

Clay Minerals Of Some So-called Degraded Paddy Soils. Part 2 : Nature of clay fractions of the Saijo soil, Hiroshima Prefecture and the Kakuto soil, Miyazaki Prefecture

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CLAY MINERALS OF SOME SO-CALLED DEGRADED PADDY SOILS. PART 2

Nature of clay fractions of the Saijo soil, Hiroshima
Prefecture and the Kakuto soil, Miyazaki Prefecture

SHIGENORI AOMINE AND TOSHIO HIGASHI

In a previous paper (1) clay minerals contained in two so-called well-known degraded paddy soils were reported. It was shown that one soil derived from alluvial deposits of the Kurobe River in Toyama Prefecture has dominant illite, and clay fraction of the other soil, derived from weathered materials of granite and siliceous sandstone in Kagawa Prefecture, was largely composed of degraded illite. The results seem to indicate that the clay fraction of the degraded paddy soil is illitic.

The purpose of this work is to make a similar investigation on other degraded paddy soils to add further knowledge to the nature of the clay in the soils.

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MATERIALS AND METHODS

Soil samples used for this study were taken from two degraded paddy soils in Hiroshima and Miyazaki prefectures. Location of the soils, depth of horizons, and brief characteristics of the horizons are presented in Table 1.

Several physical and chemical analyses of fine earths and

Table 1. Soil samples used.

Sample number	Horizon	Depth	Characteristics
Locality: Saijo-machi, Kamo-gun, Hiroshima Prefecture			
410	A ₁	0-13 cm	Light grey sandy loam, arable layer
411	A ₂	13-20	Light grey sandy loam, plowing pan layer
412	A ₃	20-27	Light grey sandy loam,
413	B ₁	27-36	Yellowish grey sand loam, with brown mottlings
414	B ₂	36-45	Yellowish grey sandy loam clay, with blackish brown mottlings
Locality: Kakuto-mura, Nishimorokata-gun, Miyazaki Prefecture			
427	A ₁	0-24	Grey loam
428	A ₂	24-36	Light grey silt loam
429	B ₁	36-66	Light grey silt loam, with brownish streaks
430	B ₂	66-86	Light grey silt loam, with little brownish streaks

Table 2. Physical and chemical analyses of the fine earths.

		Saijo soil					Kakuto soil			
		410	411	412	413	414	427	428	429	430
Coarse sand (2-0.2 mm.)	(%)	22.2	31.0	39.5	40.5	12.3	7.3	6.0	1.4	4.3
Fine sand (0.2-0.02 mm.)	(%)	41.6	34.8	31.0	26.2	27.5	26.5	21.3	18.4	20.4
Silt (0.02-0.002 mm.)	(%)	32.9	21.4	18.1	18.7	37.3	48.7	52.5	59.2	54.1
Clay (<0.002 mm.)	(%)	3.3	12.8	11.4	14.6	22.9	17.5	20.2	21.0	21.2
pH(H ₂ O)		5.8	6.2	6.9	7.8	7.6	5.3	5.6	5.8	5.9
Exchange acidity	(y ₁)	0.6	0.4	0.4	0.4	0.4	2.6	2.2	1.2	1.3
Hydrolytic acidity	(y ₁)	9.6	8.3	2.9	2.8	2.4	18.3	12.7	10.0	6.3
Total organic carbon (C)	(%)	1.28	1.09	0.43	0.29	0.42	2.32	0.92	0.5	0.44
Total nitrogen (N)	(%)	0.16	0.13	0.06	0.04	0.05	0.27	0.16	0.10	0.08
C/N		8.0	8.4	7.2	7.3	8.4	8.6	5.8	5.7	5.5
Effect of air-drying* (p.p.m.)		47	30	19	6	3	131	66	55	53
	(%)	2.9	2.3	3.2	1.5	0.6	4.9	4.1	5.5	6.6
C. E. C. § (m.e./100 gm.)		9.8	8.2	5.8	6.4	11.2	13.0	11.5	13.1	9.7
Exchangeable Ca (m.e./100 gm.)		5.6	6.2	5.1	5.6	10.0	4.5	3.4	5.9	5.6
Mg (m.e./100 gm.)		0.2	0.2	0.2	0.3	0.8	0.8	0.3	0.4	1.3

* Accumulation of ammoniacal nitrogen after incubation for 28 days in waterlogged condition at 28°C.

