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# Weathering rates and mechanisms causing changes in rock properties of sandstone

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**Abstract:** Temporal changes in colour as well as mineralogical, chemical, physical and mechanical properties due to weathering during 350,000 years were examined using sandstone gravel in fluvial terrace deposits, with a known emergence time, that were distributed in the Miyazaki Plain, Japan. The time between the age of each terrace formation (20, 70, 90, 110, 120, 250 and 350 ka) and the present can be assumed to be the weathering period. Field observations showed that younger gravels (70-, 110-, 120-ka gravels) are yellowish in colour and older ones (250- and 350-ka gravels) reddish.

The main results of the measurements are: (1) according to the observation of mineral texture in thin sections, pore volume increases with increasing weathering period, and then pores connect to each other in the matrix of the sandstone; (2) the amount of clay minerals present in the matrix gradually decreases with time; (3) chemical composition of $SiO_2$ and alkali elements gradually decrease with time, whereas $FeO+Fe_2O_3$ and $Al_2O_3$ are constant during 350 ka; (4) effective porosity, pore radius and pore volume increase with increasing weathering period; (5) changing rates of specific surface area are constant during 350 ka; and (6) the rock strength index decreases with time.

These results suggest that (1) the increase in pore volume in gravel with increasing weathering period is caused by leaching of the matrix materials; (2) iron concentration and the formation of goethite and hematite occur in older gravels; (3) connected pores are formed by leaching of matrix minerals; and (4) although both pore size and pore volume increase with time, owing to the fact that winding pores are rare, both data are constant during 350 ka on the changing rates of a specific surface area. These results lead to the conclusion that leaching of matrix materials and increasing pore volume play major roles in the weathering of sandstone.

**Résumé:** Une étude a été prise dans les prairies de Miyazaki, le Japon, pour déterminer le changement des propriétés temporelles, changement de couleurs, changement minéralogique, changement chimique, changement physique, durant une période entre 350.000 ans, du a l'érosion causée par le vent et les graviers utilisant de graviers déposés sure la terrasse fluviales

Le temps entre chaque formation de terrasse est de 20, 70, 90, 110, 120, 250 et 350 ka ans et le présent est assumé comme la période de l'érosion. Une observation a l'oeil nu montre que les graviers sont d'une date récente (70-. 110-.120 ka graviers) et ceux qui montent une période plus vieille, ont respectivement une couleur jaunâtre et rougeâtre. Les résultats de ses études sont décrits ci-dessous; Selon ces observations, la texture minérale aux sections montre que le pore accroît dès que la période de l'érosion s'accroît et qui il y a des pores entremêlés entre la matrice des roches. Les minérales argileux se forment de matrices qui s'affaiblissent graduellement avec un accroissement de la période de l'érosion. La composition chimique $SiO_2$ et les éléments alkali décroissent graduellement avec une augmentation de la période de l'érosion, alors que $FeO+Fe_2O_3$ et $Al_2O_3$ restent constants pendant 350 ka ans. La porosité effective, le rayon et le volume de pore augmentent avec une augmentation de la période de l'érosion. Une superficie spécifique de la surface a changé à un taux constant pendant 350 ka ans. L'index de la robustesse des roches décroît avec une augmentation de la période de l'érosion.

Ces résultats suggèrent que (1) La croissance de pore des graviers, qui se produit durant a une augmentation de la période de l'érosion, est causée par un lessivage des matrices minérales; (2) Une concentration de fer et une formation goéthite et hémitate se produit pour les vieux graviers : (3) Des pores entremêlés se forment due au lessivage des matrices minérales. (4) Bien que la taille et le volume des pores augmentent avec une augmentation de la période de l'érosion, et bien que le phénomène de pore du au vent est rare, le taux de changement d'une superficie spécifique d'une surface et l'érosion chimique ont resté constants pendant 350 ka ans. Pour résumer, le taux de lessivage des matrices minérales et la croissance du volume des pores ont joué une part importante dans l'érosion des roches.

