following six places—Taniuchi (Mié village), Mié (do.), Morimoto (do.), Kôbe village, Nagaoka (Tyozen village), and Ôro (Goka village). They are distributed through the area about 120 square km, as shown in Fig 2.

Any of them is of an irregular lenticular shape and the pegmatite of Ôro presents one example as shown in Fig 1.

When the mineral parageneses of all the pegmatites are compared according to Table I, the pegmatite of Nagaoka is found to be quite different from the other five in the following points: firstly, rare-earth bearing minerals are not produced in it. Secondly, it was strongly affected by the hydrothermal solution with much sulfide. And of the pegmatites producing the rare-earth bearing minerals, the three pegmatites of Mié village exceedingly resemble one another in mineral paragenesis, discriminating themselves greatly from the remaining two which have, however, mineral paragenesis of their own.

As for plagioclase, the pegmatite of Ôro alone contains its small quantity.

In most cases essential minerals of the pegmatite are zonally arranged as shown in Fig. 3 and Fig. 4.

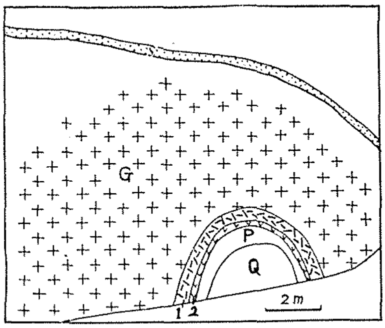


Fig. 3. Sketch of the pegmatite with zonal arrangement of graphic granite and minerals, Oro, Goka village.

Q : Quartz zone
P : Perthite zone
1 : Graphic granite zone which is rich in biotites of bamboo leaf shape.
2 : Graphic granite zone which is poor in biotites.
G : Granite
S : Soil

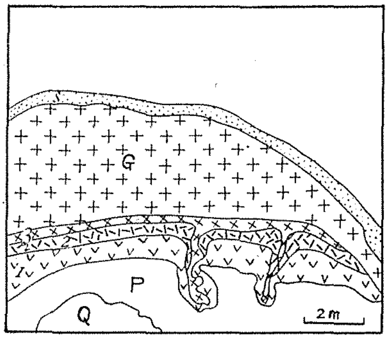


Fig 4. Sketch of the pegmatite with zonal arrangement of graphic granite and minerals, Morimoto, Mié village.

Q : Quartz zone P : Perthite zone
1 : Graphic granite zone which is poor in biotites
2 : Graphic granite zone which is rich in biotites of bamboo leaf shape
3 : Fine-grained two mica granite zone.
G : Granite S : Soil.

Table 3. Chemical analyses of feldspars.

Locality / Composition	Oro, Goka village	Taniuchi, Mié village	Taniuchi, Mié village
	%	%	%
SiO_2	65.19	65.00	64.26
Al_2O_3	20.22	20.81	19.47
Fe_2O_3	0.66	0.91	tr.
FeO	—	—	—
MnO	0.09	—	—
MgO	tr.	—	—
CaO	0.56	0.29	0.72
Na_2O	4.71	3.52	3.21
K_2O	8.53	9.62	11.93
H_2O^{+}	0.12	0.19	0.63
H_2O^{-}	0.12	0.22	0.10
total	100.20	99.84	100.32
Color	pale red	pale red	pale gray

As for the rare-earth-bearing minerals, they occur in the shape of idiomorph, or hypidiomorph in the perthite or biotite of a perthite zone in all cases except that ilmenite occurs in the quartz zone of Kôbe pegmatite.

Pyrite and copper-pyrite occur *in quartz zone or quartz veinlets* injecting the other zones; as a rule, the former is cubic in form, the latter massive.

Perthite is either pale gray or pale red and the results of the chemical analyses of those from Ôro and Taniuchi appear in the Table 3.

Fersmann, V. A[2] [3]. and Schaller, W. J[4]. expressed the pegmatite yielding from the residual solution of granitic magma with the term of "granitic pegmatite" and the pegmatite of this district just corresponds with it.

