ALTERATION MECHANISM OF BIOTITE IN GRANITIC ROCK CAUSED BY CRUSTOSE LICHEN "PORPIDIA"

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ABSTRACT

The alteration mechanism of biotite in granitic rock caused by crustose lichen "Porpidia" was investigated by means of X-ray diffraction, optical microscope, EPMA and electron microscope with EDX.

Vertical thin sections of lichen-rock interface were observed by a scanning electron microscope to make clear the physico-chemical phenomenon caused by lichens. The contact surfaces between lichen and a biotite were physically observed with this section, and were further investigated chemically by EPMA. As a result, physical destruction of biotite grains by lichen thallus was observed, and the formation of iron oxide or hydroxide was recognized.

The chemical compositions of some parts of the biotite crystal were analyzed by EPMA, such as altered parts close to lichen thallus, altered parts far from lichen thallus and fresh parts. Consequently, comparison of the different parts revealed increase in SiO₂ content on one hand and decrease in FeO and K₂O on the other hand from the fresh biotite crystal throughout the lichenic alteration. Due to the lower solubility of SiO₂ and higher solubility of Fe₂O₃ and Al₂O₃ from minerals under strong acid condition, which is formed by lichen thallus, it seems that Si⁴⁺ content in tetrahedral site is increased by cation exchange. Also due to the low pH and activity of high molecular weight polymers, the possible weakening of the iron–oxygen bonds will lead to the dissolution of Fe³⁺ ions. Consequently, it is likely that, due to the increasing negative charge of biotite layers with dissolution of Fe³⁺, dissolution of K⁺ will be disturbed. Vermiculite-like minerals in biotite formed with the action or effect of lichen were identified by quantitative chemical analysis as wt% of oxides and calculation of structural formulae on the basis of 22 oxygen atoms per unit cell.

Key words: Biological weathering, Lichen-rock interaction, Granite, Biotite

INTRODUCTION

The growth of lichens can be seen on exposed surfaces of various rocks. The study of lichen-rock interface, especially crustose lichen-rock interface, is important to understand the mechanism or process of biological rock weathering, because this interface is one of the few examples on which direct observation of boundary between organic bodies and rock minerals can be carried out.

It is generally difficult to distinguish biological weathering from geological weathering. Because in many cases, effect of biological weathering is much smaller than geological weathering. Therefore, effective study of biological weathering should deal only with the effect of one kind of microbes or higher plants. Crustose lichens usually grow and adhere to the substratum while crustose lichen-rock interface do not suffer significant effects from other organic matter (i.e. fungi or higher plant). However, it is necessary to consider the effect of bacteria, which is small enough to creep in lichen body. In addition, it is easy to observe the contact surfaces between lichen bodies and rock-forming minerals by making vertical thin section for optical microscopic examination. For those reasons mentioned above, this study deals with phenomenon or alteration process occurring at crustose lichen-rock interface.

Lichens are symbiotic fungi and algae, and their forms are similar to moss but these two biological groups are clearly different in terms of taxonomy. In the symbiotic system structure of lichen body, fungi gives water and living space to algae on one hand, while algae gives sugar produced by photosynthesis, on the other hand. These two things live and keep close relationships to each other. It has been recognized that the fungi component of the lichen body produce lichenic acid.
Biological effect of lichenic acid has been studied up to the present, e.g., Ascaso and Wierzchos (1994), Barker and Banfield (1996), Cochran and Berner (1996), Bjelland et al. (2002) and Souza-Egipsy et al. (2002).

Consequently, the question is “do lichens accelerate weathering process or, to the contrary, protect rock surfaces from weathering?” According to Lee and Parsons (1999), lichens stimulate rock weathering through biochemical and biophysical processes, and on the other hand, protect from frost effect. According to Bjelland and Thorseth (2002), lichens certainly accelerate weathering process, and the acceleration rate is different among species. However, under serious reducing environment, lichens relatively protect rock surface from weathering. According to Mottershead and Lucas (2002), in the case of soluble rocks, for example gypsum, lichens protect their surface from weathering. In summary, lichens accelerate weathering microscopically, but there is a possibility of macroscopic protection. However, it must be mentioned that acceleration or protection will depend on the kind of substratum and prevailing environmental condition.

