Walter E. Parham

HALLOYSITE-RICH TROPICAL WEATHERING PRODUCTS OF HONG KONG

REPRINTED FROM THE
PROCEEDINGS OF THE INTERNATIONAL CLAY CONFERENCE
TOKYO, 1969, VOL. I

REPRINT SERIES
OF THE
MINNESOTA GEO! OCICAL SURVEY
UNIVERSITY OF MINNESOTA

ISRAEL UNIVERSITIES PRESS Jerusalem, 1969

HALLOYSITE-RICH TROPICAL WEATHERING PRODUCTS OF HONG KONG

WALTER E. PARHAM

Minnesota Geological Survey, and Department of Geology and Geophysics, University of Minnesota, Minneapolis, Minnesota

ABSTRACT

Rock weathering products formed under humid tropical conditions in Hong Kong typically are rich in the clay mineral halloysite. The parent rock types studied consist primarily of granite and rhyolite porphyry, and commonly weather to depths in excess of 100 feet. Feldspars change first to a material that probably corresponds in composition to allophane. This weathering product occurs as small, tapered projections and as flame-shaped sheets that project upward from the feldspar surfaces. Tubular halloysite develops from the flame-shaped materials as weathering progresses. It is suggested that small pseudo-hexagonal kaolinite flakes slough off the outer surface of halloysite tubes and act as seed crystals for the site of further kaolinite growth.

Comparison of the modern rock weathering products of Hong Kong with the much older ones formed under similar conditions in Minnesota, U.S.A., suggest that the tapered projections and flame-shaped sheet material is unstable and with time is converted to other mineral phases. The minor amounts of halloysite present in the older weathering products of Minnesota suggest also that halloysite is converted to platy kaolinite in relatively short periods of geologic time.

INTRODUCTION

Chemical weathering of igneous and metamorphic rocks under tropical conditions took place in Minnesota, U.S.A., during the latter part of the Mesozoic era, developing a kaolinitic residuum up to 100 feet in thickness. The clay minerals of the residuum consist mostly of kaolinite with minor amounts of halloysite. Halloysite seems to have formed directly from the weathering of potassium feldspars, and some is preseved on partly altered feldspar cleavage surfaces toward the base of the weathered zone. Higher in the weathering profile, halloysite is sparse or absent, whereas platy

kaolinite is abundant. The relationship of the formation of halloysite and/or kaolinite to parent rocks, climate and other environmental conditions of past geologic times is not clear, and may have become more obscure with time. (Data relating to Minnesota in Clay Mineralogy and Geology of Minnesota's Kaolin Clays, W. E. Parham, Minnesota Geological Survey Special Publication, in preparation).

Modern weathering products forming in Hong Kong were selected for study for comparison with those of Minnesota in an attempt to clarify the factors relating to formation of halloysite and kaolinite. The weathered rocks in Hong Kong were selected for study for the following reasons: (a) their similarity to those weathered in Minnesota during Mesozoic times; (b) the weathering products in Hong Kong were formed under conditions of good drainage in a humid tropical environment, as were those in Minnesota; (c) weathering profiles showing the effects of deep chemical weathering in rocks in Hong Kong are numerous, and the transition from fresh parent rock to its weathered equivalent is well exposed whereas this is not the case in Minnesota; and (d) certain samples of chemically analyzed, modern, tropical weathering products from Hong Kong (Brock, 1943), preserved in the files of the Rock Analysis Laboratory (R.A.L.), Department of Geology and Geophysics, University of Minnesota, were available at the beginning of this study for preliminary mineralogic study.

A part of each chemically analyzed sample of Brock filed with the R.A.L. has been re-examined with X-ray diffraction and the electron microscope along with the samples collected by the author in Hong Kong during October-November, 1967. Brock's sample numbers (1943) are listed in Table 1. The letter "R" has been omitted from these samples on Fig. 1. Descriptions of all samples are listed in Tables 1 and 2.

