

On "Kotoite" and "Suanite"

Takeo WATANABE

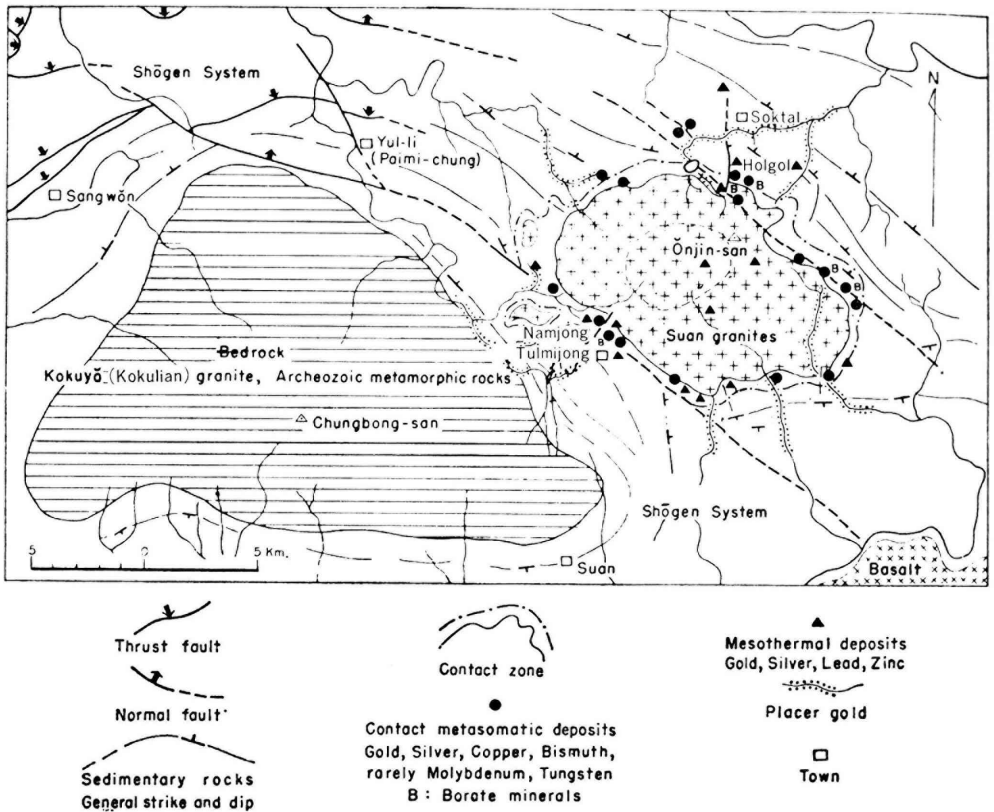


Fig. 1. Map Showing the Geologic Structure of the Mining District.
Holgol=Holkol, Tulumijong=Tul Mi Chung

Introduction

Kotoite is a magnesium borate, $Mg_3(BO_3)_2$ which occurs in abundance in the country rock of the contact-type gold-bearing copper bismuth deposit in the Holkol (Hol-gol) mine, Suan district, Korea. Since 1930 the writer has been studying the geology and ore deposits of the Suan district, and in 1932 he noticed that the

properties of the mineral did not correspond to those of any known mineral. In 1938, after further study of the ore of the Holkol mine, the writer determined the mineral to be one which had never been found before in nature. He named the mineral "kotoite" after the late Bunjirō Kotō, Professor Emeritus of the University of Tokyo.

The mode of occurrence of this mineral is interesting and important, not only in studying the genesis of the Holkol deposit, but also for prospecting the deposit. As Japan lacked boron resources in those days, the rock containing kotoite, a kind of metasomatized rock called "kotoite marble," came to be regarded as a boron ore. The process of manufacturing borax from the ore was studied by members of the Research Institute of the Hitachi Manufactory, and the attempt proved successful, not only experimentally, but also industrially. Thus, the deposit came to be mined more actively for boron.

Although the studies reported in this paper were first published in the "KATO Commemoration Volume," the writer has added some recent findings to bring the study up to date.

The geology and mineralogy in the district were first studied by Kotō (1910) and later by Tamura and Tsurumaru (1915), Higgins (1918), MacLaren (1915), and Shannon (1921 A, B) and many important facts were brought to light by them. The writer has continued his study of the remaining details of the problem. He is greatly indebted to the late Professor T. Kato for his constant encouragement and guidance in the course of the work. He is also indebted to Professors K. Uwatoko, J. Suzuki, and Z. Harada of Hokkaidō University for their valuable suggestions and help during the time when he was on the staff of that university; fortunately, with their support, the writer had many opportunities to visit the type locality and was able to continue his study for a number of years. The study on kotoite was accomplished under the direction of Prof. P. Ramdohr, director of the Institute of Mineralogy and Petrology, Berlin University, while the writer was in Berlin as a student sent abroad by the Ministry of Education in 1937-1938. The writer is indebted to Prof. P. Ramdohr and Prof. H. Strunz for their great help and he is also indebted to Prof. W. Eitel for his kindness while a part of the experiment was being carried out under his direction. In later years, when kotoite came to be regarded as a boron ore, the writer was aided by grants from the Fifty-seventh Special Committee of the Association for the Promotion of Science, Japan, to make a more detailed study on the Holkol deposit, than he had been able to do in the past.

During the field studies, the writer was afforded many facilities by the mine. Thanks are due especially to Mr. J. Nakamura of the Holkol Mining Office, Hōkō Mining Co., Ltd., as well as to the men and officials of the mine for their kindness. The writer also wishes to thank Mr. T. Yoshioka and Mr. S. Miyagi of the Research Institute of the Hitachi Manufactory, Mr. Y. Tamura of the Osaka Industrial Laboratory, Prof. S. Nagai of the Engineering Department of Tokyo University, Mr. H. Inuzuka of the Institute of the Tokyo Shibaura Electric Co.,

Ltd., Mr. T. ASAYAMA of Kyoto University, and Mr. E. ASHIDA, chief of the Kanagawa factory of the Takeda Chōbei Shōten, for their kindness in supplying data for analysis of the kotoite-marble.

The study of the X-ray analysis of kotoite was completed by Mr. R. SADANAGA at the Mineralogical Institute, the University of Tokyo, by courtesy of Prof. ITO. As their findings are cited in this paper, the writer wishes to express his grateful thanks to Prof. ITO and Mr. SADANAGA.

1. Progress on the Study of Kotoite

Early in the summer of 1930, the writer spent about two and one-half months in the Suan district, making a general survey of the area. The ore body, which was actively being worked in Holkol at that time, is a large ore pipe and was called the "new body," *i.e.* the present "north body." As the mineral association of the ore is both complex and interesting, the writer reported on it in the *Journal of the Geological Society of Japan* in 1933. In this report, the writer described an altered magnesian limestone which composes the country rock of the deposit and noted the existence of a fine-grained mineral which bears some resemblance to forsterite, showing a very small 2 V. This mineral is the kotoite described below.

The kotoite, as described on a later page, occurs as very fine crystals, and its optical properties are very close to those of olivine and diopside, so that they can hardly be distinguished from one another when the section is observed under an ordinary microscope; The mineral might be recognized as nearly uniaxial or biaxial with a very small 2 V if it could be observed successfully at a high magnification. As the writer did not obtain the conoscopic figure at the beginning of the study, he considered the mineral a fine-grained forsterite which occurs usually as relatively coarser crystals in the same rock. After the writer observed the interference figure under the high-powered microscope, he noticed that this mineral was different from the coexisting forsterite. Upon further study, its refractive indices were measured by using the immersion method. No known minerals corresponded to it, however, and judging from the mineral association, he assumed it to be a silicate mineral rich in magnesia, probably either clinoenstatite or pigeonite, which were being discussed at that time. He had many doubts about this assumption, because such an occurrence of these pyroxene had rarely been reported from the contact metamorphic rocks which contain forsterite; in addition, the mineral was observed to be so unstable, compared with forsterite, that it had completely decomposed by secondary alteration while the forsterite remained unaltered.

The writer again visited the Suan district in 1931, and consulted with his professors and seniors, but final determination of the mineral was impeded by the difficulty of isolating a pure sample of the fine-grained crystals.

While the writer was studying in Berlin in 1937–38, he studied the ore from the Suan district and attempted to describe it. He also intended to study the as yet undetermined mineral. At that time, Dr. Hugo STRUNZ, assistant at the Institute,

was engaged in an X-ray study of silicate minerals. He was especially interested in clinoenstatite-enstatite relationships, namely, the structural problems of both the rhombic and monoclinic pyroxene, and had collected various specimens. The writer considered this convenient for judging the undetermined mineral from Suan and attempted to separate a pure sample of the mineral from the ore by using the heavy-solution method. He obtained a large amount of Clerici solution, and a considerable amount of the pure sample was obtained. Using the powder method, an X-ray photograph of the mineral was prepared. By comparing it with the photographs of pyroxenes, the writer determined that the mineral was entirely different from the pyroxene group. Almost simultaneously with this examination, the writer attempted a preliminary quantitative analysis of the mineral in order to determine what chemical composition it would have if it were a member of the pyroxene group. As a result of repeated analyses, the writer recognized that the silica content of this mineral was very low, which was quite unexpected. Until that time, the writer had supposed it to be a kind of silicate mineral, so he was afraid that he had erred in preparing the sample and examined the separated material under the microscope. In addition, he observed the flame reaction of a dissolved solution of the sample. Upon testing the flame coloration of the product resulting from the quantitative analysis for silica, he found a remarkable green color. This unexpected fact attracted the writer's attention, so he investigated the cause. He first tested a part of the sample to see whether it showed a green flame reaction or not. In order to accomplish this he tried to wash out completely the Clerici solution from the sample, because the heavy solution contains Tl which produces a green flame coloration. But strangely enough, the green color could not be removed, no matter how many times the sample was washed. Therefore, the writer could not deny the existence of another material which produces a green flame reaction.

From that time the writer considered the possibility of the presence of boron; judging from the existence of ludwigite in the same rock, he should consider that the mineral was a borate mineral.