**Keywords:** weathering, sandstone, gravel, porosity, tensile strength, clay minerals

# INTRODUCTION

In order to determine the rates and mechanism of sandstone weathering, this study investigates the rock properties of sandstone gravel taken from terrace deposits in a series of dated fluvial terraces in the Miyazaki Plain, south Kyushu, Japan. The rates of long-term weathering are estimated based on the assumption that the weathering period is equal to the period between the age of emergence of these terraces and the present. This study discusses the changes in rock properties due to weathering during 350 ka. The measured sandstone properties include the following: (1) rock texture and mineralogical properties (observation of thin sections, mineral compositions using X-ray diffraction), (2) colors, (3) physical properties (effective porosity, pore-size distribution and specific surface area), (4) chemical properties (chemical compositions using X-ray fluorescence analysis) and (5) mechanical properties (point load strength). Using these data, the relationships between rock properties and weathering period are considered in an attempt to clarify the rates and mechanism of weathering of sandstone gravel.

# ROCK SAMPLES FOR ANALYSIS

All sampling points are directly covered with tephra layers, except for the recent river floodplain (0-ka rocks). All examined sandstones belong to the Paleogene Shimanto Supergroup. All sandstone gravel samples are fine- to medium-grain wacke-type with smaller amounts of rock fragments. About 20 samples of rounded sandstone cobbles and pebbles were taken from each sampling point. The time between the age of each terrace formation (20, 70, 90, 110, 120, 250 and 350 ka) and the present can be assumed to be the weathering period. Field observations showed that younger gravels (70-, 110-, 120-ka gravels) are yellowish in colour and older ones (250- and 350-ka gravels) reddish.

# ANALYSIS AND RESULTS

## *Rock Texture and Mineralogical Properties*

Microscope observation was carried out using three rock samples taken from each sampling site. Most of the rock samples are fine-grained wacke-type, consisting of phenocrysts of quartz and plagioclase, and clay and iron minerals in the matrix (Figure 1). Although the appearance of quartz phenocrysts has not changed as the weathering period has grown longer, plagioclase has been decomposed during the past 350 ka. Matrix minerals have also decomposed, as shown by the fact that the thin sections of matrix become increasingly unclear as the weathering period increases, while iron minerals with a diameter of about 10 to 100 µm have gradually concentrated in matrix during 350 ka.

## *Mineral Compositions*

Minerals were identified by X-ray diffraction analysis (XRD; Rigaku Co., Ltd., RAD-C System). Samples include quartz, plagioclase and clay minerals such as kaolinite, chlorite and illite. The intensity of diffracted peaks of plagioclase, illite and kaolinite gradually decreased during 350 ka, while a diffracted peak of chlorite cannot be recognised after 250-ka rocks. These results show that plagioclase and clay minerals decompose as the weathering period increases.

## *Colour*

The colors of rock samples were determined using a Minolta CM-500 Spectrophotometer as having components $L^*$, $a^*$ and $b^*$, i.e., $L^*$ $a^*$ $b^*$ colour space. The $L^*$-value is the degree of lightness: $L^* = 0$ corresponds to black, and $L^* = 100$ corresponds to white. A positive value of $a^*$ expresses red, and a negative value green. A positive value of $b^*$ indicates yellow, and a negative value blue. Positive $a^*$- and $b^*$-values express an increase in iron minerals such as goethite and hematite (Nakashima et al., 1992).

The results of colour measurements arranged by weathering time are shown in Figure 2. The $a^*$-value increases during the 250- to 350-ka period, and the $b^*$-value increases during the 0- to 250-ka period. These results support the argument that the colour becomes more yellowish during the 0- to 250-ka period and then more reddish during the 250- to 350-ka period. Therefore, the present data of the colour value indicate that goethite increases during the 0- to 250-ka period, and hematite increases during the 250- to 350-ka period.

