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Alteration of Plagioclase in Granite during Weathering

By

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with 6 Tables, 11 Text-figures and 2 Plates

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ABSTRACT: Secondary minerals of plagioclase in spheroidally weathered granite of the Seranishi-cho, Hiroshima Prefecture, Japan have been examined by the technique of X-ray diffraction, X-ray fluorescence, DTA, TGA, pH measurements and electron microscopic observation. Spheroidally zoning of A, B and C owing to the difference of appearance at outcrop corresponds to the secondary mineral components, that is, allophane or allophane-like substance is found only in the zone C of core-part of granite and the zone B, metahalloysite and halloysite are predominant in the zones A and B that were intensely weathered parts, and kaolinite exists in the zones A and B, but lacks in the zone C. The crystallinity and grain-size of these kaolin minerals increase gradually from near central core-part toward the outer A zone. The probable altering sequence of plagioclase in the weathered granite of this district has been roughly assumed as follows:

plagioclase → allophane → halloysite → metahalloysite → kaolinite

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I. INTRODUCTION

The alteration products and the process of constituent minerals of granitic rocks during weathering have been studied by many researchers till now. Various kinds of minerals such as vermiculite, kaolinite, gibbsite, chlorite and montmorillonite have been reported as alteration products of biotite, for instance, KASHIWAGI (1963), KATO (1965), YAMASHITA (1968), NAGASAWA et al. (1972), KAKITANI and KONO (1972) etc., while halloysite as the alteration product of plagioclase is the common and predominant mineral reported by WOLFF (1967), SHIMIZU (1972) and others. From the study of weathered granite in Hong Kong, PARHAM (1969) has reported that allophane appears to the first stage of alteration and then changes to halloysite under humid tropical condition. WOLFF (1967) has pointed out that plagioclase in woodstock granite has broken down under weathering condition, formed halloysite directly, and then changed to metahalloysite. SHIMIZU (1972) studied some weathered granitic rocks in Japan and indicated that they

had the same alteration process as that of WOLFF (1967) except for kaolinite as the end product. Gibbsite has also been reported to be detected together with some kaolin minerals as alteration products of plagioclase by some researchers, e.g. KATO (1965), etc. There are many studies up to the present as mentioned above; however, the problem of the alteration products and the process of plagioclase in weathering have not been adequately clarified yet.

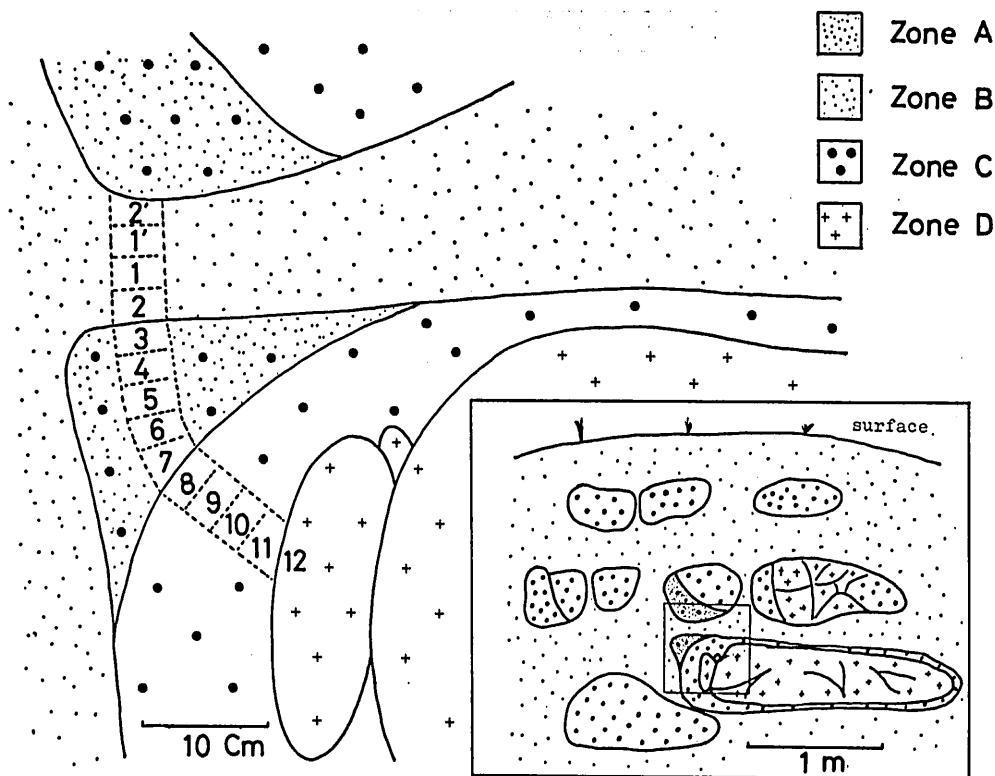


FIG. 1. Diagram showing weathering gradation (zones A, B, C and D) of granite in the Seranishi-cho outcrop in the district and localities of examined specimens.

Among some outcrops of the medium-grained granite of the Seranishi-cho district, Hiroshima Prefecture, we have chosen one outcrop as shown in Fig. 1, and studied on the successive alteration of plagioclase on the weathered zonal parts around the central core-stone. The results of mineralogical study of the alteration products and mineral sequence in the process of weathering will be discussed in this paper. Special attention has been paid on the examination of the existence of allophane as one of the alteration products of plagioclase.

