

Attapulгите from Kanetake, Fukuoka Prefecture, Japan

Okamoto, Yohachiro
Department of Geology, Faculty of Sciences, Kyushu University

Tanaka, Nobuya
Faculty of Engineering, Kyushu University

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Attapulgitite from Kanetake, Fukuoka Prefecture, Japan*

By

Yohachiro OKAMOTO** and Nobuya TANAKA***

Introduction

OKAMOTO found in 1955 a moss-like "mountain leather" occurring in the limestone at Nishiyama, Kanetake-mura, Sawara-gun, Fukuoka Prefecture.

The chemical composition and physical properties of the mountain leather are in exact accordance with those of the attapulgitite from Attapulgis, U.S.A.

Modes of occurrence of attapulgitite so far known are as follows:

(1) alteration product from pyroxene or amphibole in sedimentary rocks; (2) alteration product from montmorillonite formed in lagoon; (3) pocket or veinlet in the syenite that suffered montmorillonitization.

Thus, the attapulgitite from Kanetake shows a peculiar mode of occurrence that is worthy of the closest attention.

Mode of Occurrence

The Kanetake area is composed of the Paleozoic limestone and amphibolite-schist and the later intrusions of granodiorite and biotite granite.

Being thermally metamorphosed, the limestone was transformed into marble usually rich in skarn minerals, such as garnet, diopside, thulite, vesuvianite, wollastonite, common epidote, etc., but no skarn mineral is not with in the marble where the attapulgitite is found.

Though the amphibolite-schist and the metamorphosed limestone lie side by side, the attapulgitite is found only in the latter, occurring as veinlets either along fissure in the marble or in terra-rossa formed along the fissure. The mineral of the former occurrence is pure white, but that of the latter is stained with terra-rossa.

Microscopic Observation

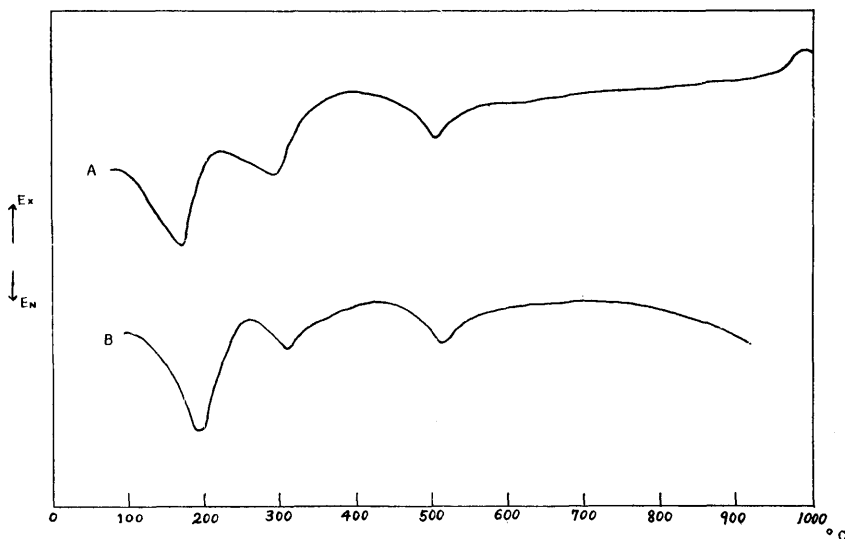
Under the microscope, the veinlets of the attapulgitite penetrate through calcite grains and the attapulgitite fibres often sit in a sub-parallel direction crossing diagonally the fissure, in which remain not a few rounded grains of calcite (Plate 5, fig. 1).

* Received October 28, 1957

** Department of Geology, Faculty of Science

*** Department of Mining, Faculty of Technology

The refractive index N_g of the fibres determined by the immersion method is 1.535, the double refraction being low.



Text-fig. 1. Differential thermal analysis curves for attapulgites

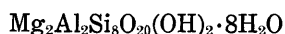
A: Kanetake, Fukuoka Prefecture

B: Rhki Chagin Gorxy Region, U.S.S.R. (STEPHEN, 1954)

Chemical Composition

The result of chemical analysis is given in Table I, together with those of some attapulgites from other countries.

BRADLEY gave the following ideal structural formula for attapulgite: $(\text{Si}_8\text{O}_{20})\text{Mg}_5(\text{OA})_2 \cdot 8\text{H}_2\text{O}$. Since Al atoms, however, are contained in a large amount in any attapulgite, it may be reasonable to consider that they should be present as one of chief constituents. Thus, on the basis of 42 anions implied in BRADLEY's structural formula, the following structural formula would be presented instead:



This formula coincides with that of the so-called pre-montmorillonite; namely, in the formula 2Al takes the place of the octahedral layer.