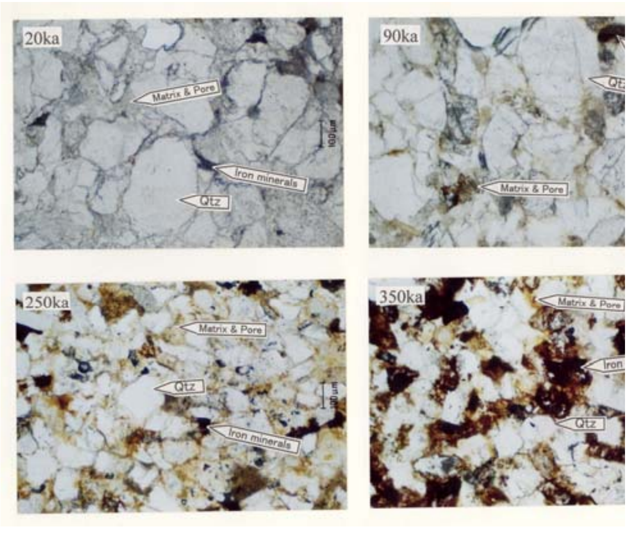

**Figure 1.** Photomicrographs of sandstone in thin section (open nicols)

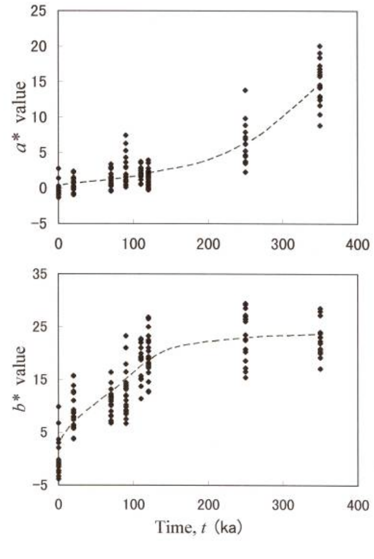

**Figure 2.** Temporal changes in colour indices ($a^*$- and $b^*$-values)

## Chemical Composition using X-ray Florescence Analysis

Chemical composition was determined using X-ray florescence analysis (XRF: Rigaku model 3270). Figure 3 shows that the chemical compositions of $SiO_2$, MgO, CaO, $Na_2O$ and $K_2O$ decrease as the weathering period increases, while those of $Al_2O_3$ and $FeO+Fe_2O_3$ are mostly constant during 350 ka. Although $SiO_2$ is a compound with low solubility in natural water, it leaches out easily due to weathering. Alkali elements such as MgO, CaO, $Na_2O$ and $K_2O$ with high solubility also leach out easily due to weathering, while $Al_2O_3$ and $FeO+Fe_2O_3$ tend to remain in the internal part of gravel because both compounds have a low solubility. Quartz, plagioclase and clay minerals such as illite and kaolinite consist of silica. Decreasing silica is considered to be caused by leaching of clay minerals because they are more soluble than is quartz. Plagioclase consists of alkali elements such as $Na_2O$, $K_2O$ and CaO. The decrease of these elements is considered to be caused by the decomposition of plagioclase.

Since the solubility of Al and Fe in natural water is lower than that of alkali elements, it remains in the matrix of sandstone. Aluminum is considered to be supplied by the decomposition of plagioclase, chlorite and kaolinite, and iron is considered to be supplied by the leaching of chlorite.

The *CIA*- and *PI*-values are excellent indices of leaching of chemical elements due to weathering (Reiche, 1943; Nesbitt and Young, 1982). The *CIA*-value expresses the degree of leaching of $SiO_2$, and the *PI*-value expresses the degree of leaching of alkali elements (CaO, $Na_2O$ and $K_2O$) because $Al_2O_3$ and $FeO+Fe_2O_3$ have a low solubility. Temporal changes in the calculated *CIA*- and *PI*-values are shown in Figure 4. The *CIA*-value ranges from 68 to 89, while *PI*-value ranges from 78 to 86. *CIA*- and *PI*-values change at a constant rate during 350 ka.

