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SMECTITE IN MARINE QUICK-CLAYS OF JAPAN

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Abstract—Clayey sediments of marine origin having sensitivities (ratio of undisturbed to remolded shear strength) as high as 1000 at a reduced salt concentration are found around Ariake Bay, Kyushu, Japan. The clay fraction of the sediments contains smectite as a principal mineral (33–42%) in contrast to the illitic composition of most previously described quick-clays. However, this smectite is a low-swelling type, and the Na-form expands only to about the same extent as the Ca-form and gives a sediment volume almost equal to that of the Ca-clay. This low-swelling smectite appears to consist of packets of unit layers even after saturation with Na, and thus reacts like the kaolinite or illite of conventional quick-clays.

Key Words—Japan, Marine, Quick-clay, Smectite, Swelling.

INTRODUCTION

"Quick-clay" was first found in Scandinavia, and similar soils are also found in several other countries (Rosenqvist, 1953; Bjerrum, 1954; Torrance, 1975). The most characteristic property of such clays and soils is a high ratio of undisturbed strength to remolded strength, referred to as the clay's "sensitivity." Quick-clays are invariably of marine origin and are subjected to leaching by fresh water due to land elevation, resulting in a lower concentration of salt in the pore water. With a reduced salt content, the shear strength of the sediment is markedly decreased by remolding, leading to high sensitivities. Mineralogically, quick-clays consist predominantly of illite (hydrous mica), quartz, feldspars, and chlorite (Rosenqvist, 1953; Bjerrum, 1954; Berry and Jørgensen, 1971; Torrance, 1974, 1975) and are characterized by a low "activity," defined as the ratio of the plasticity index to the amount of <2-μm size particles in the sediment (Bjerrum, 1954). Small quantities of vermiculite and montmorillonite have also been reported in the clay fractions of Canadian marine clays (Brydon and Patry, 1961; Soderman and Quigley, 1965), but according to Torrance (1975), the addition of as little as 4% of pure montmorillonite to a quick-clay of Norway decreased the liquid behavior of this soil upon remolding. Recently, Smalley *et al.* (1980) reported that some volcanic ash soils from the North Island of New Zealand have sensitivities of as high as 140, due apparently to the presence of spherical halloysite which inherently has a low activity. Little has been published on the mineralogy of the quick-clays in Japan. Sasaki (1974) found a thin bed of very sensitive clay resembling varved clay in the Kutcharo volcanic ash in Hokkaido, in the northern part of Japan. Here, halloysite is the principal clay mineral and is accompanied by a small amount of kaolinite.

Clayey sediments 15 to 42 m thick are widely distributed around Ariake Bay in Kyushu, Japan. The sediments are derived from recent marine mud of Ariake Bay and are drained artificially or naturally. They have extremely high sensitivities at reduced salt concentration (>50, in places near 1000), classifying them as quick-clays (Rosenqvist, 1953). However, mineralogical analysis indicates that their clay fractions contain considerable smectite, contrary to the mineralogical composition of quick-clays of Scandinavia and eastern Canada. Previously (Egashira and Ohtsubo, 1981), the authors reported abnormal values for the liquid limit of a recent marine mud of Ariake Bay which contained smectite as the main mineral; the liquid limit of the sample equilibrated with 0.01 N NaCl was lower than that of the sample equilibrated with 0.01 N $CaCl_2$ and increased with increasing NaCl concentration. Based on sediment-volume and viscosity measurements, they proposed the presence of a low-swelling smectite, different from the normal, high-swelling smectite. Therefore, the smectite in the extremely sensitive sediments studied in the present paper may be a low-swelling type.

MATERIALS AND METHODS

Profile samples of clayey sediments were taken using a thin-walled sampler at three different sites (Yamaashi, Higashi-shiroishi and Ariake-kantaku) of Shiroishi-machi, Saga Prefecture. The samples were stored in the laboratory in a field-moist condition as 10-cm pieces. Another sample (W-179-3) containing high-swelling smectite, from the sub-soil layer of paddy soil at the Saga Agricultural Experiment Station (Nanri, Kawazoe-machi, Saga Prefecture), was studied for comparison (Egashira and Ohtsubo, 1981).