Generally speaking, the chemical composition of what is called attapulgite may be represented by the above formula; thus, for example, the structural formulae for the attapulgites referred to the Table I are listed as follows:

1. $(\text{Mg}_{1.65}\text{Fe}_{0.01}'''\text{Al}_{2.10}''\text{Fe}_{0.13}''')(\text{Si}_{7.91}\text{Al}_{0.09})\text{O}_{20}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$
2. $(\text{Mg}_{2.01}\text{Fe}_{0.02}'''\text{Al}_{1.54}''\text{Fe}_{0.37}''')(\text{Si}_8\text{O}_{20}(\text{OH})_2) \cdot 8\text{H}_2\text{O}$
3. $(\text{Mg}_{1.44}\text{Fe}_{0.03}'''\text{Al}_{2.23}''\text{Fe}_{0.17}''')(\text{Si}_{7.78}\text{Al}_{0.22})\text{O}_{20}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$
4. $(\text{Mg}_{1.43}\text{Al}_{2.26}'''\text{Fe}_{0.02}'')(\text{Si}_{7.62}\text{Al}_{0.38})\text{O}_{20}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$

Table I. Chemical composition of some attapulgitites

	1	2	3	4
H ₂ O (—)	7.73	9.12	10.29	10.32
H ₂ O (+)	15.70	10.89	13.24	12.00
SiO ₂	51.07	59.64	51.17	52.35
Al ₂ O ₃	12.10	8.76	13.73	15.44
Fe ₂ O ₃	1.22	3.36	1.55	2.12
FeO	0.21	0.23	0.31	—
MnG	none	—	—	—
CaO	3.95	2.02	2.89	0.24
MgO	8.30	9.05	6.40	6.60
Na ₂ O	none	$\left. \begin{array}{l} \text{Na}_2\text{O} \\ \text{K}_2\text{O} \\ \text{MnO} \\ \text{P}_2\text{O}_5 \end{array} \right\} 2.40$	—	—
K ₂ O	none		—	—
Total	100.18	100.07	99.58	98.97

1. Nishiyama, Kanetake-mura, Sawara-gun, Fukuoka Prefecture, Japan.
Analyst, N. TANAKA (this paper).
2. Attapulgit, Georgia, U.S.A. (BRADLEY, 1940)
3. Nijni-Novgorod, U.S.S.R. (CAILLERE and HENIN, 1951)
4. Shetland Isles. (STEPHEN, 1954)

Table II. X-ray powder data on attapulgitites

1		2		3		4	
d	I/I ₁	d	I	d	I	d	I
10.5	1.0	10.4	10				
6.4	0.6	6.32	3	6.15	6		
5.42	0.5	5.40	3				
		4.96	0.5	5.09	4		
4.49	0.8	4.51	7	4.56	8		
4.18	0.3	4.15	5	4.38	10	4.39	3
3.69	0.5	3.66	2	4.03	3	4.08	3
3.50	0.3			3.63	3	3.76	3
3.23	1.0	3.20	6	3.18	1.5(Br)	3.14	10
3.03	0.1	3.03	1.5	2.78	5	3.16	2
2.61	0.8	2.604	6	2.645	3	2.869	2
2.55	0.3	2.526	6	2.495	0.5	2.862	1
2.38	0.3	2.334	0.5	2.272	1	2.517	2(Br)
2.15	0.5	2.130	2	2.187	3	2.218	1
1.82	0.1	1.675	2	2.073	0.5	2.045	1
1.62	0.1	1.609	1	1.982	0.5	1.859	5
1.56	0.3	1.566	1	1.863	1	1.764	0.5
1.50	0.5	1.500	2	1.740	1	1.695	0.5
				1.698	1	1.631	0.5
				1.616	0.5	1.602	0.5
				1.540	0.5	1.489	2
				1.500	0.5	1.435	2
						1.423	2
						1.400	2

Notes.—CoK_α radiation; d in Å; I: relative intensity (estimated visually).

- 1) Attapulgit, Attapulgit, U.S.A. (BRADLEY, 1940).
- 2) Attapulgit, Kanetake, Fukuoka Prefecture, Japan.
- 3) Ditto, heated at 750°C for 2 hours.
- 4) Ditto, heated at 1000°C for 2 hours.