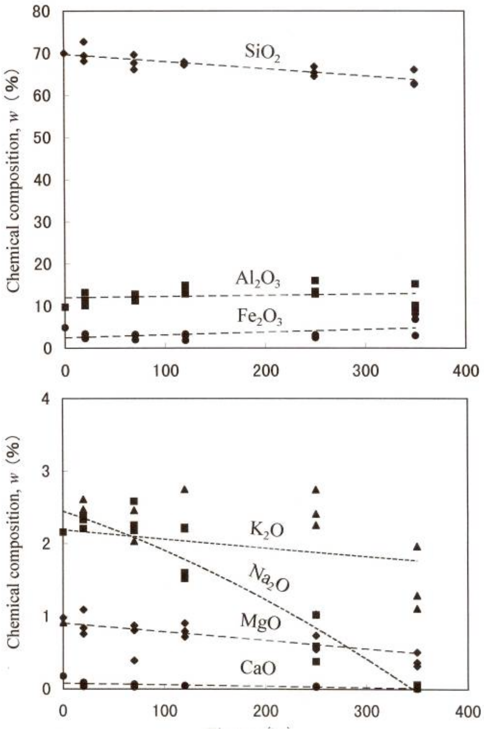

**Figure 3.** Temporal changes in chemical composition

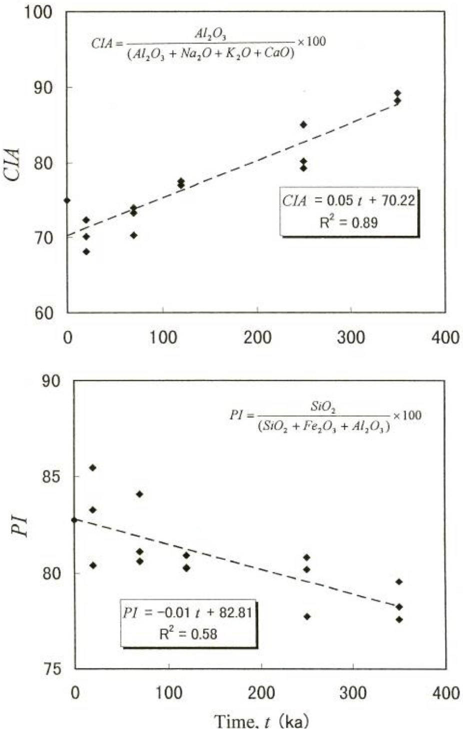

**Figure 4.** Temporal changes in chemical weathering indices (*CIA* and *PI* values)

## Effective Porosity and Pore Size Distribution

Temporal changes in the effective porosity $n_e$ (%) are shown in Figure 5. The effective porosity increases as the weathering period increases during 350 ka. The effective porosity rapidly increases during 0 to 20 ka, and 90 to 120 ka, and after that a little increase in the $n_e$-value occurs during 120 to 350 ka.

Pore-size distribution (PSD) was measured with a mercury intrusion porosimeter, the Porosimeter 2000 manufactured by the Karulo-Elva Co., Italy. A representative example of the histogram of pore size distribution is shown in Figure 6. The pattern of pore size distribution of 70- to 110-ka rocks mostly shows the normal distribution. In histograms for 120- to 350-ka rocks, the mode-value of pore volume is exceptionally rich. Both the mean pore radius and the total pore volume increase with increasing the weathering period.

The mean pore radius, *r* (μm), and the specific surface area, *S* ($m^2/g$), were calculated from the pore size distribution data from each sample. The mean pore radius increases as the weathering period increases, most rapidly during 90 to 120 ka. The specific surface area increases at a nearly constant rate during 350 ka. Thus the pore radius increases by varying amounts, but the specific surface area increases at a constant rate as the weathering period increases.