The British Crown Colony of Hong Kong occupies an area of almost 400 square miles, consists of many mountainous islands and peninsulas with irregular shorelines, and has maximum altitudes of slightly over 3,000 feet above sea level. It lies within the northern limits of the tropics and has an average annual rainfall of about 85 inches. Tree cover is sparse and most of the hillsides are covered only by grass and low scrub.

The majority of bedrock in Hong Kong falls into two general categories, granite and rhyolite porphyry. The rocks are part of batholith that trends ENE from Hong Kong across southeastern China. Parts of all rock types in Hong Kong show the effect of deep chemical weathering, and the range in pH values of many of the Colony's soils is generally between 4.5 and 5.5 (Davis, 1952; Grant, 1960). Climatic conditions throughout southeastern China are similar to those in Hong Kong, and it is inferred therefore that weathering products forming in Hong Kong characterize those forming in large areas of southeastern China. Stratigraphic names from Hong Kong used in this paper are taken from those established by Ruxton (1959).

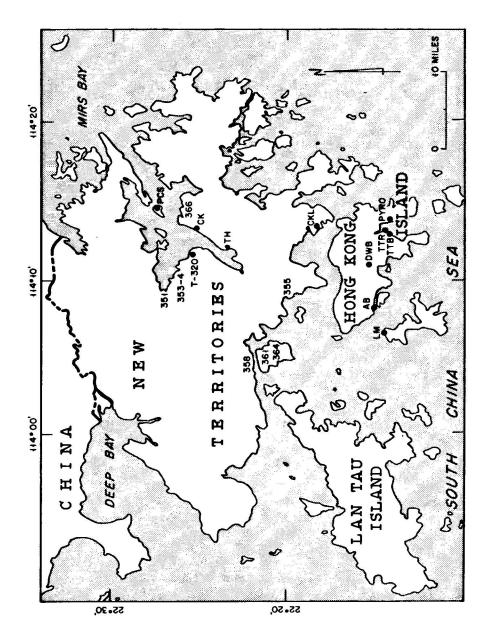


FIG. 1. Map showing sample localities of weathered rocks in Hong Kong

TABLE 1. Mineralogy of weathered rocks collected by R. W. Brock

Minn. F Analy Lab. N	sis (Brock, 1943)	Mineralogy of the $< 2\mu$ fraction*
	Plover Cove Formation	
R-369	weathered rhyolite porphyry; Tai Po, (1-X)**	mus., hal., kaol. ?***
R-359	decomposed rhyolite porphyry, 1-foot from fresh rock; Ting Kau, (6-A)	hal.; amorph. mat.; tr. amt. ill., qtz.
R-354	rhyolite porphyry decomposed brown clay; Tai Po, (7-A)	kaol., irreg. outline; tr. amt. qtz., verm., mont.
	Hong Kong Granite	
R-355	granite, partly weathered; Sham Shui Po, (4-A)	hal., gibb., mus., K-feld; tr. amt. qtz., verm. or mont.?
R-356	granite, highly weathered; near R-355,	hal.; tr. amt. gibb., qtz., verm. or mont.?
R-357	granite, completely decomposed; Gin Drinkers Bay, (4-C)	hal.; tr. amt. ill., verm.
R-367	granite, weathered to clay, granitic texture preserved, (4-D)	hal.; tr. amt. gibb.?
R-368	same as R-367 except granitic texture is absent, (4-E)	hal.
R-366	granite porphyry, weathered, texture preserved; Shap Pat Heung, (4-F)	hal.; tr. amt. ill.
R-364	porphyritic granite?, partly decom- posed, texture preserved, Tsing I Island, (5-A)	hal.; tr. amt. ill., qtz., k-feld.
R-365	same rock as R-364 except complete. decomposed to brown earth, (5-B)	hal.; tr. amt. ill., qtz.
a -	Other Rock Units	
R-351	quartz monzonite, partly weathered; Tai Po Market, (10-A)	hal.; ill. ?, irreg. outline; tr. amt. qtz.
R-352	more completely weathered than R-351, (10-B)	hal.; ill.

