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Smectite in weathered hyaloclastite of the Shakotan Peninsula, Hokkaido, Japan: significance of clay mineralogy in engineering geology

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ABSTRACT

The occurrence and chemical composition of smectite in hyaloclastite from the Toyohama rock fall site of 10 February 1996 and the Oshoro Bay area were examined by using optical microscope, X-ray diffractometer, electron probe microanalyser, and analytical transmission electron microscope. Frequently, smectite occurs as fillings and linings of pores in various forms, and sporadically as the replacement of volcanic glass in hyaloclastite. The analytical results show that smectite is of dioctahedral and Fe-rich type. Its compositional field varies extensively between montmorillonite and nontronite. Moreover, interlayer cations of smectite vary intensively in relation with the type of weathering in hyaloclastite. Na-smectite occurs characteristically in discoloured hyaloclastite affected by superficial weathering, whereas Ca- and K-smectite occurs dominantly in the hyaloclastite collected from the inner part of the rock mass. These features of smectite in hyaloclastite can be attributed to its formational and weathering environments. Mineralogical characterisation of hyaloclastite provides detailed information on engineering properties of this type of rock mass and rock materials.

INTRODUCTION

Submarine volcaniclastic rocks of late Tertiary, including hyaloclastite, are widely distributed in southwestern Hokkaido, Japan (Fig. 1). In this area along the coast, occasional accidents occurred due to rock mass collapses, especially from the cliff formed by submarine volcaniclastic rocks of hyaloclastite. In particular, a large-scale rock mass collapse on 10 February 1996 at the Toyohama Tunnel was a serious one. It is located at the eastern coast of the Shakotan Peninsula, southwestern Hokkaido. The collapsed rock mass was 70 m high, 50 m wide, and 11,000 m^3 in volume, which crushed the entrance of a tunnel and killed 20 persons (The Committee of Inspection on the Toyohama Tunnel Disaster 1996). After this disaster, many people in its related fields are showing a great interest in the stability of rock mass slope.

The stability of rock slope is controlled by the rock mass property in addition to the environmental conditions of the slope, such as geology, topography, meteorology, and hydrology. In general, the rock mass properties are intensively affected by rock alteration due to weathering and/or hydrothermal processes as well as the rock feature present in the original rock type. Thus, the changes in composition and texture of rock mass during the rock alteration can have a great influence on its geotechnical properties. Therefore, in order to understand the rock property closely related with the rock slope stability, mineralogical characterisation of rock alteration is indispensable.

In this study, the mineralogical characterisation of hyaloclastite was carried out by using optical microscope, electron probe microanalyser (EPMA), analytical transmission electron microscope (ATEM), and X-ray diffractometer (XRD). This paper describes the occurrence and mineralogical feature of Fe-rich smectite, which frequently occurs in the hyaloclastite.

HYALOCLASTITE SAMPLES

Submarine volcaniclastic formations of middle Miocene to Pleistocene of Tertiary age in the Shakotan Peninsula, are composed mainly of hyaloclastites formed by quench fragmentation in water, subaqueous pyroclastic flow deposits, and their reworked sediments (Yamagishi 1981). In addition, the submarine volcaniclastic rocks characterised by hyaloclastites are widely distributed in the coastal part of the Shakotan Peninsula, as shown in Fig. 1. Hyaloclastite samples were collected mainly from two coastal sites. These are the rock mass collapsed sites of the Toyohama Tunnel (Fig. 2) and Oshoro Bay.

The rock mass around the Toyohama rock fall site is composed of massive and stratified andesitic hyaloclastite layers (Plate 1A) of middle to late Miocene age (The Committee of Inspection on the Toyohama Tunnel Disaster 1996). The hyaloclastite frequently comprises coarse andesite/dacite blocks in fine-grained matrix (Plate 1B), while its lithofacies varies extensively even within one outcrop, e.g., from tuff to volcanic breccia. The hyaloclastite can be

