Heterogeneous biotite from Ryoke metamorphic rocks in the Yanai district, southwest Japan

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Abstract Heterogeneous biotite in regard to Ti occurs in the Ryoke metamorphic rocks in the Yanai district, southwest Japan. Where this biotite is associated with reversely zoned garnet, in which the Mn content increases towards the periphery, there is a decrease of the Ti content of the biotite towards the garnet. The change in composition of the biotite approaching the reversely zoned garnet is expressed by the substitution, $\text{Ti}_2\Box^{\text{vi}}\text{Al}^{\text{vi}}_{-2}\text{R}^{2+}_{-1}$. The presence of Ti-poor biotite in contact with the embayed parts of garnet with a highly indented form and the partitioning of Fe and Mg between biotite and garnet suggest that the formation of Ti-poor biotite involved the consumption of this garnet during retrograde metamorphism.

Key words : Ryoke metamorphism, Ti-heterogeneity in biotite, reversely zoned garnet, retrograde metamorphism

Introduction

Sheet silicates such as chlorite and biotite in metamorphic rocks have been assumed to be chemically homogeneous both for theoretical analyses of the compositional zoning of garnets (Hollister, 1966) and also in the study of the distribution of Ti between muscovite and biotite (Guidotti *et al.*, 1977). However, several authors have described sheet silicates with a heterogeneous distribution of Fe and Mg. These sheet silicates are associated with reversely zoned garnet, in which the Mn content increases towards the rim (*e.g.*, Evans & Guidotti, 1966; Kwak, 1970; Kurata, 1972; Tracy *et al.*, 1976).

Evans & Guidotti (1966) described biotite inclusions within a garnet that each has distinct chemical composition which is also different from that of the matrix biotite. In particular, they showed that the biotite included in the core of garnet is richer in Al and Mg and poorer in Ti, K and Fe than those in the matrix. They concluded that biotite included in the core of garnet

preserved the composition of biotite formed under low temperature and that biotite included in the margin has a composition formed close to equilibrium with the matrix. The reverse zoning profile of the host garnet can be explained by the consumption of garnet to produce a part of the matrix biotite. Kwak (1970) showed that the Mn content of biotite in the matrix increases with distance from a reversely zoned garnet and that the Mg content decreases. He ascribed this to an exchange reaction between garnet and biotite having taken place after their formation. Kurata (1972) distinguished two types of chlorite in one sample, one Mg-rich the other Fe-rich. Most chlorite that occurs in the pressure shadow of or as pseudomorph after garnet is richer in Fe than independent grains defining the schistosity. He considered that the Mg-rich chlorite was formed during prograde metamorphism and the Fe-rich chlorite was formed during retrograde metamorphism. Tracy *et al.* (1976) described biotite immediately adjacent to reversely zoned garnet that is more magnesian than elsewhere in the rock, and similarly ascribed this to a local retrograde exchange reaction.

In this study of Ryoke metamorphic rocks in the Yanai district, southwest Japan, I recognized

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Fig. 1. Geological map of the Yanai district, southwest Japan (compiled from Okamura, 1960, Higashimoto *et al.*, 1983, 1986 and the author's study). The asterisk shows the locality of the sample dealt within this paper in detail (sample No. YCH3).

heterogeneous biotite associated with reversely zoned garnet. It is heterogeneous with respect to Ti but homogeneous with respect to the Fe/Mg ratio. This study is concerned with the mode of occurrence and the genesis of such biotite.

Geology and metamorphic zonation

The Yanai district, southwest Japan, is underlain by Triassic-Jurassic sedimentary rocks, the Kuga Group (Kojima *et al.*, 1951), and their metamorphosed equivalents. The Kuga Group mainly consists of mudstone and chert (Fig. 1) and is intruded by Cretaceous granitic rocks, whose K-Ar ages of biotite are 82 to 103 Ma (summarized by Higashimoto *et al.*, 1983). A Rb-Sr biotite whole rock isochron gives a 95 Ma age for a schist sample approximately 2 km from the granitic intrusion (Shigeno & Yamaguchi, 1976).

In the study area, there is a clear increase in the regional metamorphic grade from north to south (Shimizu, 1983) with the exception of the highest grade, sillimanite zone, where the metamorphic conditions are still controversial (Nakajima, 1987).

