On a Radioactive Mineral, found as Crust under the Hot Spring Water of Hokuto in Taiwan.

By Yōhachirō Okamoto.

In 1906, I first mentioned the crystalline mineral crust, found on the bottom of that part of the creek of Hokuto¹ or Pak-tau in Taiwan, where water is very hot. Whether the mineral was really deposited from the hot water *now* flowing there is still disputed. Analyses by the students of Chemistry in the Science College, Imperial University of Tōkyō, show that the mineral consists of PbSO_4 and BaSO_4 . But the proportion of these two substances seem to vary a great deal, as shown by the disagreement of these analyses with that recently made in the Imperial Geological Survey. The zonal structure of sections of the well crystallized individuals of this mineral seems to produce the variable proportion of the two substances in single crystals.

The radioactivity of the mineral, which was discovered in 1908 by Mr. Sunohara² and myself, led Mr. Ishitani, physicist, and Mr. Yamakawa, physician, both belonging to the Imperial University of Tōkyō, to visit Hokuto in this year, in order to observe the emanation in the hot water itself. We await their report in connection with this interesting physical property of the water. Mr. Ishitani is now extending his investigation to the radioactivity of the mineral now under consideration.

At the same time, I had the opportunity to point out to Prof.

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1) A place near Taihoku. It is not read *Hokutō*, with a long *o* after *t*, as would naturally be expected from the two Chinese characters (北投), with which this place-name is written.

2) A family name (春原), which is always misread Haruhara.

Kotora Jimbō in the field the characteristic features of the crystallization of this mineral.

The analyses of the hot water were made in the Hygienic Laboratory of Tōkyō on his return from Taiwan. He was fortunate enough, to ascertain in this year the mode of occurrence of a similar mineral from a creek of tepid (!) water at the Shibukoro hot spring, on the southwestern flank of the volcano Yakeyama in the province of Ugo in the main island of Japan. The mineral from Shibukuro¹ (or Shikayu or Shika-no-yu) had been first brought to Tōkyō about ten years before by the late Mr. Hirosaburō Sakurai,² but left uninvestigated. His original specimen was a coarse-fibrous crust, with a dirty gray incrustation on the surface, and was known only as a sulphate containing lead. But Prof. Jimbō observed also the well crystallized specimens, which proved the similarity of the specimens from Taiwan and the main island. He was kind enough to send us pieces of the mineral from Shibukuro.

These facts have led me to attempt the following note on our present knowledge of the mineral from Taiwan.

OCCURRENCE. Hokuto is well known for its spa, which has many Japanese hotels and bathing houses, in which the hot water of the creek is chiefly used. Only in one of the houses, a quite different water from a more distant place is introduced to the basin. A remarkably straight fissure-valley running down the west flank of the volcano Shichiseizan,³ a part of the Daiton⁴ volcanic group has in it a number of solfataras with workable sulphur deposits, and an explosion crater with a perfectly flat bottom, lying on the southwestern side of Mt. Shichiseizan. On this flat ground there are springs of hot water (80° to about 90°C), which runs in very shallow streamlets over this level ground. The sample I for analysis was

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1) 澁黒. 鹿湯. 鹿ノ湯	2) 櫻井廣三郎
3) 七星山.	4) 大屯.

taken at the spring on the eastern side, and II at one lying on the middle of the foot of the southern hill slope. These and other springs from the same place unite on the western side to form a single hot water creek, not more than one foot and a half in depth, and only about 10 ft. in width. Where the bed of the creek is wide, the hot water flows in many narrow currents, and this seems to have facilitated the deposition of the mineral, if it was really formed from the water now flowing there. The explosion crater as usual shows rocks bleached and decomposed to earthy masses. Sulphur was once worked there, as is shown by a small heap of waste. As the spring waters issuing out from the level ground are all of the same kind, it will suffice if we give the following analysis of hot water (IV) at the principal bathing place, where it makes low water-falls, the gravelly creek bed suddenly descending as if traversed by a fault line. This almost perfectly coincides with the analysis of the sample III, which was obtained at the spot where the streamlets on the level ground finally unite on its western side. Analysis of the sample IV (filtered) gave per thousand.