He resolved to complete the entire analysis of the separated material. The optical constants and specific gravity were measured, and the presence of boron and magnesium was confirmed. Thus, optically, the mineral came to be recognized as having no known corresponding boron minerals, and the writer decided to compare it with artificial borate.

In March of the same year he found, through GROTH's "Chemische Kristallographie," mineral properties which almost coincide with those of magnesium ortho-borate, which were measured previously by MALLARD. Thus, the mineral was postulated to be a new mineral whose occurrence in nature had not yet been noted.

Accordingly, in addition to carrying on the complete quantitative analysis of the mineral, the writer decided to synthesize magnesium borate, as EBELMAN had done, in order to identify it by X-ray with the mineral.

After succeeding in synthesizing the artificial crystal, an X-ray photograph of its

powder was compared with that of the mineral from the Suan district, which had been photographed in February. In April 1938 the writer confirmed the coincidence of the two X-ray photographs. Soon after that, the chemical analysis was also completed, and, based on its chemical composition, the mineral was confirmed as magnesium ortho-borate.

From these data, the writer identified it as a new mineral and named it "kotoite" after Prof. Bunjirō Kotō, the "father of Japanese geology," who was the first to publish detailed descriptions of the rocks and minerals from the Suan district, especially those of the Holkol deposit.

In the summer of 1938 the name kotoite was made public before the meeting of the Mineralogical Society of Germany held at Graz, Austria (WATANABE, 1939 A,B).

Though the writer endeavored with Dr. STRUNZ to continue X-ray studies of the artificial crystal, they were not completed during his stay in Germany. However, the study was continued by SADANAGA of the Institute of Mineralogy, University of Tokyo, who recently reported their results.

In addition, the writer recognized the existence of kotoite in a rock specimen from Rézbánya, Romania, during his study on kotoite. The writer received a magnesium borate mineral and in the course of the study compared it with a fibrous borate mineral contained in a rock from Holkol in order to confirm whether the latter is szaibelyite or not. He noticed the presence of many crystals within the section of the former which correspond to those of kotoite. Immediately after that, the writer separated it from the sample and examined it by X-ray, using the powder method, and found that the X-ray photograph of this mineral coincides with that of kotoite.

The mineral szaibelyite was first reported by PETERS in 1861, and specimens were distributed among the museums in Europe and America. Nevertheless, the presence of kotoite had not been noticed, probably due to the fineness of kotoite in the rock and to the close resemblance of the optical properties of this mineral to those of forsterite or pyroxene. The old photomicrographs in Prof. Kotō's paper (1910) indicate that he regarded the kotoite from Holkol as diopside. Both HIGGINS (1918) and SHANNON (1921A,B) seem to have overlooked this mineral, describing it only as diopside. Though kotoite is now known to occur at only two localities. Holkol and Rézbánya, it is highly possible that it may be found in other regions which have geologic conditions similar to those of the two localities.

In the summer of 1939 and the spring of 1941, after the discovery of kotoite, the writer visited the Suan district and made a detailed investigation of both the surface and subsurface of the Holkol mine. Upon examining the distribution and mode of occurrence of kotoite, the writer found that there is a large reserve of the mineral, and that the genesis of the Holkol deposit is closely related to the formation of kotoite.

The writer took up the consideration of industrial uses of kotoite because of its

abundance and its relatively simple, homogeneous chemical composition. In 1938 these views were sent from Berlin to Dr. Benzō KATSURA, adviser to the Holkol mine, but the opportunity for industrial utilization of kotoite did not present itself. Later, in July 1949, Mr. Tōsaku YOSHIOKA of the Hitachi Institute read the writer's paper to the late Prof. Shintarō NAKAMURA, who perceived the possibility of utilizing kotoite in the glass industry, which was then running short of raw materials. At that time, Japan imported all her boron materials, especially from the United States, Chile, and Argentina. Most Japanese were little concerned with the boron resources, but the Hitachi group began to study this problem almost two years before World War II.

When the writer reported the existence of kotoite before the meeting of the Mineralogical Society held in Graz, the petrographer, Dr. H. STÜTZEL, who was present at the meeting, stated his views as follows: when magnesia bricks or magnesia clinkers are used for flooring and wallboard in the steel industry, boric acid is often mixed into them, and as a result, their tendency to decompose, a defect of magnesia firebrick as flooring and wallboard material, can more or less be rectified; this suggests the possibility of the production of a high temperature compound which might have a chemical composition similar to that of kotoite.

2. Mode of Occurrence

A. Holkol mine,¹⁾ Suan Hwanghae-do, Korea (WATANABE, 1943)

1. History of the mine; general views on the geology and ore deposits:

The mine has long been known as the Suan gold mine, the oldest gold mine in Korea. It was opened on a large scale by the Americans and the English about 40 years ago. The name "Suan gold mine" is a name which, in early days, included several mines around Onjin-san. The present Holkol mine was also called Suan mine because it is located in the center of the claim. The principal area of development was the Namjōng-ni deposit, a branch of the old Suan mine, and in about 1928 the claim was divided into two areas, the Holkol mine in the north and the Namjōng (Tul-Mi-Chung) gold mine in the south. The historic name of the Suan gold mine was assumed by the newly-developed Namjōng (Tul-Mi-Chung) mine, which was being operated by the Japan (Nippon) Mining Co., and the Holkol mine was given a name of its own. In 1928 the Holkol mine belonged to Chong-un Chong and three others, but after 1932 it was managed by the Japanese. More recently, the mine was worked by the Hōkō Mining Co., Ltd., and gold, silver, copper, tungsten, and boron were mined.

The office and ore-dressing plant of this mine were located at Soktal-li in the upper reaches of the U-gang, a branch of the South Taedong-gang. The principal mining area is Pogwang-ni, which is also known as Holkol, about 2 km south of Soktal-li. This region can be reached by truck in five to six hours by way of Sang-

¹⁾ The mine is called "Kotsudo" in Japanese and "Holgol" in Korean.

won and Yul-li from P'yongyang or Chunghwa stations on the railroad between Seoul and Sinuiju. Locally, the road runs across an abrupt area.

The geology of the area near the mine will be briefly described. As shown in the sketch map of the geologic structure (Fig. 1), Pre-Cambrian formations are widely distributed in the Suan district; especially in the Holkol region where Holkol dolomite, which consists chiefly of dolomite of the Shidōgu series, the middle Sangwon system, and Suan shale, which corresponds to the Kuken series, is found. These sedimentary rocks are intruded by Suan granite, which is considered to be representative of the Pulguk-sa igneous activity which took place during the Late Mesozoic. The rocks of the Sangwon system have been intensely metamorphosed, and various kinds of thermal metamorphic rocks were formed around the contact. In addition to the Namsan gold-copper-tungsten deposit northwest of Sōktal-li, the important ore bodies of the main working deposits in the Holkol mine are the pyrometamorphic gold-copper deposits. There are three important ore bodies, in the east, west and north.

The East ore body is vertical and extends from east to west along the contact between the Suan granite and the Holkol dolomite, forming skarn composed chiefly of diopside, tremolite, phlogopite, and minerals of the chondrodite group. It also contains native gold, bornite, chalcopyrite, pyrrhotite, bismuthinite, and several other minerals. In the thermally metamorphosed crystalline dolomite surrounding the skarn, we can find such borate minerals as ludwigite and kotoite.

The West ore body comprises two types of deposits; one is the skarn type, similar to the east ore body, and the other is the mesothermal metasomatic type. The west ore body is 60–100 m west of the east body. The deposit along the contact with the Suan granite is of the skarn type, similar to that of the East ore body. The deposit along the Holkol fault west of the Holkol valley and that contained in its crush zone show a dip and strike similar to the fault which strikes north to south and dips 55–60° east and contain two types of deposits, the skarn type and the mesothermal type, both of which occur in the metamorphosed dolomite. Ludwigite and kotoite have been found in the country rock of the skarn deposit, though only on rare occasions. The mineral association in the skarn ore is nearly the same as that of the East ore body. The mesothermal-type ore, on the other hand, has no skarn minerals, but some traces of replaced dolomite of the crush zone remain. Metallic minerals, other than chalcopyrite, contained in the ore are tetrahedrite, sphalerite, and galena, and the silver content of the ore is somewhat higher than that of the skarn ore.

The north ore body is located about 30–60 m north of the other two ore bodies, and it occurs as an ore pipe completely surrounded by dolomite.

Both the east and west ore bodies were found in the early days of the development of the mine, and mining began with the outcrops. The discovery of the North ore body, on the other hand, took place relatively late; that is, it was found by a Korean in the tunnel after the time of the foreign management and was for a time called "new ore body," or Pae-kum-kaeng (white metal tunnel) ore body, be-

cause of the large occurrence of white minerals, native bismuth etc., in the ore. It has become the most important ore body of the Holkol mine. The country rocks widely distributed around the ore body are dolomite-marble (crystalline dolomite) and brucite-marble. A marble-like metamorphic rock which contains kotoite and calcite as the principal mineral constituents is always found near the ore body, and the skarn consisting of diopside, phlogopite, clinohumite, etc., which may be regarded as a gold-copper ore, is completely surrounded by it.

Thus, the metamorphic rock which consists of kotoite and calcite, has been formed by replacing dolomite by the B_2O_3 in a pneumatolytic process whereby the deposit was formed. As the rock resembles ordinary marble, it will be called "kotoite-marble." In other words, the North ore body is a composite ore pipe of kotoite-marble which includes diopside skarn. This pipe stands like an inverted funnel, somewhat thick in the lower part and thinning out in the upper (see Fig. 2).

The relationship between the diopside skarn and the kotoite-marble is quite complicated and shows irregular boundaries, as seen in Fig. 3. However, the skarn is found almost in the center of the ore pipe, though it branches in three directions in the lower part. Specimens which exhibit the relationship of the three zones, that is, the dolomitic marble, kotoite-marble, and diopside skarn, are rarely found, but they can be collected from the margin of the ore body (see Fig. 4).