In this study, I divide the Ryoke metamorphic rocks into five zones by using mineral assemblages of pelitic and siliceous rocks. In order of increasing grade, these are: the chlorite zone, chlorite-biotite zone, biotite zone, cordierite zone and sillimanite zone. The appearance and disappearance of the main constituent phases in each zone are shown in Fig. 2.

Mode of occurrence of biotite and garnet

Biotite Biotite is a common phase in the regions of higher-grade than the chlorite-biotite zone (Fig. 2). Figure 3 shows the Ti content of biotite in ascending order of metamorphic grade. Chemical compositions of biotite were measured using an electron microprobe, Hitachi S550, with an energy dispersive X-ray analytical system, Kevex 8000, at Kyoto University. The analytical method essentially follows Mori & Kanehira (1984). All data are from graphite-bearing pelitic and siliceous rocks. Furthermore, all samples except Nos. YAD12 and YCC12 are also ilmenite-bearing. With the exception of the sillimanite zone, the maximum value of Ti

Phase/Zone	Chl	Chl-Bt	Bt	Crd	Sil
Chl			---		
Ms					---
Bt					
Grt		---	---		---
Crd					
Kfs				---	
And				---	
Sil					---

+Qtz, +Pl, +Gr, ±Ap, ±Zrn, ±Ilm, ±Tur

Fig. 2. Appearance and disappearance of constituent minerals of pelitic and siliceous rocks with ascending metamorphic grade. Abbreviations of minerals follow Kretz (1983), *i.e.*, Chl; chlorite, Ms; muscovite, Bt; biotite, Grt; garnet, Crd; cordierite, Kfs; K-feldspar, And; andalusite, Sil; sillimanite, Qtz; quartz, Pl; plagioclase, Gr; graphite, Ap; apatite, Zrn; zircon, Ilm; ilmenite, Tur; tourmaline.

Fig. 3. The variation of Ti content of biotite in one sample. Based on 22 oxygens. Each box corresponds to one point analysis. The solid and open boxes indicate those in contact with and apart from garnet, respectively. The boxes with a slash indicate those included in garnet. The signs of "n" and "r" in the parentheses following the sample No. mean normally zoned garnet-bearing and reversely zoned garnet-bearing, respectively.

Fig. 4. Relationship between reversely zoned garnet and heterogeneous biotite (*sample No. YCH3*). The numbers show Ti content of biotite based on 22 oxygens.

content of biotite within a single specimen increases with increasing metamorphic grade. The average values of maximum Ti content for each sample are 0.21, 0.25, 0.38 and 0.35 for the chlorite-biotite, biotite, cordierite and sillimanite zones, respectively. This tendency is consistent with the general trend of biotite composition summarized by Guidotti (1984). However, the Ti content of biotite commonly varies considerably even in one thin section (Fig. 3).

There are three distinct modes of occurrence of biotite with heterogeneous Ti content. In pelitic schist with distinct compositional banding, the Ti content in biotite in micaceous layers is higher than that in quartzose layers (*e.g.*, *sample Nos. YBB6, YCA1*). For example in *sample No. YBB6*, the five Ti-richest data are from micaceous layers and the four Ti-poorest data are from quartzose layers. In this case, the heterogeneity mimics that of the host rocks. The second type of heterogeneity is observed in biotite partly replacing cordierite, where the Ti content is lower than those of the matrix biotite (*e.g.*, *sample No. YCB8*, the lowest two data are those replacing cordierite). The third type occurs among biotite in the matrix of pelitic and siliceous gneisses that contain reversely zoned garnet (*e.g.*, *sample Nos. YFE3, YCH3*). It is a particular phenomenon that is the concern of this paper and is described in detail in the next section. This type of heterogeneity is restricted to samples containing reversely zoned garnet. Samples containing normally zoned garnet never have this type of biotite heterogeneity. However, in spite of the existence of reversely zoned garnet, some rocks do not contain Ti heterogeneity (*e.g.*, the heterogene-

ity of Ti in biotite in the sample (*No. YCA1*) is caused by compositional banding).