In addition, another question will be “how do lichens accelerate weathering?” Banfield et al. (1999) summarized the role of lichens and categorize them into two main activities, namely physical and chemical activity. The physical activity is that of fungal hyphae, which penetrate into the rock along cracks or grain boundary, and peel off mineral fragment there-from. The chemical activities can be divided further into three aspects as follows:

1) Acid production of lichens decrease pH of rock surface water, and accelerate reduction of Fe and Al, which are difficult to dissolve in neutral-pH water.

2) Low or intermediate molecular weight organic ligands weaken iron-oxygen bonds, accelerate dissolution of Fe, and form complexes with them.

3) High molecular weight polymers also form complexes with iron in solution, thereby lowering saturation state of the solution.

According to Aghamiri and Schwartzman (2002), by using experimental methods with samples from outcrops of mica-schist, it is clear that lichen compound accelerate dissolution of several cations (K, Mg, Na, Si etc.). In addition, Wierzchos and Ascaso (1996) also reported that lichenic acid production can cause serious or extensive dissolution of interlayer K in biotite of granitic rocks, and can accelerate partial alteration into vermiculite. In either case, it can be seen that both physical destruction by fungal hyphae and also chemical dissolution with lichenic acid play important role in biotic mineral alteration processes.

However, it should be pointed out that this study pays special attention on biotite in lichen-granite interface. According to Banfield et al. (1999), microbes can cause positive interaction with rock-forming mineral to scavenge essential elements. For lichens, Ca and Si content in ground or subsurface water is enough for their growth on one hand, while Fe, K and P content are not enough on the other hand. The major component minerals of granitic rocks are quartz, K-feldspar, plagioclase, and biotite. Among these, the mineral containing the most amounts of Fe, K and P is biotite. Thus, the effect of lichen activity seems to appear clearly in respect of biotite. So that is one of the reasons why this study pays special attention to biotite in the granite sample.

Many studies have been carried out on biological alteration of rock minerals with crustose lichen, as stated above, and several of them pay attention to biotite (Wierzchos and Ascaso, 1996). However, these studies does not consider the poor uniformity of biotite composition appearing among each grains, and closely resemble each composition to same in a rock sample and discuss the tendency of experimental data taking an average. It was found from chemical analysis of biotite in this study that biotite grains in a rock sample have compositions different each other. In the case of discussion about micro-scale difference of chemical composition, for example, biological weathering effect, it is likely that poor uniformity among each grain will have considerable effect on results. In this study, to focus on the effect of poor uniformity as much as possible, the experimental tendency of each grain rather than of each sample were discussed. And putting together these tendencies, the typical tendency that applied to all grains was determined. This methodological approach seems to provide a more detailed study as compared to past research works.

It has been recognized that lichen-rock interaction depends on the kind of lichen species, the type of rock unit, and the prevailing environmental condition around them. Consequently, it is necessary to carry out case studies from various regions. This study represents one of such cases.

Lastly, studies of biophysical and biochemical alteration of rocks with lichens are said to be interesting not only from mineralogical and biological viewpoint but also from archaeological viewpoint (Lamas et al., 1995; Arino et al., 1995; Romao and Rattazzi, 1996; Delalieux et al., 2001; Ascaso et al., 2002). For the preservation of cultural assets and stone buildings, lichen activities are important factor. Therefore there is the need for further investigations in this study field.