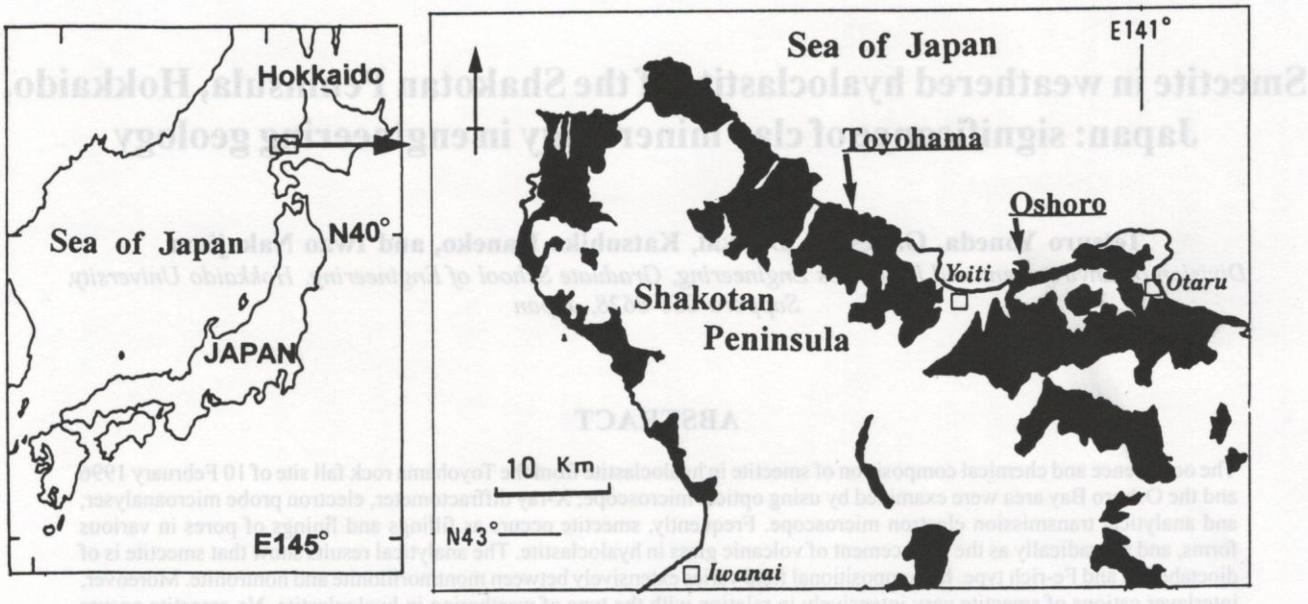

Fig. 1: Location and distribution map of the formations (black area) with hyaloclastite of middle Miocene to Pleistocene at the Shakotan Peninsula in Hokkaido, Japan. Geological data are compiled from Hasegawa and Osanai (1977) and Yamagishi (1981)

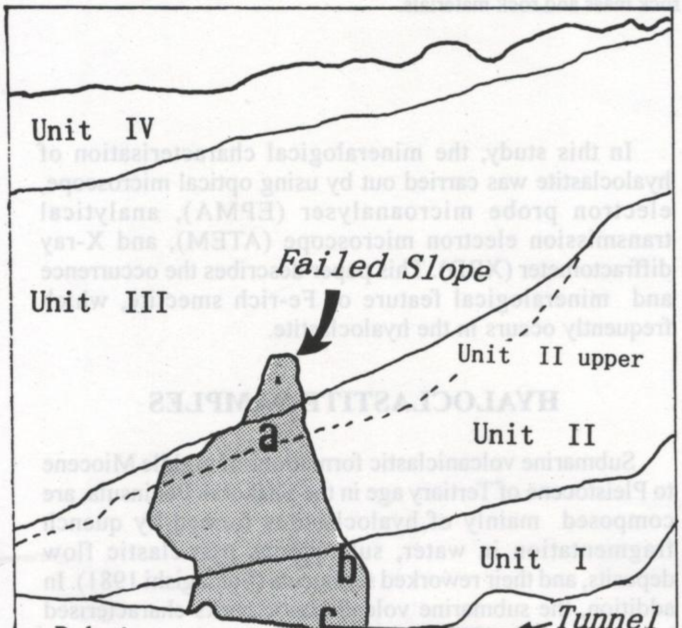

Fig. 2: Hyaloclastite formations (Unit I-IV) constituting a steep cliff at the Toyohama rock fall site of 10 February 1996. Map is simplified after the committee of inspection on the Toyohama Tunnel disaster (1996). The letters a, b, and c, which are marked on the failed slope, correspond to the photographs of A, C, and B of Fig. 3, respectively.

classified as inhomogeneous soft rock. In general, volcanic blocks of hyaloclastite are hard and apparently fresh, but the matrix is loose and slightly discoloured. The samples of hyaloclastite can be classified into two types, viz. the superficial samples of discoloured hyaloclastite affected by superficial weathering, and the inner fresh (or slightly discoloured) samples of hyaloclastite collected from the inner side of rock mass Plate 1C).

