CHEMICAL REACTIONS AT THE BOUNDARY BETWEEN GNEISS AND AMPHIBOLITE IN THE RYOKE METAMORPHIC TERRAIN AT TAKATO, JAPAN

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Introduction

In the Ryoke metamorphic terrain, anorthite-bearing amphibolites often occur enclosed in pelitic gneisses containing sodic plagioclases. At the boundary of these different rocks chemical reactions should have taken place because anorthite, oligoclase and other minerals in these rocks could not coexist in equilibrium. Actually, a sequence of zones, reaction zones, was present in all the contacts examined. Previous studies have confirmed the systematic changes of mineral assemblages and chemical gradients of plagioclase and biotite across the reaction zone (Ono, 1968 MS). It seems highly probable that these zones form by a metasomatic process of the kind discussed by Thompson (1959) and Vidale (1969).

The principal object of this study is to present the analytical results by means of an electron probe microanalyzer and to discuss the chemical reactions between amphibolites and gneisses. The reaction zone studied is the same as that described previously (Ono, 1968 MS, 1971).

Brief description of the reaction zone

Folded amphibolites ranging 2 to 50 cm thick occur enclosed in gneisses at Takato town along the Mibu river. A reaction zone consisting of orthoclase-free and amphibole-free rocks develops between gneisses and amphibolites. The investigated reaction zone is composed of the following zonal sequence from gneiss toward amphibolite: orthoclase zone, andesine zone, biotite zone, and amphibole zone. The orthoclase and the amphibole zones mainly consist of original rocks, gneiss and amphibolite respectively. Both of the andesine and the biotite zones are about 4 cm thick.

Gneissose structure of the orthoclase zone gradually disappears toward the andesine zone where granular texture well develops. The andesine zone shows very fine-grained granular texture in the domain adjacent to the biotite zone. Numerous spots of biotite showing distinct preferred orientation are characteristic of the biotite zone, however, quartz and plagioclase show granular texture. Modal amounts of biotite and opaque minerals are discontinuous at the boundary of the andesine and the biotite zones, hence this sharp boundary is considered to have been the original boundary between gneisses and amphibolites. Thin quartz-feldspathic veins often occur along this boundary. The amphibole zone shows preferred orientation of amphibole and biotite. The examined reaction zone is devoid of biotite-free amphibolite which is not uncommon in central part of other amphibolites. Biotite amphibolite examined here may be metasomatized biotite-free amphibolite.

Main constituent minerals of each zone are as follows: orthoclase zone: biotite, cordierite, garnet, sillimanite, orthoclase, plagioclase (An25), quartz, graphite, pyrrhotite, chalcopyrite; andesine zone: biotite, garnet, plagioclase, quartz, pyrrhotite, chalcopyrite;
Fig. 1 Schematic modal change of minerals across the reaction zone.

O, A, B, and Am are the orthoclase, andesine, biotite and amphibole zones respectively. The boundary between the andesine and the biotite zones is represented by zero in the scale of this and the following figures.

**Biotite zone:** biotite, garnet, plagioclase, quartz, pyrrhotite; **Amphibole zone:** biotite, actinolitic hornblende, plagioclase (An₉₀), quartz, pyrrhotite, chalcopyrite.

A schematic modal change across the four zones is shown in Fig. 1. The smooth curves in the figure were drawn on the basis of simplified modal analyses of many thin sections prepared across the reaction zone. There is a discontinuity in mode of minerals at the boundary of the andesine and the biotite zones. Sillimanite, cordierite and orthoclase in the orthoclase zone decrease their amount and grain size toward the andesine zone. Biotite in the biotite zone abruptly decreases its amount near the amphibole zone.

Mineral assemblages of the four zones are plotted onto the Al₂O₃-K₂O-(FeO+MgO)-CaO diagram as illustrated in Fig. 2. It is obvious that sequences of mineral assemblages systematically change from the orthoclase zone to the amphibole zone.

**Chemical composition of minerals**

Refractive indices, X-ray data and wet chemical analyses of constituent minerals suggest that there are continuous chemical changes of minerals across the reaction zones (ONO, 1968MS). This previous conclusion was checked by means of an electron probe microanalyzer.