MATERIALS

As a sample of this study, medium-grained granitic rock with crustose lichen “Porpidia” was picked up from local boulder in Hongo-cho, Hiroshima Prefecture (Fig. 1). The reason why the boulder was selected for this study is that boulder sample appeared to be unaltered as compared with outcrop sample. It is necessary to pick up fresh sample suffering less chemical weathering effect, because of purpose of observing biological effect. The
locality was a streamside, and showed a high percentage of humidity with partial, but bright shade. From the results of XRD analysis for the sample, it was found that the major component minerals were quartz > K-feldspar > plagioclase > biotite. Although no secondary mineral was detected from inner fresh part of the sample, some kaolin minerals and a few vermiculite and gibbsite were detected from the surface part.

For comparison with granite sample, rhyolite sample was also picked up from the outcrop in Mukaibara-cho, Hiroshima Prefecture (Fig. 2). The rhyolite sample was also covered with crustose lichen "Porpidia" like the granite sample. In this case, humidity is high, and characterized by full shade all the daylong. Besides lichens, mosses were also observed on the same surface of outcrop. From the results of XRD analysis for the rhyolite sample, it was found that major component minerals were quartz > plagioclase. Added to this, as secondary minerals, kaolin mineral > vermiculite > micaceous mineral were also detected.

"Porpidia" group is classified as Ascolichens Lecanorales Porpidiaceae on taxonomy. It has been recognized that the "Porpidia" group grow adhering to rocks or other substratum, form gray to light-gray patterns on the rock surface, and produce a dish-like apothecium. The apothecium of "Porpidia" group shows characteristic features, namely, there is no handle, and the edge rise up a little forming black bordering. Also it should be mentioned that the "Porpidia" group occur all over the place, from Hokkaido to Kyushu area in Japan, and are distributed widely throughout the northern hemisphere. From the features of apothecium, the kind of genus can be determined easily with the naked eye, but the determination of species is very difficult. Therefore, the classification of lichen species can be regarded as an immature study area in taxonomy.

Crustose lichen "Porpidia" covering the granite surface can be divided into two color types, namely, beige type and bluish gray type (Fig. 1). In this paper, only the beige type is used as the object of observations.

Lastly, the time scale of lichenic alteration seems to be several tens of years, because the ages of gravestones, which were covered by lichens as sufficiently as in the granite sample used in this study, were all 60 to 70 years. Lichens also seem to need several tens of years to form sufficient colony on rock surfaces.
EXPERIMENTAL METHODS

Drawing of sectional plans
At the beginning, to evaluate the scale of alteration by lichens in rock weathering, sectional plans of samples were drawn up. If the scale is large, depressions ought to be found on the surface of samples macroscopically. On the contrary, it is possible that lichens protect rock surface from chemical and physical weathering by coating the surface. If that is the case, then part of the area under the lichen thallus ought to rise up convexly.

In this study, granite sample was cut at perpendicular plane against sample surface as shown in Fig. 3. The sectional plans were named a-a’, b-b’, and c-c’.

Optical and electron microscopic observations
Subsequently, to understand the phenomenon occurring at lichen-rock interface, vertical thin sections of the interface were observed by an optical and electron microscope.

After the detailed observation using optical microscopy, the thin sections were stained by lead-citrate (Reynolds, 1963), and examined with electron microscopy equipped with a Back Scattered Electron (BSE) detector.

Chemical analysis by EPMA
Chemical analysis was carried out by EPMA (15.0 kV) to compare chemical compositions at three different positions from rim to center of a biotite grain as mentioned below.

In only one grain of biotite, for example ‘grain01’ (Fig. 4), three kinds of biotite compositions were analyzed, namely; composition of altered zone near the lichen thallus (A1~A6), composition of altered zone without lichen thallus (B1~B5), and composition of comparatively unaltered zone (C1~C6). Mean value of these analytical data were calculated and compared with one another in order to find out characteristic features of the biotite alteration with lichen thallus. Similarly, chemical analyses were carried out on 16 grains in all so as to extend the tendency of ‘grain01’ to general tendency.

In the BSE images, an altered zone means darker area while an unaltered zone means brighter area. Applying each analytical data to chemical formulae of biotite, it was confirmed that the brighter area, namely unaltered area are more closely similar to an ideal composition of biotite.