Na_2SO_4	1.9055
NaCl	0.9990
$\text{Al}_2(\text{SO}_4)_3$	0.6588
KCl	0.7130
NH_4Cl	0.6073
CaSO_4	0.4787
FeSO_4	trace
$\text{Fe}_2(\text{SO}_4)_3$	0.5420
MgSO_4	0.2024
$\text{Al}_2(\text{HPO}_4)_3$	0.0012
BaSO_4	0.0007
PbSO_4	trace
H_2SiO_3	0.2174
HBO_2	0.0710
Free acid (HCl)	1.9596

This water, containing free hydrochloric acid, is colourless and with a scanty amount of sediment. Its taste is astringent and strongly acid. Smell of sulphuretted hydrogen is observed. Specific gravity of the sample determined in Tōkyō, $d_{26}^{26} = 1.0150$.

The determination of BaSO_4 and PbSO_4 was made with 17 liters of a mixture of unequal amount of the samples I to IV.

The water from another spring, called "Kaikōsha-onsen," issuing from the higher part of the southern side of the explosion crater flows into the streamlets on the flat ground on its bottom, but has apparently little influence on the composition of the sample IV. The analysis of the Kaikōsha-onsen in the Hygienic Laboratory shows that it is a dilute alum spring.

The level bottom of the explosion crater shows a white efflorescence of substances soluble in hot water and found accompanied by a little quantity of flourey sulphur. There is observed no deposition of well crystallized minerals like the radioactive mineral now under consideration. Where the streamlets of the hot water unite on the western side, the mineral is already found deposited, but the gravelly part of the creek apparently suited to its abundant deposition begins about 70 ft. below this spot and extends about 2000 ft. further down. Lower down the creek, the bottom is muddy and does not show any deposit. The gravel is mostly of andesite, more or less decomposed on the surface. There are found sometimes boulders more than 3 ft. in diameter, but the stones are usually not much larger than man's head. These are cemented together by sand grains, which are consolidated by impregnation with a yellow to brown substance, which may not be simply iron oxide.

This gravel as well as the consolidated sand is covered with thin crystalline crusts, the outer part of which is formed of druses of rhombic tabular crystals. This is the mode of occurrence of our radioactive mineral. It is remarkable to observe that the crystal development is almost entirely restricted to those parts of stones now

covered with the hot water. At the water line, a dirty gray, compact crust is produced, with no crystal development and no radioactivity. The substance is also quite different in composition from the radioactive mineral, the following analysis recently made in the Imperial Geological Survey showing that the gray crust consists essentially of silica :—

SiO ₂	77.10
SO ₃	2.48
Al ₂ O ₃	2.27
Fe ₂ O ₃	2.11
CaO	0.13
MgO	0.30
Na ₂ O	0.73
K ₂ O	0.14
TiO ₂	trace
P ₂ O ₅	trace
Ign. loss	14.94
	100.20

This gray crust sometimes carries crystals of gypsum lying flat on the surface. The crust spreads out 1 in. in width and as much above the water. But the surface of the gravel as well as of the consolidated sand of the creek bed, which is exposed to the air is usually quite bald, without the covering of any new mineral formation.

Whether the radioactive mineral is really deposited by the *present* spring water can not yet be affirmed ; but the hot water forming the creek contains a trace of lead and a minute quantity of barium.

The most abundant occurrence of the mineral is observed, where the creek water shows now a temperature of about 50° to 60°C.

CHEMICAL COMPOSITION.—In the following are mentioned (I) the analysis of the coarse-fibrous crust in 1909, by the second year students of the Science College, Imperial University of Tōkyō ; and (II) that of the well crystallized, brown-coloured, quite fresh

specimen in 1911, by Mr. Shimizu of the Imperial Geological Survey. I is an average of three analyses and II that of two analyses.