R-353	lateritic clay from quartz monzonite porphyry; Lam Mian valley near Tai Po, (10-C)	kaol., irreg. outline; hal.; tr. amt. qtz.
R-361	lamprophyre, partly weathered; Tsing I Island, (12-A)	no regular morphology of minerals, diffuse x-ray peaks at 12.3Å, 8.24Å, and 3.51Å; tr. amt. ill.; diffuse peak after glycol treatment at 14.7Å.
R-362	lamprophyre, completely decomposed, (12-B)	hal; kaol., irreg. outline; tr. amt. qtz.
	tained by W. E. Parham	

^{**} Brock (1943) sample number

verm. = vermiculite ill. = illite hal. = halloysite

kaol. = kaolinite chlor. = chlorite

feld. = feldsparmont. = montmorillonite amorph. mat. = amorphousmaterial

gibb. = gibbsite mus. = muscovite qtz. = quartz

tr. amt. = trace amount

TABLE 2. Mineralogy of weathering products from Hong Kong collected by Parham

Sample number		Mineralogy of the $<2\mu$ fraction*		
1. La	Plover Cove Formation			
TTB-1	Rhyolite porphyry, fresh, pale greenish- white, 3 ft. above road level, cut on NE side of NW-SE road along Tai Tam Bay between Notting Hill and Rep Hill.	Replica shows large smooth areas on fresh rock surface, some flame-shaped films rising from rock surface, small patches on surface show alteration to hal tubes.		
TTB-2	2 feet above TTB-1, rock less glassy in appearance.	Replica mostly shows start of growth of flame-shaped films rising from rock sur- face, lesser amount of hal. tubes forming.		
TTB-3	2 feet above TTB-2, bleaching of rock notice- able, rock slightly punky.	hal.; a few flakes with irreg. outline; replica shows start of growth of hal. tubes and numerous flame-shaped films rising from rock surface.		
TTB-4	2 feet above TTB-3, mostly white and softer than TTB-3.	hal., numerous packets of tubes; ill.?; tr. amt. K-feld., qtz. ?; replica shows start of growth of hal. tubes and flame-shaped films rising from rock surface.		
TTB-5	3 feet above TTB-4, white, soft.	hal.; tr. amt. qtz., K-feld., ill.?		
ТТВ-6	3 feet above TTB-5, white, very soft.	hal; tr. amt. qtz., gibb., K-feld.		

^{***} Most abundant mineral listed first.

TABLE 2 contd.

TIR-3	Rhyolite porphyry, westhered, light buff to white, soft, $3\frac{1}{2}$ ft. above fresh rock; sample taken from roadcut at S. end of Tai Tam Reservoir on E. side of Tai Tam Road.	ill.; hal.; tr. amt. K-feld.
TTR-4	1-foot above TTR-3, rock weathered to yellow clay.	kaol., thin plates, lack hexagonal outline; hal.; tr. amt. verm., ill.
PYRO-1	Highly weathered pyroclastics, soft, clay rich, rose colored; from 25 ft. bluff on E. side of Shek O Road, S. 35°W. from Dragon Back	hal., some spherical masses of tubes; tr. amt. ill ?
AB-3	Rhyolite porphyry, highly weathered to soft, buff colored clay, taken at base of W. end of large roadcut on N. side of Shek Pai Wan Road, N. of W. end of Ap Lei Chau and W. of Aberdeen.	hal.; hexagonal kaol.; tr. amt. qtz., K-feld.; two strong unidentified x-ray peaks at 11.6Å and 3.45Å.
AB-4	Completely weathered rock, altered to very soft clay, at W. end of exposure.	ill., hal., a few bundles of tubes present; tr. amt. hexagonal kaol.

