Soil Phys. Cond. Plant Growth, Jpn.
土壌の物理性
No. 78, p. 35〜43 (1998)

Role of Iron Oxides in Changes in Soil Microstructure with Drying and Reducing Treatments

Tomoki TAKAHASHI and Kazunobu TORIYAMA

Hokuriku National Agricultural Experiment Station
1-2-1, Inada, Joetsu, Niigata, Japan, 943-0193

Abstract

We studied the mechanisms of changes in the microstructure of anaerobic rice paddy soil when it is converted to upland field crops or restored from upland field to a rice paddy, by comparing the paddy soils with a model substance consisting of smectite-Fe oxide complexes.

For the paddy soil (Epiaquepts), the sediment volume (SV) of the soil decreased when the matric potential of the soil was lower than -1.5 MPa and increased again with the flooding following drying. The amount of reduced iron with flooding suggested that the increase in the SV depended on the history of soil reduction and not on the state of reduction. The effect of the reduction of iron oxide on the SV was evaluated using the smectite-Fe oxide complex. The SV of the smectite with iron oxide decreased more than that of the smectite without iron oxide when the matric potential of the sample was lower than -1.5 MPa. When samples were reduced by adding sodium ascorbate, only the SV of the smectite that contained iron oxide increased.

SEM micrographs showed that layersilicates were not aligned in smectite containing 0.085 kg kg^{-1} iron oxide, and it seemed that iron oxides bound to the layersilicates randomly. We concluded that the aggregation of layersilicates in the presence of iron oxide with drying was one of the factors that decrease the SV, and that the decreased volume could be restored by the reductive dissolution of the iron oxides in the soil.

Key Words : clay microstructure, iron oxide, paddy-upland rotation soils, scanning electron microscopy, sediment volume

Introduction

Poor friability and drainage are the major problems of soils involved in the rotation between rice paddies and upland crops. Both of these problems are related to soil structure (Dexter, 1988). More research on the microstructure of soils is necessary before we can improve these physical properties in soils.

It is essential to dry the soil to improve the soil structure in the converted upland fields. In general, the soil in rice paddies have no obvious structure while the field is flooded (Kida and Kawaguchi, 1960 ; Saito and Kawaguchi, 1971). However, when the paddies are converted into upland fields, soil structure gradually develop (Nakano, 1978). It is expected that drying would cause development of the structure, since drying can irreversibly change the soil's dispersing properties (Kubota, 1972) and consistency (Iwata *et al.*, 1990).

Measurement of sediment volume (SV) is the simplest way to evaluate soil microstructure in the paddy-upland rotation soils. Naganoma and Moroyu (1983) found that the SV decreased when a paddy was converted into an upland field, and that the SV increased when the field was converted back into a paddy-field. They proposed an index based on the SV to evaluate the state of the soil microstructure of paddy-upland rotation soils. However, they did not investigate the mechanism of the SV changes in the paddy-upland rotation soils.

There are some studies on the reason of the decrease with drying. Egashira and Nakayama (1979) found that the decrease in the SV during drying was highly correlated with the amount of aggregate larger than 2 μm, and they concluded that the rearrangement of clay particles during the drying process enhanced the aggregation of the soil. Katou *et al.* (1985) showed that the SV decreased irreversibly when the soil was dried at a water potential (matric potential plus osmotic potential) lower than -1.5 MPa. They explained that -1.5 MPa was the critical point for the change in soil microstructure, since the clay structure of layersilicates is rearranged irreversibly and clay domain structure (Quirk, 1994) forms at this point. On the other hand, some studies have pointed out the importance of sesquioxides in the irreversible changes of soil structure produced with drying. Kubota (1972) found that Andisols aggregated irreversibly at a water potential lower than −1.5 MPa. He concluded that this was due to dehydration and olation reaction between allophanes. Furthermore, Iwata *et al.* (1990) indicated that such irreversible changes in the physical properties of soil occurred not only in allophanic soils but also in any other soils that contained significant amounts of sesquioxides.