§ Cation-exchange capacity.

mineralogical analysis of fine sand separates were carried out to learn general properties of the soil samples. The results obtained are given in Tables 2, 3, and 4.

The tables show that these degraded soils differ from normal paddy soils in that the content of Fe_2O_3 and MnO and cation-

Table 3. Chemical composition of fine earths (>2 mm.).

		Saijo soil					Kakuto soil			
		410	411	412	413	414	427	428	429	430
SiO_2	(%)	69.28	70.83	72.96	71.06	63.01	66.78	67.87	62.86	65.75
Al_2O_3	(%)	13.81	13.78	12.94	14.73	17.39	14.82	16.03	18.75	16.61
Fe_2O_3	(%)	3.38	2.14	2.42	5.17	7.32	3.03	2.41	3.82	4.41
MnO	(%)	0.01	tr.	tr.	0.05	0.12	0.01	0.01	tr.	0.01
CaO	(%)	0.90	0.77	1.11	0.71	0.91	2.30	1.89	1.22	1.30
MgO	(%)	0.31	0.34	0.66	1.21	1.41	1.32	1.37	0.74	1.13
K_2O	(%)	2.58	3.48	3.32	3.21	1.96	1.97	1.95	1.78	2.32
Na_2O	(%)	0.76	1.60	1.45	0.84	0.72	2.14	2.09	1.89	1.88
TiO_2	(%)	0.50	0.48	0.52	0.47	0.87	0.51	0.42	0.52	0.59
P_2O_5	(%)	0.04	0.10	0.04	0.01	0.03	0.20	0.15	0.07	0.07
Ignition loss	(%)	8.40	6.10	3.97	3.81	6.18	7.60	5.95	7.93	5.16
Total	(%)	99.97	99.62	99.39	101.27	99.65	100.68	100.14	99.58	99.23

Table 4. Mineralogical composition of fine sand separates (0.2-0.02 mm.).

	Saijo soil					Yakuto soil			
	410	411	412	413	414	427	428	429	430
Heavy mineral (sp. gr. >2.70) (%)	1.1	1.1	0.8	1.3	1.4	5.5	5.9	4.4	4.0
Light mineral (sp. gr. <2.70) (%)	98.9	98.9	99.2	98.7	98.6	94.5	94.1	95.6	96.0
Magnetite	±*	±	±	±	±	±	±	±	±
Rhombic pyroxene	±	±	±	±	±	+	+	+	+
Monoclinic pyroxene	-	-	-	-	-	±	+	±	±
Monoclinic amphibole	±	±	±	±	±	+	±	±	±
Zircon	±	±	±	±	±	-	-	-	-
Quartz	卐	卐	卐	卐	卐	-	-	-	-
Orthoclase	卐	卐	卐	卐	卐	-	-	-	-
Plagioclase	卐	卐	卐	卐	卐	卐	卐	卐	卐
Microcline	-	-	+	±	±	-	-	-	-
Glass	+	+	+	+	+	卐	卐	卐	卐

* 卐: Flood (60%), 卐: Abundant (30-60%), 卐: Common (10-30%),

卐: Occasional (5-10%), +: Rare (1-5%), ±: Vary rare (<1%), -: None.

exchange capacity are low, and the content of SiO_2 is rather high. The K_2O content in the Saijo soil may be largely due to orthoclase, and the considerable high content of Na_2O and CaO in the Kakuto soil may be due to the presence of glass and plagioclase.