Field-moist samples were treated with 7% H_2O_2 to remove organic matter and were dispersed at pH 10 (by the addition of NaOH) by shaking. The <2-μm clay fractions were collected by repeated sedimentation and siphoning. The <2-μm clays were washed 4 times with 1 N NaCl or $CaCl_2$ followed by dialyzing or washing with water to remove excess salt. The clays then were shaken for 5 hr to ensure their ultimate dispersion.

Table 1. Physical and geotechnical properties of the samples used in this study.

Locality	Sample	Depth (m)	Content of <2-μm clay (%)	Salt[1] concentration in the pore water (g/dm³)	Water content (%)	Consistency[3] limits (%)			Activity	Shear strength[4] (kg/cm²)		Sensitivity (c/c$_r$)
						LL	PL	PI		c	c$_r$	
Yama-ashi elevation, 0.9 m; distance from the seashore, 9 km	Y-2	2.6–2.7	38.8	0.25	79	72	35	37	0.95	0.153	0.0032	48
	Y-6	6.5–6.6	34.0	0.85	88	61	30	31	0.91	0.251	0.0020	125
	Y-8	8.5–8.6	22.3	0.77	96	68	40	28	1.26	0.132	0.0024	55
	Y-13	13.6–13.7	39.6	0.11	93	63	33	30	0.76	0.409	0.0010	410
	Y-15	15.2–15.3	39.6	0.10	87	54	31	23	0.58	0.261	0.0004	650
Higashi-shiroishi elevation, 2.2 m; distance from the seashore, 6 km	S-3	3.3–3.4	56.0	4.7	157	149	53	96	1.71	0.127	0.0043	30
	S-6	6.6–6.7	47.0	8.0	127	114	51	63	1.34	0.142	0.0076	19
Ariake-kantaku elevation, 0 m; distance from the seashore, 2 km	A-8	8.5–8.6	45.0	5.4	126	105	42	63	1.40	0.098	0.0045	22
				3.4[2]	125	89	41	48	1.07	0.090	0.0025	36
				0.23[2]	130	78	45	33	0.73	0.097	0.0001	970
	A-12	12.4–12.5	26.0	3.1	68	46	34	12	0.46	0.138	0.0060	23
				0.58[2]	58	39	30	9	0.35	0.128	0.0010	130
				0.23[2]	58	37	29	8	0.31	0.149	0.0002	750
Saga Agricultural Experiment Station	W-179-3	0.20–0.65	40.5		51	76	28	48	1.19			

[1] Salt was represented by NaCl after measuring Cl$^-$ concentration.

[2] Salt concentration was decreased by leaching the undisturbed sample with deionized water in a oedometer ring under the load equivalent to its overburden pressure.

[3] LL = liquid limit; PL = plastic limit; PI = plastic index.

[4] The shear strength was measured using unconfined compression test for the undisturbed samples of the Yama-ashi and Higashi-shiroishi samples and using vane shear test for the other samples. c = undisturbed shear strength; c$_r$ = remolded shear strength.

The surface area of Na-saturated clays which were previously freeze-dried was measured with ethylene glycol-monoethyl ether (Eltantawy and Arnold, 1973; Egashira *et al.*, 1977). The cation exchange capacity (CEC) was measured according to the method of Wada and Harada (1969) by saturating with Ca and extracted with K. The sediment volume was measured by transferring duplicate dispersed clay suspensions containing 100 mg clay into 25-cm³ cylinders. An aliquot of NaCl or $CaCl_2$ solution was added to flocculate each clay suspension which then was diluted to about 22 cm³ with water. The measuring cylinders sealed with a rubber stopper were shaken vigorously for 15 sec followed by a final dilution of the suspensions to 25 cm³. The final concentration of NaCl and $CaCl_2$ was 0.04 N and 1.0 N. The cylinders were allowed to stand at 25°C. The sediment volume was read visually every 24 hr until it became constant. The relative deviation of the measurement was less than 5%.