II. GENERAL FEATURE OF OUTCROP AND SAMPLING

The samples of weathered granite for this study have been collected from one outcrop

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of the Seranishi-cho, Hiroshima Prefecture. It is medium-grained granite mainly composed of pinkish potash feldspar, plagioclase, quartz, biotite and small amount of hornblende, and has granophyric texture. This weathered granite has spheroidal form, and there is no geologic evidence of hydrothermal activity. The spheroidally weathered granite can be divided into four zones in the order A, B, C and D from the outside to the central core-stone (Fig. 1). Zone A is stained in brownish color and weathered plagioclase grains are easily crushable by finger. Zone B is slightly stained in brownish color and biotite flakes usually have brownish color and plagioclase grains are crushable by finger. Zone C is slightly stained in light yellowish color, but holds the original texture of granite, and plagioclase grains are not crushable by finger. The granite in zone D is almost fresh.

According to the classification of weathering state by RUXTON and BERRY (1957), the weathered zone of A, B and C may correspond to those of their zone II or III. Besides, these zones A, B, C and D may be correlated with the classes 5, 4, 3 and 2 respectively, which were proposed by OLLIER (1965) and MELTON (1965).

Used samples of weathered granite for this study were collected from 12 points in the outcrop as shown in Fig. 1.

III. PREPARATION OF SAMPLES AND EXPERIMENTAL METHODS

The specimens of weathered plagioclase were carefully picked out from weathered granite under the binocular microscope and were further purified using isodynamic separator, though quartz grains of small amount as very minute inclusions (less than 1μ) could not be put away from the pulverized plagioclase. The pulverized plagioclase specimens that were carefully controlled to be 200 meshes were examined by the methods of X-ray diffraction, X-ray fluorescence, DTA and TGA analysis, pH measurements and electron microscopic observation.

X-ray diffraction powder patterns were obtained using JEOL-JTX-7 diffractometer ($\text{CuK}\alpha$, 30 KV, 15 mA) with the specimens prepared on the slide glass under the atmospheric condition. X-ray fluorescence analysis was performed using a reflection from EDDT crystal in a vacuum system. DTA and TGA experiments were done with Rigakudenki micro-differential thermal analyzer and micro-thermogravimetry analyzer, respectively. The pH-values for some specimens were measured using Hitachi-Horiba D-5 pH meter. Electron microscopic observations were performed with JEOL-7 (maximum 60 KV).

Some kaolin minerals previously examined by some authors were prepared for the reference of the identification, (1) kaolinite from Georgia, U.S.A. (KESLER, 1956), (2) kaolinite from Seto, Aichi Pref., Japan (NOZAWA 1955, TANEMURA 1964), (3) halloysite from Imaichi, Tochigi Pref., Japan (IWAO et al. 1958), (4) allophane (so-called "Kanumatsuchi") from Kanuma, Tochigi Pref., Japan (IWAO et al. 1958).

IV. EXPERIMENTAL RESULTS AND CONSIDERATION

(1) X-ray diffraction

X-ray diffraction patterns for pulverized plagioclase are illustrated in Fig. 2. As seen in Fig. 2, the reflection pattern of specimen 12 is limited only to plagioclase and