The “zone near the lichen thallus” means <50 μm away from lichen-mineral interface, while the “zone outside lichen thallus” means >400 μm away. However, even if it was >400 μm away from lichen body some area near the cracks which fungal hyphae seem to be able to creep in were exempted.

RESULTS

Sectional plans
Contrary to the hypothesis outlined earlier, the results from the sectional plans indicate no tendency as shown in Fig. 3. Rather, from the study sample, the gentle slope of sectional plans confirms the weak action of lichens.

Optical and electron microscopic observations
Physical action of lichens
The optical and BSE micrographs of lichen-biotite obtained from transverse sections across the lichen-granite interface show biophysical weathering of granitic biotite. Figures 5 and 6 reveal or show the lichen-biotite

FIG. 4. The photograph showing back scattering electron image of biotite obtained by EPMA with chemical analyzed points. A: altered points closed to lichen thallus, B: altered points and C: fresh points.

FIG. 5. Optical micrograph of lichen-biotite interface.
interface. Hyphae penetration and successive detachment, separation and exfoliation of granitic biotite plates were observed in lichen-biotite contact zones. In addition, some biotite particles were curved, bent, and surrounded by fungal hyphae. Also, biotite grains with no lichen cover on the surface or fissures were also observed with BSE (Fig. 7). Such biotite grains lacked characteristic bioalteration features mentioned above.

Reddish brown material

From the thin-sections, reddish brown aggregates were observed not only in the lichen-biotite contact zone but also in the lichen-quartz/feldspars contact zone (Fig. 8). Some reddish brown material concentrated in the minerals and others concentrated in lichen thallus. However, mineral grains with no lichen cover on the surface lacked concentration of reddish brown material,

Fig. 6. BSE image of lichen-biotite interface obtained by EPMA.

Fig. 7. BSE image of biotite grains without lichen on the rock surface.

Fig. 8. Optical micrograph of reddish brown material observed at the lichen-biotite interface.

Fig. 9. Optical micrograph and EDX images of the interface of thallus and feldspar.
except for insignificant concentration in biotite grains.

Examination by XRD, using Cu-Kα radiation generated at 100 kV, 40 mA at a scan speed of 2 theta = 1 degree min⁻¹ with a 0.5 degrees divergence slit, a 0.15 mm receiving slit and a 0.5 degrees scattering slit, of the interface material did not reveal the presence of the iron oxides.

In Fig. 9, X-ray images obtained by SEM-EDX demonstrate the spatial distribution of Fe on lichen-mineral interface. The Fe spatial distribution maps exactly record the forms and shapes of reddish brown material shown on the optical images.

In summary, the result of optical observations, X-ray diffraction, and SEM-EDX observations indicate that the reddish brown material is a poorly crystallized form of Fe oxide.

Chemical features within bioweathered biotite

Results of quantitative analysis for ‘grain01’ (Fig. 4) as wt% of oxides are displayed in Table 1 and Fig. 10. The same analyses for further 15 grains (grain02–grain16) were performed to obtain the general tendency. It was found from the result that changes in mass of Si⁴⁺, Fe⁶⁺ and K⁺ contents have significant tendencies (Figures 11 and 12).

The percentage loss of SiO₂ changes during abiotic weathering (C to B) was 1.7~8.7%. On the other hand, the percentage during bioweathering (C to A) was 0.6~12.3%.

The percentage of FeO (total Fe as FeO) changes during abiotic weathering (C to B) had no tendency (5.6% loss~6.2% gain) while the percentage loss during bioweathering (C to A) was 0.8~17.8%.

In addition, the percentage loss of K₂O changes during abiotic weathering (C to B) was 62.3~66.6% while the percentage loss during bioweathering (C to A) was, comparatively small with values of 18.0~52.8%.