Specimens for X-ray powder diffraction (XRD) were prepared by taking duplicate clay suspensions (without $Na_2S_2O_4$-Na-citrate-$NaHCO_3$ treatment) containing 50 mg clay. One was washed with 1 N $MgCl_2$ and the other with 1 N KCl. Excess salt was removed by washing with water. One cubic centimeter of water was added, and an aliquot of the suspension containing 30 mg of clay was dropped onto a glass slide (28 × 48 mm), air-dried, and X-rayed. The K-saturated specimen was heated at 300°C and 550°C, and the Mg-saturated specimen was solvated with glycerol. Filtered CuKα radiation from a Rigaku diffractometer was used for XRD analysis.

RESULTS AND DISCUSSION

As shown in Table 1, the samples from Yama-ashi are extremely sensitive and are classified as very or extra quick-clays (Rosenqvist, 1953). They have a low salt concentration compared to the samples from Higashi-shiroishi and Ariake-kantaku, suggesting that the reduced salt concentration in the pore water is responsible for the development of the quick-clays, similar to those in Scandinavia and eastern Canada (Rosenqvist, 1953; Bjerrum, 1954; Torrance, 1975). However, the activity of the Yama-ashi sample is considerably higher than that of Norwegian marine clays (including quick-clays) having equivalent salt concentrations (Bjerrum, 1954). The samples from Higashi-shiroishi and Ariake-kantaku also are classified as medium quick-clays, even

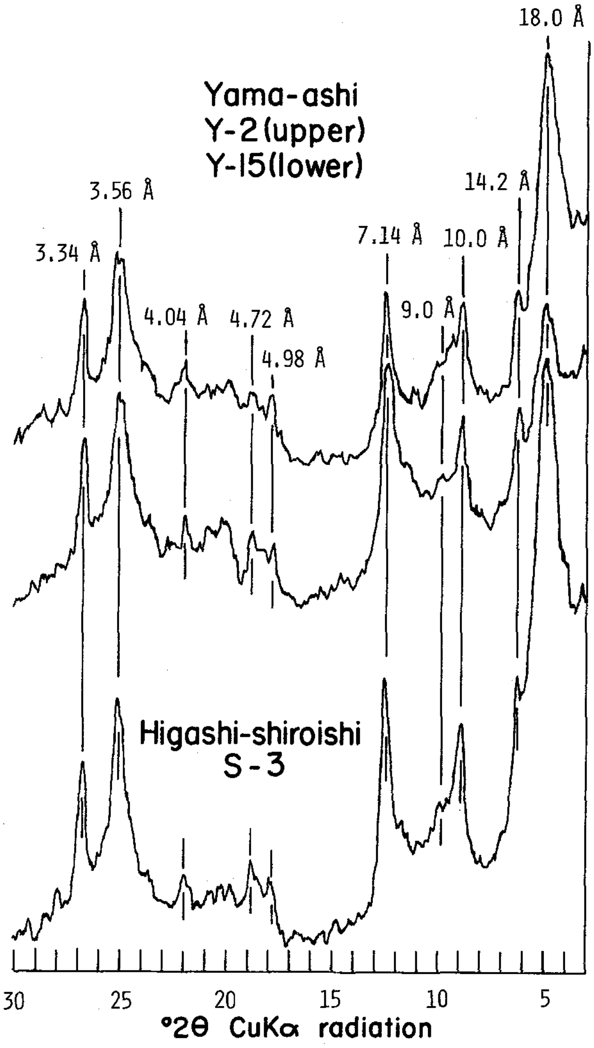

Figure 1. Selected X-ray diffraction patterns of the Mg-saturated and glycerol-solvated <2-μm fractions from Yama-ashi and Higashi-shiroishi.