The methods of preparing clay fractions and of analyses applied were similar to those in the previous work (1).

X-RAY ANALYSIS

X-ray diffraction data is given in Table 5 and 6. The diffraction pattern of any one clay sample from the Saijo soil is similar to another, as seen in table 5. The diffraction pattern shows a weak line at about 14 Å, which is replaced by a line at 10–11 Å by treatment for 30 minutes at 600°C. or with ammonium salt solution. These facts suggest that the 14 Å line is attributed to vermiculite. As a 10.2 Å line remained after heating to 600°C. for half an hour, the line is partly due to illite or relatives (1). The 7.3 Å line disappeared as a result of heating along with other characteristic lines of halloysite. The 4.83 Å line shows the presence of gibbsite. In addition to these minerals some feldspars are also recognized in a small amount.

Table 6 shows that the diffraction patterns bear resemblance to one another, indicating no difference in the types of clay minerals of every horizon examined. It is apparent that hydrated halloysite and halloysite are the most prominent minerals in the soil, and the ratio of the former mineral to the latter mineral increases with depth. As a line at 14.4 Å is lost by the treatment with ammonium salt solution, or heating at 600°C. for half an hour, this line is not attributed to chlorite, but to vermiculite. A small amount of feldspar is also present.

Table 5. X-ray diffraction date of clay fractions ($<2\mu$) saturated with ethylene glycol.

410		410*		411		412		413		414	
d(Å)	i§	d(Å)	i§	d(Å)	i§	d(Å)	i§	d(Å)	i§	d(Å)	i§
14.3	VI			14.3	VI	14.3	VI	14.3	VI	14.3	VI
10.2	III	(10.7	III	10.2	IV	10.2	IV	10.2	IV	10.2	IV
7.3	II			7.3	III	7.3	III	7.2	II	7.3	II
						5.94	VI	5.94	VI	5.94	VI

4.83 VI		4.83 VI	4.83 VI	4.83 VI	4.38 VI
(4.45 I	(4.44 II	(4.45 I	(4.45 I	(4.45 I	(4.45 I
4.16 IV	4.01 V	4.16 IV	4.16 IV	4.16 IV	4.16 IV
3.57 III		3.57 III	3.57 III	3.57 III	3.57 III
3.22 VI	3.22 VI	3.22 VI	3.22 VI	3.22 VI	3.22 VI
2.54 III	2.59 V	5.54 III	2.54 III	2.54 III	2.54 III
	2.44 VI				
2.33 IV	2.33 IV	2.33 IV	2.33 IV	2.33 IV	2.33 IV
	2.26 VI				
2.18 VI		2.18 VI	2.18 VI	2.18 VI	2.18 VI
	2.12 VI				
1.99 V	1.99 VI	1.99 V	1.99 V	1.99 V	1.99 V
1.81 V	1.81 VI	1.81 V	1.81 V	1.81 V	1.81 V
	1.67 VI				
1.53 V	1.53 VI	1.53 V	1.53 V	1.53 V	1.53 V
1.49 III	1.49 VI	1.49 III	1.49 III	1.49 III	1.49 III
1.45 VI		1.45 VI	1.45 VI	1.45 VI	1.45 VI
1.37 V	1.37 VI	1.37 V	1.37 V	1.37 V	1.37 V
1.29 IV		1.29 IV	1.29 IV	1.29 IV	1.29 IV
1.24 IV	1.25 VI	1.24 IV	1.24 IV	1.24 IV	1.24 IV

* Heated for 30 minutes at 600°C.

§ i: Intensity, I: Very strong, II: Strong, III: Medium, IV: Weak,
V: Very weak, VI: Barely visible, (: Reflection broadened.

Table 6. X-ray diffraction data of clay fractions ($<2\mu$) saturated with ethylene glycol.