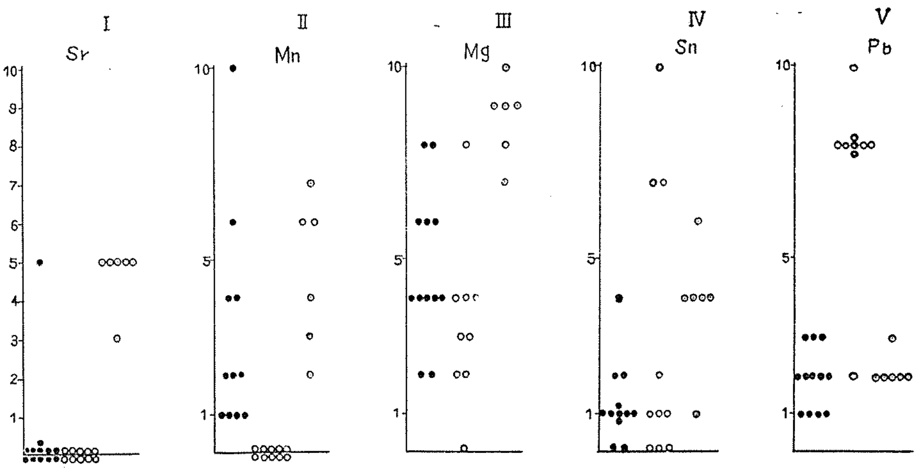
Moreover, Kemp, J. F[5]., Goldschmidt, V. M[6]., Fersmann, V. A[2][3]., Hess, H. L[7]., Schaller W. J[4]., Hosking, K. F. G[8]. &c pointed out that at the end of the stage when granite was solidified volatile components and some special rare elements were concentrated in the residual magma and it intruded into fissures and openings in the surrounding granite. The writer is of opinion that the pegmatite of this district has also the process of occurrence that can be explained by the above theory.

The results of the spectrographic analyses of trace elements contained in the essential minerals of granite and pegmatite

As regards feldspar, quartz and biotite contained in the granite and pegmatite, the results of analyses of the trace elements in these minerals appear in the Table 4 and Figs. 5, 6, and 7. The numbers given on the left side of the figures are the arbitrary scale of the concentration of the element, "O" indicating "not detected".

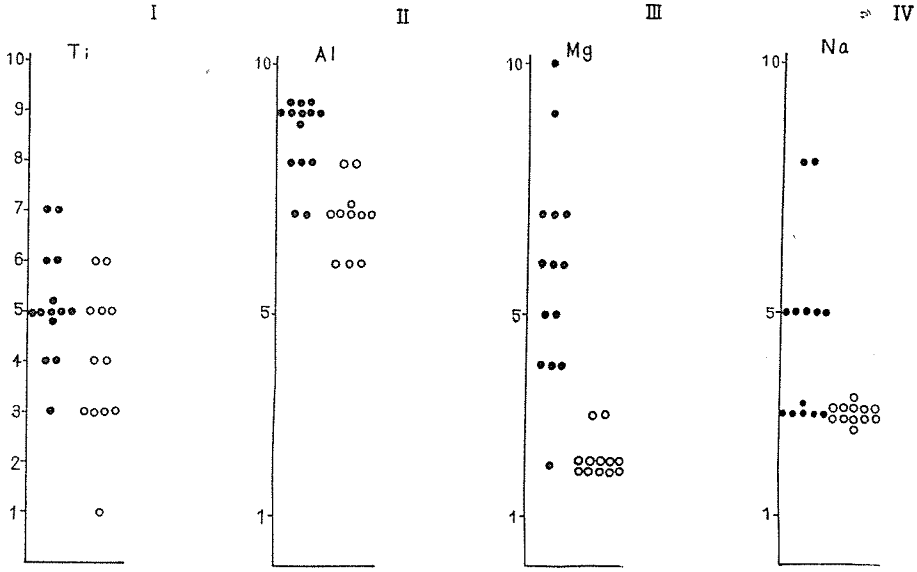
Discussion

From the results shown in Figs. 5–7, it is clear that any element is contained in a larger quantity in the minerals of granite than in the corresponding