### Mechanical Properties

The point loading tensile test is a simple method for examining the mechanical properties of rocks. The strength of about 20 samples from each terrace deposit was measured with a point load strength apparatus (Model TS-40; Maruto Co., Japan). Point loading tensile strength was calculated to be the index of point load strength,  , which is defined by Kimiya (1975) as follows:

$$\tau = \log \sigma_t \ldots \qquad (1)$$

where $\sigma_t$ is tensile strength (kgf/cm$^2$). Temporal changes in the tensile strength $\tau$ are shown in Figure 7. This figure shows that the value of $\tau$ decreases during 0 to 120 ka, but after that the value of $\tau$ is constant during 120 to 250 ka. The rock strength decreases most rapidly during 90 to 120 ka.

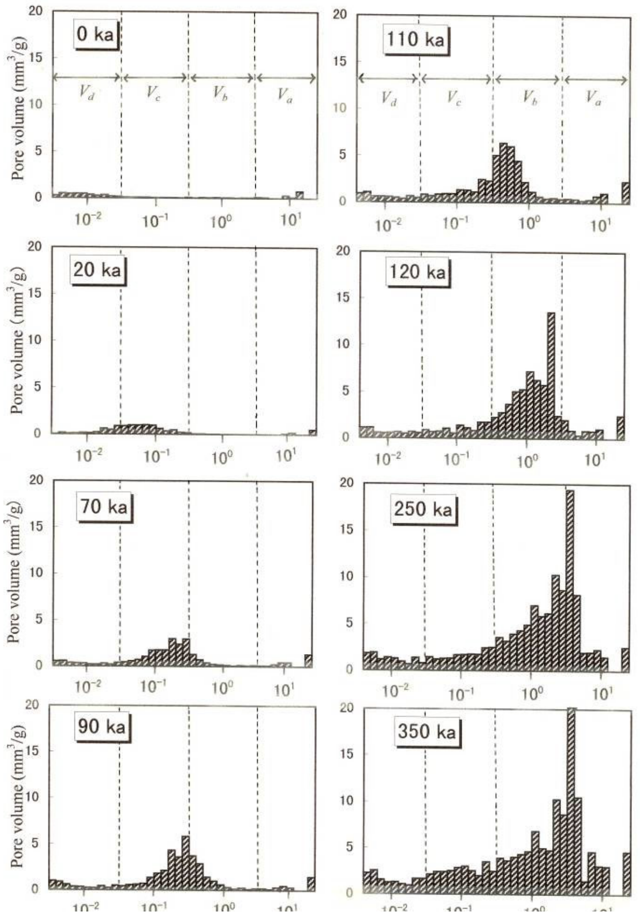

**Figure 6**. Histograms of pore size distribution measured by the mercury intrusion porosimeter

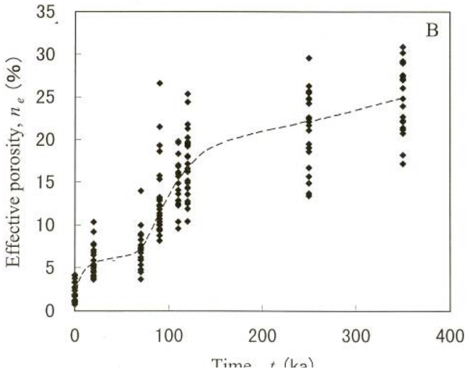

**Figure 5**. Temporal changes in effective porosity, $n_e$

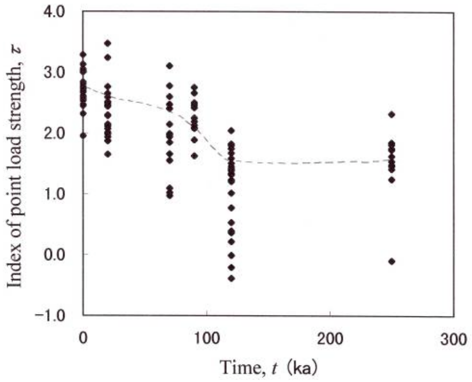

**Figure 7**. Temporal changes in index of point load strength, $\tau$.

## DISCUSSION

### Temporal Changes in Rock Properties

The temporal changes in rock properties due to weathering during 350 ka are examined using the above results. Patterns of temporal changes in rock properties can be subdivided into the following three types:

*(Type-1)*: $b^*$-value, effective porosity, $n_e$, mean pore radius, $r$, mean and index of point load strength, $\tau$. These rock properties have high rates of changing during 90 to 120 ka, while they change only slightly during 0 to 90 ka and 120 to 350 ka.

*(Type-2)*: $a^*$-value. This rock property changes rapidly during 250 to 350 ka but only slightly during 0 to 250 ka.

*(Type-3)*: Specific surface area, $S$, and two chemical weathering indices (*CIA*- and *PI*-values). These rock properties change constantly during 0 to 350 ka.

The Type-1 pattern includes physical properties except for the specific surface area, mechanical properties and the colour index of the $b^*$-value: the patterns of temporal changes in physical and mechanical properties and the colour index of the $b^*$-value are similar. The changing patterns of physical properties are similar to those of mechanical properties.

The physical properties and the colour indices change during the 0- to 120-ka period. The index of point load strength has decreased to the minimum value before 120 ka, i.e. rock strength first decreases in comparison to the other rock properties of Type-1. This result corresponds well to the results of previous studies (i.e. Oguchi et al., 1999). This relation suggests that rock strength is reduced by even slightly increasing pore volume and/or inner cracks.

The Type-2 pattern occurs only in the *a**-value. The *a**-value increases with an increasing amount of hematite and degree of mineralization of hematite (e.g. Nakashima et al., 1992). The changes of iron minerals due to weathering are discussed in detail in later.

The Type-3 pattern includes the specific surface area and the chemical weathering indices (*CIA*- and *PI*-values). The changing rates of both indices are almost constant during 350 ka. This changing pattern suggests that chemical reactions occurred actively at the surface of the materials with a large specific surface area, as indicated by Oguchi et al. (1999). Since the specific surface area of sandstone increases at a constant rate during 350 ka, the chemical weathering indices change at a constant rate during 350 ka.

## Mechanism of Weathering in Sandstone Gravel

### Change in Pore Structure in Sandstone due to Leaching of Matrix Minerals

Observations of photomicrographs and PSD data (Figs. 1 and 6) show that (1) connected and elongated pores are mainly formed in matrix, and (2) micro-pores with a diameter of less than 1μm are rare. The measurements of the effective porosity and pore-size distribution show that the pore volume and the mean pore radius of sandstone increase as the weathering period increases. These changes are explained by the connection of pores that caused changes to the pore structure. For example, if neighboring pores running parallel are connected with each other by leaching of the matrix, the pore radius would increase but effective porosity would remain constant.

Characteristic properties of sandstone weathering are that (1) the pore radius increases as the weathering period increases, and (2) the changing rate of the specific surface area is constant. In general, chemical reaction occurred at the surface of the materials. Chemical weathering rates are constant because the increasing rate of the specific surface area is constant. Changing patterns of chemical weathering of rocks correspond to that of the specific surface area of rocks. The results of XRF data confirm that the rates of chemical weathering are constant.

The weathering mechanism of sandstone can be understood as the sequential leaching of matrix. At the early stage of weathering, the porosity of sandstone is low because the matrix consists of clay minerals such as illite, kaolinite and chlorite. At the later stage of weathering, elongated and connected pores form in the matrix as a result of the leaching of clay minerals.

Chigira (1991) discussed that (1) concentrated iron minerals filled the pore space in matrix with weathering, and (2) decreasing in effective porosity and increasing in density occurred as a result of iron concentration. Photomicrographic observation and PSD data show that the effective porosity, $n_e$, increases as the weathering period increases (Figs. 1, 5 and 6), although iron minerals concentrate in the matrix of sandstone. Cementation of sand particles by iron minerals is not clear in sandstone gravel of the present study because rock strength decreases as the weathering period increases.

