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SMECTITES IN IRON-RICH CALCAREOUS SOIL AND BLACK SOILS OF TAIWAN

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Abstract—The iron-rich calcareous soil (Typic Rhodustalf) from the Penghu island group represents a volcanic area. The black soils (Typic Haplustert, Vertic Endoaquoll, Typic Hapludolls) are typical of eastern Taiwan. Four A horizons and a pedon from the iron-rich calcareous soil and four pedons from the black soils were studied to analyze soil properties and clay compositions. The objective was to compare the properties of smectites developed from different parent materials. The materials were studied by using conventional X-ray diffraction (XRD) of K- and Mg-saturated clays and involved the alkylammonium ($C = 12$) method and the Greene-Kelly test. The mean-layer charge of smectites (0.48–0.52 $cmol(c)/O_{10}(OH)_2$) in the iron-rich calcareous soil was found to be higher than the black soils (0.43–0.48 $cmol(c)/O_{10}(OH)_2$). A smectite of higher charge developed from the basalts. This smectite is enriched in Fe and Mg, and lacks Si, thereby forming beidellite and/or nontronite. In contrast, under high precipitation, elevated temperature, base saturation (*e.g.*, Na, K, Ca, Mg), and about equal wet and dry cycles per year in the black soil environments, smectites developed from the complicated geologic site of eastern Taiwan. These smectites transformed to smectite-kaolinite mixed-layer clay and thus, resulted in lower-charge smectites. The K fixation capacity of the iron-rich calcareous soil was higher than the black soils.

Key Words—Black Soils, Iron-Rich Calcareous Soil, Mollisols, Smectite, Vermiculite, Vertisols.

INTRODUCTION

The Penghu island group, located ~60 km from the west coast of Taiwan, represents a volcanic district of fissure eruptions (Figure 1). These islets are marked by low, flat-topped tablelands consisting of basalt flows and tuffs with little sand and clay intercalation (Ho, 1988). The red soils with maghemite nodules of high magnetic susceptibility are developed from this important volcanic group (Wang *et al.*, 1988a). Goethite and hematite are the dominant Fe-oxides in the soil and the mafic rock, along with laths of plagioclase, dark augite, and some olivine. The olivine basalts show spheroidal weathering, which produces a rusty brown mass. The upper surface of the basalt flows is usually weathered into red soil under the subtropical sun and poor drainage. Leung and Lai (1965) classified these soils as young, reddish-brown lateritic soils which commonly develop where the average annual rainfall is <1000 mm. Vertisols and Mollisols are not extensively found in Taiwan, but are important in agricultural soil sources in eastern Taiwan (Figure 1). The four pedons described here are believed to be representative of approximately 160 km^2 of Vertisols or Mollisols and associated black soils occurring in eastern Taiwan.

The coastal range lies between the longitudinal valley and the eastern coast of Taiwan. This range is ~135 km long, with a maximum width of 10 km, and is only 3 km wide at the northern and southern extremities. On the east, it faces the Pacific ocean (Figure 1), whereas on the west, it is separated from the central range cordillera by a narrow linear valley. The central range is the most mobile strip of the Taiwan Orogenic belt and is of exceptional tectonic interest owing to the predominance of chaotic rock or melangé, which played an important role in the history of the mountain belt. The Neogene deposits in this eastern basin are mainly marine and partly volcanoclastic, having a combined thickness of 6000–7000 m.

Layer charge is an important property of 2:1 phyllosilicates in that it determines the cation-retention capacity of clays. It influences the selectivity of various cations during ion-exchange, and it affects the ability of clays to adsorb water and various polar organic molecules (Laird, 1987). However, use and management of the high smectite-containing, iron-rich calcareous soil of Penghu and the eastern Taiwan black soils for agricultural production requires a better understanding of their properties than now exists. The primary objective of the present work was to characterize the smectites in the iron-rich calcareous soil of the Penghu island group and the black soils of eastern Taiwan and to provide detailed data on field morphology, chemistry, physical properties, and mineralogy of these soils.

MATERIALS AND METHODS

Study area

The Penghu island group, because of their basalt origin, consists of iron-rich calcareous soil. The sample sites are located between latitudes 23°09′N and 23°47′N, and longitudes 119°18′E and 119°47′E (Figure 1). The climate is hyperthermic, semi-arid, with a mean annual temperature of 23°C, a mean annual precipitation of 900 mm, and evaporation of 1600 mm, with monsoons in May and August. The natural vegetation at the sites consists of *Imperata cylindrica, Gaillardia pulchella, Miscathus floridulus*, salt-resistant, and drought-resistant species.

Table 1. Summary of selected physical and chemical properties of soils tested.