	I	II
PbO	21.96	19.38
BaO	32.04	42.27
SrO	0.93	trace
CaO	0.51	0.17
SO ₃	30.81	31.70
Fe ₂ O ₃	3.93	0.43
Al ₂ O ₃	0.88	0.48
MgO	1.04	0.28
K ₂ O	—	0.14
Na ₂ O	0.53	1.53
H ₂ O	2.53	—
Ign. loss	—	2.74
P ₂ O ₅	0.01	—
SiO ₂	1.27	0.97
	96.44	100.09

Fluorine is present in the sample I, but was not determined.

The difference of analytical results in I and II may be explained by the development of zonal structure in the crystals. Whether the mineral consists of layers of two different substances, BaSO₄ and PbSO₄, or their isomorphous mixtures in various proportions, can not yet be affirmed.

In glass tube, the radioactive mineral slightly decrepitates and fuses, becomes black, and assumes a metallic luster and variegated tarnish. With soda on charcoal, lead globules are easily obtained.

PHYSICAL CHARACTERS. The radioactive mineral crust is coarsely fibrous, with a drusy development on the surface. The colour varies from brown to brownish-yellow or almost white, while the dull white appearance of some crystals is apparently due to weathering. The pale zones in the crust are extremely thin, and

were not separately analyzed.

The crystals forming the druse are in rhombic plates with pyramidal faces, about 1/10 in. in diameter and half as much in thickness. They mostly form a subparallel growth on the basal pinacoid, if the rhombic face is taken as the base and the other four as a unit prism. The total thickness of the parallel growth is about 3/10 in. The base is bright and glassy and more lustrous than the prismatic faces, but is slightly depressed. The latter have a resinous luster and horizontal striation. The acute angle of the prism is about 72° to 78° , no exact measurement being possible, for the prismatic faces are striated and curved by subparallel union. The streak is white to brownish-white. Hardness, about 3.5. Cleavage, distinct along the base, but indistinct along the prismatic face. Mr. Kuhara¹ found the specific gravity about 6.1 at 19°C . He determined the mean index of refraction, roughly by the method of Schroeder van der Kolk, to be between barite and anglesite, the optical orientation being identical with that for these minerals. Zonal structure consists in the presence of lighter and darker coloured zones parallel to the faces of the prism.

The radioactivity was only observed by placing the mineral on a photographic plate in a dark room, and for the quantitative treatment of emanation we await the result of Mr. Ishitani's experiments.

He powdered the mineral and measured its radioactivity by an electroscope of Greinacher type, and informed us, that the available external α -ray activity of this mineral was 0.366 times or about 1/27 of that of uranium oxide.

OCCURRENCE OF A SIMILAR MINERAL AT SHIBUKURO. Soon after his return from Taiwan, Prof. Jimbō went to Shibukuro, where, to his immense satisfaction, he found a crust with a drusy surface, exactly similar to our mineral. The occurrence there, however, is distinguished by the extreme thickness of the crust, which often

exceeds two inches ; by its usually paler colour ; by its less frequently showing a drusy outgrowth, the crust being usually only covered with a gray incrustation, resembling in colour the compact crust at Hokuto ; by the want of yellow substance observed there ; and by the very low temperature of the tepid water of the creek, the gravel of which is cemented together by the crust-forming mineral. The hot water of the Shibukuro springs flows into its chief rivulet and not into the creek with the mineral. A particularly rich deposition of floury sulphur distinguishes this water from many other sulphurous waters in Japan. The occurrence of a barite-like mineral, showing a combination of the base, a pyramid and a prism, attached to a pale silicified volcanic rock of the solfatara on the side of the hot springs of Shibukuro is another interesting object of investigation. The analysis made at the Mine Inspection Bureau at Sendai, gave

BaO	64.07
SO ₃	33.82
Fe ₂ O ₃ + Al ₂ O ₃	1.81
SiO ₂	1.43
CaO	0.02
	101.15

The analyzed mineral is sometimes colourless and sometimes reddish, and almost perfectly transparent. A rhombic tabular mineral, resembling the radioactive mineral from Hokuto, is also found attached to this silicified rock.

As regards the crystal druses on the coarse-fibrous crust, no systematic analysis has yet been made, but preliminary tests by Mr. Kuhara show the presence of barium and lead, and the physical properties as well as the external appearance exactly agrees with the specimen from Hokuto.

(THE END)