Hong Kong Granite and associated rocks		
DWB-1	Syenite dikes in Hong Kong granite, surface of feldspars bleached white, exposure on NW side of Deep Water Bay Road about 1/4 mi. W. of Wong Nai Chung Gap.	Replica shows formation of thin, flame- shaped films rising from feld. surface along cleavage plane edges; small tapered projections of varied length developed over much of surface.
DWB-2	Weathered granite, white clay, pink K-feld- spars and quartz, soft; 100 yds. SW of DWB-1.	hal., some bundles of tubes; ill.; tr. amt. hexagonal kaol.
DWB-3	K-feldspar (orthoclase) fragments, pale pink, color dulled by weathering; 100 yds. SW of DWB-2.	Replica shows formation of thin, flame- shaped films and tapered projections rising from feld. surface; some hal. tubes scat- tered over surface; tr. amt. hexagonal kaol.
T-320-3	Weathered K-feldspar crystals from vug in weathered granite; exposed in artificially cut terrace at altitude of 320 ft., above Ma Liu Shui.	hal., a few bundles of tubes; ill.
T-320-5	Weathered granite.	hexagonal kaol. and kaol. aggregates; hal.; tr. amt. qtz.

TABLE 2 c	ontd.	
TH-1	4 feet wide, weathered basic dike (basalt?) in granite; green-blue; Turrett Hill Quarry on E. side of Tide Cove.	mont.; hal.
CK-1	Highly weathered granite?, no original texture preserved, very clay rich, white; exposed in barrow pit above orphanage at Cheung Kang, on E. side of Tide Cove.	hal., some in packs of tubes showing tend- ency of splitting between adjacent tubes; ill.; tr. amt. hexagonal kaol.
LM-2	Highly weathered granite from outer zone of boulder, core of boulder fresh; sample from base of cliff at N. end of Lamma Is.	hal., some spherical masses of tubes; tr. amt. hexagonal kaol.
LM-7	Decomposed granite, near LM-2	hal.
LM-8	Granite, highly weathered, stained to shades of rose-purple; along path from N. end of Lamma Is. to ferry landing at Yung Shu Wan.	hal., some spherical mass of tubes.
CKL-3	Syenite, white, weathered, lower pit of Cha Kwo Ling kaolin mine; N. of and across Hong Kong Harbor from Skau Ki Wan.	hal., some 4 micron diameter masses of tubes; tr. amt. hexagonal kaol., qtz., K-feld., plag. feld.
CKL-4	Syenite, pink, more highly weathered than CKL-3.	hal., tr. amt. K-feld.
CKL-6	Clay, white, occurs as 1/4 in. thick slicken- sided bands in weathered syenite.	hal., some 3 micron diameter spherical masses of tubes, some bundles of hal. tubes; tr. amt. K-feld., plag. feld.
CKL-7	Slickensided white clay bands in weathered granite, upper pit.	hal., hexagonal kaol.
CKL-8	Granite, weathered.	hal., some bundles of tubes; tr. amt. K-feld.
CKL-9	Weathered K-feldspar crystals in pegmatite in granite.	hal., some bundles of tubes; K-feld.
CKL-12	Sedimentary clay, white, soft; in lower pit.	hal., some bundles of tubes; hexagonal kaol.; tr. amt. gibb.
	Buff Head Format	ion
T-320-6A	Maroon siltstone, partly weathered; exposed in N. part of terrace at T-320 locality, Ma Liu Shui.	ill.; tr. amt. hal., gibb?
T-320-6D	Siltstone, buff colored, more weathered than T-320-6A.	hal.; ill.

TABLE 2 contd.

PCS-1

Siltstone, alternating beds of buff and ill.; kaol.; tr. amt. hal. maroon color, partly weathered, exposure at NE end of Pak Sha Tau Chau at south spillway of Plover Cove Reservoir.