The samples of hyaloclastite and pillow lava were collected from the coast of Oshoro Bay. The cliff along the coast is composed of andesitic hyaloclastite and pillow lava of late Miocene age. The appearance of hyaloclastite from Oshoro Bay is similar to that of the Toyohama hyaloclastite, except the fine-grained and intensively discoloured matrix. Moreover, the pillow lavas are characterised by intensively discoloured networks developed at the periphery and boundary of lava block (Plate 1D).

LABORATORY ANALYSIS

Impregnated thin sections with blue resin and polished thin sections of the rock samples were observed under the optical microscope. The polished thin sections were also prepared for EPMA (JXA-8800/8900) analysis in order to quantitatively determine the chemical composition of altered minerals (especially clay minerals). The analytical condition and quantitative correction method of EPMA are described by Yoneda (1991). In addition, textural analysis of chemical mapping and phase analysis were performed for several rock samples to understand chemical changes of mineral assemblage due to rock weathering.

XRD (JDX3500) analysis was carried out for the rock powders as well as the clay fraction (>2 μm in size) separated from the rock powder. All rock powder and some clay specimens were prepared as randomly oriented powders mounted on Al-holder. They were radiated with Cu Kα to determine mineral composition of rock and structural type of clay mineral. Meanwhile, some selected specimens of clay fraction were prepared as oriented powders mounting on a glass slide. The oriented powders were untreated, saturated with ethylene glycol, Mg-ion and K-ion, and also glycolated after Mg saturation. After these treatments Cu Kα radiations and measurements of diffraction patterns for the oriented specimens were carried out. In addition, ATEM (H-700 and EMAX2200) analysis for some representative specimens of clay fraction was carried out in order to chemically and morphologically characterise each individual particle. Nontronite (NG-1) from Hohen Hagen, Germany, was also analysed to compare with the smectite particle in the representative specimens of the hyaloclastite.

Mineral composition of hyaloclastite

Ca-plagioclase, pyroxene, and volcanic glass including cryptocrystalline material are major constituents of the Toyohama as well as Oshoro hyaloclastite. Under the microscope, plagioclase and pyroxene occur both as phenocryst and groundmass in the volcanic block of hyaloclastite, and also in its matrix. Minor constituents such as Fe-Ti oxides and hydrous Fe-Mn oxide minerals are also commonly observed.

In addition to this, smectite is characteristically observed as an alteration product in the hyaloclastite. XRD and microscopic observations reveal that the matrix part of hyaloclastite is richer in smectite than its volcanic block. Similarly, the existence of smectite in all hyaloclastite samples indicates its widespread distribution in the hyaloclastites of this area. Mineralogical features of smectite as a typical expansive clay mineral is also the main interest of this paper, which is described in the following paragraphs.

Smectite under the optical microscope

Under the optical microscope, smectite widely occurs as a light yellowish-grey or brownish-grey aggregate of fine-grained particles in volcanic blocks as well as matrix of the hyaloclastite.

The characteristic feature of smectite is that it occurs as pore infillings in various forms occasionally with colloform banding (Plate 2A), and as linings of rounded degassed pores in the groundmass of volcanic blocks. In addition, the occurrence of smectite in the matrix of hyaloclastite as interface fillings of fine-grained materials and crustings of grain surface is also characteristic (Plate 2B). Moreover,