DISCUSSION

Iron oxides

According to Banfield et al. (1999), low and inter-

<table>
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<tr>
<th>Point number</th>
<th>SiO₂</th>
<th>TiO₂</th>
<th>Al₂O₃</th>
<th>FeO*</th>
<th>MnO</th>
<th>Cr₂O₃</th>
<th>MgO</th>
<th>CaO</th>
<th>Na₂O</th>
<th>K₂O</th>
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<td>2.03</td>
<td>11.84</td>
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<td>0.00</td>
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<td>0.22</td>
<td>0.10</td>
<td>5.36</td>
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<td>26.16</td>
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<td>0.04</td>
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<td>0.11</td>
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<td>1.98</td>
<td>14.33</td>
<td>24.66</td>
<td>1.80</td>
<td>0.02</td>
<td>2.31</td>
<td>0.15</td>
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<td>26.17</td>
<td>1.99</td>
<td>0.00</td>
<td>2.17</td>
<td>0.10</td>
<td>0.04</td>
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<td>0.01</td>
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Point numbers are same to them showing in Figure 3.1. FeO is indicated as total Fe.

Fig. 10. Comparison of SiO₂, FeO and K₂O contents among three different analytical points in a biotite particle of grain01. A: average value at altered part closed to lichen thallus, B: average value at altered part and C: average value at fresh part. FeO is indicated as total Fe.
mediate molecular weight organic ligands and high molecular weight polymers can complex with ions on the mineral surface and can weaken metal-oxygen bonds. However, it seems likely that the organic acids play a key role in the enrichment of poorly-ordered phases of iron at the rock-lichen interface.

For comparative study, rhyolite thin sections with lichen cover on the surface were also observed with optical microscope. The results show that these zones lacked concentration of iron oxides.

**Chemical features within bioweathered biotite**

**Gain of Si\(^{4+}\)**

It has been recognized that transformation of biotite to vermiculite causes loss of Si\(^{4+}\) contents (Gilkes et al., 1979; Pozzoli et al., 1992) in chemically. The results of this study for the geochemical mass balance of the altered biotite (Table 1) during abiotic weathering (C to B) were in good agreement with data obtained by Gilkes et al. (1979) and Pozzoli et al. (1992). However, the gain of Si\(^{4+}\) content was observed during bioweathering (C to A), while it is necessary to consider relative change of Si\(^{4+}\) content with loss of Fe\(^{tot}\). It is likely that submicron size Si-rich secondary minerals, which could not be observed by EPMA resolution, were produced in bioweathered biotite zone. The result obtained was contrary to several studies (Aghamiri and Schwartzman, 2002; Banfield et al., 1999).

**Loss of Fe\(^{tot}\)**

It has been recognized that natural, abiotic weathering (pH ~ 7) of biotite to vermiculite does not cause significant gain or loss of Fe\(^{tot}\) content (total Fe as FeO) (Gilkes et al., 1979; Pozzoli et al., 1992). In addition, it has been recognized that lichenic bioalteration cause significant loss of Fe\(^{tot}\) throughout leading to different microsites in the rock-lichen interfaces which have their own conditions of pH, humidity and redox potential (Aghamiri and Schwartzman, 2002; Wierzchos and Ascaso, 1996; Banfield et al., 1999). The results of this study for the geochemical mass balance of the altered biotite (Table 1) were in good agreement with the above observation.

According to Banfield et al. (1999), pH value of 3–4, which could be possibly due to the dissolution of Fe\(^{3+}\) can be detected in the proximity of the bacterial cells within cleavages in biotite when the bulk solution pH was 7.0.

In addition, as mentioned earlier, high molecular weight polymers can weaken metal-oxygen bonds and can complex with octahedrally coordinated Fe\(^{3+}\) as well as with other major elements in minerals, e.g., Al and Si, on the mineral surface.

Therefore, the acid production with lowering pH and the high molecular weight polymers play a key role in
the loss of Fe$^{3+}$ at bioweathered biotite zone.

