

# ON MINERALS ASSOCIATED WITH ULTRABASIC ROCKS, FOUND IN THE VICINITY OF COMMON BOUNDARIES OF SHIMANE, TOTTORI, OKAYAMA AND HIROSHIMA PREFECTURES (III)

—PREHNITE AND TOURMALINE (DRAVITE) FROM THE HIROSE CHROMIT MINE, TOTTORI PREFECTURE—

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## Preface

As to the geology of Hirose Mine and its environs, Nichinan Town, Tottori Pref., some notes by the writer have been described in the Journal of Mineralogists, Petrologists, and Economic geologists, in the Science Report of Shimane University, etc.. In the ultrabasic rock in that district, there are frequent occurrences of gabbro- and diorite-dykes, the parent magma of which is supposed to be different from that of the former, which is a derivative from the peridotite magma, generated from the peridotite mantle, whereas the latter is the derivatives from the magma from the eclogite mantle.

Hirose Mine is well known, together with Wakamatsu Mine in the same prefecture and Takase Mine in Okayama Pref., for the rich production of chromite ore contained in the ultrabasic rock (serpentinite), and from these mines are also produced, occasionally, some minerals of special interest. Here a description will be made on the prehnite and tourmaline (dravite) from Hirose Mine.

### 1. Prehnite from the Hirose Mine, Tottori Pref.

#### Occurrence

The prehnite which will be described in this paper was occurred from the ōgiri level of Hirose Mine. The mineral was formed, in fissures or cavities in serpentinite, by the hydrothermal processes, and is associated a small amount of zeolite. The surface of it is more or less stained by mine water, etc. (see Fig. 1.), but its fracture is white.

#### Physical and Optical Properties

The mineral shows no characteristic crystal form, but the aggregated lamella form or clouded flat spherical form. Though its fracture surface is white, as stated above, the

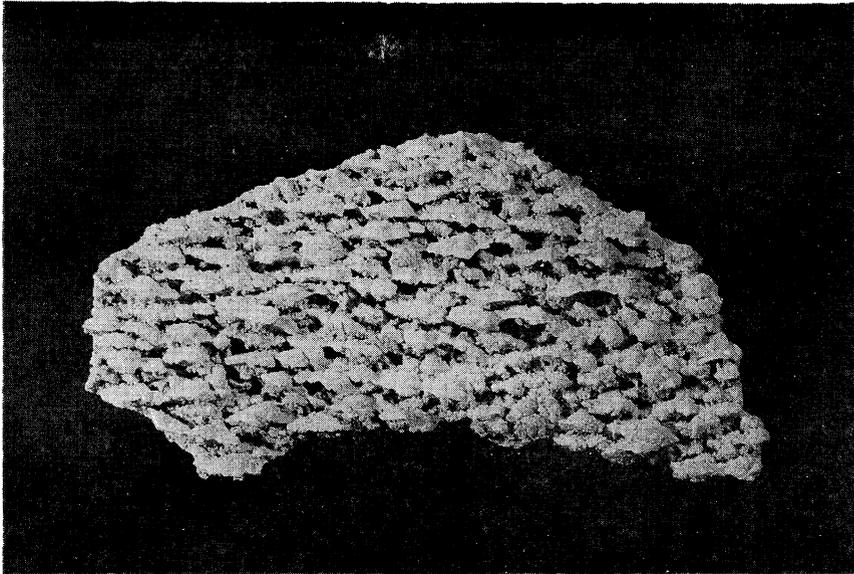


Fig. 1. Prehnite. Scale : 3/5  
34th level of Hirose Mine, Tottori Pref.

surface of the mineral is stained to light yellowish grey by mine water. The hardness of the mineral was determined to be within the range 6-7. The specific gravity, determined by pycnometer, is  $G_4^{15} = 2.920$ . Under the microscope, it occurs in aggregated crystals, that are 0.05 - 0.10 mm in the average size, colourless, and shows straight extinction and occasional optical anomaly. The optical plane is  $olo$ , and the indices are  $\alpha = 1.6137$ ,  $\beta = 1.6244$ ,  $\gamma = 1.6395$ ; double refraction is  $\gamma - \alpha = 0.0258$ . The optical axial angle is  $(+) 2V = 68^\circ 50'$ .

#### X-Ray Powder Diffraction

Powdered sample of the prehnite from which the stained portion has been removed was offered to the x-ray diffraction test, for which Shimazu Recording X-Ray Diffractometer was used. The instrumental conditions are as follows.

X-ray	35 kVp, 15 mA (Cu- $K_\alpha$ radiation)
Full scale	2000 C. P. S.
Scanning speed	$1^\circ/\text{min.}$
Chart speed	10 mm/min.
Time constant	2.5 seconds.
R. slit	0.2 mm.
D. slit	2.0 mm.