Sample	Depth (cm)	Horizon	Color (moist)	pH	CEC cmol kg^{-1}	OC g kg^{-1}	Clay g kg^{-1}	Sand g kg^{-1}	Texture	$CaCO_3$ (%)
					Penghu iron-rich calcareous soil					
TW	0–15	A	5YR3/4	8.2	29.8	11.8	390	534	CL[1]	15.1
	15–45	B1	5YR4/6	8.0	57.5	2.9	404	379	C	0.8
	45–80	B2	5YR4/6	8.8	60.3	1.2	416	374	C	7.3
AZ	0–15	A	5YR5/4	8.2	14.1	11.9	380	214	CL	14.1
SY	0–10	A	5YR3/4	8.2	11.7	5.8	220	748	CL	11.7
HC	0–12	A	2.5YR3/4	7.7	27.1	41.1	401	506	SiL	29.1
CM	0–15	A	5YR5/6	7.1	45.7	2.3	128	510	SCL	45.7
					Black soil in eastern Taiwan					
YM	0–5	Ap1	N2.5/	6.0	34.9	46.0	229	453	L	—[2]
	5–21	Ap2	N2.5/	6.3	32.6	47.0	251	484	L	—
	21–34	Ap3	N2.5/	6.6	36.9	46.0	338	383	CL	—
	34–50	AB	5YR2.5/1	6.7	31.4	23.0	302	300	CL	—
	50–70	Bw	10YR4/4	6.8	40.5	13.0	262	367	CL	—
CB	0–10	A	N2.5/	5.8	38.5	46.0	571	111	C	—
	10–44	Bss1	N2.5/	6.6	37.7	32.0	598	156	C	—
	44–64	Bss2	N2.5/	6.5	44.5	16.0	570	104	C	—
	64–102	BCss1	7.5YR3/1	6.7	44.1	8.0	490	168	C	—
	102–138	2C1	10YR4/3	6.9	35.7	1.0	166	602	SL	—
	138–	2C2	10YR5/3	7.1	32.1	1.0	90	699	SL	—
SYS	0–13	Ap1	5Y2.5/1	7.5	42.7	66.0	445	272	C	—
	13–14	Ap2	N2.5/	7.2	51.3	64.0	498	208	C	—
	40–55	2Bw	2.5YR2.5/1	7.3	48.2	30.0	458	293	C	—
	55–71	2Bg1	2.5YR4/3	7.2	41.7	15.0	358	278	CL	—
	71–83	2Bg2	2.5YR4/3	7.2	40.2	9.0	382	143	SiCL	—
	83–	2Bg3	5Y3/2	7.3	36.7	7.0	350	163	SiCL	—
CS	0–22	Ap	7.5YR3/2	5.8	19.3	39.0	238	192	SiL	—
	22–36	A1	N2.5/	6.2	20.5	27.0	246	188	SiL	—
	36–47	A2	N2.5/	6.8	14.3	8.0	331	139	SiCL	—
	47–65	Bw	5Y4/4	7.0	15.2	3.0	386	153	SiCL	—
	65–91	Bg1	10YR5/6	6.8	14.6	3.0	406	160	SiC	—
	91–	Bg2	2.5YR6/1	6.9	23.1	2.0	464	120	SiC	—

[1] C indicates clay; CL: clay loam; SiCL: silty clay loam; SiC: silty clay; SCL: sandy loam; SL: sandy loam; L: loam.
[2] —: not detected.

The research area of the four pedons in the black soils is located in Hualian and Taitung Prefectures of eastern Taiwan. The low elevation of sample sites is located between latitudes 23°02′20″N to 23°51′47″N, and longitudes 121°11′27″E to 121°32′52″E (Figure 1). The climate is hyperthermic, udic, with a mean annual temperature of 25.3°C, summer rainfall, and a mean annual precipitation of 1500 mm. Precipitation is concentrated in May and November, leading to half-year wet and dry cycles. The natural vegetation at the sites consists of *Imperata cylindrica* (Yue-mei, YM pedon), *Phragmites communis* (Chang-bin, CB pedon), *Zoysia mattrella* and *Setarria vividis* (Shi-yu-san, SYS pedon), and *Areca catechu* (Chi-shang, CS pedon). The parent materials for these sample sites consist of tuff and agglomerate (YM pedon), mudstone and tuff (CB pedon), mudstone and agglomerate (SYS pedon), and agglomerate and serpentine mixed with andesite (CS pedon) (Ho, 1988).

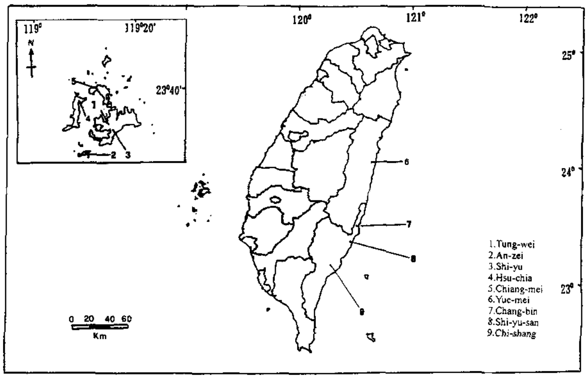

Figure 1. Location of sample sites on the Penghu island group and eastern Taiwan (Ho, 1988).

Soil material sampling

The Tung-wei (TW) pedon of the Penghu iron-rich calcareous soil represents a fine, mixed, hyperthermic, Typic Rhodustalf (Soil Survey Staff, 1996). A horizons were collected from An-zei (AZ), Shi-yu (SYY), Hsu-chia (HC) and Chiang-mei (CM) sites.

Table 1. Extended.