In contrast, there have been very few studies on the changes in the microstructure of restored paddy-field soil (restored paddy-field soil is a soil that was originally paddy-field soil, was then converted to upland field, and is now being used as paddy-field soil again). Katou *et al.* (1985) implied that the initial decrease and subsequent increase in the SV is not a simple reversible process. They suggested that iron oxides in the soil might act as a "cementing agent", and their reduction leads to deaggregation and an increase in the SV of soil. While some researchs show that paddy-field soil has no apparent structure when the field is flooded (Kida and Kawaguchi, 1960 ; Saito and Kawaguchi, 1971), the effect of the iron reduction on changes in the soil structure is still unclear. The purpose of this study is to identify the role of iron oxides in the changes in soil microstructure with drying and reducing, by using smectite-Fe oxide complexes as model substances.

Materials and methods

1) Soil

We sampled a soil (Takada soil) in an experimental field, used continuously as a rice paddy, at Hokuriku National Agricultural Experiment Station. The sampling depth was 0–10 cm. The soil that passed through a 2 mm mesh sieve was stored under the same moisture conditions as in the field at 4℃ until further use. The Taxonomic classification (USDA, 1994) of this soil is Epiaquepts.

The clay content of the Takada soil was 0.38 kg kg^{-1} and the soil texture was light clay. Smectite was the major mineralogical component of the soil, and little illite or kaolinite was present (Nakano, 1978). The citrate-dithionite extractable free iron (Holmgren, 1967) content was 0.013 kg kg^{-1}.

2) Preparation of Smectite-Fe oxide complexes

Calcium saturated smectite-Fe oxide complexes were prepared with various amounts of iron oxide, using the following modification of the method of Blakemore (1973). Ten grams (oven-dry basis) of commercial bentonite (Wako Pure Chemical Industries, Ltd.) were placed in a centrifuge tube with 500 mL of 1 mol L^{-1} NaCl, dispersed by ultrasonic treatment and mixed in an end-over-end shaker for one hour. Then, the sample was centrifuged at 2000 rpm (r=30 cm) for 10 min, and the supernatant was decanted. The procedure was repeated twice to obtain dispersed smectite. We then added various amounts of 1 mol L^{-1} ferric chloride and calcium carbonate in a molar ratio of 2 : 3 sufficient to precipitate $Fe(OH)_3$ by the following reaction.

$$2\,FeCl_3+3\,CaCO_3+3\,H_2O\rightarrow 2\,Fe\,(OH)_3 + 3\,CaCl_2+3\,CO_2 \qquad (1)$$

The samples was shaken for 16 hours and washed five times with 500 mL of 0.01 mol L^{-1} $CaCl_2$. A portion of each purified sample was air-dried for one week at room temperature for storage, and the remaining soil was kept moist at room temperature until used in the experiment. The iron content of the samples was measured by the citrate-dithionite extraction method (Holmgren, 1967). The iron content of the prepared samples ranged from 0.000 kg kg^{-1} to 0.085 kg kg^{-1}.

3) Experiment 1 : Changes in the SV of Takada soil.

Takada soil was equilibrated to various matric potentials. The matric potential was controlled by the pressure membrane method from −31 kPa to −1.0 MPa, and by the vapor equilibrium method from −2.9 MPa to −229 MPa (Katou *et al.*, 1985 ; Nakano et al., 1995). Strictly, the potential energy state of water controlled by the vapor equilibrium method was affected not only matric potential but also osmotic potential. However we considered only the effect of matric potential in this paper, since the factor of osmotic potential by the vapor equilibrium method was considered to negligible enough by comparing with that of the matric potential (Nakano *et al.*, 1995). Water content of a portion of each equilibrated sample was measured gravimetrically, and the remaining soil was used for the SV measurement. The SV was measured in the follows way : 1.0 g of the soil (oven-dried basis) was weighed in a 10-mL scaled polyethylene tube, and 9 mL of 55.5 mmol L^{-1} NaCl solution were added. The sample was shaken for 16 hours, and the volume of the suspension was brought up to 10 mL with adding distilled water. The sample was then allowed to settle for 48 hours. Then we measured the volume of the settled suspension of samples to calculate the SV.