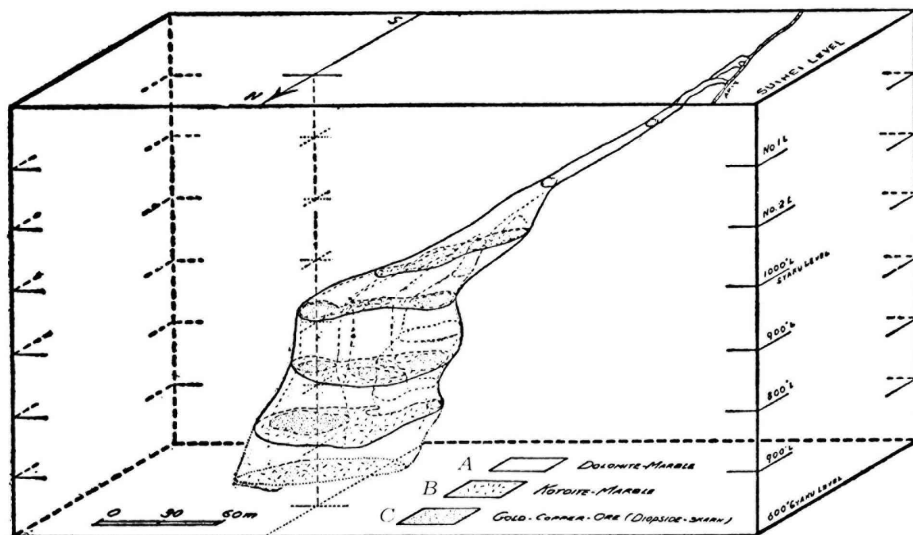


Fig. 2. Block Diagram of the North Ore Body of the Holkol Mine, Suan.

- A. Dolomite-marble
- B. Kotoite-marble
- C. Gold-copper-bismuth ore (Diopside Skarn)



- K: Kotoite-marble
- L: Ludwigite
- R: Reaction zone between diopside skarn and kotoite-marble
- D: Diopside-clinohumite-skarn containing gold and copper

Fig. 3. Underground Photograph Showing the Relation between Gold Copper-bearing Diopside Skarn and Kotoite-marble.

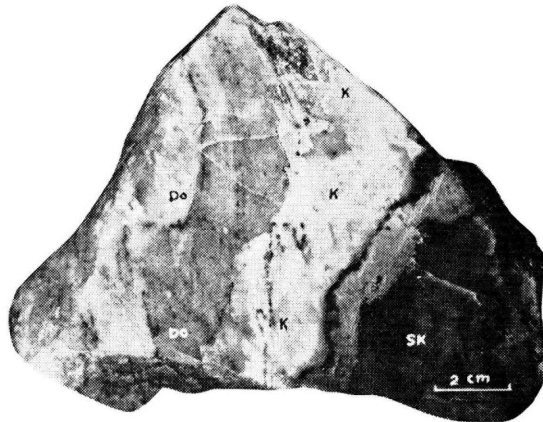


Fig. 4. Zonal Arrangement of Diopside Skarn (SK) Kotoite-marble (K) and Dolomite (Do).

Small branches of diopside skarn, round masses, or small lenticular bodies of skarn, as shown in Fig. 6, are irregularly distributed within the kotoite-marble, and almost all of the metallic minerals are contained in the skarn. Therefore, the gold-copper mine is being worked by following the diopside skarn.

The metallic minerals of the north ore body are native gold, native bismuth, bismuthinite, pyrrhotite, cubanite, chalcopyrite, bornite, and other sulfides of

bismuth or copper. As this mineral association was considered very interesting, it has already been partly reported in detail (WATANABE; 1933, 1943) and needs no further description here. In the upper part of the ore body, the associated minerals are usually poor in iron, and the ores contained in white diopside skarn are chiefly bornite and chalcopyrite; the ludwigite content of the kotoite-marble around the ore body is relatively small. In the lower part, on and below the 300 m level, the associated minerals gradually become rich in iron, and the association of such minerals as pyrrhotite, cubanite, chalcopyrite, and valleriite can be seen in many places; at the 210 m level, the ore becomes chiefly cubanite and the bornite content decreases. At the 180 m level the kotoite-marble contains a large amount of ludwigite, the diopside skarn is tinged with green, and the iron content increases slightly.

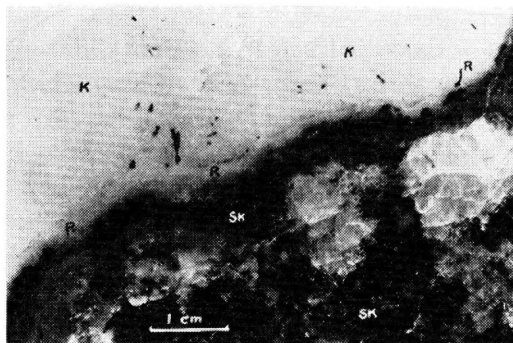


Fig. 5. Reaction Zone (R) between Diopside Skarn (SK) and Kotoite-marble (K).
(In this zone occur fluorborite, ludwigite and clinohumite.)

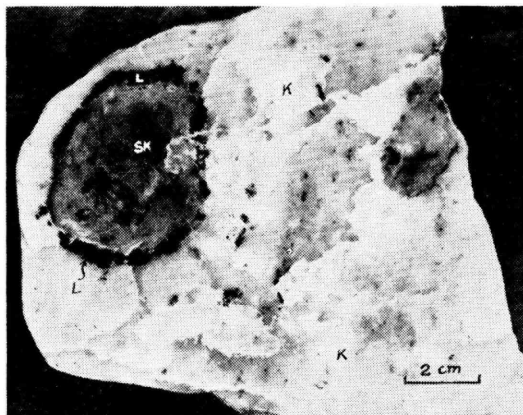


Fig. 6. Nodular Masses of Diopside Skarn (SK) with Ludwigite (L)-zone in the Kotoite-marble (K).

The relationship between the kotoite-marble and the diopside skarn, as already mentioned, is generally distinct; however, as seen in Figs. 5 and 6, a transition zone several millimeters wide occurs between them as if it were a kind of reaction zone. The mineral association in this zone differs from that of the kotoite-marble. It consists of fluoborite, szaibelyite, clinohumite, ludwigite, calcite, etc. The kotoite near this zone has been decomposed showing that the mineral was unstable at the time the zone was formed.

Judging from these relations, the writer thinks the kotoite-marble was formed earlier than the reaction zone and the formation of the skarn continued for some time after the formation of the kotoite-marble, because the diopside skarn has some branches which extend into the kotoite-marble. However, as the skarn body is always found within the kotoite-marble, they are closely related, and were possibly formed in succession, even though the stages during their formation may differ somewhat.

2. Kotoite-marble:

The kotoite-marble as described before, does not differ greatly in appearance from ordinary dolomite-marbles; however, it is somewhat hard and compact, and is difficult to crush even with a hammer. Because this rock appeared to be harder than ordinary dolomite, it was often said that the Holkol dolomite had been silicified around the ore bodies. The rock is generally white to pale yellowish brown, and sporadic black specks of ludwigite are often observed. In proportion as the clinohumite content of the rock increases, the yellowish brown color of the rock becomes progressively deeper, and brown to dark brown thin bands, com-

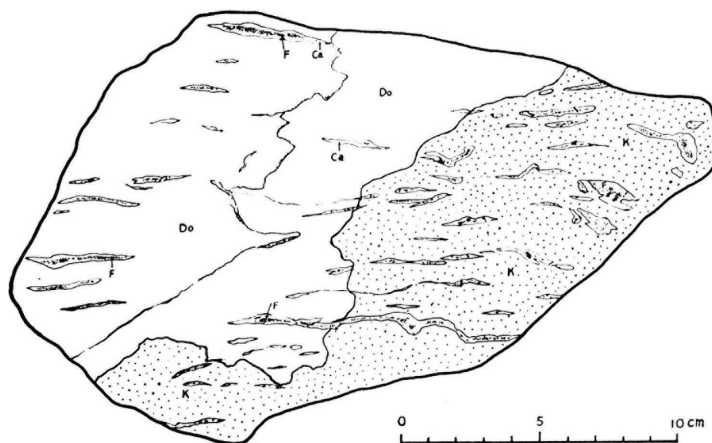


Fig. 7. Sketch showing the boundary between Kotoite-marble (K) and Crystalline Dolomite (Do).

Streaks and lenses consisting of forsterite and calcite in the dolomite-marble and kotoite-marble, corresponding to the original siliceous layers of the dolomite.

The forsterite has been formed by thermal metamorphism due to the reaction between impure silica and magnesia originally found in dolomite.

Near the forsterite is usually well developed newly formed calcite.

The kotoitization took place in the later stage of thermal metamorphism.

posed chiefly of forsterite, clinohumite, warwickite, and spinel are often found in the rock.

If the kotoite-marble is polished for a considerable length of time, the calcite of relatively low hardness may be rubbed off and the harder materials, such as grains of kotoite and tabular crystals of forsterite, will exhibit a distinct relief. From the polished specimen, one can perceive as shown in Fig. 7, that the forsterite of the kotoite-marble is distributed in the silica-rich impure zone of dolomite and, therefore, this mineral must have been formed during the recrystallization of the rock.

The boundary between kotoite-marble and the surrounding crystalline dolomite is often so indistinct that it is generally not traceable at underground; but it can clearly be observed on polished sections. Also observable is the boundary that cuts across the bedding planes, along which the forsterite is arranged. This suggests that the formation of forsterite was completed before the kotoite-marble was formed by pneumatolysis.

To distinguish between kotoite and calcite in kotoite-marble is almost impossible with the naked eye; however, if the rock had been exposed for a long time, the kotoite would be weathered and would change into a secondary mineral which is white and powdery, and could be recognized by careful observation, as innumerable fine spotty minerals (szaibelyite). Therefore, in order to confirm the presence of kotoite in kotoite-marble, it is necessary to observe a thin section under the microscope.

Under the microscope, the kotoite-marble exhibits a texture like that of penacite or predazite (WATANABE, 1935), which are known to be formed by the thermal metamorphism of dolomite or of periclase marble. But instead of periclase or brucite one finds a granular mineral which is about 0.1–0.15 mm in diameter, is highly birefringent, and is surrounded by a somewhat coarser mosaic of calcite.