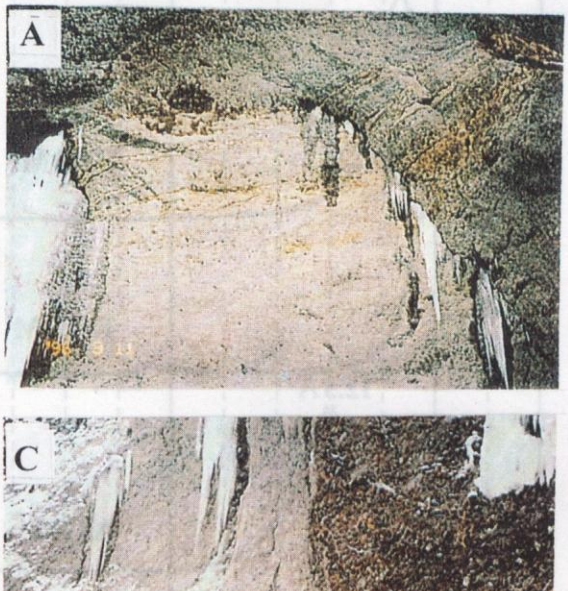

Plate 1: Photographs of hyaloclastite of Toyohama and Oshoro. A: hyaloclastite (Unit II) at the upper part of failed slope at Toyohama, B: Toyohama hyaloclastite (Unit I) composed of volcanic blocks and matrix, C: fresh rock surface observed on the slope failure (left), and rock slope discoloured during superficial weathering (right) at Toyohama, D: Oshoro pillow lava with discoloured networks.

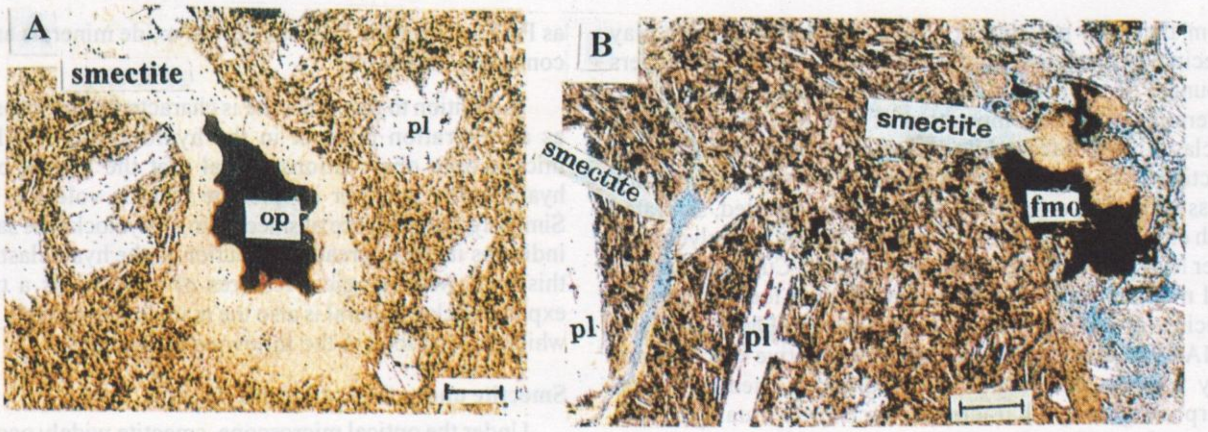

Plate 2: Optical microphotographs of hyaloclastite. A: smectite showing colloform banding and opaque mineral infilling pore in groundmass (open polar), B: vein-form smectite in volcanic block, and pore-filling smectite and Fe-Mn oxide mineral in the matrix of hyaloclastite. Bar scale: 100 μm, op: opaque mineral, pl: plagioclase, fmo: Fe-Mn oxide mineral

smectite occasionally occurs as replacement product of volcanic glass, especially in the discoloured matrix and peripheral parts of the Oshoro samples as well as in the superficial samples of the Toyohama hyaloclastite.

These microscopic observations of the hyaloclastite samples suggest that smectite was formed as a secondary alteration product, and the major occurrences of smectite as the infillings in pores can be attributed to the deposition of smectite in aqueous solution.

XRD Analysis of smectite

Smectite was detected in all samples by XRD analyses. The XRD patterns of the oriented and treated specimens of the clay fractions of a representative hyaloclastite are shown in Fig. 3. The 15Å-reflection of untreated specimen changes into 17Å-, 12.5–12.8Å-, and 18Å-reflection by ethylene glycolation, K^+-saturation, and glycolation after Mg-saturation, respectively (Fig. 3). It is indicated that the clay fractions of the hyaloclastite sample are usually composed of smectite only. In addition, XRD results of hyaloclastite samples exhibit that the matrix part is richer in smectite than the block part of the sample.

The d-spacing of 060 reflection of smectite mixed with standard silicon powders were measured. The d-spacings of smectite from the Toyohama and Oshoro hyaloclastite samples were measured as 1.508 and 1.507Å, respectively. These d(060) values lie in between the values of montmorillonite and nontronite, i.e. 1.492–1.504Å and 1.521Å (Moore and Reynolds 1997) respectively, but are close to the former. These observations suggest that smectite is of dioctahedral type.