To examine the effects of flooding on the SV, samples were incubated under flooded conditions. A 1.0 g of the soil (oven-dried basis) was weighed in a 10-mL scaled polyethylene tube and 8 mL of water were added. Samples were incubated at 20℃, 30℃, 40℃, and 30℃ with 0.1 g of dextrose to accelerate soil reduction reaction. Subsamples were drawn periodically to measure the pH, pH 2.8 acetate buffer-extractable ferrous iron content (Kumada and Asami, 1958), and the SV. To measure the SV, 1 mL of 500 mmol L^{-1} NaCl was added to the sample, shook for 16 hours, and the SV was measured. The ferrous iron content was determined by the 1, 10-phenanthroline method (Loeppert and Inskeep, 1996).

The effect of soil reduction caused by a chemical reducing agent on the SV was also examined. A 1.0 g of the soil (oven-dried basis) was weighed in a 10mL-scaled polyethylene tube, and 9 mL of a mixture of NaCl and Na ascorbate solution was added. The mixture contained 55.5 mmol L^{-1} sodium ion with various amounts of ascorbate. These samples were shaken for 16 hours, filled to a volume of 10 mL, and then the pH, pH 2.8 acetate-buffer extractable ferrous iron, and the SV were measured.

4) Experiment 2 : Changes in the SV of the smectite-Fe oxide complex.

We observed changes in the SV of smectite-Fe oxide complex with drying and the chemical reduction of iron. We used the method explained above for measuring the SV both with drying and chemical reduction, except that the sample weight was 0.5 g (oven-dried basis).

The pore size of the clay microstructure was calculated from the water retention curve using the Young-Laplace equation (Quirk, 1994).

$$r=\frac{2\gamma}{Pd}\cos\theta \qquad (2)$$

where r, γ, Pd, and θ are the assuming radius of slit with slit-shaped pores, the surface tension of water, the matric potential, and the contact angle of water respectively. We used 73×10^{-3} N m^{-1} for γ, and zero degree for θ (Nakano *et al.*, 1995).

The air-dried samples were platinum-coated and observed by using a scanning electron microscope (SEM).

5) Experiment 3 : Changes in the SV after the addition of iron oxide

We reversed the order of drying the sample and addition of iron oxide to determine the role of iron oxide in the changes in soil microstructure. The smectite and iron oxide compounds were purified and dried separately, and then they were mixed to obtain smectite with 0.085 kg kg^{-1} iron oxide. A portion of each sample was air-dried at room temperature for one week, and used to determine the SV.

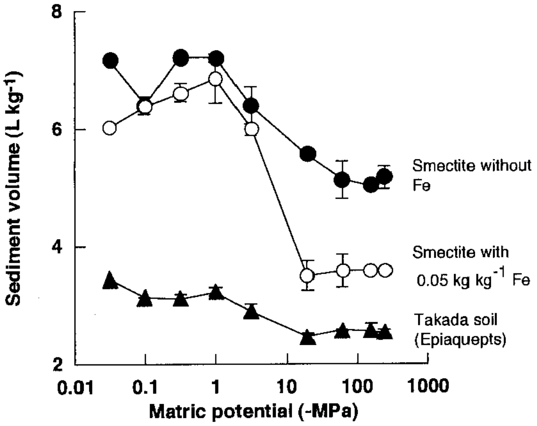

Fig. 1 Changes in the sediment volume of Takada soil (Epiaquepts) and smectite-Fe complex during drying. Data are mean value in duplicate with range.

Results and discussion

1) Changes in the SV resulting from drying

The SV of smectite-Fe oxide complex decreased with drying when the matric potential was lower than −1.5 MPa. This result is consistent with that for paddy soil (Fig. 1). The decrease in the SV was greater in the smectite with iron oxide than in the smectite without iron oxide. This tendency can be illustrated using the ratio of the SV of the dried sample to that of the fresh sample (d/f ratio, Table 1). The d/f ratio decreased gradually with increasing amounts of iron oxide.