PSD data and the results of microscope observation show that geometrically, the pores are elongated and connected to each other. The structure of pores of the present study is similar to that of an elongate pore model described by Scholle (1977). The leaching of matrix in sandstone is mostly uniform in each of the whole rock samples. A weathering rind cannot be seen in the sandstone because of this uniform leaching that has occurred. The pore shape is also in accordance with the model of Doyen (1988). He suggests that the pore space morphology of sandstone is similar to the skeleton structure, having a network of cylindrical channels. In addition, the pore volume and the mean pore radius increase as the weathering period increases, and, thus, the radius of cylindrical channels seems to increase as the weathering period increases. To summarise, the leaching of matrix and the resulting increased pore volume play a major role in the weathering of sandstone.

## Concentration of Iron Minerals in Matrix of Sandstone

The changing process of iron minerals due to weathering affects rock structure and changes in colour of sandstone. The results of observation of microphotographs show that iron minerals accumulated in the matrix as the weathering period increased. Small amounts of iron minerals are found in 0- to 20-ka rocks, and the amounts increase in 70- to 350-ka rocks. Particles of iron minerals with a maximum diameter of 100 μm are recognised in 350-ka rock. $Fe^{2+}$ is formed by leaching of chlorite due to weathering. $Fe^{2+}$ is easily oxidized to $Fe^{3+}$ in an oxidative environment such as in terrace deposits. $Fe^{3+}$ is precipitated within the rock because $Fe^{3+}$ has a low solubility in natural water. Hydroxides such as ferrihydrite are formed by precipitation of $Fe^{3+}$.

The colour of sandstone gravel changes to yellowish or reddish as the weathering period increases. The colour of rocks is affected by the iron minerals contained therein (Nakashima et al, 1992). This suggests that when sandstone becomes reddish due to weathering, it is caused by an increase in iron minerals such as hematite. The results of colour measurement suggest that iron minerals in rock samples change from ferrihydrite to goethite ($\alpha$-FeOOH) during the 0- to 250-ka period and from ferrihydrite to hematite ($\alpha$-$Fe_2O_3$) during the 250- to 350-ka period.

Experimental studies of the dehydration of ferrihydrite have been carried out by Torrent et al. (1982) and Schwertmann and Murad (1983). These studies show that hematite increases at high temperature, low humidity and neutral pH conditions, and goethite increases at low temperature, high humidity, and acidic and/or alkalinic conditions. The formation process of goethite and hematite from ferrihydrite ($Fe_5HO_8 \cdot 4H_2O$) is discussed by Torrent et al. (1982). The results of this paper show that ferrihydrite can transform to hematite and goethite at high relative humidity, about 93%. When there is a lower relative humidity and a high temperature, the transformation to hematite is highly favored over transformation to goethite.

The results of colour measurement show that goethite increased during 70 to 250 ka BP, and hematite increased during 250 to 350 ka BP (Figure 2). The behavior and crystallization of iron in sandstone gravel due to weathering are as follows: Iron originates from chlorite. Changing iron by oxidation, hydration and mineralization during weathering ultimately produces goethite and hematite. The rock structure and colour of sandstone gravel seem to be affected by changes of iron minerals in the matrix.

# CONCLUSIONS

Changes in several rock properties due to weathering have been investigated, and the weathering mechanism of sandstone has been clarified using sandstone gravel in dated terrace deposits whose formative ages are 0, 20, 70, 90, 110, 120, 250 and 350 ka. The following conclusions can be drawn from this study:

(1) Although the appearance of quartz phenocrysts does not change as the weathering period increases, plagioclase has decomposed during 350 ka. Clay minerals in the matrix also decomposed as the weathering period increased, while iron minerals have gradually concentrated in the matrix during 350 ka.