- ● Potash feldspar contained in granite
- ○ Potash feldspar contained in pegmatite
- ◎ Plagioclase contained in granite

Fig. 5. Data for concentrations of trace elements contained in feldspars of granites and pegmatites



- ● Quartz contained in granite
- ○ Quartz contained in pegmatite

Fig. 6. Data for concentrations of trace elements contained in quartz of granites and pegmatites

Table 4. Concentrations of trace elements contained in biotites and feldspars of pegmatites.

Zone Mineral Element Locality for Pegmatite	Graphic granite				Perthite			
	Potash feldspar		Biotite		Potash feldspar		Biotite	
	Ba	Sr	Ba	V	Ba	Sr	Ba	V
Ôro	— 2 2 2 2	— — — — —	* 2 **tr. **—	2 tr. —	2 2 2 2 4	— — — — —	— — —	— — —
Kōbe	4 2	4 —	4 4 4	4 4 4	4 2 4 2 4	— — — — —	3	2
Morimoto	* 8 * 8 ** 8 ** 8 8	6 6 4 8 4	6 5	5 5	4 2 2 2 2	— — — — —	4 4 7	3 3 4
Taniuchi					4 4	4 4	4	3
Mié					6 6 6 6	4 4 4 4		

* the concentration of Ba contained in the sample which occurred in the outer part of the graphic granite zone containing biotites in abundance.

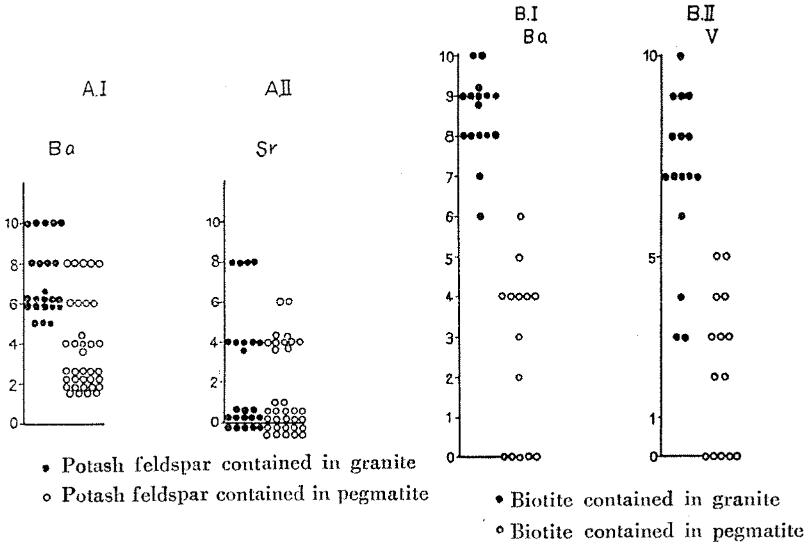
** the concentration of Ba contained in the sample which occurred in the inner part of the graphic granite zone containing biotites in a small quantity.

minerals of pegmatite. What this phenomenon denotes is that, of all the trace elements contained in magma at the stage of magmatic differentiation, most elements (except Pb. Sn) contained in the essential minerals (host minerals) which crystallized out at the same stage, were fixed more in the host minerals which had crystallized out at the earlier period of the stage i. e. the granitic stage and consequently they were fixed less in the host minerals which had crystallized out at the later period of the stage i. e. the pegmatite stage.

Fig. 8 (A, B and C) was drawn according to Table 4 to ascertain the difference of pegmatites in distribution of trace elements.