Loss of K$^+$

It has been recognized that, in the case of natural, abiotic weathering, lowering of negative charge of biotite layer with oxidation of Fe$^{2+}$ in octahedral site to Fe$^{3+}$ cause the release of the interlayer K and the transformation of biotite to hydrobiotite, further to vermiculite (Gilkes et al., 1979; Pozzoli et al., 1992). The results of geochemical mass balance of the altered biotite in this study (Table 1) were in good agreement with data obtained by Gilkes et al. (1979) and Pozzoli et al. (1992).

However, the release of the interlayer K was suppressed in the bioweathered biotite zone. Increasing of negative charge of biotite layer with significant dissolution of Fe$^{3+}$ which is formed by lichen thallus play a key role in the suppression of K release in the bioweathered biotite zone.

The result obtained from this study was contrary to those of Aghamiri and Schwartzman (2002), Wierzchos and Ascaso (1996) and Banfield et al. (1999). According to Wierzchos and Ascaso (1996), where distinct depletion of interlaminar K in adhesion zones of lichen thalli on granitic biotite sheets was observed. However, in their study, suppression of K release was significantly noted at the edge between biotite grain and hyphae cells in the deep fissure.

Comparison of alteration of biotite

Abiotic alteration of biotite

It has been generally recognized that biotite can be transformed into hydro-biotite, which is an intermediate mineral between biotite and vermiculite, and further into vermiculite, with the release of interlayer K throughout the abiotic weathering process. Through the transformation of biotite to vermiculite, a few exchangeable cation, e.g., Mg$^{2+}$ and Ca$^{2+}$ with some H$_{2}$O, enter into the interlayer site.

In this study, gain of Ca$^{2+}$ in the abiotic weathered biotite zone was observed for several grains (Table 1). It seems, therefore, that a few Ca$^{2+}$ enter into the interlayer site through the abiotic alteration process.

Lichenic alteration of biotite

From the results obtained by quantitative chemical analysis as wt% of oxides and calculation of structural formulae on the basis of 22 oxygen atoms per unit cell, it seems that vermiculite-like minerals are formed both in the case of lichenic alteration as well as in the case of abiotic weathering. However, compared with the case of abiotic weathering, loss of Fe, suppression of K release and gain of Si were detected. It is likely that increasing of negative charge of biotite layer with significant dissolution of Fe$^{3+}$, which is formed by lichen compound cause the suppression of K release and cation exchange in tetrahedral site. Depending on lower solubility of SiO$_{2}$ and higher solubility of Al$_{2}$O$_{3}$ of minerals under strong acid condition which is formed by lichen thallus, it seems that Si$^{4+}$ content in tetrahedral will be increased by cation exchange with decreasing of tetrahedral Al$^{3+}$. Slight, but not to obvious decrease in Al$^{3+}$ content can be seen in the results presented in Table 1.

According to Banfield et al. (1999), at neutral pH, secondary Fe-hydroxides and vermiculite or smectite-like silicate clays were usually formed through the biotite alteration. However, as acidity increases with lichen activity, formation of different secondary minerals, such as the alminosilicates kaolinite and halloysite, will probably occur. In this study, vermiculite-like minerals, rather than kaolinite or halloysite, were formed in the bioweathered biotite zone. However, it is quite likely that these vermiculite-like minerals will transform to kaolinite or halloysite-like minerals throughout further alteration with lichen activities.

CONCLUSIONS

From all of the above analyses, interpretation and evaluation as presented in the preceding sections, the overall summary and conclusions from this study of effects lichens on the bioweathering of biotite grains in granitic rocks can be outlined as follows:

1) Neither dramatic or effective protection nor acceleration of the weathering effects was detected with naked eye.

2) Physical destruction of biotite grains and concentration of iron oxide was recognized at the lichen-rock interface zone.

3) Vermiculite-like minerals were identified in bioweathered biotite zone as well as in abiotic weathered zone, however these chemical features were different from one another.

4) In the bioweathered biotite zone, loss of Fe content, suppression of released K and gain of Si content were also detected.

5) The observed loss of Fe content during lichenic weathering is in good agreement with data obtained by other several studies, however, suppression of K release and gain of Si content is contrary to them.

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REFERENCES


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