427	427*	428	429	430
d(Å) i§	d(Å) i§	d(Å) i§	d(Å) i§	d(Å) i§
14.4 VI		14.4 VI	14.4 VI	14.4 VI
10.2 IV	10.0 IV	10.2 III	10.2 III	10.2 I
7.3 III		7.3 III	7.3 IV	7.3 IV
4.50 II	(4.49 III	(4.50 II	(4.50 II	(4.50 II
4.01 III	4.08 IV	4.01 III	4.01 III	4.01 III
3.64 V		3.64 V	3.64 V	3.64 V
3.36 III	3.35 V	3.36 III	3.36 III	3.36 III
3.21 VI	3.21 VI	3.21 VI	3.21 VI	3.21 VI
3.02 VI		3.02 VI	3.02 VI	3.02 VI
2.84 VI		2.84 VI	2.84 VI	2.84 VI

(2.57 III 2.38 V	(2.59 VI 2.43 VI	(2.57 III 2.38 V	(2.57 III 2.38 V	(2.57 III 2.38 V
2.24 VI	2.23 VI	2.24 VI	2.24 VI	2.24 VI
2.13 VI		2.13 VI	2.13 VI	2.13 VI
2.00 VI	1.99 VI	2.00 VI	2.00 VI	2.00 VI
1.82 VI	1.82 VI	1.82 VI	1.82 VI	1.82 VI
1.67 IV	1.67 VI	1.67 IV	1.67 IV	1.67 IV
1.54 VI	1.54 VI	1.54 VI	1.54 VI	1.54 VI
1.50 III		1.50 III	1.50 III	1.50 III
1.38 V	1.38 VI	1.38 V	1.38 V	1.38 V
1.35 V		1.35 V	1.35 V	1.35 V
1.30 V		1.30 V	1.30 V	1.30 V
1.25 V		1.25 V	1.25 V	1.25 V

* Heated for 30 minutes at 600°C.

§ i: Intensity, I: Very strong, II: Strong, III: Medium, IV: Weak,
V: Very weak, VI: Barely visible, (: Reflection broadened.

DIFFERENTIAL THERMAL ANALYSIS

Differential thermal curves obtained are shown in Figure 1. In general, all curves are similar to each other and have three endothermic peaks and one exothermic peak. But some differences are observed in detail between the two soils. In the Kakuto soil, the temperature of the peak of the first endothermic reaction is 10–20°C. higher than in the Saijo soil, and on the contrary, that of the third reaction is 5–15°C. lower than in the latter. Furthermore, the thermal reaction between 100°C. and 200°C. (the first endothermic reaction) is more noticeable than that between 500°C. and 600°C (the third endothermic reaction) in the Kakuto soil; but the opposite is true in the Saijo soil. These facts suggest that the clay fraction of the Kakuto soil contains some amount of allophane with hydrated halloysite and probably halloysite (4).

The curves of the Kakuto soil have a small endothermic peak at about 300°C., which is probably due to gibbsite. The exothermic peak at about 900°C. of the Kakuto soil is rather dull and has a narrow amplitude, which may be due to contamination with allophane (4). It is worthy of notice that the exothermic peak of the curves of the Saijo soil is at an evidently low temperature (865–885°C.) as compared with that of halloysite.

DEHYDRATION ANALYSIS

Dehydration curves of the Na-clays brought to equilibrium with humidity above 50 per cent H_2SO_4 at room temperature are given in Figure 2. The curves are divided into four parts, viz. below 100°C . between 100°C . and 400°C ., between 400°C . and 500°C ., and above 500°C .

The difference between the amounts of weight loss in each stage of the two clays is apparently observed except in the fourth stage, but the total loss is nearly equal in the two clays and shows about 17 per cent in weight. The clay fraction of the Kakuto soil lost more water in the first and second stages than that of the Saijo soil, whereas the circumstance is the reverse in the third stage. The curve of the Saijo soil bears close parallel to that of hydrated halloysitic clay, although the x-ray analysis indicates a considerable amount of non-expanding three-layer mineral with halloysite.