The arrangement of smectite was considered to be the dominant factor that decreased the SV with drying. Katou *et al.* (1985) interpreted changes in the SV that occurred with drying as the result of the interaction of layersilicates. They stated that the decrease resulted from an irreversible orientation of the layersilicates that could not be affected by subsequent rewetting. However −1.5 MPa is also the critical point for the change in the soil hydrophobic properties of sesquioxide (Kubota, 1972) and soil organic matter (Nakaya, 1977). Our results confirmed that both layersilicate and layersilicate with iron oxide decreased the SV with drying at lower than −1.5 MPa. Furthermore, the addition of iron causes a greater decrease in the SV.

The mechanism for this decrease in the SV with addition of iron oxide is not clear, but iron oxide might affect paddy-field soil. Takada

Table 1 The effect of the addition of iron on the change in pH, sediment volume (SV), and pH 2.8 acetate buffer-extracted ferrous iron (Fe (II)) in air-dried and fresh smectite-Fe oxide complex containing various amounts of ferric iron

Fe content kg kg^{-1}	air-dried sample			fresh sample			d/f ratio
	pH	SV L kg^{-1}	Fe (II) $\times 10^{-3}$kg kg^{-1}	pH	SV L kg^{-1}	Fe (II) $\times 10^{-3}$kg kg^{-1}	
0.000	6.9	5.39	0.10	6.9	6.39	0.13	0.84
0.013	7.5	4.69	0.61	7.6	6.15	0.57	0.76
0.027	7.0	3.18	1.60	7.3	4.54	0.70	0.70
0.047	7.2	3.65	2.48	7.1	6.36	2.42	0.57
0.085	7.5	3.25	2.42	7.5	5.91	1.85	0.55

soil contains 0.034 kg kg^{-1} free iron per clay content. With this ratio of Fe to clay, the iron oxide could decrease the SV in the model substance (Table 1).

2) Change in the SV resulting from flooding or chemical reduction

The SV of soil increased under flooded conditions. The increase in the SV was affected by soil incubation conditions. The order of the increase in the SV was 30℃ with dextrose > 40℃ = 30℃ > 20℃ (Fig. 2-a). These results implied that soil reduction resulting from microbial activity affected the increase in the SV. Furthermore, although the change in pH at 20℃ was the same as those with pH at 30℃ and 40℃ (Fig. 2-b), the rate of the SV increase at 20℃ was significantly lower than that at 30℃ and 40℃. The pH did not seem to be a major factor in the change in the SV. The amount of reduced ferrous iron increased early in the incubation period and decreased later (Fig. 2-a). Re-oxidation of ferrous iron might be caused by oxygen leaking into the capped tubes. The increase in reduced ferrous iron early in the incubation period was roughly correlated with the SV (Fig. 2-c), but the SV at 30℃ with dextrose was higher than that at 40℃ while the amount of reduced ferrous iron was lower.

The increase in the SV seemed to be affected by soil reduction, but the contribution of reduced iron to the increase in the SV was not clear. The SV should not be a function of the state of soil reduction, because the decrease of the SV occurred with drying at matric potential lower than −1.5 MPa (Fig. 1). This supposition did not contradict our data, because the SV did not decrease later in the incubation, when the ferrous iron was re-oxidized. Hence, the increase in the SV is a function of the history of the changes in soil reduction, but not a function of the state of reduction at a specific time. The ferrous iron content in these experiments was determined by the rate of reduction of iron oxides by microbial activity and the re-oxidation by oxygen leaking into the system. It did not indicate the history of iron reduction directly. Thus, it is difficult to evaluate the effect of iron reduction on the SV in these experiments.

The addition of ascorbate to the mixture of smectite and iron oxide increased the SV (Table 2). This also occurred with paddy soil (Table 2). In the presence of 0.085 kg kg^{-1} of Fe, the d/f ratio increased significantly with ascorbate. The increase in the SV of the dried sample was attributed to the presence of iron oxides, because the addition of iron oxide was the only difference between the two samples. On the other hand, in the control, the d/f ratio was 0.84 without additional ascorbate and it decreased gradually with the addition of increasing amount of ascorbate. The addition of ascorbate decreased the SV of the clay suspension, but the mechanism for this was not