From these figures the following relationship may be summerized:

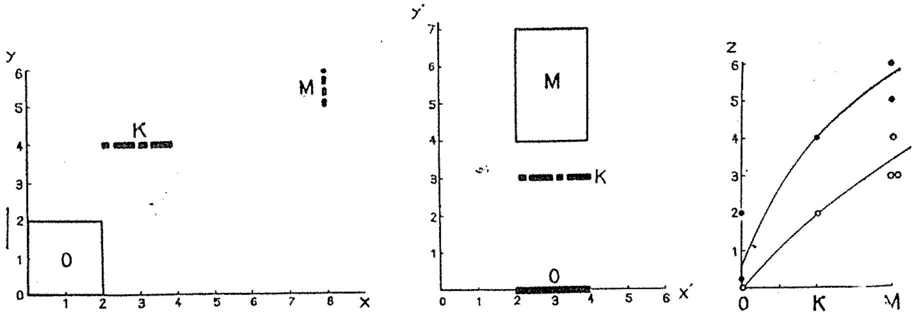
(1) *Positive correlation is noticeable between the concentrations of Ba and V,*



• Potash feldspar contained in granite
○ Potash feldspar contained in pegmatite

• Biotite contained in granite
○ Biotite contained in pegmatite

Fig. 7. Data for concentrations of trace elements contained in potash feldspars and biotites of granites and pegmatites.



A B C

Fig. 8. A. Concentration of Ba contained in the potash feldspar of the graphic granite zone is plotted against the one contained in the biotite of the same zone.

Fig. 8. B. Concentration of Ba contained in the potash feldspar of the perthite zone is plotted against the one contained in the biotite of the same zone.

Fig. 8. C. Concentration of V contained in the biotite of the pegmatite. Each solid circle represents that of the graphic granite zone; each open circle, that of the perthite zone.

x : Concentration of Ba contained in the biotite of the graphic granite zone.
y : Concentration of Ba contained in the potash feldspar of the graphic granite zone.
x′: Concentration of Ba contained in the biotite of the perthite zone.
y′: Concentration of Ba contained in the potash feldspar of the perthite zone.
z : Concentration of V contained in the biotite of the perthite zone.
O: The pegmatite at Ôro, Goka village.
K: 〃 〃 at Kōbe village.
M: 〃 〃 at Morimoto, Mié village.

while biotites produced from the same zone of the same pegmatite show almost the same concentration respecting those two elements and on the other hand, the concentrations of the above elements decrease in the order of the pegmatites of Morimoto, Taniuchi, Kôbe and Ôro.

So far as the concentration of V contained in the biotite of each pegmatite is concerned, a graphic granite zone shows a higher concentration than a perthite zone. (Table 4; Fig. 8. C)

(2) Positive correlation is noticeable between the concentrations of Ba contained respectively in the biotite and the potash feldspar of the graphic granite zone of pegmatite (Table 4; Fig. 8. A)

(3) No correlation is noticeable between the concentrations of Ba contained in the potash feldspar of a perthite zone of each pegmatite. (Table 4; Fig. 8. B)

(4) No difference is noticeable respecting the concentrations and kinds of trace elements contained in the quartz of the quartz zone of each pegmatite.

(5) In regard to all these pegmatites, except Taniuchi and Mié, every concentration of Ba and V contained respectively in the potash feldspar and in the biotite of a graphic granite zone of each pegmatite is not lower than that of a perthite zone. And as to the pegmatites of the three places except Taniuchi and Mié, the concentrations of Ba contained in the potash feldspar of a perthite zone are nearly equal to one another, though those of Taniuchi and Mié are greater than the others.

Moreover, in the synthesis of all the above relations, the following interrelations respecting each pegmatite may be perceived.

V and Ba contained in the biotite and Ba contained in the potash feldspar of the graphic granite of each pegmatite decrease in concentration in the order of Morimoto, Kôbe and Ôro.

And furthermore, if the pegmatites of Taniuchi and Mié are set against the others in point of the matter of (5) and the concentrations of Ba and V contained in biotite, they may be the pegmatites showing the highest concentrations side by side with that of Morimoto.

Considering that there is no great difference among all the pegmatites in regard to the kinds and modal percentages of their essential minerals, it is thought that what caused such differences must depend upon the difference of the concentrations of Ba and V contained in the original pegmatitic magma.