Abrupt loss of water in the third stage (between 430°C . and 470°C .) of the clay fraction of the Kakuto soil shows the presence of kaolin mineral. Unlike the clay of the Saijo soil, the curve of the clay of the Kakuto soil indicates much loss in the first and second stages. It suggests the presence of a considerable amount of allophane in the sample. On the whole the clay fraction of the

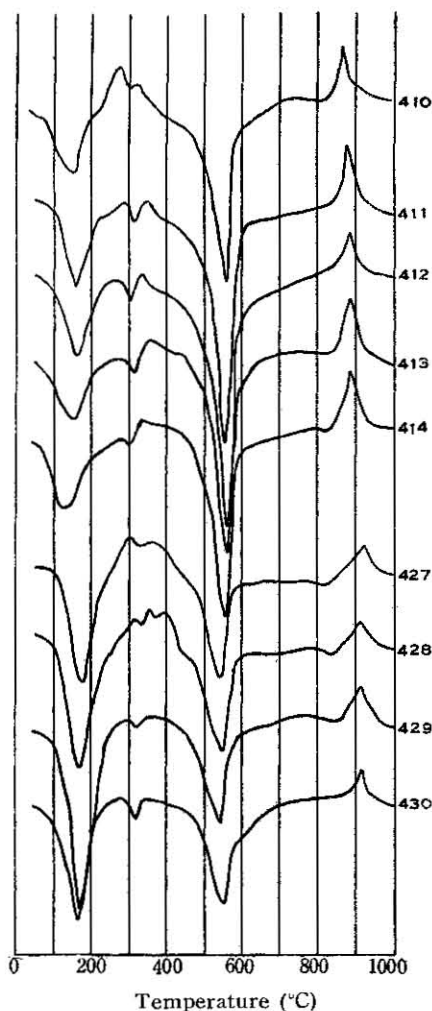


Fig. 1. Differential thermal curves of the Na-clays ($<2\mu$).

Kakuto soil is believed to be largely composed of kaolin and allophane.

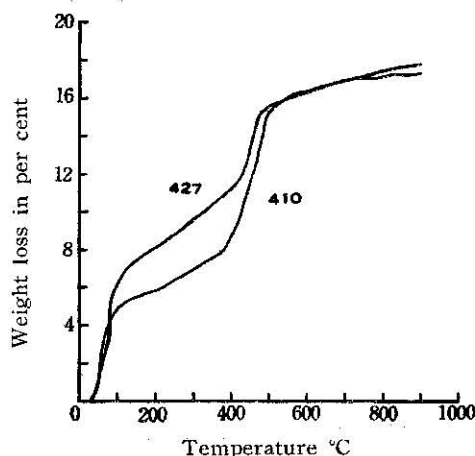


Fig. 2. Dehydration curves of Na-clays ($<2\mu$).

CHEMICAL ANALYSIS

Table 7 gives results of fusion analysis and cation-exchange capacity of the Na-clays ($<2\mu$). The data shows that the clay fractions differ from clays of common paddy soils (3) in that the content of iron is markedly low. These results, together with those of the previous work (1), suggest that the low iron is a general characteristic of the clay of the degraded paddy soil. It seems that this is more noticeable in the upper horizon.

The content of K_2O and MgO is less than 1 per cent in every clay sample with the exception of sample no. 430. This suggests that the samples do not contain much three-layer clay mineral.

SiO_2/Al_2O_3 ratio of the Saijo clay fractions is nearly equal to 2, which corresponds with that of kaolin. The ratio is well above 3 in the clays of the A horizon of the Kakuto soil. As shown later, this high ratio is partly due to the presence of diatom skeletons.

Cation-exchange capacity of the clays was usually low in the samples of the Saijo soil, and rather high in the samples of the Kakuto soil,

Table 7. Chemical composition of Na-clays ($<2\mu$).