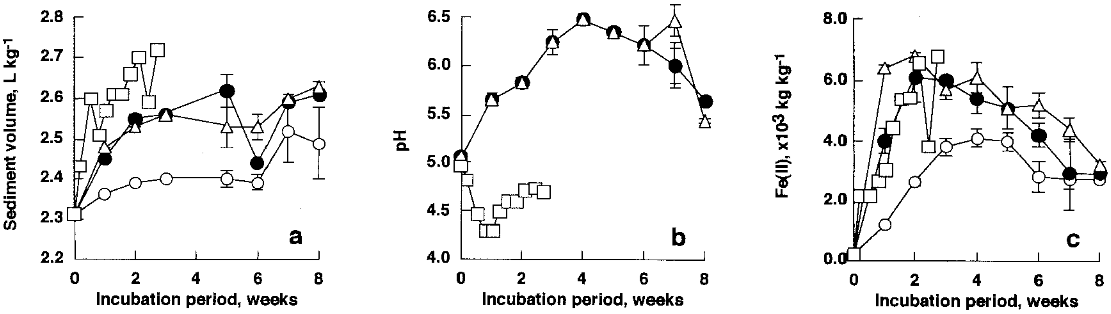

Fig. 2 Time course for sediment volume (SV), pH, pH 2.8 acetate buffer-extractable ferrous iron of Takada soil (Epiaquepts) under flooded conditions at 20℃ (○), 30℃ (●), 40℃ (△) and 30℃ with 0.1 kg kg^{-1} dextrose (□). The plots for 20℃ conditions in Fig. 2-b are hidden by the plots for 30℃

Table 2 The effect of ascorbate concentration on the change in pH, sediment volume (SV), and pH 2.8 acetate buffer-extracted ferrous iron (Fe (II)) in air-dried and fresh soil and smectite-Fe oxide complex containing various amounts of ferric iron

	ascorbate content mmol L^{-1}	air-dried sample			fresh sample			d/f ratio
		pH	SV (d) L kg^{-1}	Fe (II) $\times 10^{-3}$kg kg^{-1}	pH	SV (f) L kg^{-1}	Fe (II) $\times 10^{-3}$kg kg^{-1}	
Smectite without iron oxide	0	6.9	5.39	0.10	6.9	6.39	0.13	0.84
	10	5.8	4.43	0.27	6.0	5.40	0.31	0.82
	20	5.8	4.28	0.30	5.8	5.71	0.40	0.75
	25	5.7	4.47	0.31	5.8	5.80	0.38	0.77
	30	5.7	4.35	0.32	5.8	5.50	0.43	0.79
	40	5.7	4.24	0.32	5.8	5.44	0.41	0.78
	50	5.7	4.36	0.34	5.8	5.59	0.44	0.78
Smectite with 0.085 kg kg^{-1} iron oxide	0	7.5	3.25	2.42	7.5	5.91	1.85	0.55
	10	7.0	3.58	19.48	6.8	4.50	14.49	0.80
	20	7.1	3.64	37.25	6.9	4.60	32.65	0.79
	30	7.1	3.61	51.79	6.9	4.29	38.52	0.84
	40	7.1	3.84	72.60	6.9	4.54	45.77	0.85
	50	7.1	3.84	75.44	7.0	4.65	47.28	0.83
Takada soil	0	5.0	2.71	8.91	5.1	3.38	14.29	0.80
	10	5.0	3.29	27.69	5.3	3.30	23.48	1.00
	20	5.8	3.39	57.75	5.7	3.59	52.91	0.94
	30	5.8	3.56	62.53	5.9	3.69	54.30	0.96
	40	5.9	3.69	68.86	5.9	3.68	59.25	1.00
	50	5.9	3.83	69.30	6.0	3.80	69.52	1.01

clear.

We could not establish a quantitative relationship between the ratio of reduced iron oxide and the changes in the d/f ratio (Table 2).

3) Implication of the smectite-Fe oxide interactions during the drying process.

We compared two samples that were prepared by different procedures. For the "mixing-after-drying" sample (MaD), the smectite and iron oxide compound were purified and dried separately, prior to mixing. For the "drying-after-mixing (DaM)" sample, ferric iron was added to the smectite suspension and precipitated, and then the mixture was purified and dried.