Two polished sections of the andesine zone were prepared perpendicular to the boundary of the andesine and the biotite zones. Chemical compositions of biotite, garnet and plagioclase were checked as a function of distance from this boundary which was the original boundary between gneisses and amphibolites. One section contained a lenticular garnet of 4×10 mm across that developed along the zone boundary, but the two sections provided the same result. The analytical results described below are from the another section which does not contain
large garnet crystals.

Another hand specimen was collected from the biotite zone near the specimen stated above. Biotite and plagioclase were analyzed as a function of distance from the boundary of the andesine and the biotite zones.

**Plagioclase:** Line analyses along three traverses revealed continuous changes of An, Ab and Or contents in plagioclases in the andesine zone. Ab and Or contents decrease in the direction toward the amphibole zone. The change of An content is illustrated in Fig. 3.

The An content of plagioclase in the andesine zone increases from core to edge of the crystal except a few homogeneous grains. Antiperthite is often observed in the core of the zoned plagioclases. It should be noted that gradual chemical change of plagioclase is obvious in spite of frequent occurrence of strong reverse zoning.

Plagioclase in the biotite zone shows normal zoning, an increase of Ab content from core to edge. Anorthite (An$_{90}$) is very often in contact with more sodic plagioclases (An$_{70-80}$). Though relict anorthite is common in the biotite zone adjacent to the andesine zone, the most sodic plagioclase in the biotite zone is found in this domain. The An content of newly formed plagioclase in the biotite zone increases slightly toward the amphibole zone as shown in Fig. 3.

**Biotite:** FeO and MgO contents of biotites in the andesine zone were analyzed three times at different position of traverse. The same compositional profiles were obtained. One of them is illustrated in Fig. 4. Biotite in the biotite zone is constant in chemical composition in so far as the biotite within 2 cm from the andesine zone are concerned. However, magnesian biotite may exist in the biotite zone near the amphibole zone, because biotite in this domain has lower refractive index. Representative analytical data of biotite in the andesine zone are given in Table 1 together with those of garnet. Al$_2$O$_3$ content and FeO/MgO ratio of biotite decrease continuously in the direction toward the amphibole zone. Biotite in the amphibole zone has the lowest values of Al$_2$O$_3$ and FeO/MgO. Fe/Fe+Mg ratio of biotite is shown in Fig. 3.

**Garnet:** Nine garnet grains are contained in a thin section from the andesine zone. All the grains have rather homogeneous composition at central part of the crystal, showing increase of MnO content and decreases of MgO and CaO contents from core to edge. A large garnet in another section, however, is normally zoned at central part and reversely zoned at marginal part of the crystal. Volume percentage of central part of small garnets where MnO content is
Table 1. Representative analytical data of biotite and garnet in the andesine zone.

<table>
<thead>
<tr>
<th>Biotite</th>
<th>B6</th>
<th>Am6</th>
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</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>35.20</td>
<td>35.70</td>
</tr>
<tr>
<td>TiO₂</td>
<td>2.26</td>
<td>2.22</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>18.13</td>
<td>18.11</td>
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<tr>
<td>FeO</td>
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<tr>
<td>MnO</td>
<td>0.27</td>
<td>0.31</td>
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<tr>
<td>MgO</td>
<td>10.71</td>
<td>10.59</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>K₂O</td>
<td>8.74</td>
<td>8.74</td>
</tr>
<tr>
<td>Total</td>
<td>95.88</td>
<td>94.56</td>
</tr>
<tr>
<td>mm</td>
<td>16.16</td>
<td>12.49</td>
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Garnet