Sample	Horizon	Dithionite		NH_4-Oxalate			Exchangeable cations				Base saturation (%)
		Fe_d	Al_d	Fe_o	Al_o	Fe_d-Fe_o	K	Na	Ca	Mg	
		g kg^{-1}					cmol kg^{-1}				
				Penghu iron-rich calcareous soil							
TW	A	68.0	5.6	4.6	2.3	63.4	0.3	1.3	49.8	7.1	100
	B1	53.0	6.3	4.4	2.3	48.6	0.2	2.7	45.1	14.5	86
	B2	107.0	8.4	3.2	2.1	103.8	0.2	2.7	47.5	15.0	90
AZ	A	71.5	9.7	6.0	2.3	65.5	0.9	0.6	33.2	7.9	100
SY	A	11.6	6.6	1.9	1.2	9.7	0.3	1.0	32.7	6.3	100
HC	A	10.2	8.5	3.3	1.9	6.9	1.0	0.5	10.7	4.2	100
CM	A	8.4	4.1	2.4	0.9	6.0	0.6	0.5	14.8	2.7	100
				Black soil in eastern Taiwan							
YM	Ap1	7.3	2.7	4.7	1.4	2.6	1.0	1.4	15.7	7.3	73
	Ap2	7.2	3.1	5.0	1.6	2.2	0.7	1.2	15.8	7.0	78
	Ap3	8.7	3.3	5.5	2.2	3.2	0.4	1.6	15.9	8.7	72
	AB	7.7	3.1	4.8	2.2	2.9	0.2	1.9	15.8	9.0	84
	Bw	11.3	3.3	4.5	1.3	6.8	0.2	2.1	15.8	9.2	67
CB	A	14.9	5.5	6.9	3.5	8.0	0.5	0.7	20.6	10.8	85
	Bss1	12.2	4.4	6.1	2.8	6.1	0.1	0.9	21.9	12.3	94
	Bss2	11.1	4.1	4.9	4.4	6.2	0.1	1.3	23.4	14.3	88
	BCss1	11.3	3.2	2.5	3.8	8.8	0.1	1.5	23.4	14.5	90
	2C1	6.4	2.9	1.7	2.3	4.7	0.1	1.6	21.8	12.5	100
	2C2	4.5	1.7	1.4	0.7	3.1	0.1	1.6	20.5	11.3	100
SYS	Ap1	6.5	4.1	2.9	1.8	3.6	0.7	0.7	24.5	12.0	89
	Ap2	9.1	4.0	4.5	4.0	4.6	0.3	0.8	24.1	11.9	76
	2Bw	12.9	6.9	4.4	3.9	8.5	0.4	1.0	24.5	11.8	78
	2Bg1	11.6	5.0	2.7	3.1	8.9	0.4	1.0	24.4	11.2	88
	2Bg2	9.9	4.8	2.7	2.6	7.2	0.3	0.9	23.7	10.9	89
	2Bg3	8.7	4.7	2.7	2.2	6.0	0.3	0.8	23.1	10.4	94
CS	Ap	21.0	2.4	5.4	1.9	15.6	0.1	0.1	11.1	4.6	83
	A1	22.0	3.9	4.9	2.3	17.1	0.1	0.1	11.5	5.6	84
	A2	18.8	2.7	1.1	1.1	17.7	0.1	0.1	10.9	6.0	100
	Bw	22.7	3.5	1.3	1.5	21.4	0.1	0.1	9.0	5.6	100
	Bg1	28.2	6.1	0.7	1.5	27.5	0.1	0.1	9.4	5.9	100
	Bg2	27.5	5.6	0.8	1.4	26.7	0.2	0.2	13.0	7.5	89

The CB and SYS soils exhibited slickensides in the subsoil and exhibited cracks <1 cm wide in the solum, which may extend into the transitional horizon. A high overburden pressure is associated with the soil undergoing changes from an extremely wet to a very dry state and because it is more than 60 cm thick (Soil Survey Staff, 1996). The CB and SYS soils have a high clay content and characteristic features that allow them to be classified as a fine, montmorillonitic, hyperthermic, Typic Haplustert and a fine, mixed, hyperthermic, Vertic Endoaquoll, respectively, according to the criteria of Graham and Southard (1983) and the International Committee on Vertisols (1990). The YM and CS soils, found at a slightly higher elevation in the landscape, cannot be considered as Vertisol, since they do not have slickensides. Thus, they can be classified as fine-loamy, mixed, hyperthermic, Typic Hapludoll due to their lack of vertic properties, confirming the limited thickness and clay-content common to medium-textured soil developed on crystalline hard rocks. These soil profiles consist of various horizons, each of which was separately collected and analyzed.

Soil sample studies

The soil samples were air-dried and crushed to pass through a 2 mm sieve. Organic matter and MnO_2 was removed with a 30% H_2O_2 solution by heating on a hot plate. To improve the identification of the other soil minerals by X-ray diffraction (XRD) analysis, the samples were dispersed and fractionated to clay fractions (<2 μm) (Jackson, 1979). The silt was separated from the sand by wet-sieve (53 μm sieve) techniques. The clay fraction was dialyzed against distilled water and freeze-dried.

Soil pH was determined with a 1:1 soil to water suspension using a pH meter. Organic C was determined by wet oxidation (Walkley and Black, 1934). Soil calcium carbonate was determined by a modified Van Slyke manometric method (Van Slyke and Folch, 1940). Aluminum and iron oxides were extracted with dithionite-citrate-bicarbonate (DCB) solutions (Mehra and Jackson, 1960) for free sesquioxides (Al_d and Fe_d), and with NH_4-oxalate solutions of pH 3 (0.2 M) for non-crystalline sesquioxides (Al_o and Fe_o) (McKeague *et al.*, 1971). The extracts of sesquioxides were determined by atomic absorption (AA) spectrophotometry (Hitachi, AAS 180-30).

Cation exchange capacity (CEC) of the clay fractions was determined according to the method described by Jackson (1979), and the values of CEC were used to determine the quantity of vermiculite and montmorillonite (Alexiades and Jackson, 1965; Coffman and Fanning, 1974). The quantity of illite was estimated from the K_2O content. In clay samples, kaolinite was estimated by the XRD peak intensity at 0.72 nm (Brindley, 1980). The quantity of mixed-layer clay was estimated by subtracting the amount of other clays from total clay. The potassium fixation capacity was calculated by the following equation [(CaEC) − (KEC)]/(CaEC)] × 100 (Senkayi *et al.*, 1983; Wang *et al.*, 1988b) where CaEC = the CEC of Ca/Mg exchange and KEC = the CEC of K/NH_4 exchange.

The deferrated clay samples (*i.e.*, samples with free-iron oxides removed) were saturated with Mg and K, and mounted as slurries on glass slides for XRD analysis (Jackson, 1979). The Mg-saturated clays were examined at 25°C before and after glycerol solvation. The K-saturated clays were examined at 25°C and after heating at 110, 350, and 550°C for 2 h. The oriented clay mineral aggregates were examined with an X-ray diffractometer (Rigaku Miniflex) with CuKα radiation. The XRD patterns were recorded in the range of 2–50 °2θ with a scan rate of 2 °2θ min^{-1}.