The procedure of addition of iron oxide affected the SV (Fig. 3). While the SV of the mixing-after-drying samples did not change significantly, the SV of the drying-after-mixing samples decreased. Furthermore, the ad-

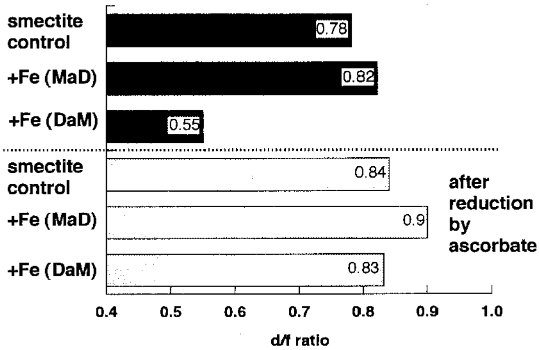

Fig. 3 Effect of the methods of Fe addition on the d/f ratio. MaD indicates the samples that were prepared by mixing dried Fe oxides and dried smectite (Expt. 3). DaM indicates the samples that were prepared by drying the mixture of smectite and Fe oxides first (Expt. 2). The iron content of all the "+Fe" sample is 0.085 kg kg^{-1}.

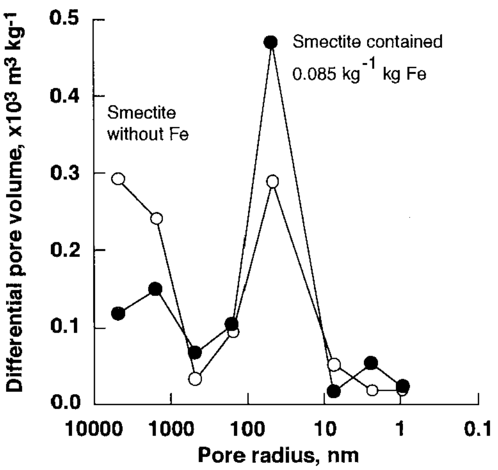

Fig. 4 The effect of addition of Fe oxides on the distribution of differential pore volume during the drying process. The pore radius were calculated by equation (2) in the text.

dition of ascorbate only increased the SV in the latter samples. The behavior of the SV of the mixing-after-drying samples was similar to that of smectite without iron oxide. These results suggest a mechanism for the increase in the SV. The increase in the SV does not result solely from the reductive dissolution (Sparks, 1995) of iron oxide. The interaction between iron oxide and the clay particles decreased the SV more effectively with drying and this could be the indispensable process for the re-increase in the SV with the reduction of iron.

4) Identification of mechanisms for the changes in the SV

To identify clay particle-iron oxide interaction, we determined changes in the soil microstructure following the addition of iron oxide, both by calculation and SEM observation. From Eq. (2), we determined that pores with a radius larger than 150 nm were predominant in the smectite without iron oxide, while more of the pores had a radius between 8 nm and 48 nm in smectite with 0.085 kg kg^{-1} iron oxide (Fig. 4). The addition of iron oxide reduced the pore size in the clay microstructure. The SEM micrographs supported this. In the smectite without iron oxide, we observed that the clay domain structure was developed well. Many quasi-crystals stacked together forming larger structures. According to Ben Rhaïem *et al.* (1987), drying increases the numbers of quasi-crystals that make up the clay domain structure in pure smectite. Plate 1a agreed with their results. On the other hand, in the sample with iron oxide, the quasi-crystals were arranged randomly and smaller pore became predominant (Plate 1 b). Oades and Waters (1991) showed a similar SEM micrographs obtained from Oxisol, in which the dominant clay fraction was kaolinite and iron oxides. They showed that the microstructure of their Oxisol was very stable and resisted to disaggregating treatments involving vigorous shaking. This is similar to the method we used to determine the SV of our sample. Though, we have no data on the interaction of iron oxides and smectite while drying, it is well known that smectite and iron oxides can interact and form stable aggregations that resist dispersion (Blakemore, 1973 ; Frenkel and Shainberg, 1980). It seemed that this interaction might have occurred in our samples.