X-ray Analysis

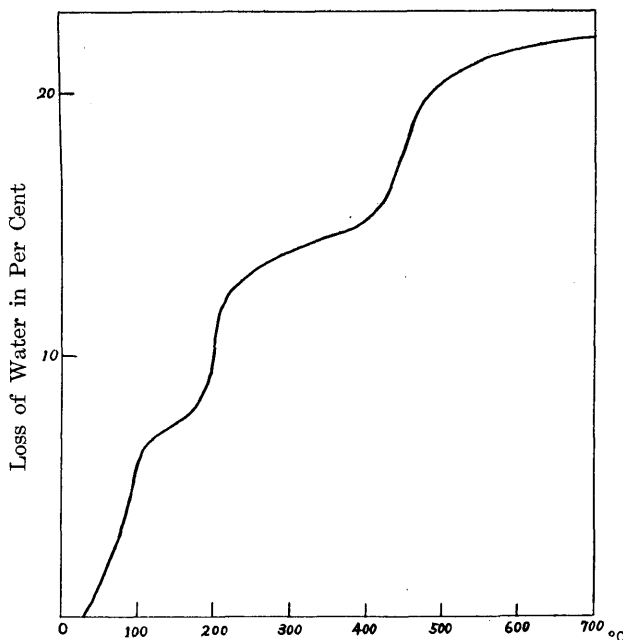
X-ray powder diffraction data on our attapulgite, which are in exact accordance with those on the mineral from Attapulgius, U.S.A., together with those on our materials heated at 100, 300, 500, 700 and 1000°C are given in Table II and their diffraction diagrams are shown in Plate 6.

As is clearly seen from the diagrams, the crystal structure of our attapulgite shows no change by heating up to 300°C. By heating the mineral up to the temperatures at 500 and 700°C, the strongest line for attapulgite, 10.4Å (110), disappeared, and no such phenomenon as the replacement of the 10.4Å line by hole that has been reported by BRADLEY (1940) was observed, but the heated specimens gave the diffraction lines that are entirely different from those of unheated specimen; namely, the lines shift to those of Longchambon's palygorskite II (CAILLERER and HENIN, 1951).

Differential Thermal Analysis and Dehydration Investigation

The reaction temperatures of attapulgite vary with the locality. For instance, according to STEPHEN (1954) the specimen from Shetland Isles has three endothermal peaks, one at 200°C, the second at 300–350°C, and the third at 500–560°C; while CAILLERE and HENIN (1951) determined them on the specimen from Nijni-Novgorod at 150, 350, 800°C respectively and an exothermal peak at 800–1000°C as well. The D.T.A. curve for our attapulgite shows three endothermal peaks and one exothermal peak (text-fig. 1), and that no change in the structure was occurred during the thermal analysis was confirmed by the piperizin method.

As is evidently clear from the dehydration curve shown in Text-fig. 2, dehydration passes through three processes



Text-fig. 2. Dehydration curve for attapulgite from Kanetake, Fukuoka Prefecture

which is comparable to those shown by the D.T.A. curve. The dehydration from the room-temperature to 470°C is confined to hygroscopic water and free water, giving no influence to the structure as is evident from Plate 6 and Table II. Therefore,

the temperature at which the diffraction lines shift, as already referred to, to those of palygorskite II (attapulgitite II) should be between 470 and 500°C.

Electron Microscopic Observation

The electron micrographs of the specimen from Shetland Isles (STEPHEN, 1954), which are useful for the determination of attapulgitite, illustrate that the specimen consists of a bundle aggregate of long and thin forms, while such a tubular form characteristic to hydrated halloysite and chrysotile is not found. The micrograph of our attapulgitite demonstrates not only a bundle form, but also a long, slender tubular one (smaller than $10 \times 0.05 \mu$), which resembles hydrated halloysite (Plate 5, fig. 2). Since neither hydrated halloysite nor chrysotile is mixed in our specimen, the mineral of tubular form may possibly be attapulgitite. It may here be remarked incidentally that the existence of tubular attapulgitite was proved by Honjo, of Tokyo Institute of Technology (T. SUDO's oral communication).

Summary

Attapulgitite in the form of "mountain leather" is found at Nishiyama, Kanetake-mura, Sawara-gun, Fukuoka Prefecture, Japan.

The area is composed of the Paleozoic limestone and amphibolite-schist and the later granitic intrusions which metamorphosed the limestone to marble, in which are found skarn minerals, such as garnet, diopside, thulite, vesuvianite, common epidote, wollastonite, etc.