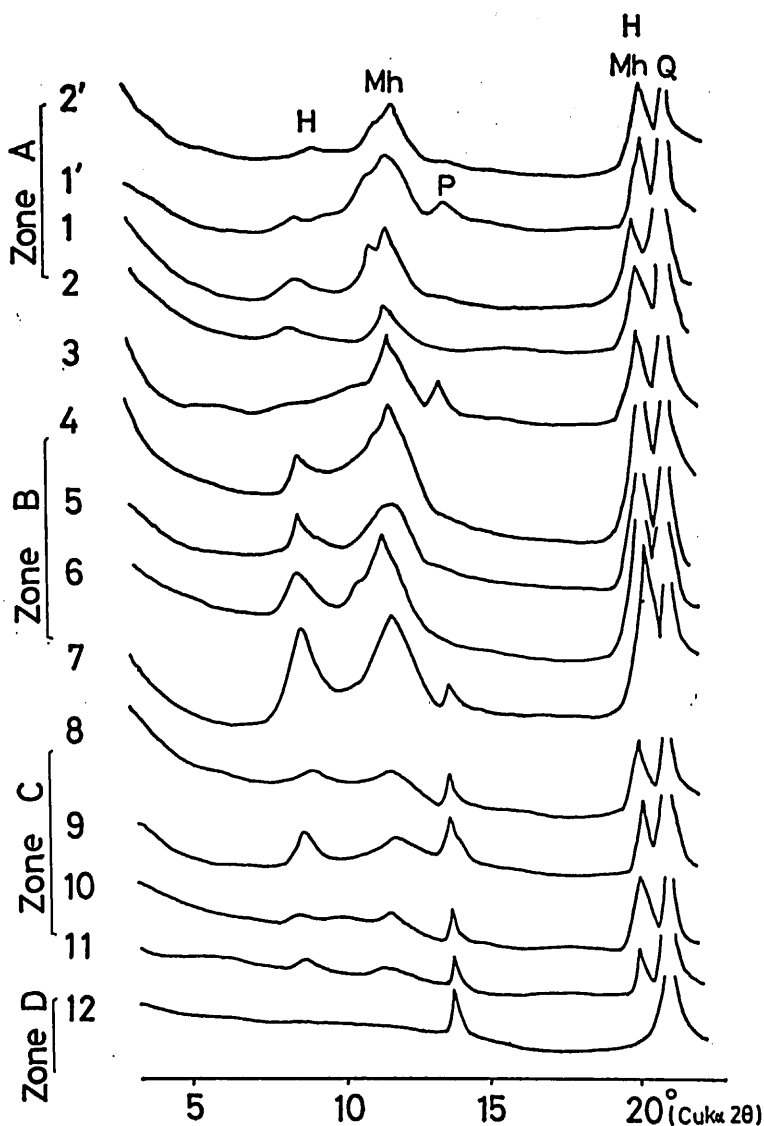


FIG. 2. X-ray powder diffraction patterns of plagioclase and its alteration products in the weathered granite of the Seranishi-cho outcrop.

H: Halloysite Mh: Metahalloysite P: Plagioclase Q: Quartz

quartz. The reflection intensities of specimens decrease from the corestone (specimen 12 in zone D) toward the zone B, and in the zone A the reflections of plagioclase are scarcely observed. But (100) reflection of quartz appears in patterns of all specimens.

The basal reflections (001) at about 10 \AA of halloysite are recognized in the specimens of the zones C and B, while 7 \AA basal reflections (001) of metahalloysite are predominant in the specimens of the zones A and B. Around 10 \AA spacing contracts to 7 \AA with heating

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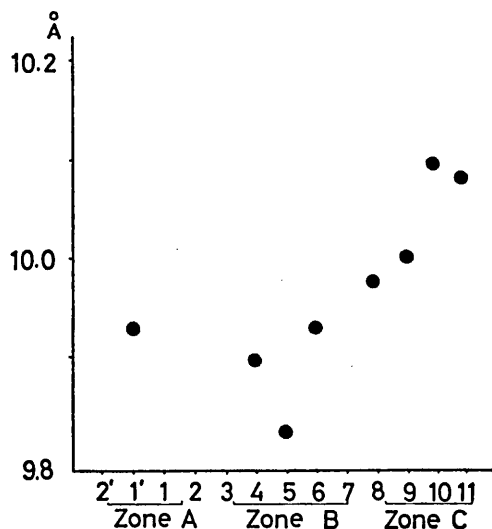


FIG. 3. Diagram showing the relationship between the basal d-spacings (001) of halloysite and sampling numbers.

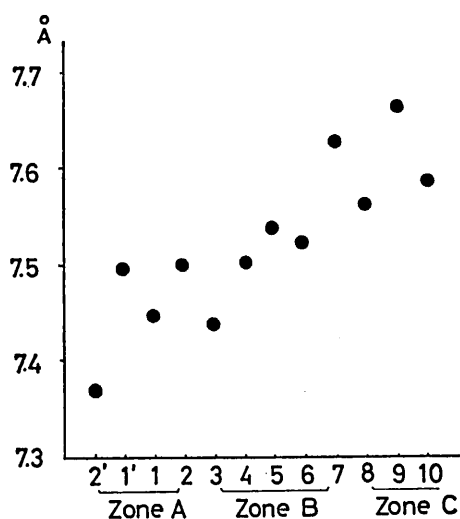


FIG. 4. Diagram showing the relationship between the basal d-spacings (001) of metahalloysite and sampling numbers.

TABLE 1. THE BASAL d-SPACINGS (001) OF HALLOYSITE AND METAHALLOYSITE IN ZONES A, B AND C.

| Specimens | Halloysite (001) | Metahalloysite (001) | |
|--------------|------------------|----------------------|--------|
| Zone A | 2' | 9.93 Å | 7.37 Å |
| | 1' | — | 7.50 |
| | 1 | — | 7.45 |
| | 2 | — | 7.50 |
| Zone B | 3 | — | 7.43 |
| | 4 | 9.90 | 7.50 |
| | 5 | 9.83 | 7.54 |
| | 6 | 9.94 | 7.52 |
| | 7 | — | 7.62 |
| Zone C | 8 | 9.80 | 7.55 |
| | 9 | 10.00 | 7.65 |
| | 10 | 10.12 | 7.59 |
| | 11 | 10.08 | — |
| Kaolinite | — | 7.10 | |
| from Georgia | — | 7.23 | |
| Kaolinite | — | 7.23 | |
| from Seto | — | — | |
| Halloysite | 10.20 | — | |
| from Imaichi | — | — | |

of 1 hour at 110°C, which fact may show the existence of halloysite as pointed out by WARSHOW (1961) and others. According to the definition proposed by DOUILLET and NICOLAS (1969), the mineral that has about 7 Å basal reflection may be called metahalloysite.

The amount of halloysite increases from the zone C toward the zone B and decreases again in the zone A. Metahalloysite is characteristically formed in the zones B and A and the amount decreases in the zone C.

Figs. 3 and 4 show the relationship between the values of basal spacings and sampling numbers due to the order of zoning for halloysite and metahalloysite respectively. Table 1 shows the d(001)-spacing of halloysite and metahalloysite in the zones A, B, C and of kaolinite (Georgia, Seto), halloysite (Imaichi).

As seen in Table 1, the d-spacings (001) of halloysite are within range of between 9.9 Å and 10.1 Å in the zone C, and gradually contract to the value in the zone B (9.8–9.9 Å).