Quantitative chemical analysis of smectite

Quantitative EPMA analysis of smectite was carried out for the hyaloclastite samples. The representative

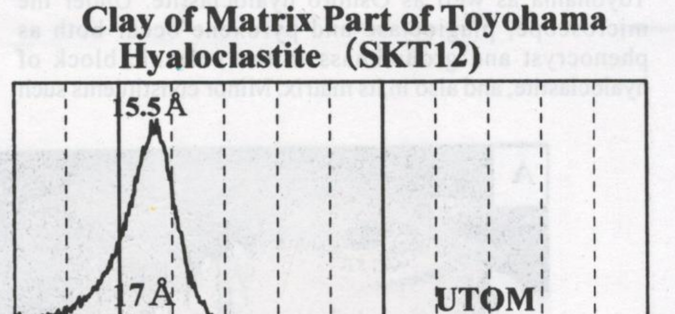

Fig. 3: XRD patterns of clay specimens from the Toyohama hyaloclastite.

analytical results are listed in Table 1 together with the structural formulae, which were calculated on the basis of $(O_{10}(OH)_2)$. Total iron is expressed as Fe_2O_3. The analytical results show that smectite is of Fe-rich type with the extensively variable content of Fe.

Mole ratios of SiO_2-Al_2O_3-Fe_2O_3 of smectite by EPMA analysis were plotted in a ternary diagram (Fig. 4A). Montmorillonite, beidellite, and nontronite of ideal chemical compositions were also plotted in the diagram. Smectite of the hyaloclastite shows a high SiO_2 ratio. The compositional variation in Fe_2O_3 is inversely correlated with Al_2O_3. These plots indicate that the analysed smectite has Fe-Al composition in between that of ideal montmorillonite and nontronite. Regarding the tetrahedral composition of smectite, in many cases, the number of Si ion in the tetrahedral sheet of analytical data fall within a range of 4.0–3.9 (Table 1). It suggests that the tetrahedral composition of smectite is montmorillonitic. However, some smectites of the Oshoro hyaloclastite samples show the tetrahedral composition close to that of nontronite.

ATEM analysis was also done for the clay particles of hyaloclastite samples (Fig. 5). Quantitative chemical composition of the clay particles was calculated by using the thin-foil correction method. Mole ratios of SiO_2-Al_2O_3-Fe_2O_3 of clay particles were plotted in a ternary diagram (Fig. 4B). The compositional variation after ATEM analysis for the clay particles is nearly concordant with that after EPMA analysis for clay aggregate, while the former shows more extensive variation. In addition, the clay particles are richer in Al_2O_3 composition than nontronite of standard sample (NG-1). The ATEM analysis suggests that Al_2O_3-Fe_2O_3 composition of the clay particles ranges within a composition field of montmorillonite and nontronite.

Table 1: EPMA analysis of smectite. Total Fe is expressed as Fe_2O_3. Structural formula is based on $O_{10}(OH)_2$ (22 Oxygen)

Wt. %	Toyohama			Oshoro	
	1	2	3	4	5
SiO_2	53.06	53.28	57.04	54.52	41.40
TiO_2	0.66	0.03	0.94	0.78	3.38
Al_2O_3	8.46	2.99	14.93	7.75	5.57
Fe_2O_3	20.07	29.47	11.31	20.37	28.91
MnO	0.01	0.00	0.01	0.03	0.09
MgO	3.29	2.36	3.28	3.97	1.28
CaO	1.58	1.53	0.94	1.32	1.22
Na_2O	0.03	0.23	0.44	0.62	0.24
K_2O	1.60	0.23	0.61	1.62	0.62
Sum	88.76	90.11	88.36	90.98	82.71
Si	3.83	3.89	3.89	3.85	3.39
Ti	0.04	0.00	0.05	0.04	0.21
Al	0.72	0.26	1.20	0.64	0.54
$Fe^{(3+)}$	1.09	1.62	0.58	1.08	1.78
Mn	0.00	0.00	0.00	0.00	0.01
Mg	0.35	0.26	0.33	0.42	0.16
Ca	0.12	0.12	0.07	0.10	0.11
Na	0.00	0.03	0.06	0.08	0.04
K	0.15	0.02	0.05	0.15	0.06
Sum	6.30	6.20	6.23	6.36	6.29

1: pore-linings in volcanic block, 2: lenticular aggregate in ground mass, 3: matrix fillings of tuff, 4: pore-linings in volcanic block, 5: replacing of matrix glass.