<table>
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<tr>
<th></th>
<th>1</th>
<th>1'</th>
<th>2</th>
<th>2'</th>
<th>3</th>
<th>3'</th>
<th>4</th>
<th>4'</th>
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<td>SiO₂</td>
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<td>37.13</td>
<td>37.53</td>
<td>37.58</td>
<td>37.62</td>
<td>37.22</td>
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<td>FeO</td>
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<td>28.47</td>
<td>27.67</td>
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<td>26.80</td>
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<td>MnO</td>
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<td>5.17</td>
<td>7.52</td>
<td>5.35</td>
<td>7.70</td>
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<tr>
<td>MgO</td>
<td>3.39</td>
<td>4.18</td>
<td>3.64</td>
<td>5.35</td>
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<td>4.35</td>
<td>3.34</td>
<td>4.47</td>
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<tr>
<td>CaO</td>
<td>1.38</td>
<td>1.59</td>
<td>1.96</td>
<td>2.31</td>
<td>2.15</td>
<td>2.79</td>
<td>2.75</td>
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<tr>
<td>Total</td>
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<td>98.99</td>
<td>99.19</td>
<td>99.26</td>
<td>99.30</td>
<td>98.25</td>
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</table>

mm: the distance from the boundary of the andesine and the biotite zones. ( ) : number of analyzed points. Minerals in other zones are also listed.

rather constant is about 40% in average, though true center of each crystal may be not always observed in thin section.

Chemical composition at the edge of each garnet shows maximum variation of 1.5 wt% in MgO and MnO contents (Fig. 5). Chemical variations of CaO and FeO contents are small compared with those of MgO and MnO contents. Average composition of the edge of four garnet crystals is listed in Table 1 together with the composition at the core. MnO and CaO contents increase in the direction toward the amphibole zone, and FeO content shows the opposite change. Chemical changes in all the analyzed garnets from the andesine zone are shown in Fig. 6 as a function of distance from the original boundary between the gneisses and the amphibolites.

Complete chemical change of plagioclase was ascertained for a thin reaction zone formed at the boundary between quartzo-feldspathic vein and amphibolite, as shown in Fig. 7. This reaction zones was observed in Komagane region, south of Takato. Plagioclase grains in the amphibole zone and in the core of the vein are rather homogeneous in composition respectively, but zoned plagioclases are common in the biotite zone and marginal part of the vein. A few plagioclase crystals in the vein showed increase in An content.

Fig. 5 Compositional variation (wt %) at edges of garnet crystals.

mm: the distance of measured garnet crystals from the original boundary between gneisses and amphibolites.
Fig. 6 Average chemical composition at the edge of garnet crystals in the andesine zone.

Table 2. Bulk chemical composition of the reaction zone.

<table>
<thead>
<tr>
<th></th>
<th>J1</th>
<th>J2</th>
<th>J3</th>
<th>H1</th>
<th>H2</th>
<th>H3</th>
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<tr>
<td>O</td>
<td>6.85</td>
<td>6.48</td>
<td>6.29</td>
<td>6.53</td>
<td>6.32</td>
<td>6.15</td>
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<tr>
<td>A</td>
<td>0.77</td>
<td>0.75</td>
<td>0.66</td>
<td>0.73</td>
<td>1.28</td>
<td>1.29</td>
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<tr>
<td>B</td>
<td>16.93</td>
<td>17.76</td>
<td>18.37</td>
<td>16.87</td>
<td>17.04</td>
<td>15.93</td>
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<tr>
<td>Am</td>
<td>3.23</td>
<td>3.07</td>
<td>3.12</td>
<td>0.53</td>
<td>0.87</td>
<td>0.48</td>
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<td>Cr</td>
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<td>2.48</td>
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<tr>
<td>Mn</td>
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<td>0.07</td>
<td>0.10</td>
<td>0.13</td>
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<tr>
<td>MgO</td>
<td>2.43</td>
<td>2.43</td>
<td>2.43</td>
<td>2.94</td>
<td>5.21</td>
<td>7.27</td>
</tr>
<tr>
<td>CaO</td>
<td>0.38</td>
<td>1.39</td>
<td>1.60</td>
<td>3.78</td>
<td>5.42</td>
<td>8.86</td>
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<tr>
<td>Na2O</td>
<td>1.69</td>
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<td>0.55</td>
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<tr>
<td>K2O</td>
<td>5.04</td>
<td>2.69</td>
<td>2.48</td>
<td>1.86</td>
<td>3.48</td>
<td>1.67</td>
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<td>H2O+</td>
<td>2.06</td>
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<td>0.98</td>
<td>1.50</td>
<td>2.13</td>
<td>1.83</td>
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<td>H2O-</td>
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<td>0.50</td>
<td>0.53</td>
<td>0.20</td>
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<tr>
<td>P2O5</td>
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<td>0.12</td>
<td>0.17</td>
<td>0.09</td>
<td>0.42</td>
</tr>
</tbody>
</table>

Analysis by H. Ikenari

O, A, B and Am are the same as those in Fig. 1.