This fine crystal, as mentioned previously, bears some resemblance to olivine or pyroxene, but a detailed investigation will disclose many differences between them. When the crystal is very fine, the cleavages are not distinct, but when somewhat coarse, it shows distinct cleavages on fairly thin sections, and extinction is straight and parallel to the *c*-axis. When the section is perpendicular to the *c*-axis, the cleavages (110) are clearly seen to cross each other at an angle of about 60° and the extinction is not parallel (Fig. 8).

Kotoite crystals are usually somewhat rounded, but hypidiomorphic crystals with developed faces are common. The arrangement of kotoite in kotoite-marble is shown in Figs. 8, 10, 11, and 12; a few dozen neighboring fine crystals are distributed with the same orientation and extinction, as can be seen almost simultaneously under crossed nicols. The space between the kotoite crystals is filled with calcite mosaic which is somewhat coarser than kotoite. The composing ratio of the two minerals measured on thin sections is not very different in any portion, *i.e.* the volumetric ratio of kotoite to calcite is nearly 2:7. This ratio shows a genetically restricted value, so the occurrence of a rock consisting completely of kotoite cannot be expected in the dolomite-contact zone. For the same reason, the

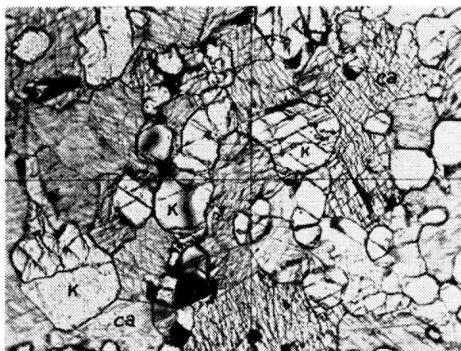


Fig. 8. Photomicrograph of the Kotoite-marble
(Notice the cleavage in kotoite grains.) $\times 60$.
K: Kotoite Ca: Calcite

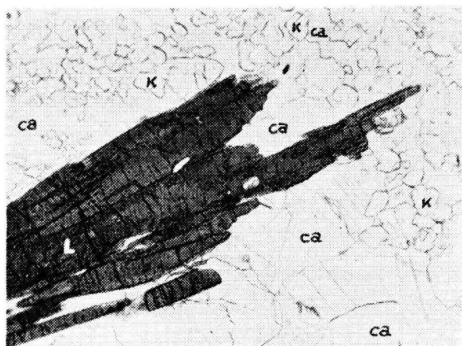


Fig. 9. Ludwigite in Kotoite-marble $\times 30$.
(In the area of calcite surrounding the ludwigite granular kotoite is not usually found.)



Fig. 10. Clinohumite in Kotoite-marble
($\times 90$, crossed nicols).
K: Kotoite Ca: Calcite h: Clinohumite

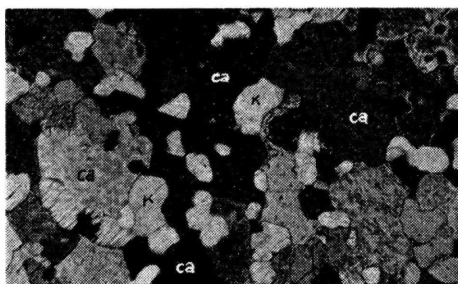


Fig. 11. Photomicrograph of Kotoite-marble from Holkol
($\times 90$, crossed nicols).
K: Kotoite Ca: Calcite



Fig. 12. Photomicrograph of Kotoite-marble from Rézbanya, Romania
($\times 90$, crossed nicols).
K: Kotoite Ca: Calcite Sz: Szaibelyite

volumetric ratio of periclase to calcite in periclase marble and that of brucite to calcite in pentacite or predazzite exhibit similar conditions. These ratios are also within the limits of 2:7 to 3:7.

In addition to the above-mentioned two minerals, several accessory minerals are contained in kotoite-marble. The important ones are forsterite, clinohumite, spinel, such borate minerals as ludwigite, suanite, warwickite, and a few other sulphide minerals. In order to study the composing ratios of minerals in kotoite-marble, two typical thin sections were selected, and the results of study by using the Rosiwal micrometric method are presented as (17) and (18) in Table 1. The results of the chemical analyses of kotoite-marble are also given in Table 1. These results, especially when comparing the values of (19) with those of the chemical analyses, show that the value for the kotoite-marble cannot be more than 14.19 per cent.

Each component mineral of the rock will be described briefly as follows:

Kotoite: This mineral is usually found as fine-grained and allotriomorphic crystals about 0.1 to 0.15 mm in diameter; attaining a maximum diameter of 0.5 mm. Occasionally it occurs as hypidiomorphic crystals with well-developed faces. Other properties of this mineral will be described later in detail.

Table 1. (a) Chemical Analyses of Kotoite-marble from Holkol.

No.	1	2	3	4	5	6	7	8	9
d	—	—	—	—	—	—	—	—	—
SiO ₂	3.56	3.38	2.56	14.24	3.60	2.46	3.64	2.71	2.24
TiO ₂	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.03	n.d.
Al ₂ O ₃	0.21	0.21	0.09	0.31	0.32	0.07	—	0.67	0.48
Fe ₂ O ₃	0.31	0.29	0.23	0.37	0.46	0.29	0.26	0.27	0.62
MnO	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.05	n.d.
CaO	34.10	34.72	33.50	34.02	34.24	34.42	33.94	33.23	33.43
MgO	21.28	20.30	21.62	14.52	21.20	22.68	21.14	22.66	24.32
B ₂ O ₃	11.98	10.59	14.63	7.36	12.02	12.71	12.39	12.49	12.28
K ₂ O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.05	n.d.
Na ₂ O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.63	n.d.
F	tr.	tr.	tr.	tr.	tr.	—	tr.	n.d.	n.d.
Ig. loss	27.82	29.78	27.10	28.38	27.42	27.90	27.54	27.41	26.76
Total	99.26	99.27	99.73	99.20	99.26	100.53	98.91	100.20	100.13

No.	10	11	12	13	14	15	16	17	18	19
d	—	—	—	—	—	—	2.86	2.87	2.86	2.85 (cal.)
SiO ₂	7.64	5.11	1.98	2.96	2.98	2.39	3.63	2.0	2.2	—
TiO ₂	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	—	—	—
Al ₂ O ₃	1.55	0.14	0.22	0.61	0.68	1.10	0.82	0.4	0.4	—
Fe ₂ O ₃	0.25	1.08	0.65		0.80	0.25	0.33	2.8	1.1	—
MnO	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	—	—	—
CaO	34.73	29.26	34.01	33.70	32.03	34.90	34.43	39.2	38.4	34.28
MgO	23.93	28.01	25.00	15.75	21.58	22.18	22.75	17.3	18.8	24.64
B ₂ O ₃	9.83	9.60	11.34	11.26	15.12	9.50	9.39	8.3	9.0	14.19
K ₂ O	n.d.	n.d.	n.d.	2.06	n.d.	0.42	n.d.	—	—	—
Na ₂ O	n.d.	n.d.	n.d.		n.d.		n.d.	—	—	—
F	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	—	—	—
Ig. loss	21.76	26.89	26.69	34.21	26.86	29.23	28.71	30.0	30.1	26.89
Total	99.69	100.09	99.89	100.55	100.05	99.97	100.06	100.00	100.00	100.00

(b) Mineral Composition of Kotoite-marble (Percentage by weight; calculated values)

No.	1	2	3	4	5	6	7	8	9	
Calcite	60.7	61.9	59.8	—	61.1	61.4	60.6	59.3	59.7	
Kotoite	32.5	28.8	33.8?	—	32.5	34.5	33.7	34.1	33.2	
Ludwigite	0.5	0.5	0.6	—	0.8	0.5	0.5	0.5	1.1	
Forsterite	8.3	7.9	6.0	—	8.4	5.8	8.5	6.3	5.3	
Spinel	0.3	0.3	0.2	—	0.4	0.1	—	0.9	0.7	
Total	102.3	99.4	100.4	—	103.2	102.3	103.3	101.1	100.0	
$\frac{\text{CaO}}{\text{MgO}}$ (Mol. rat.)	1.15	1.23	1.11	1.68	1.16	1.09	1.15	1.05	1.00	
No.	10	11	12	13	14	15	16	17	18	19
Calcite	49.5	61.2?	60.7	60.1	57.2	62.2	61.4	69.5	68.5	61.17
Kotoite	26.7	25.6	30.5	30.8	40.8	25.8	25.4	21.4	23.9	38.83
Ludwigite	0.4	1.9	1.2	—	1.4	0.4	0.5	3.2	1.9	—
Forsterite	17.9	12.0	4.6	6.9	7.0	5.6	8.6	5.3	5.1	—
Spinel	2.2	0.2	0.3	—	1.0	1.5	1.1	0.6	0.6	—
Total	96.7	100.9	97.3	97.8	107.4	95.5	97.0	100.0	100.0	100.0
$\frac{\text{CaO}}{\text{MgO}}$ (Mol. rat.)	1.04	0.75	0.98	1.55	1.06	1.13	1.09	1.63	1.46	1.00
(1) Kotoite-marble	182-m level, north ore body									
(2) Kotoite-marble	Do.									
(3) Kotoite-marble	North drift on 212-m level north ore body							According to		
(4) Kotoite-marble	(Contains impurities) 212-m level, north ore body							Yamazaki and		
(5) Kotoite-marble	No. 4 breast, 303-m level, north ore body									
(6) Kotoite-marble	No. 4 breast, 242-m level, north ore body									
(7) Kotoite-marble	No. 4 breast, 273-m level, north ore body									
(8) Kotoite-marble	North ore body; analyzed by the Ceramic Inst. of the Dept. of Commerce and Industry (According to Mr. Tetsuji ASAYAMA)									
(9) } Kotoite-marble	North ore body; according to Prof. Shōichiro NAGAI of the Engineering Dept., Tokyo Univ.									
(10) }										
(11) }										
(12) }										
(13) Kotoite-marble	1,000-shaku level, north ore body. (Analyzed by the Takeda Chōbei Shoten)									
(14) Kotoite-marble	North ore body; analyzed by the Inst. of the Hitachi Manufactory.									
(15) Kotoite-marble	(S. 1563); 1,000-shaku level, north ore body. (Analyzed by the Industrial Inst. of Osaka)									
(16) Kotoite-marble	(S. 260); north ore body; analyzed by the MATSUDA Inst.									
(17) Kotoite-marble	(S. 260); north ore body; computed value by ROSIHAL method. (10 shaku=3.03m)									
(18) Kotoite-marble	(S. 839); north ore body; computed value by ROSIHAL method									
(19) Theoretical value for kotoite marble ($\text{Mg}_3\text{B}_2\text{O}_6+3\text{CaCO}_3$) derived from pure dolomite ($\text{Mg, Ca}(\text{CO}_3)_2$)										

Calcite: This mineral composes the largest part of the rock. When it fills the space around the kotoite crystals, it occurs as mosaic crystals about 0.2 mm in diameter, i.e. about twice the size of the kotoite crystal. Around the crystals of such minerals as ludwigite and forsterite, the calcite tends to form a somewhat coarser mosaic, and in many cases no kotoite crystals are found among them.