The deferrated clays were treated with LiCl solution (3 M) to test for the different species of smectite (Greene-Kelly, 1953). The alkylammonium saturated clays were obtained through treatment with acqueous solutions of alkylamine (C = 12) hydrochlorides. The alkylammonium was prepared by mixing alkylamine and HCl solutions (Lagaly, 1979; Laird, 1987). The 30 mg of deferrated clay was heated at 65–75°C in a water bath for 24 h. The organo-clay was washed with a 1:1 ethanol and water solution several times and subsequently analyzed by XRD, using *d*(001)-values to calculate the mean layer charge distribution (Egashira *et al.*, 1981; Laird, 1987; Olis *et al.*, 1990; Xing and Dudas, 1994; Burras *et al.*, 1996). After DCB treatments, 0.5 g of clay was successively treated with a sodium citrate solution (pH 7, 0.3 M) and heated at 80°C for different time periods to remove the interlayer materials (Matsue and Wada, 1988). The time periods for sodium citrate treatments were 0.5, 1, 2, 4, 8, 12, and 16 h. Al, Fe, and Mg contents in these solutions were determined by atomic absorption (AA). After each treatment of sodium citrate solution, the clay was saturated with K and heated at 110, 350, and 550°C, while the Mg-saturated clay was treated with glycerol solvation and then analyzed by XRD (Tamura, 1958).

RESULTS AND DISCUSSION

The pH of the iron-rich calcareous Penghu soil is above 7.0 (Table 1) with calcium carbonate and shell nodules. High contents of Fe_d and low Fe_o indicate high concentrations of crystalline Fe-oxides (Fe_d − Fe_o). The soil texture ranges from clay to sandy clay. The magnetic susceptibility of the Penghu iron-rich calcareous soil shows a large variation with soil depth. This variation is proportional to the amount of Fe_d and ferromagnetic minerals such as maghemite and magnetite. The low Fe (II) content (5–14%) with respect to total Fe indicates the predominance of maghemite over magnetite (Wang *et al.*, 1988a).

Each pedon study of black soils in eastern Taiwan showed texture ranging from clay, silty clay, silty loam, and silty clay loam to sandy loam. The range of soil color depending on moisture content was from N 2.5/ to 10YR 5/6. The pH of the soils increases from 5.8 to 7.5 with depth and diminishing organic carbon content. The CEC of pedons was >31 cmol kg^{-1} except for the CS pedon. The base (Na, K, Ca, and Mg) saturation of all pedons was >67% with high exchangeable Ca and Mg (Table 1) and low exchangeable Na and K. In general, the CS pedon contained high quantities of Fe_d (18.8–28.2 g kg^{-1}), which increased with depth. A high Fe_o content was present in the B horizon of the CB pedon. However, the Fe_o, Al_o, and Al_d, and Al_o content in the YM, CB, and SYS pedons had no discernable trend (Table 1).

X-ray diffractograms of K-saturated clays at 25°C in the B1 horizon (TW pedon) showed a 1.29 nm *d*(001)-value. In the K-saturated clay heated at 110°C and 550°C, the *d*(001) peaks were 1.23 and 1.01 nm, respectively. In the Mg-saturated clay, the *d*(001)-value was 1.42 nm and shifted to 1.74 nm with glycerol solvation (Figure 2A). These results indicated smectite, illite, and kaolinite (Table 2). The Greene-Kelly (1953) test of heating Li-saturated clay at 220°C with subsequent glycerol solvation resulted in a 1.70 nm XRD peak, indicating a dioctahedral smectite with tetrahedral-substitution (Wang *et al.*, 1988b; Borchardt, 1989). The Chiang-mei (CM) paleosol profiles also had nontronite nodules, which developed on the basalts of the Late-Miocene volcanic group (Wang, 1997; Wang *et al.*, 1998). The *d*(001) peak of illite (AZ pedon, A horizon) was at 1.01 nm in K-saturated clay heated at 110 and 550°C. No significant expansion occurred in Mg-saturated clay solvated with glycerol. However, the *d*(001) peak at 0.72 nm was not observed after heating the K-saturated clay at 550°C (Figure 2B). The XRD patterns indicate that kaolinite, smectite, and illite are present at the A horizon of the SY pedon (Table 2; Figure 2C). No significant chlorite peaks were detected by XRD analysis. The basal interplanar *d*(001)-values of the alkylammonium-clay complexes (Wang *et al.*, 1988b) were from 1.86 to 1.93 nm (C = 12) (Table 3). Soil smectite extracted by treatment of successive hot sodium citrate solutions for 16 h and further treated with alkylammonium slightly changed the *d*(001)-values. The mean-layer charge per formula unit of smectite