Garnet Garnet occurs in the higher grade zones than the chlorite-biotite zone (Fig. 2). In the chlorite-biotite and biotite zones, it occurs sporadically in siliceous rocks, while in the cordierite and sillimanite zones, it appears commonly both in pelitic and siliceous rocks. Three distinct zoning patterns of Mn are recognized in garnet. The first type, in which the Mn content decreases with distance from the core, is referred to here as normal zoning. The associated garnet grains are euhedral or subhedral. The second type of garnet has a uniform Mn content and is likewise euhedral or subhedral. The last type shows an outward increase of Mn content in the peripheral margin of the crystal and is called reverse zoning. This type of garnet has a rounded or highly-indented form. Heterogeneous biotite treated in this study only occurs associated with reversely zoned garnet.

Heterogeneous biotite associated with reversely zoned garnet

A remarkable heterogeneity in the Ti content of biotite associated with reversely zoned garnet was recognized in a pelitic gneiss (*sample No. YCH3*, for locality see Fig. 1), occurring as a block captured in granitic rocks from the cordierite zone. The specimen contains garnet, cordierite, biotite, ilmenite, K-feldspar, plagioclase, quartz and graphite as matrix mineral. It also contains small amount of flaky muscovite associated with cordierite and biotite. Cordierite includes fibrous sillimanite.

In the specimen, biotite is the main constituent

Table 1. Analyses of biotite shown in Fig. 4.

sample no. point	YCH3 Bt1●	YCH3 Bt2●	YCH3 Bt3●	YCH3 Bt9●	YCH3 Bt10●	YCH3 Bt17△	YCH3 Bt18△	YCH3 Bt19●	YCH3 Bt20●	YCH3 Bt21●	YCH3 Bt22●	YCH3 Bt23●	YCH3 Bt24●	YCH3 Bt25●	YCH3 Bt26●	YCH3 Bt27●
SiO ₂	35.16	34.74	35.05	35.52	35.25	35.44	34.82	34.87	34.71	35.20	35.41	35.66	35.00	35.70	34.69	35.01
TiO ₂	1.80	2.14	1.82	2.60	3.99	3.10	3.71	2.62	2.71	2.82	3.35	3.24	3.32	3.79	2.31	3.18
Al ₂ O ₃	20.05	19.73	19.85	20.04	19.61	18.40	18.59	19.81	19.34	19.34	19.26	19.53	18.67	19.23	20.52	19.47
FeO	20.21	19.73	19.85	20.04	19.61	21.08	19.18	20.05	19.74	20.22	19.46	18.41	18.18	19.44	19.44	19.07
MnO	0.18	0.16	0.09	0.12	0.15	0.15	0.12	0.14	0.11	0.11	0.24	0.16	0.18	0.23	0.21	0.24
MgO	7.59	7.49	7.33	7.29	6.96	7.72	9.22	7.15	7.18	7.18	7.13	7.43	7.89	7.67	6.81	7.12
Na ₂ O	0.11	0.11	0.11	0.14	0.15	0.16	0.23	0.08	0.00	0.13	0.19	0.22	0.65	0.42	0.28	0.26
K ₂ O	8.71	9.14	8.95	9.32	9.38	9.35	9.27	9.22	9.27	9.26	9.21	9.32	9.18	9.29	9.09	9.08
total	93.91	93.62	92.36	94.39	94.12	95.40	95.14	93.94	93.06	94.06	94.25	93.97	93.27	94.51	93.35	93.43
Si	5.428	5.382	5.503	5.465	5.443	5.435	5.316	5.394	5.420	5.443	5.445	5.468	5.426	5.444	5.383	5.423
Ti	0.209	0.250	0.215	0.301	0.463	0.358	0.426	0.304	0.319	0.304	0.388	0.374	0.411	0.435	0.269	0.371
Al	3.648	3.673	3.546	3.511	3.391	3.325	3.344	3.613	3.558	3.525	3.490	3.529	3.412	3.456	3.753	3.554
Fe	2.609	2.557	2.606	2.579	2.532	2.703	2.449	2.595	2.577	2.615	2.502	2.561	2.317	2.318	2.523	2.470
Mn	0.024	0.020	0.011	0.015	0.020	0.020	0.015	0.019	0.015	0.014	0.032	0.021	0.024	0.030	0.028	0.032
Mg	1.747	1.730	1.716	1.671	1.603	1.764	2.099	1.648	1.672	1.656	1.634	1.688	1.823	1.743	1.574	1.644
Na	0.032	0.032	0.033	0.043	0.045	0.047	0.067	0.025	0.000	0.038	0.057	0.085	0.194	0.124	0.085	0.078
K	1.714	1.807	1.792	1.830	1.849	1.830	1.806	1.820	1.845	1.827	1.807	1.824	1.815	1.808	1.800	1.793
total	15.411	15.451	15.422	15.415	15.346	15.482	15.522	15.418	15.406	15.422	15.355	15.340	15.462	15.358	15.415	15.365