Forsterite: This mineral usually occurs as fine-grained idiomorphic crystals flattened parallel to (100). Specific gravity=3.22. Under the microscope it shows straight extinction, and the refractive indices, measured by the immersion method, are: $\alpha=1.638$, $\beta=1.651$, $\gamma=1.669$; $\gamma-\alpha=0.031$; (+)2V=84°. These values indicate that the mineral almost belongs to the forsterite endmember. It is distributed within kotoite-marble, assuming such forms as lenses, fine bands, and layers. It is also often found in association with clinohumite.

Clinohumite Minerals: The minerals of the chondrodite group are irregularly distributed in the rock; however, they may occur as veinlets in association with forsterite, spinel, and calcite. When the rock is relatively rich in these minerals, it is pale yellow or pale yellowish brown. Study of the optical and other properties of most of the minerals of this group shows that it corresponds to clinohumite ($4\text{Mg}_2\text{SiO}_2 \cdot \text{Mg}(\text{F}, \text{OH})_2$). The clinohumite occurs as somewhat coarser crystals than the kotoite and forms idiomorphic crystals, most of which are about 0.2 to 1.0 mm in diameter. Specific gravity=3.20. Optical properties are: $\alpha=1.625$, $\beta=1.631$, $\gamma=1.655$; $\gamma-\alpha=0.030$; (+)2V=71°; extinction angle (angle between the twinning plane and X')=9-10°; more or less pleochroic, changing from colorless to pale yellow; however, colorless crystals may also be found among the thin sections. The minerals of this group are characterized by their lamellar twinning, as shown in Fig. 10.

Spinel: This mineral is found in only a few places in kotoite-marble. Since it is fine-grained and sporadically distributed, it may be overlooked. The crystals are generally idiomorphic and occur mostly in octahedral form. However, a spinel twinning is also frequently found.

The spinel, in some cases, forms irregular brown layers several millimeters to several centimeters wide. It is associated with forsterite, clinohumite, ludwigite, etc. in kotoite-marble. The spinel found in the kotoite-marble composing the upper part of the North ore body, where little ludwigite is found associated with it, is colorless in thin section; when concentrated by using heavy solution the crystals are pale pink. The spinel in the lower part of the North ore body, where much iron-rich ludwigite is associated with it, is somewhat greenish, even in thin section.

The crystals are optically isotropic, and the refractive index of the colorless ones is $n=1.718$; the measured specific gravity is 3.54.

Ludwigite ($(\text{Mg}, \text{Fe}^{+2})_2\text{Fe}^{+3}\text{BO}_5$): This mineral is a borate mineral which has long been a subject of discussion. Prof. Kōtō described it as lievrite (ilvaite), because its optical properties in thin section bear a close resemblance to those of lievrite. After that, D. F. HIGGINS (1918) regarded it as an iron-rich pyroxene, and

gave it a new name, "collbranite." Still later, E. V. SHANNON (1921, A,B) confirmed it to be ludwigite.

The mineral appears black, and often occurs as a radial aggregation of acicular crystals several millimeters, occasionally 2–3 cm, in length. It is scarcely contained in the kotoite-marble of the upper part of the north ore body. As shown in Fig. 2 it forms black minutely striated zones which appear as if they were the bedding planes of the original rock.

This mineral also occurs sporadically in kotoite-marble, and is especially concentrated on and near the 182 m level in the lower part of the North ore body. It is also found in abundance in the kotoite-marble of both the East and the West ore bodies.

Under the microscope, the thickest part of the thin section of this mineral is opaque but the thinnest part is semi-transparent and distinctly pleochroic, being deep brown parallel to the elongation of the crystal (z'), and deep grass green perpendicular to it (x'); absorption $z' > x'$; straight extinction. As shown clearly in Fig. 9, the ludwigite crystals in kotoite-marble are usually surrounded by calcite that is not associated with kotoite.

Suanite ($2\text{MgO} \cdot \text{B}_2\text{O}_3$ or $\text{Mg}_2\text{B}_2\text{O}_5$): This, a new mineral, (WATANABE, 1953) is a magnesium borate mineral which occurs extensively in the kotoite-marble of the North ore body. As the mineral appears white, it can barely be seen with the naked eye, but it can clearly be recognized by its silky luster when it occurs as an aggregate of fibrous bundles several millimeters long. In a part of the east ore body, this mineral occurs as somewhat large crystals in kotoite-marble which contains ludwigite. Some of them attain 2–3 cm in length, but the mineral has long been undetected due to its lack of color.

Under the microscope, this mineral is observed to be coarser than the nearby kotoite, and aggregates radially exhibiting very high birefringence. The direction of x' is parallel to the elongation of the crystal; $\alpha = 1.596$, $\beta = 1.639$, $\gamma = 1.670$; $(-)$ $2V = 70^\circ$; $r > v$; specific gravity = 2.91. Chemical tests have clearly indicated the presence of Mg and B and X-rays method has confirmed that this mineral differs from similar known borate minerals such as camsellite and szaibelyite has already been confirmed by X-ray.

The sample from the east ore body was analyzed by Mr. Kiyoshi ISONO through the kindness of Dr. YAMANE, the former director of the Geological Survey of Japan. The results of the analysis are given in Table 2.

Since the analyzed sample clearly contained small quantities of calcite and forsterite as impurities, the chemical composition of the mineral must be considered by removing CaO , CO_2 , SiO_2 , etc. from the results of the chemical analysis. The value for H_2O is obtained by deducting the value for CO_2 from that for ignition loss, so the mineral must be examined to find out whether this was part of its original content of the mineral or not. However, this mineral can be thought to have a composition of almost $2\text{MgO} \cdot \text{B}_2\text{O}_3$, as shown in Table 2.

Warwickite ($3(\text{Mg}, \text{Fe})\text{O} \cdot \text{TiO}_2 \cdot \text{B}_2\text{O}_3$): (WATANABE, 1954) A brown, very

fine-grained mineral, this occurs as spots in the kotoite-marble. When these "spots" were observed under the microscope, they were found to be an aggregate of fine acicular crystals 0.2–0.5 mm in length. x' is parallel to the elongation of the crystal; pleochroism is more or less observable as x' =pale yellow and z' =light brown; the index of refraction is very high.

Table 2. Chemical Analysis of Suanite From Holkol.
(Analysed by K. Isono, Geol. Survey, Dept. of Commerce and Industry)

	I	II	III
SiO ₂	0.70%	-%	-%
Al ₂ O ₃	0.97	-	-
Fe ₂ O ₃	0.33	-	-
MgO	46.48	54.89	53.66
CaO	3.70	-	-
Na ₂ O	0.90	-	-
K ₂ O	0.00	-	-
B ₂ O ₃	38.20	45.11	46.34
CO ₂	5.70	-	-
H ₂ O \pm	3.50	-	-
	100.48	100.00	100.00

- I: Chemical analysis of suanite; the sample contains a few impurities of calcite, spinel, and forsterite.
 II: Calculated values for MgO + B₂O₃ in the chemical analysis of suanite reduced to 100.
 III: Theoretical values for 2 MgO • B₂O₃ (by weight percentage)

Though this mineral is widely distributed in the kotoite-marble, its properties were hard to confirm because of the fineness of the crystal and the difficulty in separation.

Since warwickite occurs only in kotoite-marble and not in the surrounding dolomite, it was also assumed to be a kind of borate mineral. An X-ray photograph of the powder was prepared by using a minute amount of the sample separated from the rock, and this was identified as warwickite. This was only the second occurrence of warwickite which had been found in the world.

Associated sulfide minerals: Large amounts of opaque metallic minerals are generally contained in the diopside skarn, although very fine spotty sulfides are also found in kotoite-marble. Under the reflecting microscope, two types of mineral association can be seen. When there is little iron bornite, chalcopyrite, sulphosalts of copper and bismuth, and native bismuth are observed; when there is much iron, such minerals as pyrrhotite, chalcopyrite, cubanite, valleriite, bismuthinite, native bismuth, and native gold are present.

Chemical composition: Since kotoite-marble first attracted attention as a boron ore, many chemical analyses of it have been completed. The data which the writer received are listed in Table 1, and the mineral composition computed with them is given as (2) in Table 1.

The table shows that the rock has a remarkably well-defined chemical composi-

tion. Kotoite-marble is very narrowly defined in composition, samples were collected from various parts of the ore body, because the rock is derived from relatively pure dolomite supplied with boron; its outstanding characteristic of the rock. By considering the constituent minerals detected under the microscope, one should be able to compute the composing ratio of the minerals. According to these results, the ratio is almost constant in all specimens.

3. Reaction zone between kotoite-marble and diopside skarn: The boundary between the skarn, which is mined as gold and copper ore, and the surrounding kotoite-marble is generally very distinct. Although a narrow reaction zone occurs between them, the mineral association in this zone differs somewhat from those of the other parts. Microscopic examination of the rock of this zone

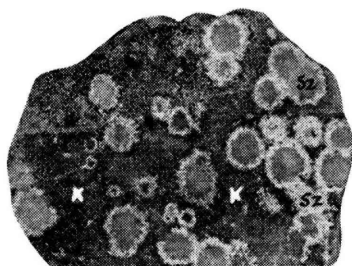


Fig. 13. Polished Surface of Kotoite-marble Containing Spherical Masses of Szaibelyite from Rézbánya, Romania $\times 1.6$.