Table 2. Bulk chemical composition of the reaction zone.

Table 2 shows that the CaO and MgO contents are considerably lower and the K2O content is higher in the biotite zone (H2) than in the amphibole zone (H1). Na2O content in the biotite zone appears to be larger compared with that of the amphibole zone. Chemical compositions of J1, J2 and J3 indicate increase of CaO and decrease of K2O and H2O contents toward the biotite zone. Na2O content is higher in the andesine zone than in the orthoclase zone.

Because migration of one component from a given rock to others accompanies the changes in volume and weight percentage of the other components, it is very difficult to determine slight increase or decrease of each component in the metasomatized rocks. However, compositional changes of K2O, MgO, H2O and CaO are remarkable in the reaction zone, i.e. the andesine and the biotite zones, compared with chemistries of original gneisses and amphibolites here represented by the orthoclase and the amphibole zones respectively. Hence, it is evident that these components have migrated across the reaction zone.

Chemical reaction

There are systematic changes in mineral assemblages and chemical composition of minerals across the studied reaction zone as shown in the attached figures. Migration
of chemical species is also confirmed by bulk rock analysis. These facts suggest that chemical reactions had taken place at the boundary between gneisses and amphibolites.

1. Decomposition of K-feldspar

K-feldspar is absent in the andesine zone which was originally orthoclase zone (gneiss). K-feldspar, cordierite and sillimanite in the orthoclase zone are very small in amount in the domain adjacent to the andesine zone. These facts are reasonably explained by decomposition of these minerals near the andesine zone and removal of $K_2O$ component from the orthoclase zone. Increase of $Na_2O$ content and decrease of $K_2O$ content are remarkable in the andesine zone. $K_2O$ component migrated from gneisses toward amphibolites, and the opposite movement of $CaO$ component occurred. The following chemical reactions are considered to be representative ones:

1. $2KAlSi_3O_8 + CaO \rightarrow CaAl_2Si_2O_6 + 4SiO_2 + K_2O$
2. $2KAlSi_3O_8 + Al_2SiO_5 + 2CaO \rightarrow 2CaAl_2Si_3O_8 + 3SiO_2 + K_2O$
3. $3Mg_2Al_5Si_3O_{10} + 6CaO + 2KAlSi_3O_8 + 2H_2O \rightarrow 6CaAl_2Si_3O_8 + 3SiO_2 + K_2Mg_3Al_2Si_6O_{20}(OH)_4$

2. Decomposition of amphibole and formation of biotite in the biotite zone

Rocks of the biotite zone contain relict anorthite, suggesting that it was originally amphibolite. Amount of amphibole in the amphibole zone adjacent to the biotite zone is poor compared with central part of the amphibole zone. $CaO$ content is poorer and $K_2O$ content is richer in the biotite zone than in the amphibole zone. Thus, decomposition of amphibole and formation of biotite have taken place in the amphibole zone near the biotite zone. $K_2O$ component of biotite may be derived from the orthoclase zone according to the reactions (1) and (2). $FeO$ and $MgO$ components of biotite have originated from amphibole and pyrrhotite which were decomposing in the amphibole zone. $Al_2O_3$ in biotite may be derived from decomposing anorthite.