There are analogous relationships for metahalloysite also, that are 7.6–7.5 Å in the zone C and 7.5–7.4 Å in the zone A.

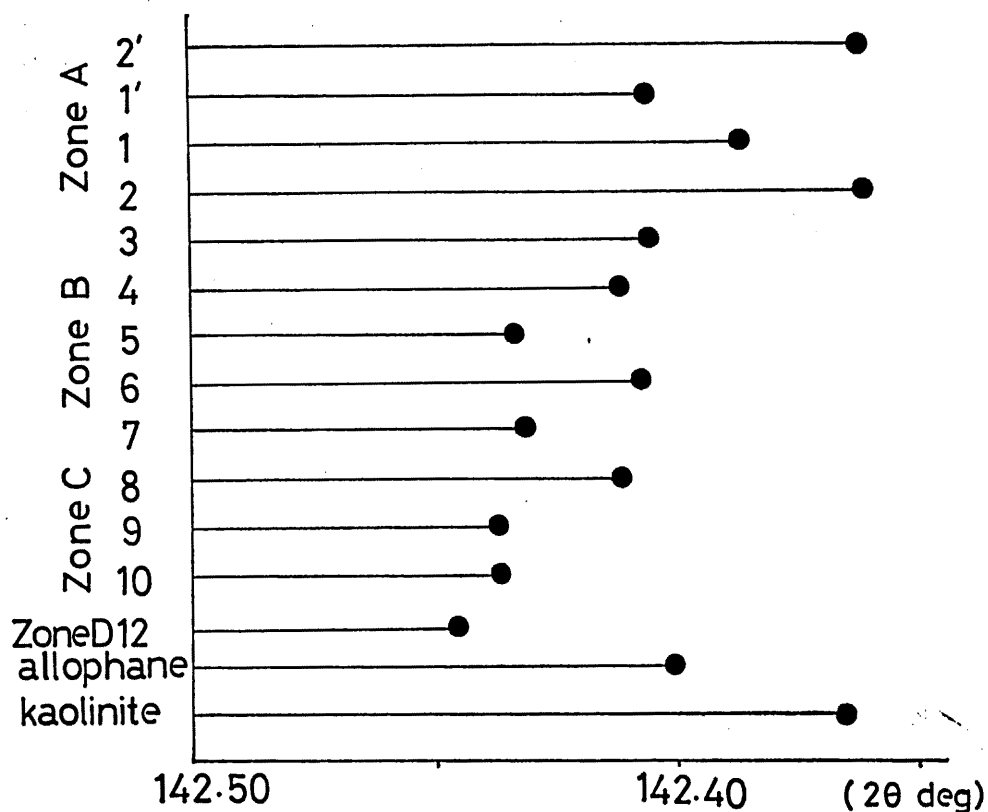


FIG. 5. Diagram showing variation in the wavelength of $Alk\alpha$ between alteration products of plagioclase (in 2θ values of EDDT).

Allophane: Kanumatsuchi, Kaolinite: Georgia kaolin, Zone D 12: Fresh plagioclase.

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TABLE 2. THE WAVELENGTH OF $AlK\alpha$ (2θ) BETWEEN ALTERATION PRODUCTS OF PLAGIOCLASE IN ZONES A, B AND C.

| Specimens | Wavelength (2θ) | |
|---------------|--------------------------|--------|
| Zone A | 2' | 142.36 |
| | 1' | 142.40 |
| | 1 | 143.39 |
| | 2 | 142.36 |
| Zone B | 3 | 142.40 |
| | 4 | 142.41 |
| | 5 | 142.48 |
| | 6 | 142.41 |
| | 7 | 142.43 |
| Zone C | 8 | 142.41 |
| | 9 | 142.44 |
| | 10 | 142.44 |
| pure Al metal | 142.50 | |
| plagioclase | 142.45 | |
| kaolinite | 142.36 | |
| allophane | 142.40 | |

TABLE 3. THE VALUES OF pH IN ZONES A, B AND C.

| Specimens | pH |
|-------------------------------|------|
| Zone A (1'-2' 1 -2) | 5.78 |
| Zone B (3-7) | 6.48 |
| Zone C (8-11) | 6.77 |
| Allophane (Kanumatuchi) | 7.45 |
| Kaolinite (Georgia kaolin) | 5.60 |
| Halloysite (Imaichi clay) | 5.65 |
| Distilled water +NaF | 5.60 |

(2) X-ray fluorescence analysis

In order to confirm the existence of allophane, the coordination number of Al in the alteration products of plagioclase were examined using X-ray fluorescence. The angular positions (wavelength) of $AlK\alpha$ are shown in Table 2 for alteration products of plagioclase, together with aluminium metal, plagioclase, kaolinite and allophane for comparison. The values of wavelength for the alteration products of plagioclase have those between fresh plagioclase and kaolinite.

The values of specimens in the zone C are nearly the same as those of fresh plagioclase, and those in zone A are almost the same as those of kaolinite. The values of the intermediate zone B are nearly equal to those of allophane which has Al 4- and 6-coordination numbers (Table 2 and Fig. 5).