The attapulgitite is not found in the amphibolite-schist, but in the veinlets along the fissures of the metamorphosed limestone or in the terra-rossa formed along the fissures.

Chemical composition, differential thermal analysis curve, X-ray diffraction diagrams and dehydration curve confirm the mountain leather to be attapulgitite which has, as presumed by BRADLEY, nearly the same constitution as that of Pre-montmorillonite.

Acknowledgements

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Plates

Plate 5

Explanation of Plate 5

Fig. 1. Photograph showing the mode of occurrence of the attapulgite from Kanetake, Fukuoka Prefecture. $\times 100$.

C, Calcite; A, Attapulgite; H, Hollow

Fig. 2. Electron micrograph of the attapulgite from Kanetake, Fukuoka Prefecture. $\times 12,000$.

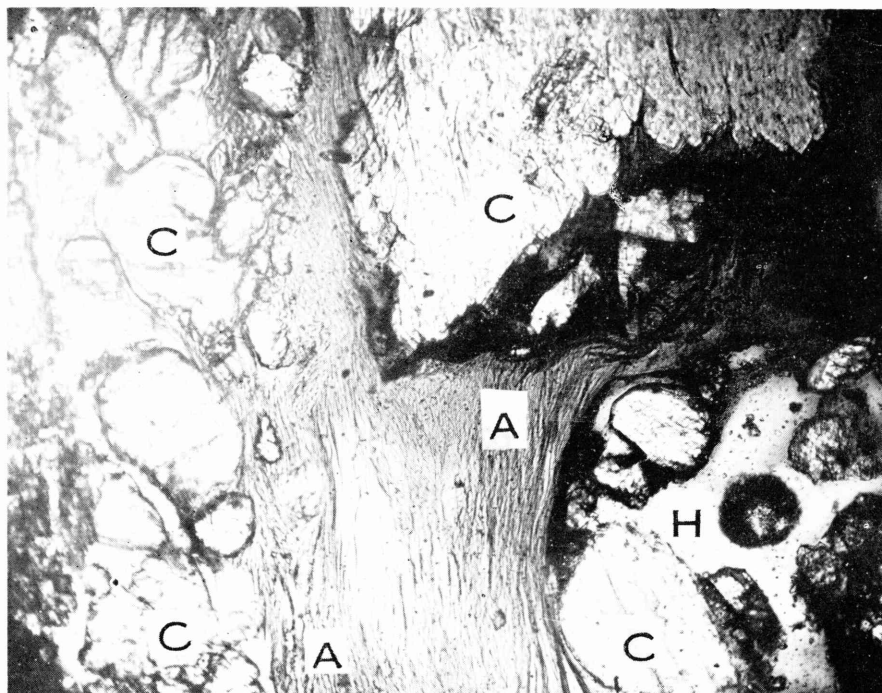


Fig. 1

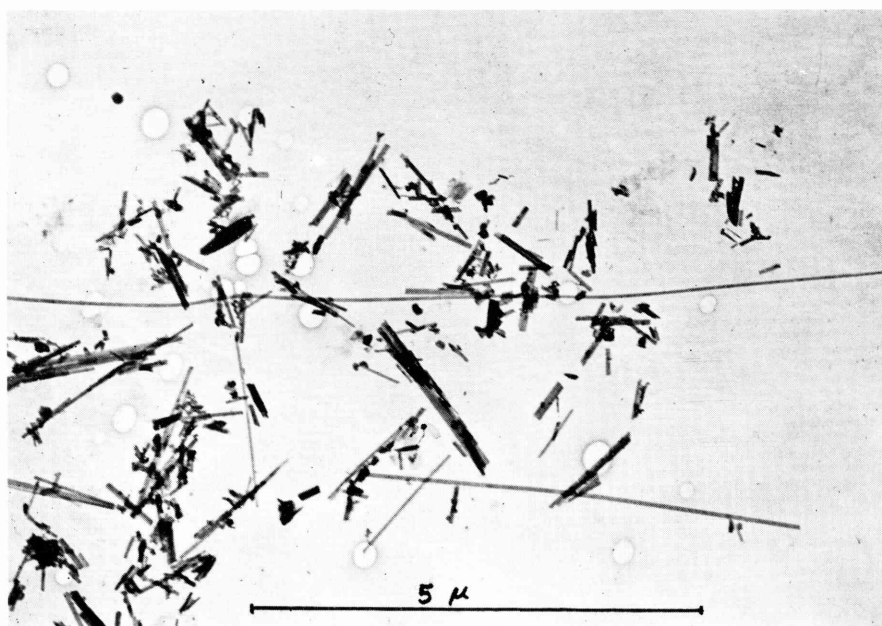


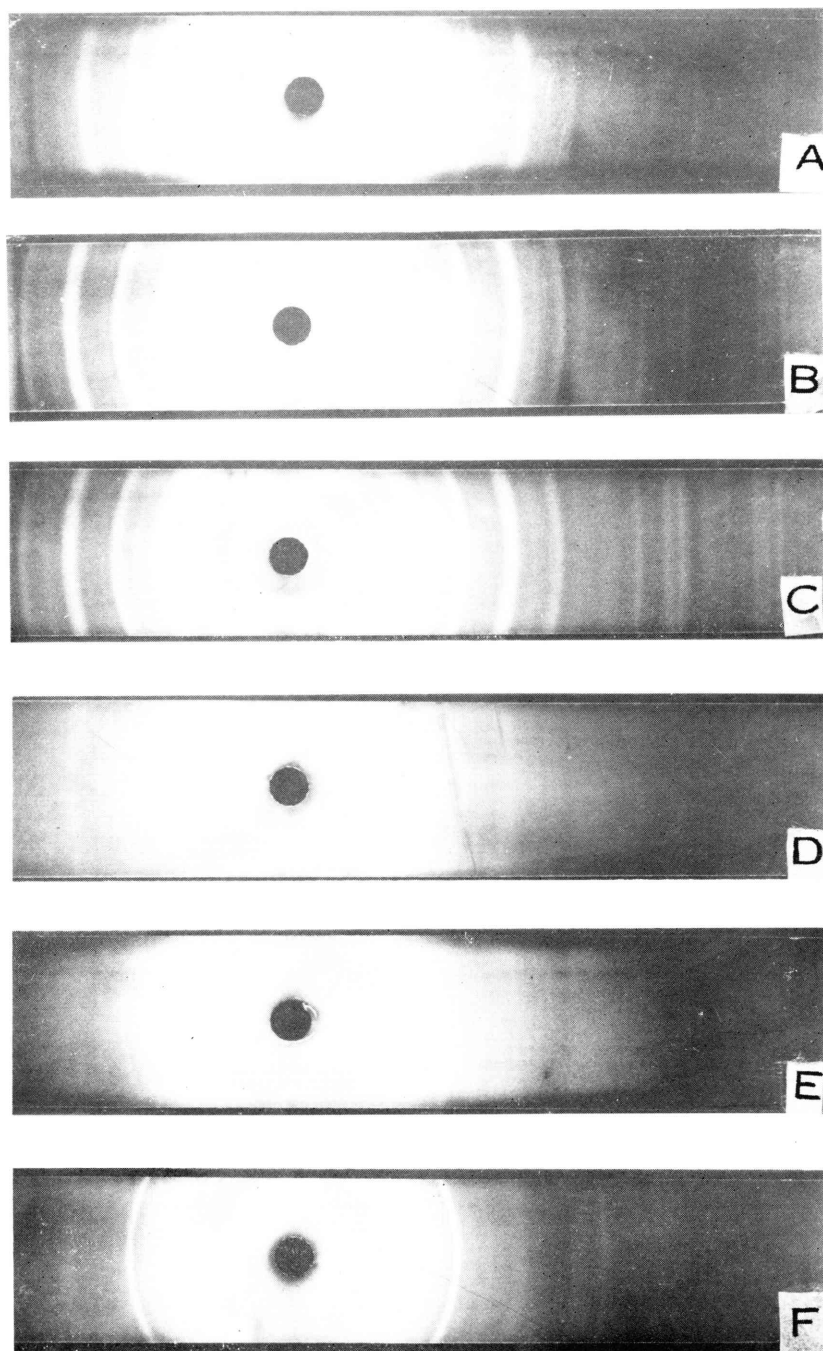
Fig. 2

Plate 6

Explanation of Plate 6

X-ray powder diffraction diagrams of the unheated and heated attapulgites from Kanetake (filtered CoK_α radiation; camera radius 40.3 mm).

- A: unheated
- B: heated at 100°C for 2 hrs
- C: heated at 300°C for 2 hrs
- D: heated at 500°C for 2 hrs
- E: heated at 750°C for 2 hrs
- F: heated at 1000°C for 2 hrs



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