PROCEDURE

The weathered rock samples were examined by X-ray diffraction and with the electron microscope. X-ray diffraction data were obtained on oriented aggregates of the $< 2\mu$ fraction for all samples and nickel-filtered copper radiation was used throughout the study. Transmission electron micrographs were made also on the claysize fraction with a Phillips 100-B electron microscope. Electron micrographs of replicas of freshly broken surfaces were made for certain sequences of fine-grained igneous rock samples taken across the weathering profile. In addition, replicas were made from surfaces of individual feldspar crystals of some of the granites and syenites showing various stages of weathering. Replicas, prepared from acetate peels, were shadowed with platinum-palladium and backed with a carbon film.

PUBLISHED CLAY MINERAL DATA OF HONG KONG

The clay mineralogy of weathered rocks in Hong Kong has not been studied in detail. Brock (1943) studied the chemical changes that take place during the tropical weathering of various rock types in Hong Kong. Although he included a certain amount of X-ray work and data from petrographic microscope examination in his report, Brock commented that the mineral changes that take place in rock weathering in Hong Kong are not well known. Kaolinite seemed to be the most common clay mineral in the weathered materials, but chemical analyses showed alumina to be more soluble than would be expected for kaolinite. His petrographic work with index oils did not indicate the presence of halloysite or allophane; however, he did suggest that there was considerable amorphous material in many samples. Later, halloysite was identified in one sample of weathered Hong Kong granite from the Sham Shui Po area of Hong Kong (Davis, 1952, p. 103), and electron micrographs indicated that tubular halloysite was the only clay mineral present in the clay-size fraction of a few additional kaolin samples from the Colony (Honjo et al., 1954).

^{*} Most abundant mineral listed first. hal. = halloysite, kaol = kaolinite, gibb = gibbsite, ill = illite, verm. = vermiculite, mont. = montmorillonite, qtz. = quartz, feld. = feldspar, plag. = plagioclase, tr. amt. = trace amount.

DISCUSSION

The most common clay mineral in the modern weathering products of Hong Kong is tubular halloysite. It forms from all rock types that have been examined, including granite, syenite, quartz monzonite, rhyolite porphyry, lamprophyre, pyroclastics, and siltstone. Kaolinite is generally absent or is a minor component in the weathering products, and seems to be forming from halloysite rather than feldspar. Electron micrographs of weathered rock surfaces suggest that halloysite does not form directly from the weathered rocks, but instead evolves from an intermediate weathering product that occurs as tiny, tapered projections and as flame-shaped films on the surfaces of feldspars that are undergoing weathering. It appears that in the Hong Kong samples studied, halloysite and kaolinite probably give way to gibbsite under advanced weathering conditions close to the top of the weathering profile.

Halloysite \cdot 4H₂Q or 2H₂O occurs in tubular forms that have maximum diameters of about $0.3\,\mu$ and lengths that may exceed $12\,\mu$. An electron micrograph of typical forms of halloysite from Hong Kong weathering products is shown in Fig. 2. Cross-sections of halloysite observed show polygonal rather than circular outlines.

Examination of the weathered products by a combination of X-ray diffraction and electron microscopy seems to be necessary if mixtures of halloysite and kaolinite are suspected (Brindley et al., 1963). Identification of single mineral components in mixtures of halloysite, kaolinite, and amorphous alumino-silicate material by wet chemical analysis is more difficult, as indicated by the work of Brock (1943). Though he considered kaolinite to be the common clay mineral in the weathering products of Hong Kong, he noticed, as mentioned earlier, that chemical analyses showed alumina to be more soluble than would be expected if kaolinite were in fact present. It has been shown since then, however, that 50 to 90% of the total alumina in halloysite can be extracted by sulfuric acid treatment, whereas only 3% could be extracted from kaolinite (Grim, 1968, p. 436-437).