Moreover, quantitative analytical data by EPMA shows that the interlayer cations of smectite extensively vary with the type of hyaloclastite sample as shown in Fig. 6. Usually, Na-smectite occurs in discoloured hyaloclastite, which is affected by superficial weathering, whereas Ca- and K-smectite occur dominantly in hyaloclastite collected from the inner part of the rock mass. While taking their microscopic occurrences into consideration, this difference in occurrence of smectites in hyaloclastite may be attributed to their formational environments and/or weathering conditions.

Chemical mapping of smectite

As mentioned earlier, the secondary formation of smectite characterises micro-texture of hyaloclastite, which would have a close relationship with the engineering properties of the rock mass and rock materials. Chemical mapping and

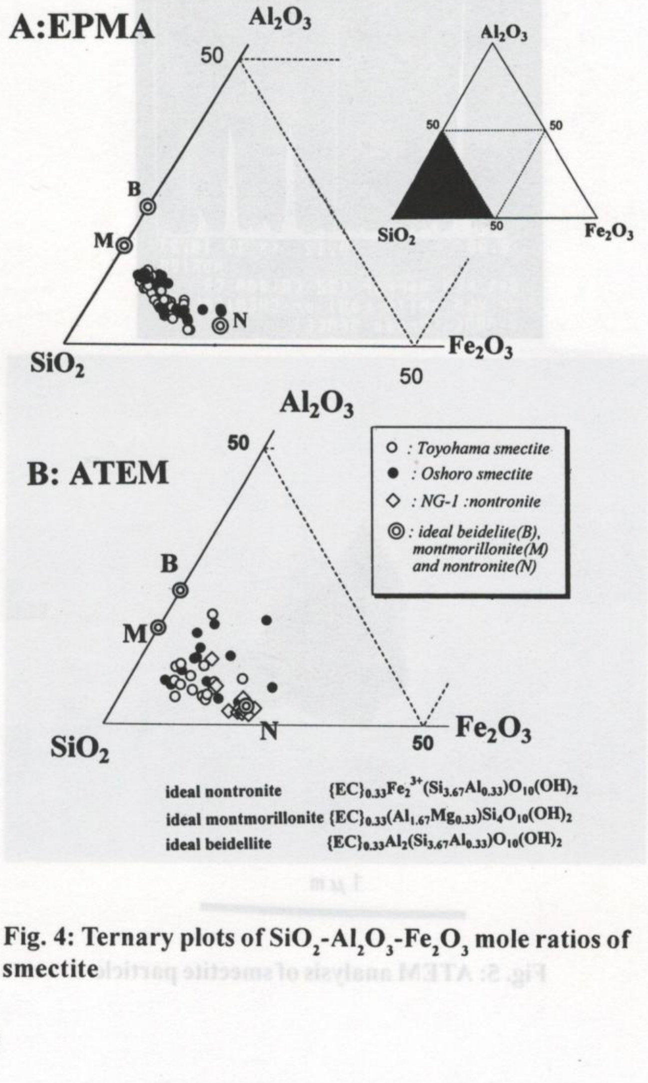

Fig. 4: Ternary plots of SiO_2-Al_2O_3-Fe_2O_3 mole ratios of smectite

phase analysis of the micro-texture including smectite was performed for many hyaloclastite samples by using EPMA.

A secondary electron image and chemical maps calculated from the characteristic X-ray intensities of major elements for a matrix part of a hyaloclastite sample are shown in Plate 3. The analytical part of the hyaloclastite samples free from superficial weathering exhibits a typical micro-texture. It is shown that smectite is richer in Fe and Mg than coexisting minerals such as columnar/acicular plagioclase, pyroxene, and volcanic glass. Besides, it is clearly indicated that smectite occurs as interspace-fillings or crustings of grains constituting the matrix.