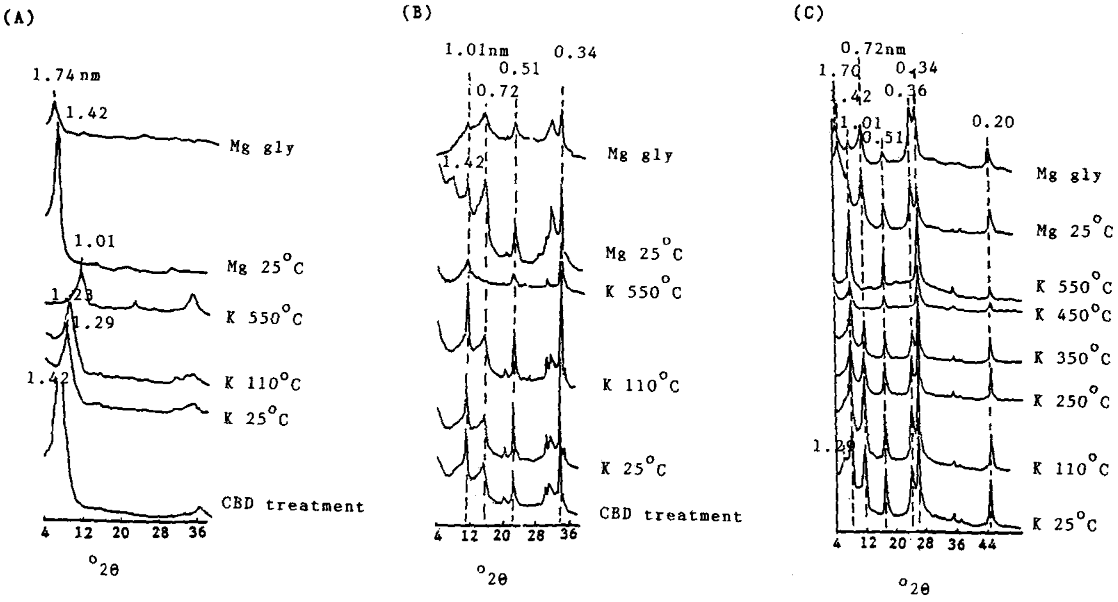

Figure 2. X-ray diffractograms of clay of the (A) TW pedon (B1 horizon), (B) AZ pedon (A horizon), and (C) SY pedon (A horizon).

was calculated from the *d*(001)-values of the reference clays (n = 13) using the linear equation of Y = 1.7023X + 1.0435 (R^2 = 0.9795) (Table 3) where Y is the *d*(001)-value from the alkylammonium exchange method and X is the mean-layer charge cmol(c)/$O_{10}(OH)_2$. The results of alkylammonium exchange indicate that the layer charge of smectite from Penghu island is between 0.48–0.52 cmol(c)/$O_{10}(OH)_2$ (Table 3). However, after the hot sodium citrate treatment, the mean-layer charge of the Penghu smectite showed a slight increase in the range between 0.49–0.54 cmol(c)/$O_{10}(OH)_2$ (XRD data not shown).

Using the Greene-Kelly test (Kantor and Schwertmann, 1974; Glasmann, 1982; Bühmann and Grubb, 1991), it was determined that beidellite or nontronite is the common weathering product of the basalts. However, in the central Coastal Range of Oregon, beidellite or nontronite is metastable with respect to kaolinite when soil is subjected to more active leaching conditions. The smectite to kaolin transformation may involve a solid-state reaction with kaolin-smectite interstratifications as an intermediate phase(s). This reaction is reported to proceed through expulsion of certain interlayer and layer cations, the most important being Fe (Wildman *et al.*, 1968). A corresponding increase in the Al/Si ratios in the clay mineral ensues, with the release of Fe precipitating as goethite and/or hematite, and thus imparting the characteristic red color to the soil (Herbillion *et al.*, 1981; Bühmann and Grubb, 1991).

Because the basalts have low Si content, tetrahedral substitutions produce high-charge beidellite in the Penghu iron-rich calcareous soil. Different weathering systems exist in these districts, in places only a few meters away from the area of shallow red soils. Limited leaching in small, partially enclosed basins resulted in the production of montmorillonite-dominant clay soil (Beckmann *et al.*, 1984; Wang *et al.*, 1988b; Ahmad, 1996).

The Mg-saturated clays with glycerol solvation have a very intense XRD peak of 1.41 nm in the Ap2 horizon of the SYS clay fractions. The 0.72 nm peak, indicating the presence of kaolinite, was observed in all clay samples but this peak was removed by heating at 550°C. The smectite, which was slightly expanded with glycerol solvation, re-expanded after Li-saturation and heating at 280°C. The basal *d*(001)-values of the alklyammonium-clay complexes were in the range of 1.84–1.85 nm in clays saturated with alkylammonium ion (C = 12) (Figure 3). In contrast, successive hot sodium citrate treatments essentially eliminated the weak reflections at 1.24 nm. The quantity of Al, Fe, and Mg extracted by sodium citrate solution sharply increased with extraction time for the first 16 h (Table 4). The hot sodium citrate may first dissolve the loosely bonded interlayer materials and then slowly dissolve the relatively resistant matrix (Xing and Dudas, 1994). In the Mg-saturated clay with glycerol solvation, the *d*(001)-value of the XRD peak shifted to 1.85 nm. The characteristic XRD peaks of the YM and CB soil clays were similar to that of the SYS pedon (Ap2 horizon) (Pai, 1997).

With the exception of the CS pedon, the XRD diffractograms of specimens treated with alkylammonium ion (C = 12), displayed reflections in the range of 1.78–1.86 nm (Table 3), suggesting bilayers of straight-chain hydrocarbons (Olis *et al.*, 1990; Lagaly, 1992; Reid *et al.*, 1996). The monolayer-bilayer transition is consistent with the presence of smectite (Sen-

Table 2. Clay mineralogy of selected horizons of soil tested.

Sample	Horizon	Smect	Verm.	Ill.	Kao.	Mixed-layer kao-smect.	Quar.	Feld.
				Penghu iron-rich calcareous soil				
TW	A	++	+	+++	++	—	—	+
	B1	+++	+	+++	+	—	—	+
	B2	++	+	++	+++	—	—	—
AZ	A	+	—	—	+++	—	—	+
SY	A	+++	—	+++	++	—	—	—
HC	A	++	+	+++	+++	+	—	—
CM	A	+++	—	++	+++	—	—	—
				Black soil in eastern Taiwan				
YM	Ap1	++	—	+	+	+	+	+
	Ap2	++	—	+	+	+	+	+
	Ap3	++	—	+	+	+	+	+
	AB	++	—	+	+	+	+	+
	Bw	++	—	+	+	+	+	+
CB	A	+++++	—	—	—	+++	—	+
	Bss1	+++++	—	—	—	+++	—	+
	Bss2	+++++	—	—	—	++	—	+
	BCss1	+++++	—	—	—	+	+	+
	2C1	++	—	—	—	+	+	+
	2C2	+	—	—	—	—	+	+
SYS	Ap1	++++	—	++	++	—	+	+
	Ap2	++++	—	++	++	—	+	+
	2Bw	++++	—	++	++	—	+	+
	2Bg1	+++	—	++	++	—	+	+
	2Bg2	+++	—	++	++	—	+	+
	2Bg3	+++	—	++	++	—	+	+
CS	Ap	—	++	—	++	—	+	—
	A1	—	++	—	++	—	+	—
	A2	—	+++	—	+++	—	+	—
	Bw	—	+++	—	+++	—	+	—
	Bg1	—	+++	+	+++	—	+	—
	Bg2	—	+++	+	+++	—	+	—