Kaolinite in the Hong Kong samples is present as both pseudohexagonal plates and as irregular flakes. The minimum diameter of pseudohexagonal kaolinite in samples AB-3, DWB-2 and 3, T-320-5 (Fig. 3), CK-1, LM-2, CKL-3, 7 and 12 is less than the diameter of the associated halloysite tubes. Halloysite tubes that appear to have lost small patches or flaps of material from their outer surfaces, thus exposing part of the surface of a narrower tube within, are relatively common in many of the weathering products in Hong Kong. The fact that the diameter of the smallest pseudohexagonal kaolinite flakes and the diameters of patches or flaps of missing material from the halloysite tubes are essentially the same may suggest that thin, small kaolinite plates may initially have sloughed off the flat sides of halloysite tubes of polygonal cross-section. If after the thin platelet of kaolinite is



FIG. 2. Transmission electron micrograph of halloysite from weathered syenite, sample CKL-4

released, it may act as a seed or a site for furter growth, eventually leading to development of large pseudohexagonal crystals of kaolinite.

Kaolinite having an irregular outline has been identified in several other samples (Tables 1 and 2). If kaolinite had developed in certain rocks in the Hong Kong region as a result of past hydrothermal alteration, it probably could not be distinguished from kaolinite formed during the more recent weathering period. There is no direct evidence to support such an inference, but it is possible that kaolinite of irregular outline was formed by some geological process prior to the period of modern weathering.

Halloysite and kaolinite do not seem to form directly from the weathering of feldspar, but rather develop from an intermediate phase (Fig. 4). The intermediate phase grows outward from feldspar surfaces during weathering in the form of tiny bumps and tapered projections. Material of similar morphology has been identified as allophane by Yoshinaga et al. (1968). The bases of the projections commonly



FIG. 3. Halloysite tubes and pseudohexagonal kaolinite from weathered Hong Kong granite, sample T-320-5. Tube showing partly formed pseudohexagonal flaps is indicated by arrow

merge laterally with one another during their growth to form thin, flame-shaped sheets. This weathering product most likely originates at points of structural weakness on feldspar surfaces (Parham, 1969). Fig 4. shows the development of flame-shaped films along the intersection of two cleavage planes on a surface of partly weathered K-feldspar. The forms exhibited in these electron micrographs are representative of those seen in this study.

Replicas were not made for surface alteration products forming directly on micaceous minerals and, therefore, it is not certain that the flame-shaped sheets would be typical morphology for surface weathering products on these minerals. However, electron micrographs of samples T-320-6A and D, a partly weathered, micaceous, maroon siltstone, show that through progressive weathering illite decreases in abundance as halloysite increases. This might suggest that the micaceous

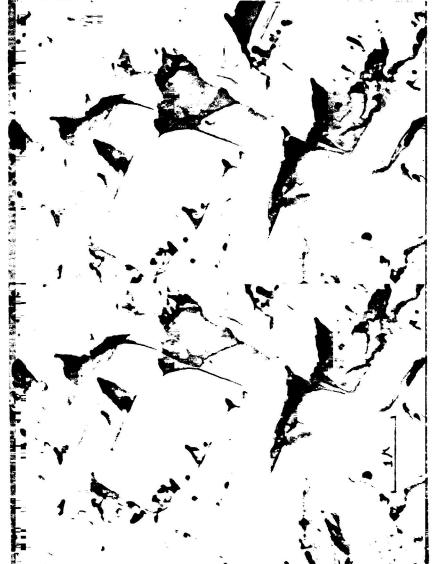


FIG.4. Stereographic set of electron micrographs of surface replica of weathered K-feldspar from syenite, sample DWB-1 showing tapered projections and flame-shaped sheets

minerals probably follow the same path as the feldspars on weathering to halloysite. There is no indication from the study of the micaceous samples that they alter directly to kaolinite.

Gibbsite occurs in Hong Kong in the weathering products of granite and rhyolite porphyry where drainage has been good for long periods of time. It probably develops from the breakdown of halloysite and kaolinite under advanced weathering conditions. It is present in some near-surface zones of various weathering profiles (Tables 1 and 2).