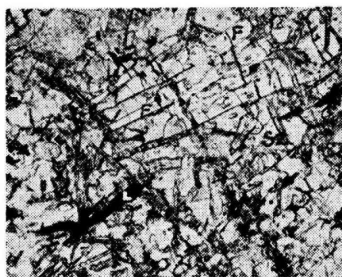


Fig. 14. Photomicrograph of the Reaction Zone between Kotoite-marble and Diopside Skarn, Holkol $\times 30$.

F: Fluorborite
L: Ludwigite
Sz: Szaibelyite

shows that it retains the original texture of kotoite-marble; but the kotoite has already decomposed and faded away, and, as shown in Fig. 14, other borate minerals—fluorborite, szaibelyite, and ludwigite—come out in place of it and the silicate minerals, especially those of the chondrodite group increase. This obviously suggests that the formation of the kotoite-marble preceded that of the diopside skarn and the special association of such minerals may have been established by obtaining a supply of F, OH, etc. which makes the kotoite unstable on the surface of the diopside skarn. Each of these minerals are described briefly below.

Fluorborite ($Mg_3B_2O_6 \cdot 3Mg(OH, F)_2$): This mineral occurs as prismatic crystals 0.5–1 mm long. The section perpendicular to the axis is clearly hexagonal. The crystal is optically uniaxial and negative; $\omega=1.550$, $\varepsilon=1.522$, $\omega-\varepsilon=0.028$; that *i.e.* it shows almost the same properties as the fluorborite which was discovered by GEIJER (1926) in the ludwigite-bearing rock from Sweden.

Szaibelyite (HMgBO_3): This mineral also occurs as colorless and very fine acicular crystals rarely larger than 0.2 mm long. The birefringence measures as high as 0.074, and $x' = 1.650$, $z' = 1.576$; x' is parallel to the direction of the elongation of the crystal.

Other minerals: Other minerals, c.g. ludwigite, clinohumite, and calcite which occur in the same reaction zone, are nearly equal to those already described, so detailed descriptions of them have been omitted.

B. Kotoite from Rézbánya, Romania (KOCH, 1888, PETERS, 1861)

Various kinds of ore deposits are known from Transylvania, or Siebenbürgen, in the Balkans. Rézbánya, especially, has long been known for the occurrence of various kinds of minerals, as well as copper and zinc deposits. These minerals and copper-zinc deposits are found where the Mesozoic limestone is in contact with diorite and green dikes. A kind of marble of special appearance, which was collected by SZAIBLYI, a mining engineer, has long been known to mineralogists, and has been studied extensively by PETERS and others. PETERS found a fibrous mineral in the rock and named it *szaibelyite*, and by chemical analysis STROMEYER showed it to be a kind of borate.

While he was studying borate minerals in Berlin, the writer found a mineral corresponding to *szaibelyite* among the component minerals of the rock from Holkol. In order to compare it with the *szaibelyite* from Rézbánya, he obtained a piece of the Rézbánya specimen through the kindness of J. SCHROETER who was at the Berlin Technical University at the time. This specimen, as shown in Fig. 13, was a marble containing spherulites about 3–4 mm in diameter, corresponding to PETERS' old description, that is, the specimen was judged to be similar to that studied by PETERS.

Under the microscope, the spherulites are seen to be chiefly of *szaibelyite*; the outer part of the spherulites consists of a somewhat longer fibrous mineral than the inner part, and the former crystals radiate from the surface of the latter and are cemented by calcite.

Both the fibrous minerals constituting the outer and the inner parts of the spherulites are obviously of the same kind, that is, the two have the same refractive indices: $\gamma' = 1.651$, $\alpha' = 1.578$, and x' is found equally in the direction of elongation. Needless to say, this is the same mineral that was studied and called *szaibelyite* by PETERS and investigators after him.

However, the marble-like part surrounding the spherulite was microscopically seen to contain a granular mineral which is associated with *szaibelyite* and a few opaque minerals. This granular mineral, as shown in Fig. 12, has distinct cleavage and straight extinction, small 2V, and is characterized by the simultaneous extinction of several crystals. Since its properties are quite similar to those of the Holkol kotoite, the mineral was studied in detail optically; then it was also separated from calcite with dilute hydrochloric acid and studied by chemical and X-ray methods. The results of these studies proved this mineral to be kotoite.

The mineral composition of the marble-like part containing kotoite in the Rézbánya specimen was studied by using the ROSIWAL method. The results are shown in Table 3.

Table 3. Mineral Composition of the Marble like Part Containing Kotoite in the Rézbánya Specimen.

	Sp. gr.	Volume, %	Weight, %
Calcite	2.71	81.0	79.1
Kotoite	3.06	16.7	18.5
Szaibelyite	2.70	2.0	2.1
Sulfides (Chiefly sphalerite)	4.0	0.3	0.3

The lower kotoite content of this rock, as compared with that of the kotoite-marble from Holkol, may be attributed to the decomposition and change into szaibelyite of some kotoite.

Thus, kotoite was discovered almost simultaneously in specimens from both Holkol and Rézbánya. The confirmation of this mineral enables us to solve various problems which were not fully clarified earlier when PETERS and STROMEYER (1863) studied szaibelyite. For instance, the values reported for the chemical analysis of szaibelyite should have aroused suspicion if they were a mixture of szaibelyite and kotoite, because STROMEYER did not recognize the existence of kotoite in his specimen, though it certainly contained it. The meaning of previous chemical analyses may be understood if they are examined again from such a point of view.

In short, the kotoite of Rézbánya is found in the country rock of a contact deposit as it is in Holkol and similarly forms kotoite-marble which contains szaibelyite. Similar conditions probably generated both cases.

3. Properties of Kotoite

A. Chemical Composition

As already described in the section on mode of occurrence, collection of a pure sample of kotoite is difficult. However, as kotoite is not very soluble in cold hydrochloric acid, it will be left in the residue when the kotoite-marble is treated with cold dilute hydrochloric acid, while the calcite is dissolved. If 100 gm of kotoite-marble are treated, about 30 gm of residue will be obtained, and many fine grains of kotoite will be found in it, though it may be mixed with many other insoluble minerals.

In treating the sample with acid, the original specimen should not be finely

crushed; little pieces should be used. When such pieces are dissolved, the forms of the relatively coarser minerals—forsterite, clinohumite, ludwigite, and suanite—will remain unchanged.

During the first step, the writer used a 0.15 mm sieve to separate the coarser minerals from the fine colorless sandy grains. Under the microscope, this sandy material was observed to consist mainly of kotoite grains, though mixed with a few fine crystals of forsterite, spinel, and other borate minerals. During the second step, in order to reject these fine impurities, he prepared Clerici heavy solutions, specific gravity 2.95 and 3.2 and attempted to collect pure kotoite by using a centrifugal separator. The sample remaining within the limit of specific gravity between 2.95 and 3.2, was composed almost entirely of pure kotoite. As further separation was considered quite difficult, this same sample was used for chemical analysis.

The sample was first tested for the presence of boric acid, iron, magnesia, etc., and then was carefully fused with sodium carbonate. During the course of the quantitative analysis, special care was taken to separate the boric acid, and afterwards, the quantity of silica, alumina, iron oxide, lime, and magnesia were measured in the ordinary way. The ferrous oxide and water contents of another sample were measured; the boric acid was separately measured by the Chapin method. The results are shown in Table 4, of which SiO_2 , Al_2O_3 , and Fe_2O_3 are considered to have been derived from the impurities. They may be divided and distributed between forsterite and spinel. If these are deducted from the molecular ratio, we arrive at the following:

$$(\text{Mg, Fe, Ca})\text{O}:\text{B}_2\text{O}_3 = 1.5209 : 0.5055 = 3:1$$

As the value for $\text{FeO} + \text{CaO}$ measures only about 1 per cent of the MgO content, the formula for the Holkol kotoite can be stated as $3\text{MgO} \cdot \text{B}_2\text{O}_3$.

Table 4. Chemical Analysis of Holkol Kotoite.

	Anal. value (%)	Mol. ratio	$-0.0220 \text{ Mg}_2\text{SiO}_4$ $-0.0039 \text{ Mg (Al, Fe)}_2\text{O}_4$	Ratio	$\text{Mg}_3\text{B}_2\text{O}_6$ Theo. value (%)
SiO_2	1.32	0.0220	—	—	—
Al_2O_3	0.26	0.0026	—	—	—
Fe_2O_3	0.20	0.0013	—	—	—
FeO	0.61	0.0085	} 1.5209	3.00	63.46
CaO	0.18	0.0032			
MgO	62.78	1.5571			
B_2O_3	35.20	0.5055	0.5055	1.00	36.54
$\text{H}_2\text{O}^{+110^\circ}$	0.05	0.0028	—	—	—
Total	100.60				100.00

B. Form and Physical Properties

Kotoites from both Rézbánya and Holkol occur as very fine grains. The crystal form has not been clarified, though it can usually be recognized as allotriomorphic or hypidiomorphic; the crystal grains are 0.1–0.5 mm in diameter. The hypidiomorphic crystals seem to have well-developed pyramids or domes, and they are occasionally found to be somewhat elongated in the direction of the *b* axis. Distinct cleavage is found parallel to axis *c*, and in some cases straight extinction is observable. These characteristics may serve to identify this mineral. In the section perpendicular to the *c* axis, a pair of cleavages parallel to (110) which cross each other at an angle of about 65° can be seen.