[1] Smect.: indicates smectite; Verm.: vermiculite; Ill.: illite; Kao.: kaolinite; Quar.: quartz; Feld.: feldspar.
Symbols of —: trace or not detected; +: <50 g kg^{-1} soil; ++: 100 g kg^{-1} soil; +++: 100–200 g kg ^{1} soil; ++++: 200–300 g kg^{-1} soil; +++++: 300–400 g kg^{-1} soil.

kayi *et al.*, 1983; Laird, 1987; Laird *et al.*, 1989; Lagaly, 1992). Alkylammonium ion (C = 12) exchange (Table 3) indicated that the mean-layer charge of the smectite from the black soils [0.43–0.48 cmol(c)/$O_{10}(OH)_2$] is less than the Penghu smectites, (0.48–0.52). However, when the black soil clays are treated with hot sodium citrate solutions, a slight increase in their mean layer charge was observed in the range of 0.45–0.51 cmol(c)/$O_{10}(OH)_2$ (XRD data not shown).

XRD patterns of the CS black soil clays (Bw horizon) showed weak and broad peaks at 1.41–1.01 nm with K-saturation. However, a sharp 1.41-nm peak occurs with Mg-saturation and glycerol solvation (Figure 4A). These results may indicate that a large quantity of interlayer material produces these XRD peaks of weak intensity. The XRD diffractograms of the CS clays are similar to that of vermiculite. However, the successive hot sodium citrate treatments essentially eliminated the broad reflections at 1.41–1.01 nm and produced an intense peak at 1.01 nm with K-saturation clay and shifted the peak to 1.81 nm with Mg-saturation by glycerol solvation. The *d*(001)-value shifted to 1.01 nm with K-saturation when heated at 110°C (Figure 4B). These values suggest the presence of a high-charge smectite or an expandable vermiculite (Egashira *et al.*, 1981; Senkayi *et al.*, 1985; Badraoui and Bloom, 1990; Lagaly, 1992; Xing and Dudas, 1994). With the exception of the CS clays, re-expansion of smectite after Li-saturation, heating to 280°C, and subsequent glycerol solvation (Greene-Kelly test), indicates a predominantly iron-rich beidellite or nontronite (Wildman *et al.*, 1968). After alkylammonium (C = 12) adsorption, *d*(001)-values were 1.90–1.95 nm, and the calculated mean layer charges were 0.50 and 0.53 cmol(c)/$O_{10}(OH)_2$ with respect to A and Bw horizons of the CS pedon (Table 3).

A semi-quantitative estimation of the phases present is based on integrated peak XRD intensities. Table 2 summarizes the clay mineralogy showing that smectite, vermiculite, kaolinite, mixed-layers of smectite-kaolinite clay, and illite are dominant in the black soils. Under high precipitation, elevated temperature,

Table 3. The $d(001)$-value dodecylammonium-clay complexes and mean-layer charge of smectites.

Sample	Mean layer charge cmol(c)/ $O_{10}(OH)_2$	DDA $d(001)$
Montmorillonite (Wyoming Bentonite, Swy-1)[2]	0.27	1.50
Montmorillonite (Marnia, Algeria)[2]	0.35	1.68
Montmorillonite (Camp Berteau, Morocco)[2]	0.34	1.63
Montmorillonite (Greek White, Greece)[2]	0.31	1.60
Montmorillonite (Greek Yellow, Greece)[2]	0.31	1.52
Montmorillonite (Moosburg, Germany)[2]	0.31	1.51
Beidellite (Unterrupsroth, Germany)[3]	0.38	1.76
Vermiculite (Transvaal, South Africa)[4]	0.75	2.25
Vermiculite (Beni-Buxera, Spanish)[4]	0.63	2.05
Montmorillonite (Arizona (Cheto), SAz-1)[5]	0.39	1.70
Vermiculite (Libby, Montana)[5]	0.70	2.28
Illite (Silver Hill, Montana, IMt-I)[5]	0.72	2.31
Vermiculite (Transvaal, South Africa)[5]	0.74	2.33
Penghu iron-rich calcareous soil		
Tw pedon (A horizon)	(0.52)[1]	1.93
(B1 horizon)	(0.52)	1.93
SY pedon (A horizon)	(0.50)	1.90
CM pedon (A horizon)	(0.48)	1.86
Black soil in eastern Taiwan		
Ym Pedon (Ap3 horizon)	(0.43)	1.78
(Bw horizon)	(0.47)	1.84
CB Pedon (Bss2 horizon)	(0.48)	1.86
(2C1 horizon)	(0.48)	1.86
SYS Pedon (Ap2 horizon)	(0.47)	1.84
(2Bg1 horizon)	(0.47)	1.84
CS Pedon (A1 horizon)	(0.50)	1.90
(Bw horizon)	(0.53)	1.95

[1] Values in parenthesis are the mean layer charge per formula unit calculated from reference clays from the equation $Y = 1.7023X + 1.0435$ ($n = 13$). $R^2 = 0.9795$.
[2] Stul and Mortier (1974).
[3] Lagaly (1979).
[4] Lagaly (1982).
[5] Malla *et al.* (1993).

basic conditions and strong wet and dry cycles of the soil environments, smectite appears to transform into a smectite-kaolinite mixed-layer clay. In general, the values of CaEC are greater than KEC. The quantity of smectite and vermiculite calculated by CaEC and KEC (Alexiades and Jackson, 1965) shows a similar trend with quantities determined from peak intensities by XRD. However, the K fixation capacities of the Penghu iron-rich calcareous soil and the CS of black soil clay fractions were greater than the YM, CB, and SYS black soils. The K-fixation capacity is proportional to the vermiculite and smectite contents and layer charge (Table 5). The K fixation capacity of black soils was less than in high charge smectite of the Penghu pedons (Badraoui and Bloom, 1990; Bouabid *et al.*, 1991; Xing and Dudas, 1994).