Experimental work on rock weathering and the subsequent formation of either kaolin minerals or of the hydrous-aluminum-oxide minerals (Wollast, 1967) indicates that, in the leaching environment, if the concentration of silica in downward moving waters is more than 5 mg/l, kaolin minerals will form; however, if the dissolved silica is below 5 mg/1 they will not form and bauxitization of the feldspars will occur instead. His findings suggest that gibbsite or boehmite, for example, can form directly from allophane, where leaching rates are continually high and the silica in solution is below 5 mg/1. The kaolin minerals should form, however, where leaching rates are slower and where there is a greater likelihood of higher amounts of dissolved silica in the leaching waters. It is clear that the original abundance of silica and aluminum in the rock-forming minerals undergoing chemical weathering must be considered along with leaching rates in understanding the origin of the newly formed weathering products (Parham, 1969). The presence of abundant halloysite in the weathering products of Hong Kong probably reflect good but not extreme leaching rates, as well as parent rocks in which silica is abundant.

Comparison of the modern rock-weathering products of Hong Kong with the much older weathering products in Minnesota suggests several points regarding the stability of halloysite, kaolinite, and the flame-shaped sheets. Firstly, no intermediate phase has been seen in the older weathered products of Minnesota that corresponds in any way to the tapered projections and flame-shaped sheets so characteristic of the Hong Kong samples. Only feldspar, kaolinite and small amounts of halloysite are present in the weathered residuum of Minnesota. This might suggest that the intermediate phase has a disordered structure and that with geologic time it changes to a more highly ordered structure, perhaps to halloysite or kaolinite. The intermediate phase of tapered projections and flame-shaped sheets probably corresponds in composition to that of allophane. Secondly, the modern rockweathering products of Hong Kong are rich in halloysite and poor in kaolinite, whereas the reverse is true for the Mesozoic weathered products in Minnesota. This suggests that halloysite is converted to kaolinite in relatively short periods of geologic time. It is inferred, therefore, that under humid tropical conditions of weathering where good drainage exists, a general sequence of mineral alterations

occurs, in which feldspar alters first to allophane, then allophane to halloysite, and finally, with sufficient time, halloysite to kaolinite.

ACKNOWLEDGEMENTS

Financial suport for this investigation was given by the Office of International Programs and the Graduate School of the University of Minnesota. Many of the faculty and graduate students of the Department of Geography and Geology, University of Hong Kong, contributed time and valuable information to assist me in the field sampling program in that Colony. Mr. C. M. Guilford and Mr. David Coffee of the engineering firm of Scott and Wilson, Kirkpatrick and Partners, Hong Kong, guided me to numerous rock cuts throughout the area of the coastal inlet of Plover Cove where reservoir construction was in progress. The assistance of all of the above persons and organizations is deeply appreciated.

REFERENCES

Brindley, G.W., Souza Santos, P. de and Souza Santos, H. de, (1963) Am. Miner., 48:897-910.

Brock, R.W., (edited by F.F. Grout and C.O. Swanson), (1943) Bull. geol. Soc. Am., 54:717-738.

Davis, S.G., (1952) The Geology of Hong Kong, 210 p., The Government Printer, Hong Kong.

Grant, C.J., (1960) Soils and Agriculture of Hong Kong, 154 p., The Government Press, Hong Kong.

Grim, R.E., (1968) Clay Mineralogy, Second edition, 596 p., McGraw-Hill Book Co., New York.

Honjo, G., Kitamura, N. and Mihama, K., (1954) Clay Miner. Bull., 2 (12):133-141.

Parham, W.E., (1969) Clays Clay Miner., 17, in press.

Ruxton, B.P., (1959) Q. Jl. geol. Soc. Lond., 115:233-260.

Wollast, R., (1967) Geochim. cosmochim. Acta, 31:635-648.

Yoshinaga, N., Yotsumoto, H. and Ibe, K., (1968) Am. Miner., 53:319-323.