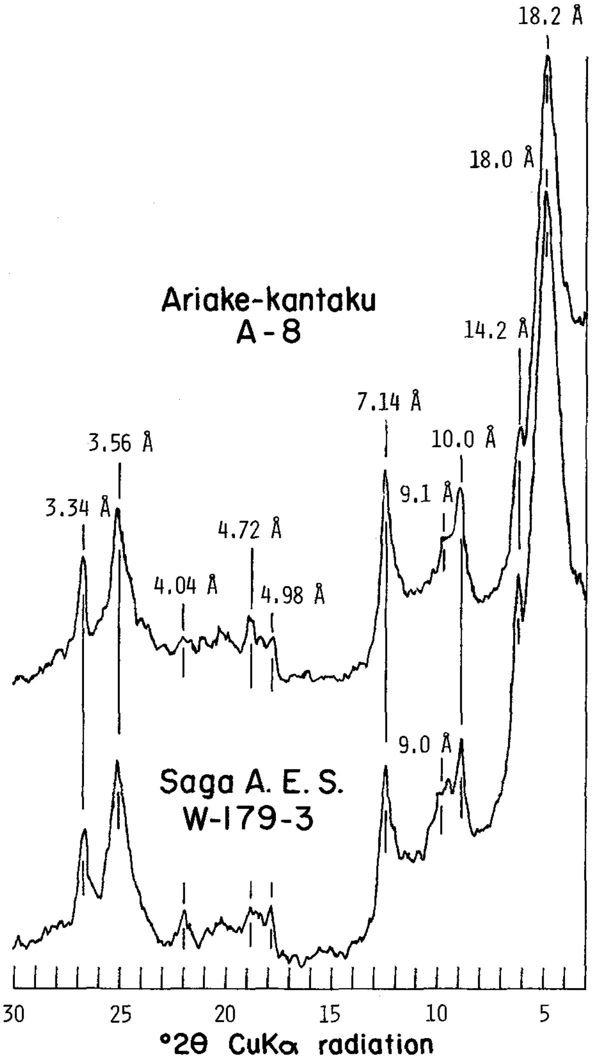

Figure 2. Selected X-ray diffraction patterns of the Mg-saturated and glycerol-solvated <2-μm fractions from Ariake-kantaku and Saga Agricultural Experiment Station.

with their relatively high salt concentrations in the natural condition. The sensitivity of the samples from Ariake-kantaku increased markedly to near 1000 by leaching out the original salt. The increased sensitivity in the reduced salt concentration is due to the decrease in the remolded shear strength while the unremolded shear strength is fixed. The reduction in salt concentration is also accompanied by a decrease in the liquid limit and activity, similar to quick-clays of other countries (Rosenqvist, 1953; Bjerrum, 1954; Torrance, 1975). Thus, the sediment samples used in this study are indeed quick-clays and have characteristics similar to those of quick-clays in Scandinavia and eastern Canada.

Figures 1–4 show the XRD patterns of the <2-μm clay fractions of the Yama-ashi, Higashi-shiroishi, Ariake-kantaku, and W-179-3 samples. Only the patterns of the Mg-saturated and glycerol-solvated specimen (Figures 1 and 2) and of the K-saturated and air-dried specimen (Figures 3 and 4) are reproduced here. The strong peak at 18.0–18.2 Å in the Mg-saturated and glycerol-solvated specimen and at 12.6–13.0 Å in the K-saturated and air-dried specimen reveals clearly that all of the clay fractions contain considerable smectite. Other clay minerals present are kaolinite, mica, and vermiculite; chlorite was detected only in the Yama-ashi sample. The silt and fine-sand fractions contain quartz, feldspars, and cristobalite.

Table 2 gives the specific surface area and CEC of the <2-μm clay fractions. The specific surface area is greater than 400 m^2/g, suggesting a predominance of smectite (Egashira, 1981). The charge density also is in