(3) The pH measurements

With the object of further confirmation of allophane in the specimens, the pH measurements were performed. Detection of allophane using phenolphthalein was not successful because of the limited amount, and so the pH measurements were done in the following way. Each specimen of 50 mg was put in 100 cc distilled water and after it was stirred up the initial pH value was measured. Then a few more drops of saturated sodium fluoride solution were added to the dispersed water of the specimens, and after ten minutes the pH value was measured again. The pH measurements of allophane, kaolinite and halloysite also were carried out for comparison. There is almost no change in the pH values of dispersed kaolinite or halloysite with water regardless of addition of NaF.

When NaF was added to allophane dispersed water, initial pH value increased slowly. This phenomenon of the deviation of the pH value may be related to the result of the reaction between $\text{Al}(\text{OH})_3$ in allophane and NaF which forms NaOH. The averages of pH values of specimens in zones A, B and C which were obtained from examination of this method are shown in Table 3.

The pH deviation (ΔpH) of the specimen in zone C, in Fig. 6, has large value relatively, while the value in zone A is small (Fig. 6).

From the result of this examination, it could also be said that allophane or allophane-like substance is recognized to exist in the specimens in the zones B and C.

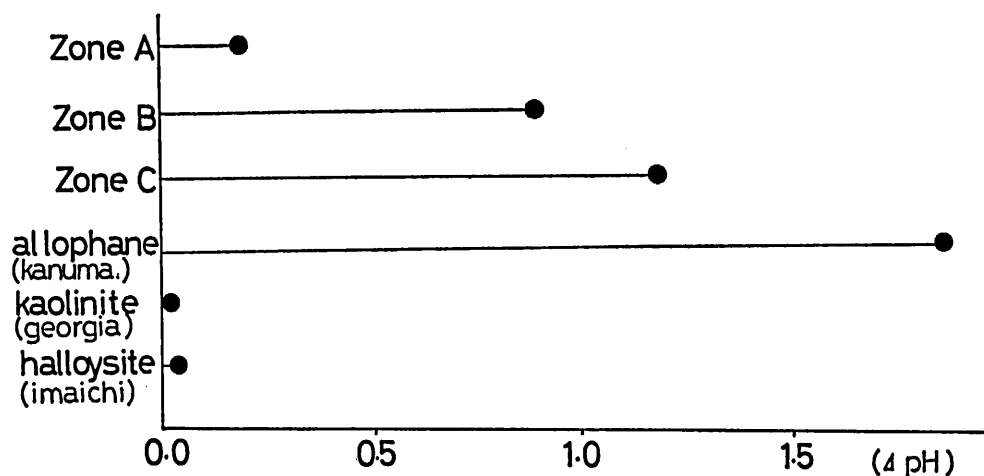


FIG. 6. The variation in the range of pH deviation in zones A, B and C.

(4) DTA experiments

KATO (1971) and SHIMODA (1971) pointed out the relation between crystallinity due to the results of X-ray diffraction and the position of differential endothermic peak at 450–500°C. Fig. 7 shows the thermal analysis curves that were measured under the following condition: the rate of heating is 10°C per minute, chart speed is 4 mm per minute. As seen in Fig. 8 and Table 4, the specimens in the zone A have high maximum temperature (485–490°C), and these temperatures decrease gradually toward the zones B and C. From the results of this experiment, it has been confirmed that the crystallinity of the alteration products in the zone A is higher than those in the zones B and C.

On the other hand, BRAMAO et al. (1952) proposed the method with the slope ratio of the endothermic peak at about 500°C, for the recognition of the crystallinity. The variation in the slope ratios of the specimens in the zones A, B and C is shown in Fig. 9.

The slope ratios for zone A have the small value (1.00–1.14), while that for zone C has large one (1.23–3.14) (Table 5). Such a tendency in the slope ratio may be caused by the difference of the crystallinity for the kaolin minerals as is pointed out by CARTHEW (1955), that is, the specimens of the zone A indicate the better crystallinity than those of the zones B and C.

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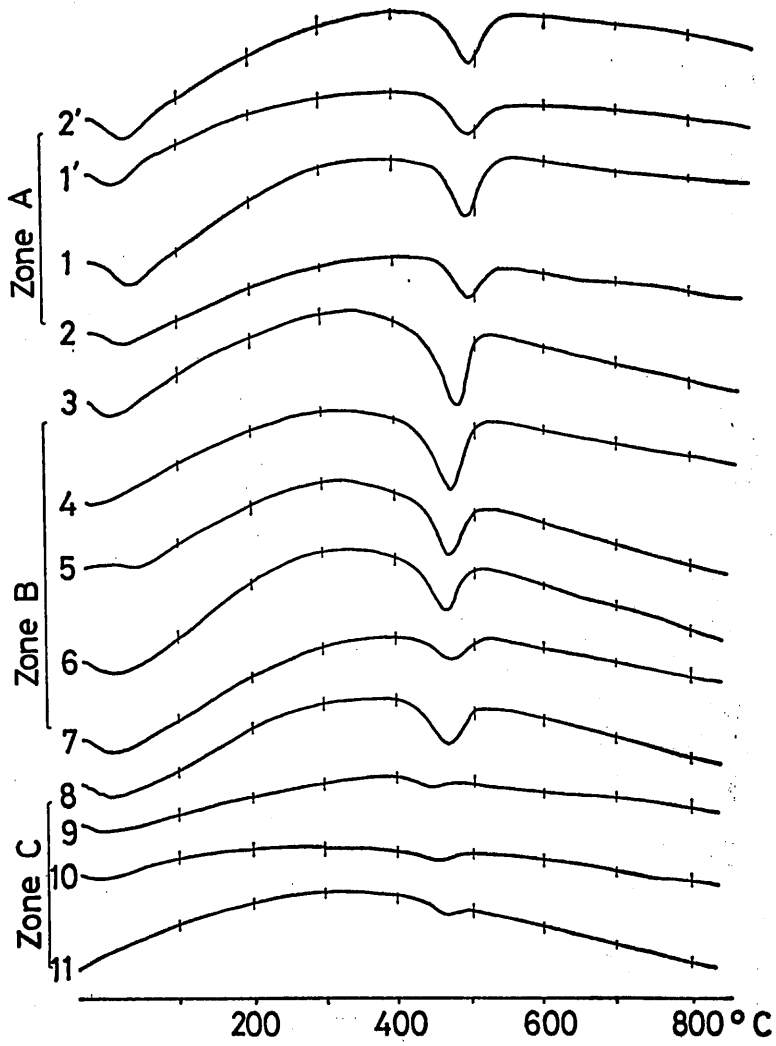