		Saijo soil					Kakuto soil			
		410	411	412	413	414	427	428	429	430
SiO ₂	(%)	42.73	41.62	40.90	38.37	39.76	53.12	49.58	40.96	41.96
Al ₂ O ₃	(%)	33.68	35.85	35.54	34.84	33.59	22.86	26.45	29.51	30.06
Fe ₂ O ₃	(%)	3.06	1.60	3.46	5.75	3.80	3.12	2.01	2.61	3.59
MnO	(%)	0.06	0.07	0.09	0.15	0.20	0.03	0.02	0.05	0.11
CaO	(%)	0.29	0.20	0.33	0.29	0.41	0.45	0.66	0.53	0.49
MgO	(%)	0.79	0.77	0.90	0.88	0.91	0.67	0.77	0.86	1.27
K ₂ O	(%)	0.94	0.75	0.78	0.71	0.87	0.96	0.78	0.76	1.70
TiO ₂	(%)	0.70	0.47	0.53	0.72	0.77	0.69	0.36	0.72	0.77
P ₂ O ₅	(%)	0.04	0.08	0.07	0.06	0.04	0.09	0.08	0.04	0.04
H ₂ O(+)	(%)	12.78	11.65	11.84	12.07	12.51	10.57	10.93	10.00	9.74
H ₂ O(-)	(%)	4.06	4.37	3.96	3.98	4.30	7.13	6.66	9.52	6.40
Total		99.13	97.43	98.40	97.82	97.16	99.69	98.30	95.56	96.13
Molecular ratio (SiO ₂ /Al ₂ O ₃)		2.15	1.97	1.95	1.87	2.01	3.94	3.18	2.36	2.37
C. E. C. (m.e./100 gm.)*		32.4	48.5	30.9	30.3	32.6	47.9	50.5	50.1	39.4

* Cation-exchange capacity (oven dry basis).

ETHYLENE GLYCOL RETENTION

Ethylene glycol retention data is shown in Table 8. Internal surface calculated from the ethylene glycol retention is 109–147

Table 8. Ethylene glycol retention by Ca-clays ($<2\mu$).

Sample number	Ethylene glycol retention after 24 hours			Internal surface
	Unheated	Heated at 600°C.	Difference	
	mg./gm.	mg./gm.	mg./gm.	sq. m./gm.
410	55.4	9.9	45.5	146.7
411	47.8	13.7	34.1	110.0
412	45.3	11.5	33.8	109.0
413	51.7	7.3	44.4	143.2
414	61.0	25.7	35.3	113.8
427	76.0	15.9	60.1	193.8
428	76.3	17.5	58.8	189.6
429	85.6	18.9	66.7	215.1
430	59.4	13.4	46.0	148.3

square meters per gram of the clay of the Saijo soil, and 148–215 square meters per gram of the Kakuto clay fraction. The latter

figures appear to be too large for kaolin. They are probably due to not only hydrated halloysite, but also allophane (2).

ELECTRON MICROSCOPY

The electron micrographs of the Ca-clays ($<2\mu$) are shown in Figures 3 and 4. Some elongated tubular particles are commonly