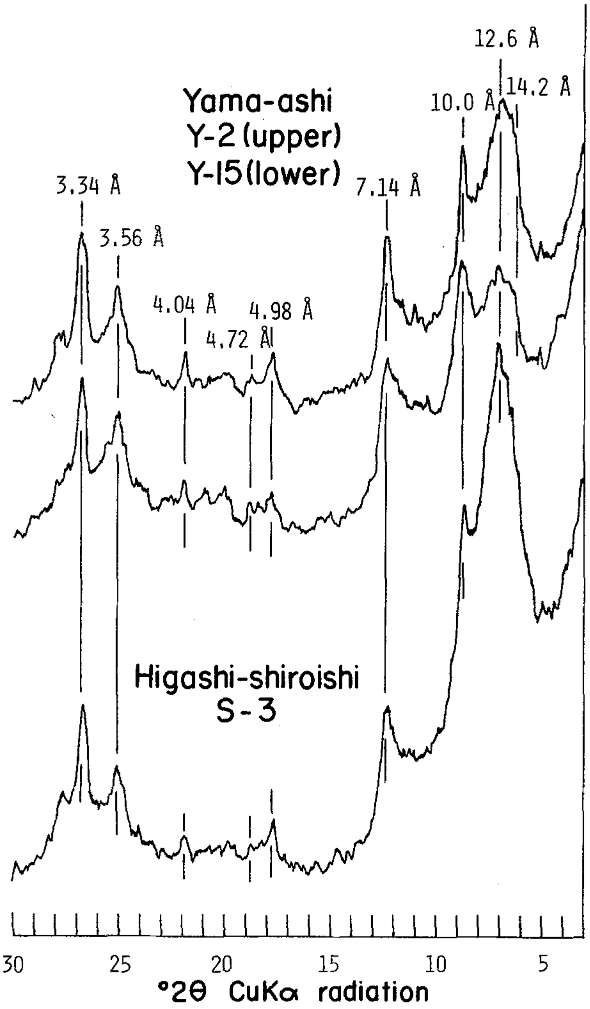

Figure 3. Selected X-ray diffraction patterns of the K-saturated and air-dried <2-μm fractions from Yama-ashi and Higashi-shiroishi.

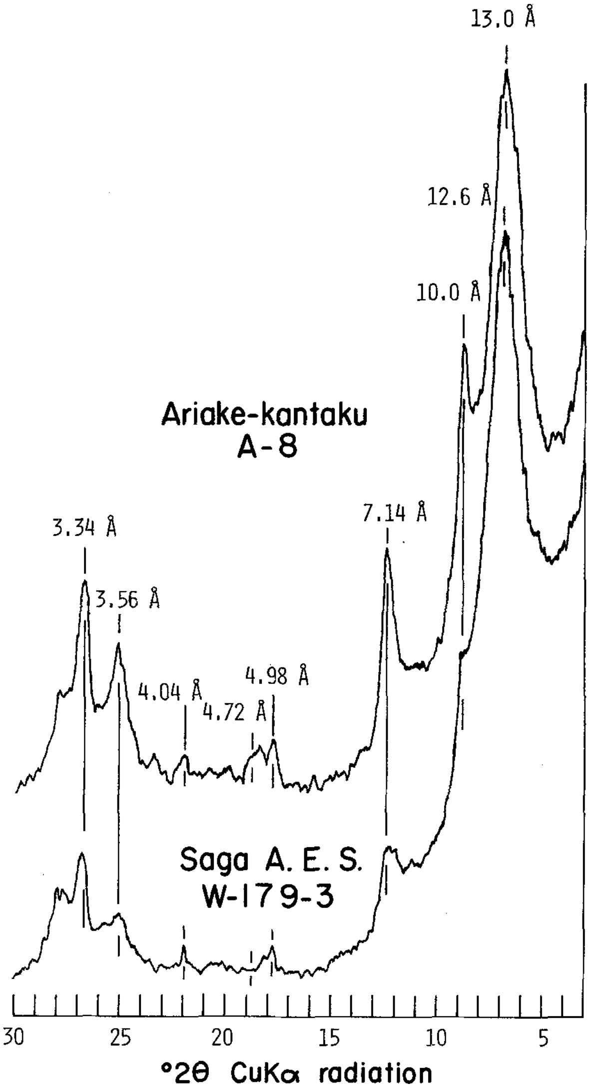

Figure 4. Selected X-ray diffraction patterns of the K-saturated and air-dried <2-μm fractions from Ariake-kantaku and Saga Agricultural Experiment Station.

the range of smectite. The smectite content of the clay fractions was estimated from their specific surface areas. A value of 800 m^2/g was assumed as the specific surface area of smectite and a value of 200 m^2/g as the average specific surface area of the other minerals. The smectite content of each sample was calculated by $800x + 200(100 - x) = 100S$, where x is the smectite content (%) and S is the specific surface area (m^2/g), and is listed in Table 2. The value of 200 m^2/g for non-smectite clays may be too low; however, this value was chosen for the following reason. One of the authors (Egashira, 1981) separated <2-μm clay fractions from about 30 soil samples that differ in their clay mineral composition and measured their specific surface areas by the same method as that described above. The values measured for these soil clays ranged from 200 to 500 m^2/g. The lowest value for soil clays devoid of smectite was 200 m^2/g.