The mechanism for the increase in the SV with the reduction of iron oxide might depend on the microstructure formed with drying. As interpreted by Katou *et al.* (1985), Ca-clay domains that are formed with drying will not separate unless exchangeable calcium ions are replaced by monovalent cations. However, in the case that the aggregation that occurs with drying was due to cementing by iron oxide, the domain structure did not improve since each domains were bound by the iron oxides. Such aggregates can be separated by the reductive dissolution (Sparks, 1995) of the iron oxides. In the rice paddy-upland crops rotation field, the cementing function of iron oxides might be more important factor to change the SV because once a domain formed without iron oxides, it will never be restored with reduction of the soil. Further research should be carried out elucidate the role of iron oxides in the

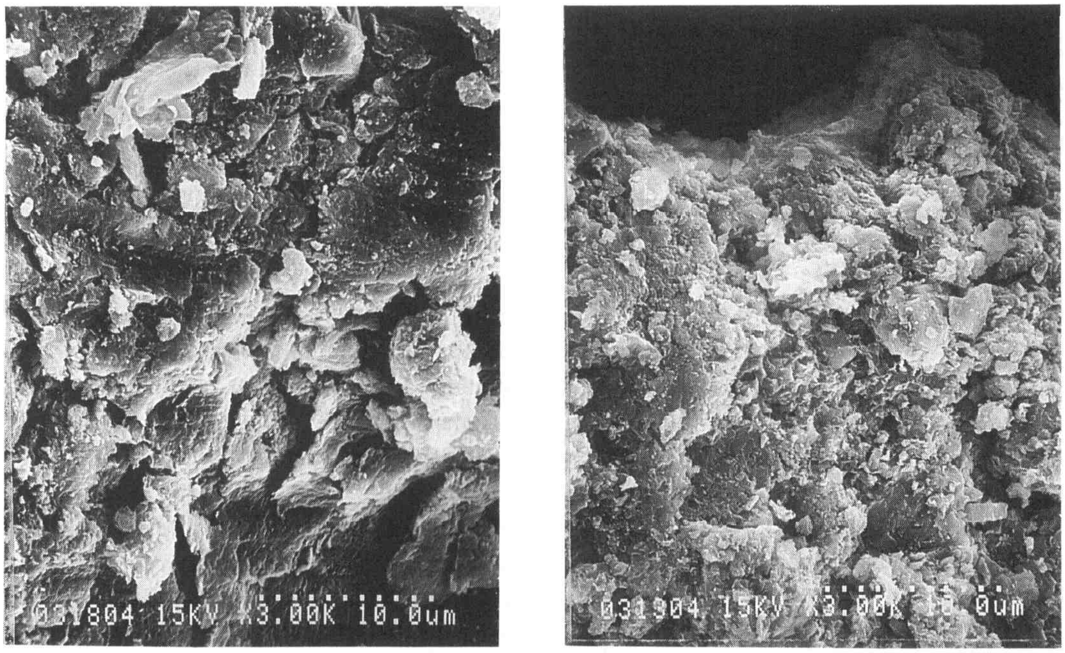

Plate 1 a

Plate 1 b

Plate 1 SEM micrographs of air-dried smectite with or without iron oxide. Plate 1 a : smectite without iron oxide. Plate 1 b : smectite with 0.085 kg kg^{-1} iron oxide.

change of soil microstructure in rice paddy-upland crops rotation fields.

Acknowledgments

We express our gratitude to Dr. M. Arai for providing scanning electron migrographs, Mr. H. Nakajima, Mr. H. Sekiya and Mr. Y. Hara for helpful discussion and advice, and to Ms. Y. Takeuchi for technical assistance.

References

Ben Rhaïem, H., Pons, C.H., and Tessier, D. (1987) : Factors affecting the microstructure of smectites : role of cation and history of applied stresses, Proc. Int. Clay Conf., Denver, 1985 (L. G. Schultz, H. van Olphen, and F. A. Mumpton, eds.), p. 292–297, Clay Miner. Soc., Indiana.