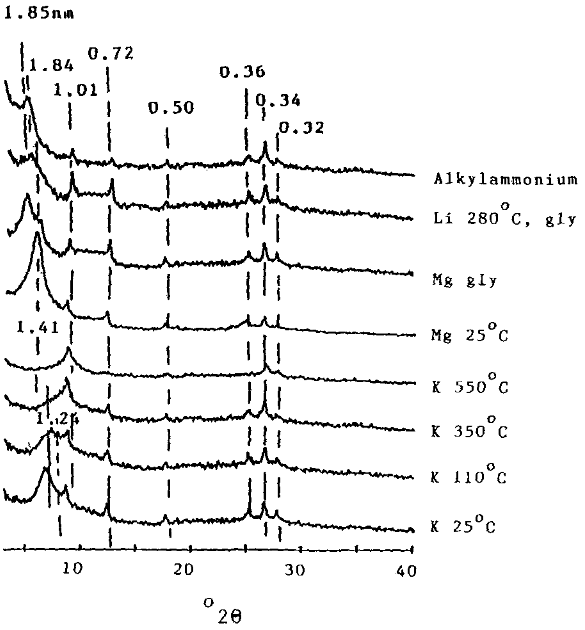

Figure 3. X-ray diffractograms of the SYS clay (Ap2 horizon) without hot sodium citrate treatments.

Smectite in the upper soil horizons is related to subsoil materials from the Penghu volcanics in colluvium. The smectite forms predominantly under wet conditions during alteration of the basaltic cobbles from colluvium. Differences in the internal drainage between the soil and the altered lithics create variations in their respective chemical environment. Weathered clasts contain different levels of micro-porosity, with fluid movement between the cobble and the soil, as well as between individual weathering domains within the cobbles. Thus, stagnant conditions favoring retention of silica and cations probably exist on a micro-scale within the altering basaltic clasts, whereas the bulk soil is leached via the wet environment (Glasmann, 1982).

Smectite genesis follows a two-stage sequence, showing an initial formation of aggregate spheroids or void walls which is followed by fibrous smectite growth (Glasmann, 1982; Keith and Staples, 1985). The formation of smectite is favored by the poor drainage of the micro-environment of the altering basaltic clasts and the soil environment. Augite phenocrysts were altered by congruent dissolution, leaving voids which were subsequently filled with smectite. Plagioclase was also altered to produce micrometer-size spheroidal aggregates of smectite. As cobbles decompose and alter the soil matrix, micro-drainage conditions may change, causing silica and bases (Ca^{2+} and Mg^{2+}) to leach, favoring the transformation of smectite to kaolinite or halloysite (Keith and Staples, 1985; Wang, 1997; Wang *et al.*, 1998). The higher-charge

Table 4. Extractable aluminum, iron and magnesium (g kg^{-1}) treated by hot sodium citrate solutions of selected black soil clays.

Sample	Horizon	Element	Extracted time (h) 1	2	4	8	16
YM	Ap3	Al	1.13	5.24	8.29	15.93	29.44
		Fe	0.08	0.19	0.64	0.89	3.62
		Mg	0.01	0.02	0.12	0.20	0.54
	Bw	Al	1.75	3.31	4.86	8.95	38.08
		Fe	0.26	0.32	0.97	1.79	4.20
		Mg	0.02	0.03	0.14	0.40	0.69
CB	Bss2	Al	3.52	5.52	8.75	12.09	27.5
		Fe	0.19	0.27	0.84	1.43	3.21
		Mg	0.01	0.03	0.05	0.24	0.85
	2C1	Al	1.91	4.22	8.31	12.09	25.92
		Fe	0.28	0.54	1.31	2.01	4.20
		Mg	0.02	0.07	0.13	0.39	0.96
SYS	Ap2	Al	2.74	3.35	10.14	19.44	32.74
		Fe	0.25	0.41	1.18	2.46	4.28
		Mg	0.05	0.10	0.24	0.67	1.56
		Al	2.46	2.74	10.74	20.44	30.53
	2Bgl	Fe	0.11	0.19	0.85	1.59	4.14
		Mg	0.03	0.04	0.17	0.71	1.45
CS	Al	Al	1.71	4.43	7.17	11.55	23.84
		Fe	0.19	0.55	0.98	1.32	3.72
		Mg	0.03	0.11	0.26	0.70	1.54
	Bw	Al	1.49	2.56	3.94	7.78	27.84
		Fe	0.11	0.31	0.76	1.32	4.67
		Mg	0.01	0.17	0.30	1.04	2.24

beidellite is believed to be developed from basalts of enriched Fe and Mg and lacking Si, thereby producing Si-deficient tetrahedral sites.