3. Compositional change of plagioclase

Plagioclase changes continuously from $An_{23}$ to $An_{40}$ within the orthoclase and the andesine zones. If sodium is fixed component, chemical change in plagioclase is due to formation of calcic plagioclase by the reactions (1), (2) and (3). New calcic plagioclase may have reacted with preexisting sodic one, and plagioclase become calcic at the end of decomposition of K-feldspar. Since mode of K-feldspar ($Ab_{25}$) and plagioclase ($An_{25}$) in gneiss are 12 and 25 wt % respectively, plagioclase could become about $An_{40}$ after the complete decomposition of K-feldspar. Presence of sodic plagioclase less than $An_{40}$ in the andesine zone may be an evidence for mobility of $Na_2O$ component across the reaction zone. Calcic plagioclase more than $An_{40}$ in the andesine zone may be explained by decomposition of albite component and migration of $Na_2O$ toward the amphibole zone: $2NaAlSi_3O_8 + CaO \rightarrow CaAl_2Si_2O_6 + 4SiO_2 + Na_2O$ (4). Calcic plagioclases in the biotite and the amphibole zones changed to sodic one by introduction of sodium from gneiss and outflow of calcium toward gneiss.

Discussion

Metamorphic minerals should have the same chemical composition within each crystal and between given mineral species under condition of complete chemical equilibrium. Incompatible minerals can not occur in an exposure if chemical equilibrium is attained within a large volume of rocks. In the investigated gneissic terrain zonal structure of plagioclase and garnet are common, and minerals have various chemistries among different rocks in a small exposure, thus it proves that widespread chemical equilibrium was not attained at all. The extent of equilibrium may be determined if the boundaries of different kinds of rocks are examined. It is, therefore, important to investigate minerals at the boundaries of two different rocks.

If anorthite-bearing rock is in direct contact with albite-bearing rock, a chemical reaction accompanied with migration of materials may take place because of steep chemical potential gradient of anorthite or albite components across the rock boundary. Con-
Chemical reactions at the boundary

Continuous chemical change of plagioclase takes place until the mineral becomes homogeneous in composition everywhere. Actually, there is a sequence of zones at the boundary between gneisses and amphibolites examined here, and plagioclases show gradational change of chemical composition across the reaction zone. Evidences for chemical reaction at the rock boundary are continuous chemical changes in biotite, garnet and plagioclase as illustrated in Fig. 3 and systematic change of mineral assemblages as shown in Fig. 2. These regular changes are assignable to systematic change of chemical potentials of diffusing components.

It is pointed out that $K_2O$, $CaO$, $MgO$, $H_2O$ and $Na_2O$ components have transferred across the reaction zone. These components must have diffused mainly through grain boundaries, because diffusion in crystals was very slow process. This is supported by common occurrence of zoned plagioclase and garnet. At grain boundaries activity of $H_2O$ component may be high, and this is preferable to diffusion of chemical species along the grain boundaries. Diffusion of Ca and Na through plagioclase crystal, however, took place in a few crystals which showed continuous change of anorthite content and amount of quartz inclusion within a single grain. The following reaction must have taken place in the plagioclase crystal:

$$2 \text{NaAlSi}_3\text{O}_8 + \text{CaO} \rightarrow \text{CaAl}_2\text{Si}_2\text{O}_8 + 4 \text{SiO}_2 + \text{Na}_2\text{O}.$$  

The rate of formation of the reaction zone may be determined by the rate of diffusion of components across the reaction zone or by the rate of chemical reactions at the zone boundaries. Because diffusion is a rate-controlling process of solid state reactions between oxide crystals, the rate of formation of the reaction zone may be also controlled by diffusion of chemical species through the growing reaction zone. The reaction zone in the experimental capsules made by Vidale (1969) suggests that diffusion is rate-limiting step in the process.

Many reaction zones in the Ryoke gneisses were examined in detail to obtain informations on the rate process. The most striking feature obtained is that biotite, plagioclase and garnet show similar degree of chemical gradients. For example, biotite in some reaction zone shows small change of $Fe/Mg$ ratio across the reaction zone, and then coexisting plagioclase and garnet also show small gradients of An % and $Fe/Mg$ ratio, respectively. Similar tendency is also observed within a reaction zone. In Fig. 3 gradient of An content of plagioclase is high where gradient of $Fe/Mg$ ratio of bitoite is steep. These facts indicate that chemical species did move intimately associated with each other. Therefore, it is very difficult to find out the diffusion of special component which was the rate-limiting process.