sample no. point	YCH3 Bt28●	YCH3 Bt29●	YCH3 Bt30●	YCH3 Bt31●	YCH3 Bt32●	YCH3 Bt33●	YCH3 Bt34●	YCH3 Bt35●	YCH3 Bt36●	YCH3 Bt37●	YCH3 Bt38●	YCH3 Bt39△	YCH3 Bt40●	YCH3 Bt41●	YCH3 Bt42●
SiO ₂	35.41	34.77	35.06	35.43	35.46	35.41	34.47	35.74	34.98	35.26	35.87	35.49	35.17	35.26	35.27
TiO ₂	3.79	3.42	3.75	3.66	3.75	2.41	1.89	2.03	3.42	3.11	1.39	3.23	2.23	2.33	2.35
Al ₂ O ₃	19.22	18.80	19.47	19.26	19.25	19.91	19.69	20.10	19.41	19.58	19.91	19.31	19.93	20.41	20.01
FeO	19.11	19.01	18.59	19.56	20.19	19.32	17.92	19.25	18.91	19.24	20.47	19.88	20.17	19.33	19.32
MnO	0.17	0.20	0.19	0.19	0.16	0.11	0.08	0.14	0.21	0.18	0.21	0.15	0.16	0.19	0.14
MgO	6.52	7.11	7.07	6.27	6.77	7.24	6.89	7.37	6.54	6.76	7.64	7.21	7.28	7.10	7.70
Na ₂ O	0.16	0.13	0.20	0.26	0.10	0.13	0.63	0.52	0.16	0.11	0.00	0.19	0.17	0.10	0.22
K ₂ O	9.13	9.30	9.02	9.23	9.28	9.30	8.83	9.01	9.08	9.06	9.25	9.19	9.38	9.18	9.06
total	93.51	92.74	93.35	93.86	94.96	93.83	90.40	94.16	92.71	93.30	94.74	94.66	94.49	93.90	94.07
Si	5.473	5.439	5.418	5.473	5.428	5.458	5.489	5.479	5.454	5.462	5.495	5.440	5.413	5.425	5.419
Ti	0.441	0.402	0.436	0.426	0.432	0.279	0.226	0.234	0.401	0.363	0.160	0.373	0.258	0.270	0.271
Al	3.502	3.466	3.545	3.505	3.473	3.615	3.696	3.613	3.567	3.575	3.595	3.489	3.615	3.702	3.623
Fe	2.470	2.487	2.402	2.527	2.584	2.490	2.386	2.468	2.466	2.492	2.623	2.548	2.597	2.488	2.482
Mn	0.022	0.025	0.025	0.025	0.021	0.011	0.011	0.019	0.028	0.023	0.028	0.019	0.021	0.025	0.019
Mg	1.503	1.657	1.628	1.444	1.545	1.664	1.636	1.683	1.520	1.661	1.745	1.647	1.671	1.629	1.763
Na	0.047	0.039	0.060	0.079	0.029	0.079	0.195	0.156	0.048	0.033	0.000	0.055	0.051	0.030	0.066
K	1.800	1.857	1.779	1.819	1.813	1.829	1.794	1.763	1.806	1.790	1.807	1.799	1.841	1.802	1.776
total	15.258	15.373	15.294	15.298	15.325	15.389	15.433	15.433	15.290	15.299	15.453	15.370	15.467	15.371	15.419

Total Fe is treated as FeO and the formula is calculated based on 22 oxygens (anhydrous). ○ : as matrix, ● : in contact with garnet, △ : as inclusion in garnet.

Fig. 5. The X-ray image maps in respect to Ti, Mn and Mg contents of box part of Fig. 4, using Kevex Advanced Image software. The bright part shows high content of each element.