The occurrence of smectite in soil was reviewed extensively by Dudal and Eswaren (1988), Borchardt (1989), and Ahmad (1996). Smectite in soil may exist in basins, B horizons of Vertisols or poorly drained bed rock fissures, and below basic rocks. Each of these environments produces high Si and basic cations with the potential for smectite formation (Jackson, 1968). Minimum leaching in poorly drained basins is the common occurrence for smectite. The black soils in eastern Taiwan are good examples of drainage conditions required for the formation and preservation of smectite. Solutions containing large numbers Mg and Ca may contribute to the formation of a high-Mg smectite. Alternatively, montmorillonite may form through pedogenesis from solutions with high Si, Al, and Mg contents. Smectites apparently crystallize from solutions only if the pH is <6.7, in which case exchangeable Al is present (Borchardt and Hill, 1985). Montmorillonite can coexist with both beidellite and environments lacking mica and chlorite (Borchardt, 1989). However, lower-charge montmorillonite is thought to be inherited from parent materials (Bouabid *et al.*, 1996). The Vertisols or Mollisols occur on many parent materials. Weathering intensity of soil environments in eastern Taiwan is higher than for Penghu island, hence kaolinite-smectite, kaolinite, and smectite occur in the eastern black soils.

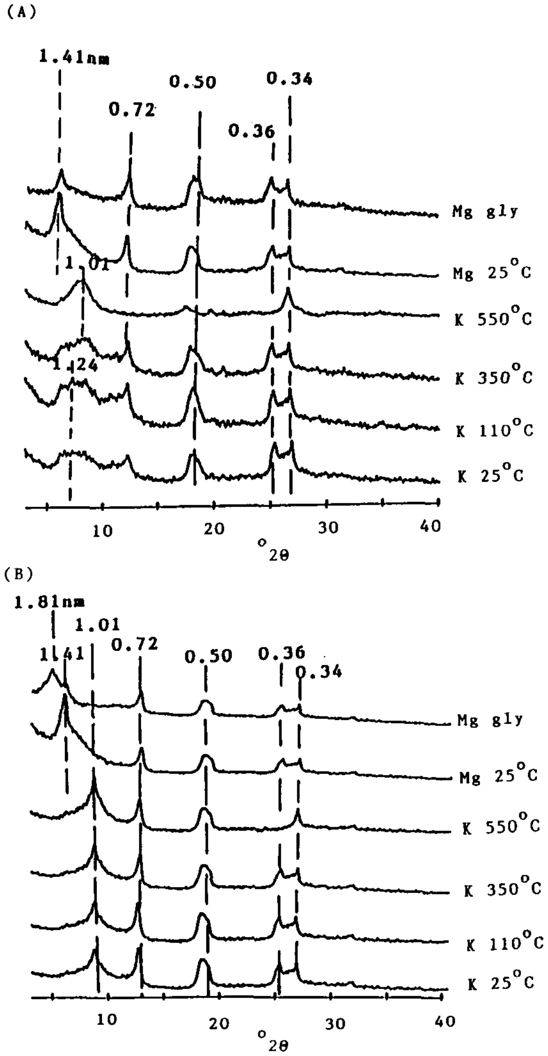

Figure 4. X-ray diffractograms of the CS clay (Bw pedon) (A) without hot sodium citrate treatments and (B) with 16 h of hot sodium citrate treatments.

CONCLUSIONS

Beidellite and/or nontronite is the most common weathering product of basalts in the Penghu island group. Beidellite or nontronite is metastable with respect to kaolinite when the soil environment is subject to more active leaching conditions. A higher-charge smectite, with the tetrahedral character of beidellite, was identified by XRD analysis using the Greene-Kelly test. However, in eastern Taiwan, in Vertisols/Mollisols and poorly drainage bed rock fissures and basic rocks, solutions with basic (*e.g.*, Ca and Mg) cations are necessary for smectite formation. The alkylammonium (C = 12) method indicated that the mean-

Table 5. KEC and CaEC values, K-fixation capacity of clay fractions, and smectite and vermiculite contents.

Sample	Horizon	KEC	CaEC	K-fixation	Smect.	Verm.	Smect.	Verm.
		cmol kg^{-1}		(%)	(%) (in clay)		g kg^{-1} (in soil)	
			Penghu, iron-rich calcareous soil					
TW	A	82	112	27	19	20	74	78
	B1	82	150	45	35	44	141	178
	B2	88	136	35	28	31	117	129
AZ	A	64	73	12	4	6	15	23
HC	A	71	105	32	25	22	100	88
			Black soil in eastern Taiwan					
YM	Ap1	64	75	15	28	7	64	16
	Ap2	65	73	11	28	5	70	13
	Ap3	68	77	12	38	6	129	20
	AB	78	90	13	32	8	97	24
	Bw	68	83	18	36	10	94	26
CB	A	72	86	16	54	9	308	51
	Bss1	71	85	17	57	9	341	54
	Bss2	79	83	5	21	3	120	17
	BCss1	85	97	12	69	8	338	39
	2C1	95	103	8	73	5	121	8
	2C2	98	112	13	81	9	73	8
SYS	Ap1	72	83	13	52	7	231	31
	Ap2	73	98	26	55	16	274	80
	2Bw	77	95	19	49	12	202	55
	2Bg1	71	78	9	20	5	72	18
	2Bg2	71	85	17	38	9	145	34
	2Bg3	76	86	12	46	7	161	25
CS	Ap	29	53	45	6	16	14	38
	A1	27	54	50	6	18	15	44
	A2	29	57	49	5	18	17	60
	Bw	20	56	64	4	23	15	89
	Bg1	20	57	65	4	24	16	97
	Bg2	20	57	65	3	24	14	111

layer charge of black soil smectites was less than the Penghu smectite. The Penghu iron-rich calcareous soil and CS pedon of black soil clay fractions possesses a K-fixation capacity.

Higher-charge smectite was developed from basalts enriched in Fe and Mg elements and lacking in Si. Thus, tetrahedral substitutions to form a high-charge beidellite occur in the Penghu iron-rich calcareous red soil. In contrast, a lower-charge smectite is also present in the black soils and this is thought to be inherited from the parent materials. Under high precipitation, high temperature, high Ca and Mg contents, and every half year wet and dry cycles of soil environments, smectite was transformed to smectite-kaolinite interstratified and kaolinite clays. The smectites produced are lower-charge smectite.

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