The smectite content in the clay fraction of the samples from Yama-ashi ranged from 42% to 33% and decreased with depth, consistent with the decrease in relative intensity of the smectite peak with depth in the XRD patterns. The samples from Higashi-shiroishi and Ariake-kantaku exhibit smectite contents of 42% and 38–39%, respectively. The smectite content of sample W-179-3, which was employed for comparison, is 39%, almost equal to that of the samples from Higashi-shiroishi, Ariake-kantaku, and the upper two samples from Yama-ashi, irrespective of the higher peak intensity of smectite. Based on these surface area, CEC, and

Table 2. Specific surface area, cation exchange capacity, charge density, and estimates of smectite content of the <2-μm clay fractions.

Locality	Sample	Specific surface area (m²/g)	Cation exchange capacity (meq/100 g)	Charge density (μe/m²)	Smectite content (%)
Yama-ashi	Y-2	450	51.2	1.14	42
	Y-6	437	47.9	1.10	40
	Y-8	423	45.2	1.07	37
	Y-13	402	44.7	1.11	34
	Y-15	396	44.5	1.12	33
Higashi-shiroishi	S-3	451	56.4	1.25	42
	S-6	453	50.5	1.11	42
Ariake-kantaku	A-8	431	53.8	1.25	39
	A-12	425	55.4	1.30	38
Saga Agricultural Experiment Station	W-179-3	432	52.8	1.22	39

Table 3. Sediment volume of the <2-μm clay fractions.

		Sediment volume (cm³/100 mg)			
		at 0.04 N salt concentration		at 1.0 N salt concentration	
Locality	Sample	Na-clay	Ca-clay	Na-clay	Ca-clay
Yama-ashi	Y-2	3.08	2.97	3.06	2.81
	Y-6	2.63	2.69	2.82	2.62
	Y-8	2.49	2.50	2.69	2.46
	Y-13	2.43	2.37	2.40	2.31
	Y-15	2.41	2.37	2.48	2.32
Higashi-shiroishi	S-3	2.56	2.93	3.20	2.97
	S-6	2.50	2.66	n.d.[1]	n.d.[1]
Ariake-kantaku	A-8	2.32	2.78	3.07	2.97
	A-12	1.61[2]	2.77	2.69	2.78
Saga Agricultural Experiment Station	W-179-3	4.10	2.58	4.09	2.49

[1] n.d. = not determined.

[2] Flocculation rate was very slow, and the supernatant liquid was slightly turbid even after the end of standing.

XRD estimates of mineral content, the clay fraction of the sediment samples used in this study appears to contain smectite as the main mineral.

As stated above, the smectite in these extremely sensitive sediments may be a low-swelling type. Table 3 lists the sediment volume of clay fractions measured after saturation with Na and Ca. The sediment volume of Na-clays of the samples from Yama-ashi, Higashi-shiroishi, and Ariake-kantaku is almost equal to or lower than that of the Ca-clays from these same localities at 0.04 N and 1.0 N salt concentrations. On the other hand, the sediment volume of sample W-179-3 is clearly greater for the Na-clay than for the Ca-clay at both salt concentrations.

The smectite of sample W-179-3 is a high-swelling clay, the property characteristic of montmorillonite (Egashira and Ohtsubo, 1981). The difference in sediment volume of the Na- and Ca-clays from sample W-179-3 is probably due to the difference in the dominant interparticle force between Na-smectite and Ca-smectite in the dispersed suspension. The dominant interparticle force of Na-smectite is repulsion arising from osmotic activity of the diffuse double-layer around the unit layers. As a result, Na-smectite readily swells and separates into unit layers. The separation into unit layers is still preserved in the flocculated suspension, resulting in a higher sediment volume. On the other hand, Ca-smectite consists of packets of unit layers separated by 9 Å within the packet even in the dispersed suspension, because the interparticle attraction through the Ca ion dominates over the double-layer repulsion. The formation of packets results in a lower sediment volume of Ca-smectite.