Fig. 6. Ti content versus Fe/(Fe+Mg) ratio of biotite from sample No. YCH3.

mineral of the gneissosity. It also occurs in contact with the embayed parts of garnet with a highly-indented shape and is parallel to the gneissosity. This mode of occurrence of biotite suggests that it was formed in association with the consumption of garnet. The biotite in contact with garnet, which will be referred to as contact biotite herein, is locally pale-brown but is commonly reddish brown and is not distinguishable from matrix biotite in color. The Ti content of biotites in the vicinity of garnet differs from one grain to another and generally decreases towards the garnet from 0.46 to 0.16, as shown in Figs. 4 and 5, which enlarges a box part of Fig. 4. All analyses of biotite shown in Fig. 4 are listed in Table 1. Furthermore, the variation of Ti content of all analyses of biotites in the specimen has no clear relationship with Fe/(Fe+Mg) ratio (Fig. 6).

In this specimen, garnet has a rather homogeneous inner part but is zoned at the extreme periphery (Fig. 5). In the zoned part, the Mn content increases and the Mg content decreases towards the margin causing a corresponding increase of Fe/(Fe+Mg), *i.e.*, 0.88 and 0.82 for the rim and inner part, respectively.

The garnet includes such minerals as biotite, cordierite, ilmenite and quartz. No obvious compositional heterogeneity of garnet can be detected around these inclusions.

The Fe-Mg partitioning between the garnet rim and contact biotite indicates lower temperature than that between the inner part of the garnet and the matrix biotite. Thus I consider that the decomposition of garnet to produce biotite took place under retrograde conditions.

In the present study, both contact biotite and matrix biotite have a similar Fe/Mg ratio, although the garnet is zoned. This observation suggests the validity of the assumption that biotite was homogeneous during the exchange reaction in terms of Fe and Mg, which is required for the modelling of the relaxation of garnet zoning (Chakraborty & Ganguly, 1991). This model is based on a constant biotite composition because this is the dominant phase compared with garnet.

The origin of heterogeneous biotite

The biotite dealt with here belongs to the system $K_2O-FeO-MgO-MnO-Al_2O_3-TiO_2-SiO_2-H_2O$. In the presence of excess quartz and H_2O -

Fig. 7. Composition of biotite in *sample No. YCH3* in the chemical system, $K_2O-R^{2+}O-Al_2O_3-TiO_2-SiO_2-H_2O$, with excess quartz and H_2O -dominant fluid. a) A-K-R²⁺ diagram projected from ilmenite. b) A-Ti-R²⁺ diagram projected from K-feldspar. Arrow corresponds to the substitution, $Ti_2\Box^{vi}Al^{vi}_{-2}R^{2+}_{-1}$. East' : $K_2R^{2+}_5AlSi_5Al_3O_{20}(OH)_4$.

Fig. 8. The mole fraction of biotites shown in Fig. 4 against the distance from garnet in the five biotite endmembers system proposed by Ikeda (1990). Phl : $K_2R^{2+}_6Si_6Al_2O_{20}(OH)_4$, East : $K_2R^{2+}_4Al_2Si_4Al_4O_{20}(OH)_4$, Tlc : $\square^{xii}_2R^{2+}_6Si_8O_{20}(OH)_4$, Ms : $K_2\square^{vi}_2Al_4Si_6Al_2O_{20}(OH)_4$, Ti-Bt : $K_2\square^{vi}_3R^{2+}_3Ti_2Si_4Al_4O_{20}(OH)_4$.

dominant fluid, the biotite can be treated as a six-components system. In order to visualize the behaviour of TiO_2 , the FeO , MgO and MnO components can be combined to define $R^{2+}O$, which makes it possible to deal with the formation of biotite during retrograde metamorphism in the system $K_2O-R^{2+}O-Al_2O_3-TiO_2$.

Figures 7a and 7b show the A-K- R^{2+} plane projected from ilmenite, and the A-Ti- R^{2+} plane projected from K-feldspar, respectively. As shown in Fig. 7a, contact biotite is poorer in "K" component and richer in " R^{2+} " component than the matrix biotite. Furthermore both modes of biotite have a significant range within Phl-East' direction. Figure 7b shows that matrix biotites have higher "Ti" component than contact biotites.