The digital image analysis can provide useful information regarding quantity and morphology of individual constituents including the pores. In this case, microscopic distribution of smectite in the hyaloclastite can be characterised by 'network veinlets' and/or 'crusting'. Furthermore, the mode of smectite in the analysed area

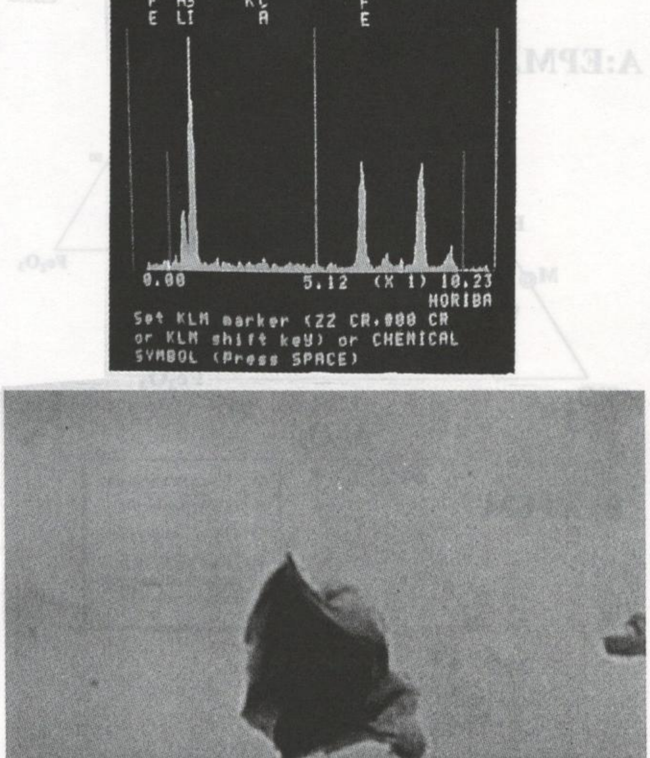

Fig. 5: ATEM analysis of smectite particle

(Plate 3) can be measured as ca. 12%. It is inferred that the microscopic distribution pattern of smectite as well as pores in hyaloclastite can have a great influence on the geotechnical properties, such as extension of micro-fracture and reduction of strength in rock mass during weathering.

DISCUSSIONS

The mineralogical features of smectite present in the hyaloclastite of this area represent a close relationship with the origin of smectite and the alteration process after its formation. Two different conditions for smectite formation in the hyaloclastite of this area are: by terrestrial weathering and in the submarine environment.

The occurrence of discoloured superficial part of hyaloclastite can be attributed to recent terrestrial weathering. In addition, the occurrence of local discoloured parts in the inner side of the hyaloclastite found at the Toyohama rock mass collapsed site, is also attributable to terrestrial weathering. Based on this viewpoint, it is obvious that smectite had formed as a terrestrial weathering product due to chemical interaction of surface and groundwater with the hyaloclastite. In this case, the macroscopic distribution of smectite in the hyaloclastite rock mass would be controlled by topographic, meteorological, and hydrological factors during the weathering process.

On the other hand, the formation of smectite in the hyaloclastite can be related to submarine processes such as hydrous alteration of volcanic glass in volcanic rocks (paragonitisation), submarine weathering (halmyrolysis), and hydrothermal alteration/precipitation associated with submarine volcanic activity (Chamley 1989; Hiller 1995). In