FIG. 7. Differential thermal analysis curves for the specimens in zones A, B and C.

TABLE 4. THE TEMPERATURES OF MAIN ENDOOTHERMIC PEAK IN ZONES A, B AND C.

| Specimens | Temperatures of main endothermic peak | |
|----------------------|---------------------------------------|--------|
| Zone A | 2' | 490 °C |
| | 1' | 490 |
| | 1 | 485 |
| | 2 | 490 |
| Zone B | 3 | 493 |
| | 4 | 480 |
| | 5 | 475 |
| | 6 | 473 |
| | 7 | 475 |
| Zone C | 8 | 473 |
| | 9 | 453 |
| | 10 | 460 |
| | 11 | ? |
| Kaolinite (Georgia) | 500 | |
| Kaolinite (Seto) | 490 | |
| Halloysite (Imaichi) | 480 | |

TABLE 5. THE SLOPE RATIOS OF MAIN ENDOOTHERMIC PEAK IN ZONES A, B AND C.

| Specimens | Slope ratios of main endothermic peak | |
|----------------------|---------------------------------------|------|
| Zone A | 2' | 1.11 |
| | 1' | 1.00 |
| | 1 | 1.05 |
| | 2 | 1.14 |
| Zone B | 3 | 1.39 |
| | 4 | 1.31 |
| | 5 | 1.40 |
| | 6 | 1.59 |
| | 7 | 1.45 |
| Zone C | 8 | 1.23 |
| | 9 | ? |
| | 10 | ? |
| | 11 | 3.14 |
| Kaolinite (Georgia) | 1.06 | |
| Kaolinite (Seto) | 1.15 | |
| Halloysite (Imaichi) | 1.59 | |

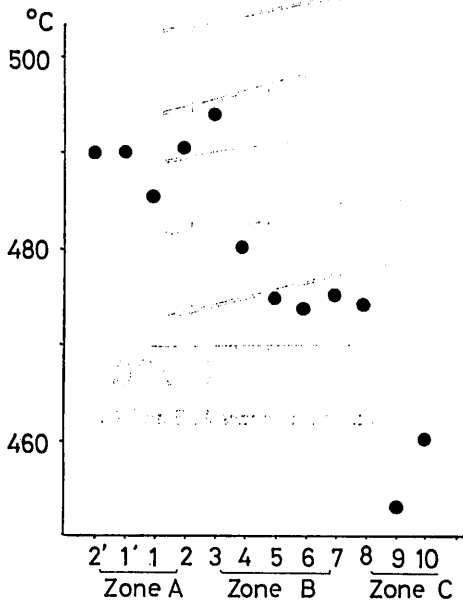


FIG. 8. The variation in the temperatures of the main endothermic peak between kaolin minerals in zones A, B and C.

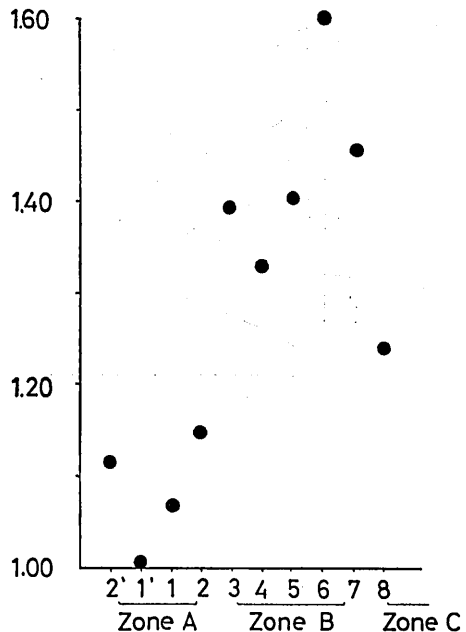


FIG. 9. The values in the slope ratio between the alteration products of plagioclase in zones A, B and C.

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(5) TGA experiments

The activation energy for the dehydration of structural water by heating was calculated, using the method of FREEMAN and CARROL (1957), from the results of thermal gravimetric analysis. The particle-sizes of the examined materials are the same as those used in the DTA experiments.