The composition of biotites shown in Fig. 4 can be expressed with the mole fraction of five endmembers as proposed by Ikeda (1990), i.e., phlogopite-eastonite-talc-muscovite-Ti-biotite ($K_2\square^{vi}_3R^{2+}_3Ti_2Si_4Al_4O_{20}(OH)_4$), where $\square^{vi}$ denotes a vacancy in the octahedral site. Each of the mole fractions is plotted against the distance from garnet (Fig. 8). With decreasing distance from the garnet, the Ti-biotite component decreases and the eastonite component increases. The components of phlogopite, muscovite and talc do not show systematic change with distance from garnet. The substitution that connects Ti-biotite and eastonite is $Ti_2\square^{vi}Al^{vi}_{-2}R^{2+}_{-1}$, which is the main substitution observed within these heterogeneous biotites (shown as an arrow in Fig. 7). The expression of substitution follows

Thompson (1982). The similarity in the proportion of the talc component between the matrix and contact biotites means that the poverty of the "K" component in contact biotite shown in Fig. 7a is not due to the real depletion of K_2O but to the enrichment of the octahedral occupancy.

In the retrograde stage, Al and R^{2+} released from resolved garnet are supplied to the existing biotite and occupy the octahedral site, causing a decrease of the octahedral vacancy. However, this process cannot bring about the observed distinct decrease of Ti content of biotite towards the garnet. Furthermore, Ti-poor biotite does not associate closely with Ti-bearing phases such as ilmenite and rutile, as shown in Fig. 4, suggesting that contact biotite was not formed by the depletion of Ti from existing biotite to compensate the increase of octahedral occupancy. Thus, not all of the materials constituting Ti-poor biotite have originated from the matrix biotite. It follows that the contact biotite was formed by retrograde metamorphism and matrix biotite and contact biotite were not homogenized after their formations. The reaction to form Ti-poor biotite may be expressed as

Ti-rich biotite + Mn-poor garnet + K-feldspar
+ fluid

= Ti-poor biotite + Mn-rich garnet + muscovite
+ ilmenite + quartz,

$27 K_2Fe_{2.4}Mg_{1.65}Al_{0.8}Ti_{0.5}Si_{5.4}Al_{2.6}O_{20}(OH)_4 + 12$
 $(Fe_{0.77}Mn_{0.07}Mg_{0.16})_3Al_2Si_3O_{12} + 4.1 KAlSi_3O_8$
 $+ 4.1 H_2O$

$= 28.1 K_2Fe_{2.62}Mg_{1.74}Al_{1.1}Ti_{0.2}Si_{5.4}Al_{2.6}O_{20}(OH)_4$
 $+ 5.1 (Fe_{0.76}Mn_{0.14}Mg_{0.1})_3Al_2Si_3O_{12} + K_2Al_2Si_2Al_2O_{20}-$
 $(OH)_4 + 7.9 (Fe_{0.95}Mn_{0.05})TiO_3 + 21.4 SiO_2.$

The formation of Ti-poor biotite around garnet is not restricted to the Ryoke metamorphic rocks. Higashino *et al.* (1982, p. 89) described biotite in contact with garnet that has a lower content of Ti than the matrix biotite in some Sanbagawa pelitic schists.

I conclude that a part of the chemical heterogeneity of biotite in the study area can be explained by the plural stages of formation of biotite, one is prograde and the other is retrograde.

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* : in Japanese with English abstract

** : in Japanese

(要 旨)

Ikeda, T., 1991, Heterogeneous biotite from Ryoke metamorphic rocks in the Yanai district, southwest Japan. *Jour. Geol. Soc. Japan*, 97, 537-547. (池田 剛, 1991, 柳井地域, 領家変成岩中の不均質な黒雲母. 地質雑, 97, 537-547.)

柳井地域の領家変成岩中の黒雲母に不均質がみられた。Mn 含有量が縁辺部に向かって増加する逆累帯構造を示すザクロ石を含む試料では、ザクロ石に向かって黒雲母の Ti 含有量が減少している。この組成変化は $Ti_2\Box^{vi}Al^{vi}-_2R^{2+}-_1$ の置換で説明される。凹凸の外形を呈するザクロ石の凹部に Ti に乏しい黒雲母がしばしば接すること、および黒雲母とザクロ石の Fe, Mg の分配から、この Ti に乏しい黒雲母は後退変成作用時にザクロ石が分解して生成したと考えられる。