A study on the crystal form of artificial kotoite was prepared by EBELMAN with measurements by MALLARD in 1887. According to MALLARD, the crystal belongs to the orthorhombic system, and crystal planes such as (110), (011), (101), (403), and (100) occur. MALLARD also computed the axial ratio $a:b:c=0.6412:1:0.5494$ by using the values $(110):(110)=65^\circ 20'$ and $(110):(011)=74^\circ 56'$; and he recognized that the cleavages on (110) are distinct. The writer also tried to synthesize artificial kotoite; he obtained a somewhat large crystal, but it did not have good form. By using the cleavage planes of this crystal, the writer determined the value $(110):(110)=65^\circ 20'$. A native crystal shows almost the same value. A noteworthy property of kotoite is the formation of secondary twinning by shock; that is, when the refractive index of kotoite is measured by the immersion method, the sample is usually crushed and lamellar twinning may often be observed. This property, which is also found in artificial kotoite, comes from the phenomenon of the so-called *Einfache Schiebung*, or slipping in a certain direction within kotoite. In order to study this direction, the writer cemented the mineral particles with bakelite and observed a thin section by using the Leiz universal stage. By studying the various relations between the optical orientation and other properties, he confirmed the angle between *C* and the direction perpendicular to the twinning plane as about 40°. He also measured the angle between the twinning plane and the cleavage plane and obtained the value 56° for it. On comparing these values with those measured from artificial kotoite, that is $(101):(001)=40^\circ 36'$, $(101):(110)=56^\circ 48'$, the former was found to almost correspond with the latter, and, hence, the twinning plane is assumed to be possibly (101). Such a twinning plane can be seen in very thin sections, although only in very rare cases.

The mineral is relatively hard. It was measured by rubbing the fine grains, which were collected from the rock, on the standard test plate; artificial kotoite was also used for this purpose, and its hardness was determined to be almost $6\frac{1}{2}$.

When a polished section of kotoite-marble is observed under a reflecting microscope, the relative hardness of kotoite can be perceived from the relief of the polished surface; it is equal to forsterite, much higher than calcite, and lower than spinel.

The specific gravity of the mineral was determined by placing the fine grains or crystals in Clerici heavy solution and measuring the specific gravity of the Clerici

solution at the point in which the suspended mineral grains remained in a state of equilibrium.

Thus, the following values were obtained: 3.106 for the kotoite from Holkol, 3.07–3.10 for that from Rézbánya, and 3.092 for artificial kotoite. The specific gravity of the old artificial kotoite measured by MALLARD was 2.987. This value is somewhat lower than that given above, so it is assumed that the old artificial kotoite may have been mixed with some other crystals.

C. Optical Properties

The pure separated grains of kotoite are colorless and transparent. The crystal is biaxial and optically positive under a conoscope, however, and occasionally appears almost uniaxial because of the smallness of $2V$. Therefore, under the microscope it closely resembles pyroxenes of small $2V$ such as pigeonite and enstatite. Optical measurements of the kotoite from the two districts and of the artificial kotoite are well in accordance with one another, as shown in Table 5.

Table 5. Optical Properties of Kotoite.

Name	WATANABE	WATANABE	WATANABE	WATANABE	MALLARD
Method	Immersion	Immersion	Prism	Immersion	Prism
Specimen	Holkol	Rézbánya	Artificial; cleavage flake	Artificial	Artificial; parallel to C
Color	Colorless	Colorless	Colorless	Colorless	Colorless
α	1.652	1.652	1.6514	1.652	1.6527
β	1.653	1.653	1.6521 (Calc.)	1.652	1.6537
γ	1.673	1.674	1.6725	1.673	1.6748
$\gamma-\alpha$	0.021	0.022	0.0211	0.021	0.0221
$2V$	$21^\circ (+)$	small (+)	$22^\circ (+)$		$24^\circ 30' (+)$ (calc.)
$2E$	36° (calc.)		38° (calc.)		$43^\circ 18'$ (obs.)

Dispersion $r > v$
Optical plane (010)

Orientation $z = c$
Extinction $z' // \text{cleavage plane}$

D. General Physicochemical Properties

An experiment to determine the melting point of artificial kotoite was made by the writer under Prof. ERTEL at the Kaiser Wilhelm Silicate Institute, but no convincing results were obtained, probably because of the volatility of B_2O_3 . Judging from the temperature curve obtained during the experiment, the writer determined that the melting point of this mineral was near $1,340^\circ C$.

When the Holkol kotoite is highly heated in air, it becomes somewhat yel-

low. It is easily soluble in warm, concentrated hydrochloric acid, although it is almost insoluble when the acid is dilute. The presence of iron, magnesium, boron, etc. can be detected in this acid solution.

Synthesis of artificial kotoite: EBELMAN (1900) succeeded in synthesizing artificial kotoite as early as 1851. He placed magnesia and boric acid anhydride in a stone crucible and fused them into magnesium orthoborate, or artificial kotoite. Both the crystallographic and optical properties of this crystal were studied by MALLARD in 1887. In 1904, GUERTLER (1904, 1909) synthesized the borates of alkali earth metals and studied them systematically, clarifying the equilibrium relations in the $\text{MgO-B}_2\text{O}_3$ system to some extent. In this system, he recognized the formation of such borates as $\text{Mg}_3\text{B}_2\text{O}_6$ ($3\text{MgO}\cdot\text{B}_2\text{O}_3$) and $\text{Mg}_2\text{B}_2\text{O}_5$ ($2\text{MgO}\cdot\text{B}_2\text{O}_3$).

The writer also obtained a good crystal of artificial kotoite during his experiments at the University of Berlin. To do so he placed a mixture of 1.27 gm MgO and 0.73 gm B_2O_3 in a small platinum crucible and heated it at about $1,200^\circ\text{C}$ in a small electric furnace.

A portion of the crystal was quite pure, but the rest contained a few crystal grains of periclase and $\text{Mg}_3\text{B}_2\text{O}_6$. The pure portion was isolated and used for X-ray study and measurement of optical properties.

Kotoite is a relatively stable mineral, but it is unstable under hydrothermal conditions or when it is exposed to the air; it weathers and decomposes easily. In the thin sections of a weathered specimen, one can observe the secondary fibrous minerals which are arranged parallel to the *b* axis. On weathered surfaces of kotoite-marble a powdery mineral is observed to have formed secondarily from the kotoite. These secondary minerals were collected from the rock and examined under the microscope, and the presence of a mineral corresponding to szaibelyite confirmed. Kotoite alters partly to brucite.

E. Results of the X-ray Study of Kotoite

An X-ray study of the Holkol kotoite was made primarily to compare it with pyroxene. As it was later clarified as a borate, the X-ray photograph was used to determine whether it coincides with that of the artificial borate $\text{Mg}_3\text{B}_2\text{O}_6$. The X-ray photographs were prepared by the powder method, and the three photographs of the carefully purified kotoite from Holkol, the kotoite from Rézbánya and the artificial crystal were found to coincide perfectly, as shown in Fig. 15. The diameter of the camera used for the photographs was 57.3 mm; the X-ray tube was fitted with an iron anticathode; β -ray was not removed. The grain size of the sample was 0.3 to 0.4 mm.

The writer studied the artificial crystal by means of the rotation method under the guidance of Dr. STRUNZ. The same specimen was later studied by SADANAGA, and others of the Mineralogical Institute of the University of Tokyo, who also analyzed the structure of the crystal. The results obtained by these Institute researchers, together with the writer's, are cited here through the kindness of SADANAGA.

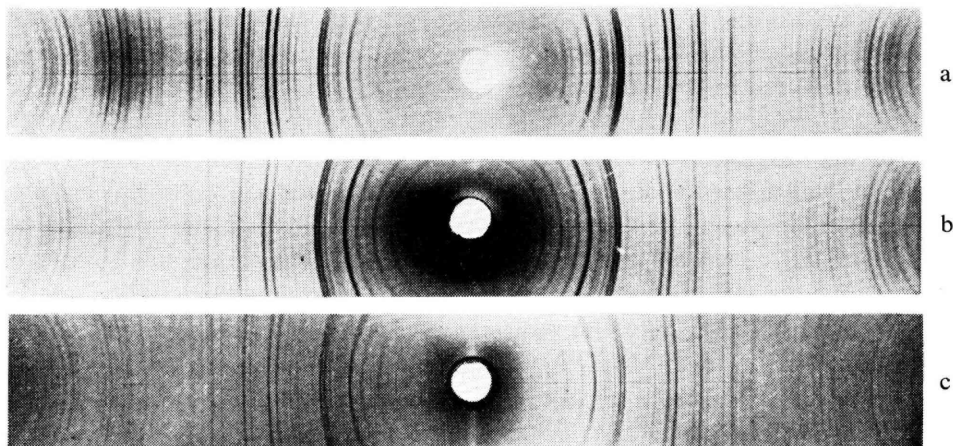


Fig. 15. X-ray Diffraction Patterns of Kotoite and Artificial Magnesium Orthoborate.

- a. Kotoite, Holkol
- b. Kotoite, Rézbánya, Romania
- c. Artificial kotoite ($\text{Mg}_3\text{B}_2\text{O}_8$)

Lattice constants (WATANABE)

$$a_0 = 5.41 \text{ \AA}$$

$$b_0 = 8.42 \text{ \AA}$$

$$c_0 = 4.51 \text{ \AA}$$

$$a_0 : b_0 : c_0 = 0.642 : 1 : 0.536$$

$$a : b : c \text{ (artificial crystal)}$$

$$= 0.6412 : 1 : 0.5494 \text{ (MALLARD)}$$

$$Z = 2$$

Space group : Undetermined

(SADANAGA)

$$a_0 = 5.38_5 \text{ \AA}$$

$$b_0 = 8.40_0 \text{ \AA}$$

$$c_0 = 4.48_7 \text{ \AA}$$

$$a_0 : b_0 : c_0 = 0.646 : 1 : 0.534$$

$$Z = 2$$

D_{2h}^{12} —Pnmm

The study on the crystal structure of kotoite was reported later by SADANAGA (1948).

4. Considerations on the Origin of Kotoite

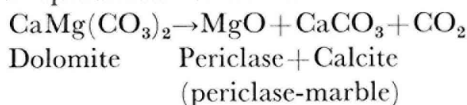
As clarified in the foregoing descriptions, kotoite occurs in a special country rock of a contact deposit. In both occurrences at Holkol and Rézbánya, kotoite is associated with calcite and forms a kind of marble which may be called kotoite-marble. The mode of occurrence of this rock at Rézbánya has not been observed by the writer. However, according to his observations at Holkol, it is found only in the dolomite of the region, and, therefore, may be considered genetically closely related to this dolomite.