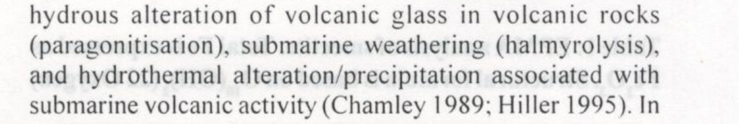

Fig. 6: Ternary plots of Ca-Na-K of smectite

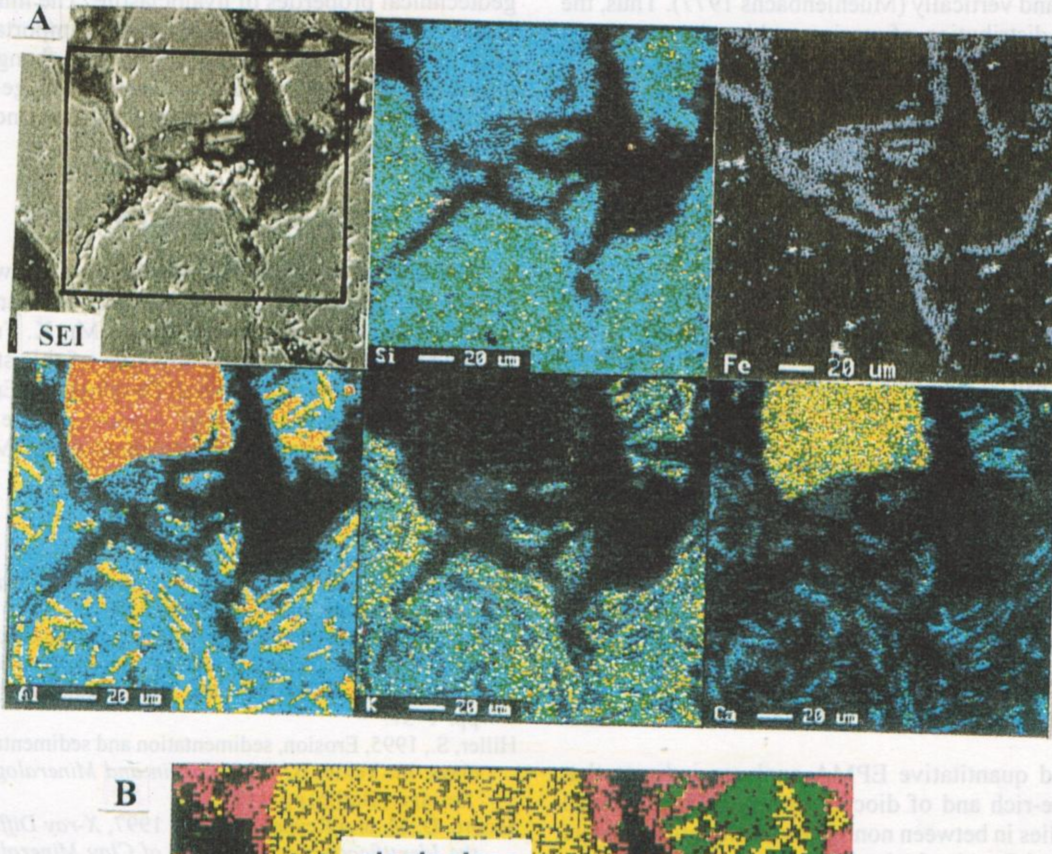

Plate 3: Mapping of characteristic X-ray images (A) and phase map (B) of a Toyohama hyaloclastite. Smectite characteristically shows network and crusting patterns. Smectite (red) and volcanic glass (green) in phase map look like the same in monochrome. See text for details. SEI: secondary electron image

these cases, the processes of forming the authigenic smectite would have a large-scale effect on submarine volcanics both horizontally and vertically (Muehlenbachs 1977). Thus, the macroscopic distribution of marine authigenic smectite in hyaloclastite should be wider and deeper than that of smectite formed in terrestrial weathering environment as mentioned above.

Taking into account the widespread distribution of smectite in hyaloclastites of this area, smectite might be considered to be marine authigenic origin, i.e., the products in the geological period through the processes such as solution and deposition, and replacement of volcanic glass in submarine environment after volcanic activity. However, mineralogical feature of smectite could have been influenced by recent terrestrial weathering related to surface water as well as groundwater.

As mentioned, the formational, diagenetic and/or weathering environments of smectite in the hyaloclastite can control its macroscopic/microscopic occurrence and detailed mineralogical properties. Such features of smectite have significant effects on the geotechnical properties like swelling, slacking, permeability, fracturing, and reduction of strength for this type of rock mass and rock materials.

CONCLUSIONS

XRD and quantitative EPMA analyses indicate that smectite is Fe-rich and of dioctahedral type. Its chemical composition lies in between nontronite and montmorillonite with octahedral Fe-Al substitution. Moreover, EPMA data show that the interlayer cations of smectite extensively vary with the type of hyaloclastite sample. Usually, Na-smectite occurs in discoloured hyaloclastite, which is affected by superficial weathering, whereas Ca- and K- smectite occur dominantly in hyaloclastite collected from the inner part of the rock mass. This difference in occurrence of smectite in the hyaloclastite may be attributed to its formational environments and/or weathering conditions. Mineralogical properties of smectite, such as structural type, chemistry, and interlayer cation-type, can have direct influence on the geotechnical properties of hyaloclastite. The mineralogical characterisation of hyaloclastite plays an important role not only in understanding the compositional changes in rock alteration process, but also in estimating geotechnical properties of rock mass and rock materials under present weathering environment.

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