Another striking feature is that there are often very thin reaction zones where the width of the biotite zone is less than 5 mm, although chemical potential gradient of anorthite component is very steep across the reaction zone. These thin reaction zones occur within K-feldspar-poor gneisses. This fact suggests that steep chemical potential gradient was not a sufficient condition for chemical reaction or diffusion across the reaction zone. Small chemical potential gradient of anorthite component in thick reaction zone could be formed with the aid of other mobile components. It seems highly probable that diffusion of $CaO$ component was not the rate-limiting process.

Since large amount of $K_2O$ must have transferred from gneiss to amphibolite for the formation of biotite in the biotite zone, numerous K-feldspar in gneiss had decomposed to produce enough amount of $K_2O$ component. If diffusion along grain boundaries was rapid enough, K-feldspar remote from amphibolite was available for producing $K_2O$ component, and thick biotite zone could be developed in spite of small amount of K-feldspar in gneiss. We can not expect well-developed reaction zone at the boundary between K-feldspar-poor gneiss and amphibolite under condition of slow diffusion of $K_2O$. The occurrence of thin biotite zone appears to indicate that the diffusion rate of $K_2O$ component was very slow and the rate-limiting process.
The reactions at rock boundaries have been described by many investigators. Most common reaction zones are those formed between ultramafic or calcareous rocks and acidic rocks. Matthews (1967) showed systematic variations in mineral assemblages and FeO/MgO ratios of minerals across the reaction zone formed between ultrabasic and acidic rocks. Appreciable amount of SiO$_2$, MgO, K$_2$O, Al$_2$O$_3$ and CO$_2$ were transferred across the reaction zone of about 0.5 m thick Ramberg (1956) described a reaction zone between dunite and quartzofeldspathic pegmatite. Hypersthenes zone and diopside-hornblende zone are formed between the boundary of the two rocks. SiO$_2$, Al$_2$O$_3$ and CaO appear to have migrated from pegmatite to dunite. Though SiO$_2$ and Al$_2$O$_3$ are practically immovable in the reaction zone formed between quartzofeldspathic vein and amphibolite (Ono, 1970), they are main mobile components in other reaction zones. Kinds of moving materials depend on chemical reactions taking place at the zone boundaries of the reaction zone.

Conclusion

Mineral assemblages change systematically across the reaction zone formed at the boundary of gneiss and amphibolite. An content of plagioclase and Mg/Fe ratio of biotite and garnet increase from gneiss toward amphibolite. Or content of plagioclase and Al$_2$O$_3$ content of biotite show the reverse changes. Main components diffusing through grain boundaries are K$_2$O, CaO, MgO, H$_2$O and Na$_2$O. The width of biotite zone is controlled by both of the amount of K-feldspar in gneiss and the diffusion rate of K$_2$O across the reaction zone.

References


——, 1971: Chemical reaction between amphibolite and gneiss, Takato district, Ryoke region. Ibid., 66, 147–155.


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関家変成帯塩化地方にみられる角閃岩と片麻岩の化学反応について

小野晃

(要旨)

片麻岩と角閃岩との境界部にはアンデシン帯と黒雲母带が生成しており、構成鉱物の化学組成が反応帯の内部で規則的に変化していることが知られている。この研究では片麻岩の境界付近について黒雲母・斜長石・ぎくろ石を電子線マイクロアナライザーによって分析し、化学組成の規則的変化を十分に確認した。斜長石のZoningとぎくろ石の結晶表面でのMnOとMgOのばらつきが認められると、斜長石は角閃岩へ向かってCaOが増大し、K$_2$O、Na$_2$Oが減少している。ぎくろ石は角閃岩へ向かってCaOとMgOが増大しており、FeOが減少している。このような規則性は局部的な化学平衡が近似的に成立していることを示している。反応帯形成時には黒雲母片麻岩に存在することが重要であり、この鉱物がないと斜長石の組成変化が大きくとも反応帯ができていない。化学ポテンシャルの大きな条件は多量の物質移動の必要条件であるが十分条件ではない。