Arrhenius equation is expressed in the following formulas:

$$k=A \exp (-E/RT) \dots\dots\dots (1)$$

$$kX^n=dX/dt \dots\dots\dots (2)$$

From the formulas (1) and (2), the next formula is introduced:

$$\log (dX/dt)=n-\frac{E}{R}(1/T)/\log X \dots\dots\dots (3)$$

The relation between mole fraction and weight is shown as follows:

$$-dX/dt=-(X_0/W_c) (dW/dt) \dots\dots\dots (4)$$

$$W_r=W_c-W \dots\dots\dots (5)$$

In the combination of (3), (4) and (5), the equation (6) is obtained:

$$-\frac{E}{R}(1/T)/\log W_r=-n+\log (dW/dt)/\log W_r \dots\dots\dots (6)$$

k: velocity constant, A: frequency factor, E: energy of activation, R: gas constant, T: absolute temperature, n: order of reaction, X: mole fraction, X₀: initial mole, W: total weight loss up to (t) time, W_c: weight loss at the completion of reaction, W_r: W_c-W.

The values of the activation energy of the specimens increase with the order zones A, B and C as shown in Table 6 and Fig. 10. According to the hypothesis proposed by TSUZUKI and NAGASAWA (1957), the results may show that the crystal sizes of alteration products of plagioclase become coarse gradually from the zone C to the zone A.

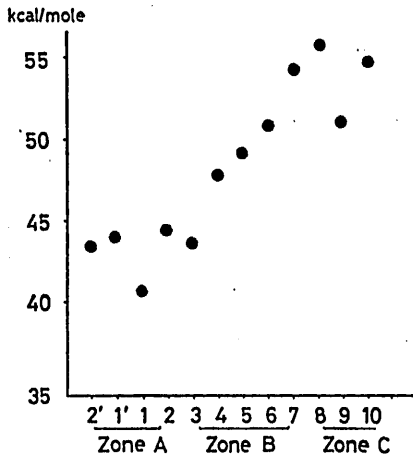


FIG. 10. The variation of the activation energy in zones A, B and C.

TABLE 6. THE ACTIVATION ENERGY BETWEEN ALTERATION PRODUCTS OF PLAGIOCLASE IN ZONES A, B AND C.

| Specimens | The activation energy (kcal/mol) | |
|-----------|----------------------------------|------|
| Zone A | 2' | 43.6 |
| | 1' | 43.7 |
| | 1 | 40.5 |
| | 2 | 44.0 |
| Zone B | 3 | 43.5 |
| | 4 | 47.8 |
| | 5 | 48.5 |
| | 6 | 50.1 |
| | 7 | 54.1 |
| Zone C | 8 | 55.1 |
| | 9 | 50.1 |
| | 10 | 54.6 |
| | 11 | — |

(6) Electron microscopic observation

Some of the electron microphotographs of the specimens are shown in Plate I. As seen in Plate I-a, the spherical particles are observed in very small amount in the specimens of the zone C. Such spherical forms are the one regarded as characteristic for allophane by BEUTELSPACHER and VAN DER MAREL (1968). We can see the tubular forms of halloysite and metahalloysite in Plate I a-f. The average diameter of tubes increases from the zone C (0.04μ) to the zone B (0.12μ) and further to the zone A (0.15μ). This tendency indicates good harmony as compared with the change in crystal-sizes of kaolin minerals in the zones A, B and C, which was assumed from the calculated activation energy. Platy flakes of kaolinite (Plate I-f) are found in the zones A and B, but never observed in the zone C.

V. CONCLUSION AND REMARKS

An alteration of plagioclase in granite during weathering has been examined on some specimens collected from one outcrop of the Seranishi-cho, Hiroshima Prefecture, Japan. The spheroidally weathered granite of this outcrop has been divided into four zones A, B, C and D.

The alteration products of plagioclase in zones A, B and C consist of kaolin minerals and small amount of allophane or allophane-like substance.

The crystallinity and crystal-sizes of kaolin minerals increase from central zone C to zone A, although very small amount of pseudohexagonal flakes of kaolinite were observed among the halloysite and/or metahalloysite of the specimens in zones A and B.

The existence of allophane or allophane-like substance were also recognized from the results of X-ray fluorescence analysis, the pH measurements and electron microscopic observation. The specimens, especially in the zone B of intermediate degree of weather-

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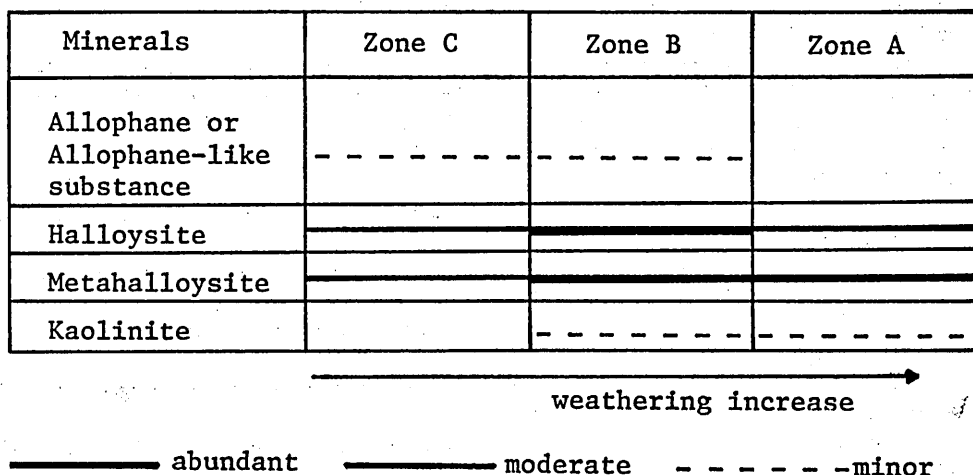
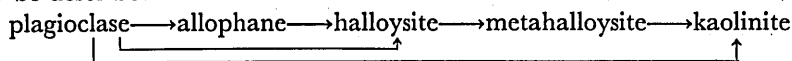