The dolomite near the Holkol deposit has undergone thermal metamorphism over a considerably wide area and has changed into crystalline dolomite (dolomite marble), while a part of the rock very near the contact has changed into brucite-marble. The crystalline dolomite adjacent to the kotoite-marble is chemically almost pure dolomite, as $\text{SiO}_2 = 2.5 - 2.7\%$, $\text{Al}_2\text{O}_3 = 1.0$ to 1.5% , $\text{Fe}_2\text{O}_3 = 0.3\%$,

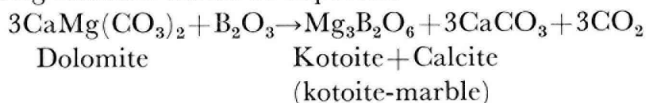
CaO=30–32%, MgO=20–21%, and CO₂ (or ignition loss)=about 42–45%. The molecular ratio of CaO to MgO is within the limit of roughly 1.0 to 1.2. Mineralogically, crystalline dolomite is composed chiefly of mineral dolomite and contains accessory forsterite (occasionally chondrodite), spinel, and calcite.

The chemical composition of kotoite-marble is observed to be roughly equal to dolomite in content of SiO₂, Al₂O₃, and Fe₂O₃; the molecular ratio of CaO to MgO is mainly within the limit of 1.0 to 1.2; however, the CO₂ content decreases 26–28% in exchange for B₂O₃, of which as much as 9–12% can be contained. Mineralogically, there is a marked difference between the two rocks; that is, no mineral dolomite is found in the kotoite-marble but, instead, calcite and kotoite accompanied by a few other minerals. Therefore, the process of the formation of kotoite-marble from dolomite can be explained very easily, if the minor constituents are not considered.

That is, it is necessary in the formation of kotoite-marble from dolomite that much CO₂ be expelled from the latter and that new B₂O₃ enter the rock. In general, dolomite becomes periclase-marble when it undergoes intense thermomorphism. This may be represented as follows:



The dolomite undergoes dissociation, and MgCO₃ is decomposed into MgO and CO₂. A chemical reaction similar to this can be postulated for the formation of kotoite-marble. If pure dolomite acquired a sufficient amount of B₂O₃ under high temperature, the following reaction would be expected.



That is, the magnesium carbonate in the dolomite dissociates into MgO and CO₂, and B₂O₃ combines with MgO to form kotoite. If such a relationship were to exist, pure kotoite-marble derived from pure dolomite should have a composition of Mg₃B₂O₆+3CaCO₃. Working from such a postulate, the writer computed the chemical composition of pure kotoite-marble; the results are given in the last line of Table 1. It is a matter of interest that these results are nearly coincident with those of the chemical analysis of the kotoite-marble from Holkol. This proves that the formation process of kotoite-marble may be represented by the reaction given in the above formula. Such a process can be termed "kotoitization." In this case, the difference in composition between natural kotoite-marble and "pure" kotoite-marble may come from the fact that the original dolomite is mixed with impurities and, therefore, the molecular ratio of CaO to MgO is not exactly one-to-one.

As previously mentioned, the dolomite had undergone thermal metamorphism over an extensive area before it changed into kotoite-marble. If the original rock contained impurities like SiO₂ and Al₂O₃, these might combine first with MgO from the dolomite molecule, such minerals as forsterite, spinel, calcite, etc. would form,

and the remaining MgO in the mineral would form crystalline dolomite. If kotoitization took place after that, the mineral dolomite which remained in the original rock would change into kotoite, and the kotoite content of the resultant would be less than that of "pure" kotoite-marble. Therefore, the value for B_2O_3 should also be less than 14.19%, the amount thought to be contained in "pure" kotoite-marble. In many cases, the ratio of CaO to MgO in natural dolomite is more or less larger than one-to-one. Since kotoite content, and therefore the B_2O_3 content, will become less than those of "pure" kotoite-marble, 14.19% may be said to be the maximum value for B_2O_3 that kotoite marble can possess. Although it is not clear in what state B_2O_3 was supplied, it is very possible that materials like B, F, and Cl emanated from the magma and migrated into the surrounding rocks when the volatile matter—especially those elements mentioned above—was concentrated in the residual magma during the pegmatitic to pneumatolytic stages of the consolidation of granite magma. Many contact minerals or contact deposits are thought to have been formed in such a manner. If the surrounding rock was dolomite and the magmatic emanation was rich in B_2O_3 , the formation of kotoite-marble from dolomite logically resulted in kotoitization.

SiO_2 , K_2O , and other metallic elements are often contained also in the magmatic emanation and form the Holkol gold-copper-bearing diopside skarn. If the original dolomite is almost pure, the resulting kotoite-marble will also be almost pure, but if it contains a small amount of SiO_2 , Al_2O_3 , etc., forsterite-spinel-bearing kotoite-marble will be formed. When the original rock is somewhat rich in iron, ludwigite is found in place of kotoite and gives rise to ludwigite-bearing kotoite-marble. If the original rock is much richer in iron, or if the rock is supplied with excessive iron in addition to B_2O_3 , kotoite will not be found in the rock, and magnetite and ludwigite are present instead (GEIJER, 1928, 1939). This means that kotoite and magnetite cannot occur in association with each other.

Summary

1. In 1930–32, while studying the geology and ore deposits of Holkol mine in Suan, a gold mining district in Korea, the writer noticed the occurrence of a very fine granular mineral in the country rock of the deposit; and later the writer proved it to be a new mineral which has a chemical composition of $Mg_3B_2O_6$, and which had never before been found in nature. He named it kotoite in memory of Professor Bunjirō Kotō. Almost simultaneously, the writer recognized that an identical mineral is contained in the szabilyite-bearing rock from Rézbénya in Transylvania in the Balkans.

2. Kotoite has the following properties:

Chemical formula: $Mg_3B_2O_6$

Chemical composition of the Holkol kotoite: $SiO_2=1.32\%$, $Al_2O_3=0.26\%$, $Fe_2O_3=0.20\%$, $MgO=62.78\%$, $FeO=0.61\%$, $CaO=0.18\%$, $B_2O_3=35.20\%$, $H_2O=0.05\%$, Total=100.60%; Theoretical value:

$\text{MgO}=63.46\%$, $\text{B}_2\text{O}_3=36.54\%$

Form: Orthorhombic system

Mode of occurrence: The mineral is found in a kind of marble (kotoite-marble) which was formed from dolomite by pneumatolytic replacement. It always occurs as fine granular microscopic crystals, hardly distinguishable by the naked eye.

Physical properties: Cleavage is perfect and parallel to (110); lamellar twinning is produced upon striking the crystal, and the plane of this twinning is assumed to be on (101); $H=6.5$; $D=3.11$ (Holkol specimen)

Optical properties: Colorless; refractive index, $\alpha=1.652$, $\beta=1.653$, $\gamma=1.673$, $\gamma-\alpha=0.021$ (Holkol specimen), $(+)2V=21^\circ$, $r>v$, optical plane parallel to (010), and $z=c$

Physicochemical properties: Can hardly be fused by means of a blowpipe.

The melting point is approximately $1,340^\circ\text{C}$; the value has not been determined exactly because of the presence of volatile matter such as B_2O_3 . The mineral is relatively unstable when it is placed in hydrothermal conditions or exposed to air, and it alters to fibrous borate minerals. The mineral is almost insoluble in cold hydrochloric acid, but is easily soluble in warm concentrated acid. An artificial crystal can easily be obtained by direct fusion of MgO and B_2O_3 when mixed in fixed proportion in a platinum crucible.

Properties studied by X-ray: The following values were obtained by measuring the artificial crystal.

$a_0=5.41\text{\AA}$, $b_0=8.42\text{\AA}$, $c_0=4.51\text{\AA}$ (measured by WATANABE)

$a_0=5.385\text{\AA}$, $b_0=8.40\text{\AA}$, $c_0=4.48\text{\AA}$ (measured by SADANAGA)

$a_0 : b_0 : c_0=0.642 : 1 : 0.536$ (WATANABE)

$a : b : c=0.6412 : 1 : 0.5494$ (Artificial crystal: measured by MALLARD)

$Z=2$, $D_{2h}^{12}-\text{Pnmm}$.

3. Kotoite-marble occurs as the country rock of the Holkol deposit, and the main gold-copper ore (or skarn consisting of diopside, clinohumite, phlogopite, etc.) is found in this marble. Therefore, the occurrence of this rock indicates the presence of gold-copper ore, and the rock plays an important role during the prospecting of the ore body.

4. Kotoite-marble consists chiefly of kotoite and calcite; the former makes up 25–35 per cent of the weight of the rock, and the latter about 60 per cent. A few accessory minerals, ludwigite, suanite, forsterite, and spinel are also contained. The chemical composition of this rock is as follows:

The principal components are $\text{B}_2\text{O}_3=10-12\%$, $\text{CaO}=32-35\%$, $\text{MgO}=20-23\%$, and $\text{CO}_2=26-28\%$; the minor components are $\text{SiO}_2=2-4\%$, $\text{Al}_2\text{O}_3=0.2-0.5\%$, and $\text{Fe}_2\text{O}_3=0.3-0.4\%$. The molecular ratio of CaO to MgO is usually within the limits of 1.0–1.2.

5. Suanite is a new mineral which has the following composition:

$2\text{MgO} \cdot \text{B}_2\text{O}_3$.

6. A large amount of kotoite-marble occurs in the Holkol mine; it was mined as the boron ore by which borax was produced.

7. Kotoite has never been found alone, but always in association with the calcite that composes kotoite-marble; it occurs as very fine grains. Kotoite-marble is found surrounded by crystalline dolomite which suggests that the rock was formed from the latter by acquiring a supply of B_2O_3 . As the kotoite has been formed from the mineral dolomite $CaCO_3 \cdot MgCO_3$ by replacing the CO_2 of $MgCO_3$ and by leaving $CaCO_3$ as calcite, the ratio of kotoite to calcite in the kotoite-marble is nearly constant.

8. Kotoite-marble is a rock which has been derived from dolomite by the introduction of volatile materials such as B, F, and Cl which were concentrated in the residual magma and emanated into the surrounding dolomite at a later stage of consolidation of the granite in the Suan district.

Additional note: In 1956 the third occurrence of kotoite was discovered by the author and A. KATO in Kamineichi dolomite quarry, Miyako, Iwate Prefecture, Japan. Some other occurrences in USSR and USA have recently been reported.

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