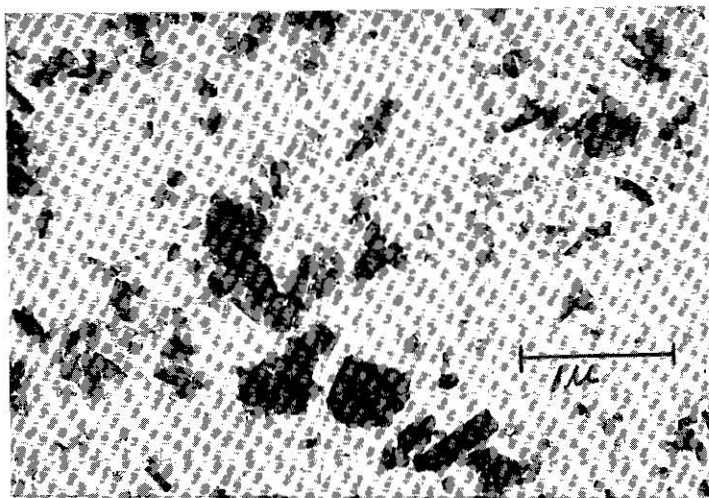


Fig. 3. Electron micrograph of Ca-clay ($<2\mu$) of Saijo soil 410.



Fig. 4. Electron micrograph of Ca-clay ($<2\mu$) of Kakuto soil 430.

found in all micrographs. The particles are undoubtedly halloysite or hydrated halloysite. Besides the particles, well-outlined flakes and aggregates are observed in the Saijo soil, and the Kakuto soil has numerous hair-like particle and a few large skeletons of diatom with a few flakes and aggregates.

DISCUSSION

From the results obtained in this work and those of the previous paper (1), it is apparent that the clay fractions of so-called degraded paddy soils do not always have a similar mineralogical composition. For instance, illite is prominent in the Nyuzen soil. On the other hand, the clay of the Kakuto soil is largely composed of hydrated halloysite and allophane. Of course these minerals are not confined to the degraded paddy soil. Special characteristics were not observed in the properties examined with the exception of low amounts of iron and manganese.

There is no doubt that the degraded paddy soil has developed by suffering from severe leaching while in a water-logged condition. It means that iron and manganese are noticeably eluviated from reduced A horizons and some of them are deposited in oxidized B horizons. The eluviation from the surface soil is clearly observed not only on a profile, but also in the chemical composition of the fine earths or of the clay fractions. Such a phenomenon is also usually recognized in the normal paddy soil which has the low level of the permanent water-table, and it is not confined to the degraded paddy soil. The difference between the degraded and normal soils is a problem of the amount of iron content in the A horizon.

A low content of iron in parent materials allows the paddy soil to easily deteriorate into the degraded soil. The soil samples examined contain noticeably less iron in the fine earths than in the normal soil. This suggests that the low iron content in the parent material has been one of the most important factors in the degradation of the sample soils. The iron content of the clay fractions in per cent was found lower than that of the fine earths suggesting that the iron had been severely leached from the soil mass.

A low content of iron in the clay fractions means the presence

of less iron or iron rich mineral, such as hydrous mica, chlorite, and montmorin. Indeed, no appreciable montmorin at all was recognized in the clay fractions of the sample soils examined. This may be another characteristic of the degraded paddy soil.

SUMMARY

Soil samples examined in this work were taken from two well-known degraded paddy fields which are located at Saijo-machi in Hiroshima Prefecture and Kakuto-mura in Miyazaki Prefecture. The former soil was derived from weathered granite, and the alluvial deposits of valcanic ejecta have developed into the latter soil.

Clay fractions less than 2 microns in diameter were separated by the sedimentation method after removing organic matter with hydrogen peroxide. The clay fractions were examined for minerals by means of the x-ray, thermal, chemical, electron microscopic, and ethylene glycol retention methods.

The clay of the Saijo soil was found to be largely composed of halloysite and degraded illite with a small amount of vermiculite, gibbsite, illite, and feldspar.

Predominant hydrated halloysite, halloysite, and allophane were found in the clay fractions of the Kakuto soil. In addition, some vermiculite, gibbsite, feldspar, and skeletons of diatoms are also contained in small quantities. A large amount of hydrated halloysite is contained in lower horizons compared with halloysite, but this relation is the reverse in the surface soil. A suggestion is offered that this may be due in part to drying in the surface soil.

No special characteristics were found in the clay fractions of the degraded paddy soils with the exception of the low iron content. However, low manganese and no appreciable montmorin may be other characteristics of the soils.

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