FIG. 11. Mineralogical zoning of weathering granite in the Seranishi-cho outcrop based on alteration products of plagioclase.

ing, have 4- and 6-coordinations state of Al in the crystal structure. KURABAYASHI et al. (1960, 1965) pointed out that allophane changes to halloysite of poor crystallinity during the process of weathering. Then also halloysite and metahalloysite of poor crystallinity are considered to keep 4- and 6-coordinations state of Al in the crystal structure.

In the zone A, the alteration products of plagioclase consist mainly of metahalloysite with small amount of halloysite. In the zone B, halloysite and metahalloysite are predominant with accompanying small amount of allophane or allophane-like substance. Analogous mineral assemblage is also observed in zone C (Fig. 11).

For the conclusion of the facts mentioned above, it is considered that allophane or allophane-like substance, halloysite and/or metahalloysite changed to kaolinite during progressive weathering. Metahalloysite will tend to be spontaneously transformed to kaolinite through geologic time as the consideration of HUANG (1974). This alteration sequence of kaolin minerals agrees with the opinions of progressive weathering after PARHAM (1969) and SHIMIZU (1972) etc. Then, the sequence of the alteration products of plagioclase may be described as follows:



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EXPLANATION OF PLATE XIX

1. Well developed spheroidal forms of weathering
(cf. Fig. 1 in the text).
2. Detailed profile of the sampling points.

EXPLANATION OF PLATE XX

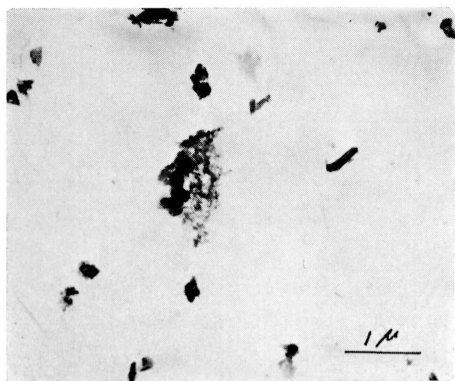
- a. Allophane or allophane-like substance showing spheritic particle in zone C (No. 11).
- b. Short tubular halloysite and/or metahalloysite in zone B (No. 11).
- c. Tubular halloysite and/or metahalloysite in zone B (No. 5).
- d. Tubular halloysite and/or metahalloysite in zone B (No. 7).
- e. Elongated tubular metahalloysite in zone A (No. 2).
- f. Elongated tubular metahalloysite and pseudo-hexagonal kaolinite in zone A (No. 2').



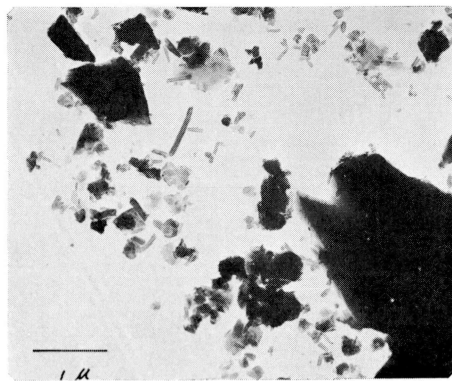
1



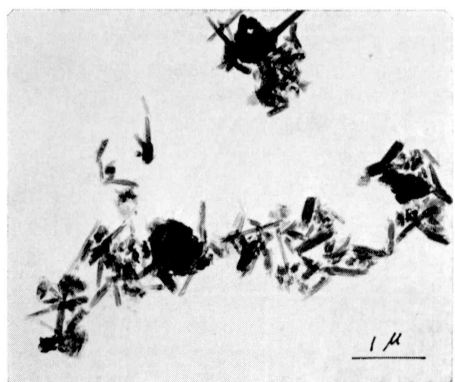
2



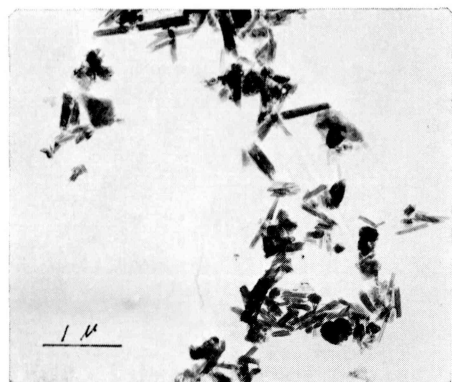
a



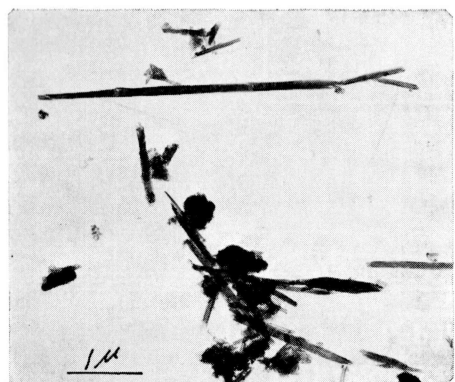
b



c



d



e



f