(2) The *a**-value increased during 250 to 350 ka, and the *b**-value increased during 0 to 120 ka. This result suggests that iron minerals in rock samples changed from ferrihydrite to goethite between 0 and 250 ka and from ferrihydrite to hematite between 250 and 350 ka.

(3) Effective porosity and pore volume increases as the weathering period increases. These results show that the pore diameter of sandstone increases as the weathering period increases, and the increased pore size is mostly more than 1 μm. The mean pore diameter increases as the weathering period increases, particularly rapidly during the 90- to 120-ka period. On the other hand, the specific surface area increases at a constant rate during 350 ka.

(4) The chemical composition of $SiO_2$, MgO, CaO, $Na_2O$ and $K_2O$ decreases as the weathering period increases, while that of $Al_2O_3$ and $FeO+Fe_2O_3$ is mostly constant during 350 ka. These results show that $SiO_2$ is leached easily due to weathering, even though it has low solubility. Highly soluble compounds such as MgO, CaO, $Na_2O$ and $K_2O$ also leach easily due to weathering, but $Al_2O_3$ and $FeO+Fe_2O_3$ tend to remain in the internal part of gravel because both compounds have low solubility.

(5) The index of point load strength decreases during 0 to 120 ka, but after that remains constant during 120 to 250 ka. In particular, rock strength decreases during 90 to 120 ka.

(6) The rock structure and colour of sandstone gravel seem to be affected by changes in the iron minerals in the matrix. The behavior and crystallization of Fe due to weathering are caused by oxidation, hydration and mineralization during weathering, which ultimately produces goethite and hematite.

(7) Weathering causes the leaching of matrix materials in sandstone, which increases pore volume and pore radius. In addition, pore volume and mean pore radius increase throughout the gravel as the weathering period increases. These findings lead us to conclude that changes in the rock properties of sandstone due to weathering are the result of increased pore volume and the concentration of iron minerals in the sandstone matrix.

# REFERENCES

Chigira, M. (1991) Chemical weathering mechanisms and their effects on engineering properties of soft sandstone and conglomerate cemented by zeolite in a mountainous area. *Engng. Geol.,* **30**, 195-219.

Doyen, P. (1988) Permeability, conductivity, and pore geometry of sandstone. *Jour. Geophy. Res*., **93**, 7729-7740.

Kimiya, K. (1975) Tensile strength as a physical scale of weathering in granitic rocks. *Jour. Geo. Soc. Japan*, **81**, 349-364 (in Japanese with English abstract).

Nakashima, S., Miyagi, I., Nakata, E., Sasaki, H., Nittono, S., Hirano, T., Sato, T., and Hayashi, H. (1992) Color measurement of some natural and synthetic minerals: *Rep. Res. Inst. Natural Resources, Mining College, Akita Univ*., **57**, 57-76.

Nesbitt, H. W. and Young, G. M. (1982) Early proterozoic climates and plate motions inferred from major element chemistry of lutites. *Nature*, **299**, 715-717.

Oguchi, C., Hatta, T. and Matsukura, Y. (1999) Weathering rates over 40,000 years based on changing in rock properties of porous rhyolite. *Phys.Chem. Earth (A)*, **24**, 861-870.

Reiche, P. (1943) Graphic representation of chemical weathering. *Jour. Sediment. Petrol*., **13**, 58-68.

Scholle, P. (1979) *A color illustrated guide to constituents, textures, cements, and porosities of sandstone and associated rocks*. The American Association of Petroleum Geologists, Oklahoma, 201p.

Schwertman, U. and Murad, E. (1983) Effect of pH on the formation of goethite and hematite from ferrihydrite. *Clays and Clay Minerals*, **31**, 277-284.

Torrent, J., Guzman, R. and Parra, M. (1982) Influence of relative humidity on the crystallization of Fe (•) oxides from ferrihydrite. *Clays and Clay Minerals*, **30**, 337-340.