Blakemore, A.V. (1973) Aggregation of clay by the products of iron (III) hydrolysis, Aust. J. Soil Res., **11** : 75–82.

Dexter, A.R. (1988) : Advances in characterization of soil structure, Soil Tillage Res., 11 : 199–238.

Egashira, K. and Nakayama, M. (1979) : Effects of drying and disaggregating pretreatments on the sedimentation volume of a clayey deposit from Ariake Bay, Kyushu, J. Sci. Soil Manure, Jpn., **50** : 98–102, (in Japanese).

Frenkel, H. and Shainberg, I. (1980) : The effect of hydroxy-Al and hydroxy-Fe polymers on montmorillonite particle size, Soil Sci. Soc. Am. J., **44** : 626–629.

Holmgren, G.G.S. (1967) : A rapid citrate-dithionite extractable iron procedure, Soil Sci. Soc. Am. Proc., **31** : 210–211.

Iwata, S., Yamamoto, K. and Sato, T. (1990) : Amorphous inorganic material contents and changes in plasticity indexes by air drying, Soil Sci. Soc. Am. J., **54** : 558–559.

Katou, H., Nakaya, N., and Maeda, K. (1985) : Changes in sediment volume, liquid limit, and plastic limit of alluvial soils upon drying, Soil Sci. Plant Nutri., **31** : 215–226.

Kida, T. and Kawaguchi, K. (1960) : Effect of flooded, reducing and drying on soil structure. part 1. Structural stability, J. Sci. Soil Manure, Jpn., **31** : 375–379 (in Japanese).

Kubota, T. (1972) : Aggregate-formation of allophanic soils : Effect of drying on the dispersion of the soils, Soil Sci. Plant Nutri., **18**, 79–87.

Kumada, K and Asami, T. (1958) : A new method for determining ferrous iron in paddy soils, Soil Plant Food, **3** : 187-193.

Loeppert, R.H. and Inskeep, W.P. (1996) : Iron In Methods of soil analysis, part 3. chemical methods, ed. D. L. Sparks, p. 639-664., Soil Science Society of America, Inc., Madison.

Naganoma, H. and Moroyu, H. (1983) : A new index to estimate the degree of "soil uplandization" in terms of soil microstructure, Integrated researches on the developing of higher technology for upland cropping in paddy field No. 1 (Tenkanhata wo shutai to suru koudohata-sakugijutsu no kakuritsu ni kansuru sogokenkyu ; Tanaka, I. *et al.* eds.), pp 46-53, National Agricultural Research Center, Tsukuba (in Japanese).

Nakano, K. (1978) : Changes in soil physical properties of clayey soil by conversion from Ill-drained paddy field into upland field, Bull. Hokuriku Natl. Agric. Expt. Stn, **21** : 63-94 (in Japanese with English summary).

Nakano, M., Miyazaki, T., Shiozawa, S., and Nishimura, T. (1995) : Water potential measurements In Physical and environmental Analysis of Soils, p. 79-87., University of Tokyo press, Tokyo (in Japanese).

Nakaya, N. (1977) The moisture characteristics of soils in relation to organic matter with special reference to the condition of soil organic matter and water repellency, Bull. Natl. Inst., Agric. Sci. Ser. B, **32**, 1-74 (in Japanese with English summary)

Oades, J.M. and Waters, A.G. (1991) : Aggregate hierarchy in soils, Aust. J. Soil Res., **29** : 815-828.

Quirk, J.P. (1994) : Interparticle forces : A Basis for the interpretation of soil physical behavior, Adv. Agron., **53** : 121-183.

Saito, M and Kawaguchi, M. (1971) : Coagulation properties of paddy soil. part 1. changes in rice soil's physical properties in plow layer during rice cropping season., J. Sci. Soil Manure, Jpn., **42** : 1-6 (in Japanese).

Sparks, D.L. (1995) : Redox chemistry of soils, In Environmental soil chemistry, p. 187-202., Avademic press, Inc., San Diego.

USDA (1994) : Keys to soil taxonomy. sixth edition, United States Department of Agriculture, Washington.

受稿年月日 : 1998年2月2日
受理年月日 : 1998年6月4日