As shown in Table 3, the sediment volume of the Na-clays is almost equal to or less than that of the Ca-clays for the samples from Yama-ashi, Higashi-shiroishi, and Ariake-kantaku. This tendency is clearly different from that observed for sample W-179-3 but essentially the same as that of the sample containing low-swelling smectite reported by Egashira and Ohtsubo (1981). This low-swelling smectite consists of packets of unit layers even after saturation with Na, and like Ca-smectite, the dominant force of the Na-smectite is interparticle attraction. In this sense, this smectite is a low-swelling clay, similar to kaolinite and illite. However, the reason why this smectite exhibits such low-swelling characteristics has not been clear until now. Because both Na- and Ca-smectite consist of packets of unit layers, the sediment volume of Na-saturated clays containing low-swelling smectite is low and almost equal to that of Ca-saturated clays. The lower sediment volume of Na-clays compared to that of Ca-clays at a 0.04 N salt concentration suggests that particles of Na-clays take more of a face-to-face arrangement under the condition of slow flocculation.

Rosenqvist (1962) showed that an artificially sedimented and consolidated swelling smectite clay greatly increases in sensitivity when the sediment is percolated by diluted potassium solutions. However, this replacement of Na by K does not seem to explain the high sensitivity of the sediments containing low-swelling smectite, because the K-substituted smectite of Rosenqvist is a high-swelling clay and probably regains its original high swelling after saturation with Na.

In conclusion, the high sensitivity of the clayey sediments of marine origin which contain smectite as the main mineral is due to the fact that the smectite is a low-swelling type, having swelling properties not unlike those of illite, the principal clay mineral in most quick-clays.

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Резюме—Глиняные осадки морского происхождения, имеющие чувствительности (отношение несмущенной и смущенной прочностей при срезе) такие большие как 1000 при уменьшенной концентрации соли, находятся в районе бухты Ариаке, Кыусху, Япония. Глинистая фракция осадков содержит смектит как основной минерал (33–42%) в противоположность к иллитовому составу большинства ранее описанных зыбучих глин. Однако, этот смектит является мало-набухающего типа, и Na-форма расширяется только до примерно такого же самого уровня как Ca-форма и приводит к почти равному объему осадков как и Ca-глина. Кажется, что этот мало-набухающий смектит состоит из пакетов элементарных слоев даже после насыщения натрием, и тогда он реагирует как каолинит либо иллит обычных зыбучих глин. [E.C.]

Résumé—Des sédiments argileux d'origine marine ayant des sensitivités (proportion de la force non dérangée à la force remoulée) atteignant 1000 à une concentration de sel réduite sont trouvés autour de la Baie Ariake, Kyushu, Japon. La fraction argile des sédiments contient de la smectite comme minéral principal (33–42%) contrairement à la composition illitique de la plupart des argiles rapides décrits précédemment. Cette smectite est cependant du type à bas gonflement, et la forme-Na s'épand seulement approximativement autant que la forme-Ca et donne un volume de sédiment presqu'égal à celui de l'argile-Ca. Cette smectite à bas gonflement semble consister de paquets de couches unitaires même après avoir été saturée de Na, et par conséquent elle réagit comme la kaolinite ou l'illite d'argiles rapides conventionnels. [D.J.]

Resümee—Tonige Sedimente mariner Entstehung, die bei reduzierter Salzkonzentration Sensitivitäten (Verhältnis der ungestörten zur Scherfestigkeit der durchkneteten Probe) von etwa 1000 haben, wurden um die Ariake Bay, Kyushu, Japan, gefunden. Die Tonfraktion der Sedimente enthält Smektit als häufigstes Mineral (33–42%) im Gegensatz zur illitischen Zusammensetzung der meisten früher beschriebenen Quicktone. Dieser Smektit quillt jedoch nur wenig, und die Na-Form quillt etwa nur so viel wie die Ca-Form und ergibt ein Sedimentvolumen, das dem von Ca-Tonen entspricht. Dieser schlechtquellende Smektit scheint aus Paketen von Einzellagen zu bestehen, sogar nach der Sättigung mit Na, und verhält sich daher wie der Kaolinit oder Illit von herkömmlichen Quicktonen. [U.W.]