On the other hand, Fersmann, V. A[2]. expressed his view regarding the system of granite-pegmatite as follows:

> "Aus den Mineralassoziationen können gesetzmäszig die Elementmigration und deren Gesetze festgestellt werden, die recht eigentümlich sind und auch einen allgemeinen Charakter besitzen, da eigentlich die Kristallisationsbahnen der Pegmatitlösungen durch eine ziemlich konstante Zusammensetzung der Granitrestlösungen angedeutet und nur durch eine Anderung im Gehalt der Akzessorien gekennzeichnet werden. Somit erhalten wir ein recht interessantes Bild der Granitpegmatite, wodurch eigentlich auch die Systematik derselben angedeutet wird.

Ein bestimmter Typus der Pegmatite (z. B. Muscovit-pegmatit) soll nicht als etwas ganz Selbständiges betrachtet werden, sondern als ein Teilstrück eines ganzen, einheitlich fortlaufenden Prozesses."

From this view-point he advocated, on the paragenesis of the accessory minerals of pegmatites, "Genetische Linien der reinen Serie der Granitpegmatite", and indicated the temperatures at which the pegmatite of each type began to occur.

According to this theory of his, all the pegmatites of this district belong to "Reine Linie" and the pegmatite of Morimoto (Taniuchi and Mié) to "Typus 1. Protopegmatite", that of Ôro to "Typus 2. Pegmatite seltene Elemente", that of Kôbe to the type between the above two and that of Nagaoka to "Typus 9. Sulfidpegmatite". And it could be safely inferred that the temperatures at which all these pegmatites began to occur, gradually fall in the order from Type 1 to Type 9.

Therefore the writer may come to the conclusion about the relation between the difference of the temperatures at which pegmatites began to occur and the difference of the concentrations of trace elements perceivable among the pegmatites, which was referred to, thus simply:

"The pegmatite which began to occur at a higher temperature, has a higher concentration of V and Ba portioned out than the pegmatite at a lower temperature."

Finally let it be allowed to make a few supplementary remarks on the causes that gave such characteristic differences in the distribution of trace elements as was mentioned above.

These phenomena, needless to say, occurred controlled not only by the rule Goldschmidt, V. M9). proposed on the idea of ionic radii and ionic charge but also by the rule Rüngwood, A. E10). suggested on the idea of electronegativity, both controlling the variation of the concentration of several elements contained in the magma during magmatic differentiation; and yet just as it was pointed out in the thesis J. Takubo1) and the writer11) had made public, the temperature at which minerals crystallized out can not be ignored as one of the factors. That is to say, the minerals which crystallized out at a high temperature had full capacity as the host minerals of trace elements; consequently it is seen that at this stage (of crystallization) a large quantity of trace elements is fixed in their host minerals and that the concentration of trace elements gradually reduce in the magma at a later stage. In all probability it is thought that the above may be one of the chief reasons.

Summary

The above may be summarized as follows:

(1) The concentration of every element (except Pb and Sn) contained in the

essential minerals of the granite is generally higher than that in the corresponding minerals of pegmatite.

(2) Respecting each pegmatite there is a peculiar difference in the distribution of Ba and V in its biotite and the potash feldspar of a graphic granite zone and this difference is parallel to that of the paragenesis of the accessory minerals of each pegmatite.

(3) As for V in biotite, it is more abundantly distributed in the biotite of an earlier stage than in that of a later stage even in the same pegmatite.

(4) Taking into consideration Fersmann's genetic systems of the pegmatites, there is a marked tendency for us to find a larger quantity of Ba and V in the pegmatite of this district which began to occur at a higher temperature.

Acknowledgement

The writer offers his hearty thanks to the late Prof. Jitsutaro Takubo who gave me much helpful guidance and advice not only in all my study but in equipping and improving the spectrographic apparatus, and the writer also expresses no less gratitude to Prof. Atsuo Harumoto who has given me a great deal of help and instruction.

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