The powder diffraction results are shown in Fig. 2 and Table 1. The lattice constants were calculated, using the atomic planes 111, 106, and 119 with high intensities of reflection. The d-values of these reflections are 3.6422, 2.5530, and 1.7702, respectively, which were obtained from the exact measurement with the standard of quartz crystal,

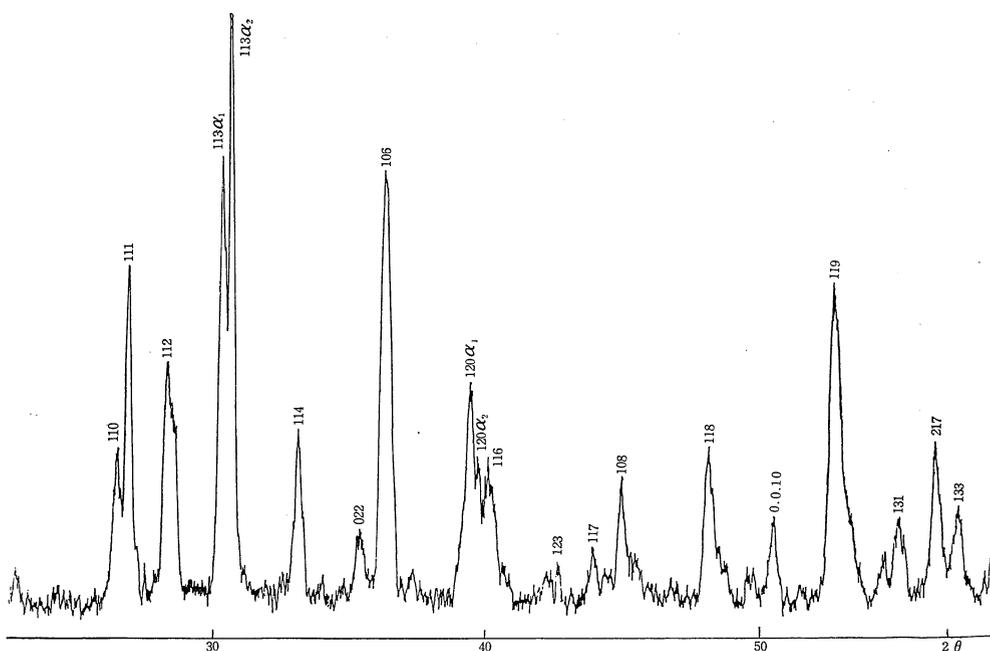


Fig. 2. X-ray diffraction pattern of prehnite from Hirose Mine, Tottori Pref.

Table 1. X-Ray Powder Data for Prehnite from Hirose Mine

$2\theta$	d	$I/I_1$	hkl	$2\theta$	d	$I/I_1$	hkl
16.9	5.2461 $\alpha_1$	5	011	48.4	1.8806 $\alpha_1$	3	126, 109
17.1	5.1852 $\alpha_2$			48.6	1.8733 $\alpha_2$		
19.2	4.6225 $\alpha_1$	7	100, 004	49.4	1.8448	13	0.0.10
19.3	4.5988 $\alpha_2$			51.6	1.7712		
21.6	4.1140	5	102, 013	53.4	1.7157 $\alpha_1$	3	130
25.3	3.5201	48	110	53.5	1.7127 $\alpha_2$		
25.7	3.4662 $\alpha_1$	65	111	54.0	1.6980	7	131, 223
26.3	3.3885 $\alpha_2$			55.3	1.6612		
27.2	3.2784	56	112, 104	56.2	1.6367	11	133
29.2	3.0582 $\alpha_1$	88	113, 015	57.5	1.6027	5	0.1.11
29.6	3.0178 $\alpha_2$			58.8	1.5703		
31.9	2.8053	28	114	59.8	1.5465 $\alpha_1$	25	129, 135
34.1	2.6299 $\alpha_1$	5	022	60.0	1.5418 $\alpha_2$		
34.2	2.6217 $\alpha_2$			8	106, 115	60.3	1.5348
35.2	2.5495	95	106, 115	61.6	1.5055 $\alpha_1$	4	302
36.1	2.4880	2	023	61.7	1.5032 $\alpha_2$		
38.2	2.3559 $\alpha_1$	44	120, 024	63.6	1.4629 $\alpha_1$	6	312
38.5	2.3382 $\alpha_2$			15	116, 008		
38.9	2.3151	12	116, 008	64.4	1.4467 $\alpha_1$	8	12.1.0
41.1	2.1961 $\alpha_1$	3	123	64.6	1.4427 $\alpha_2$		
41.4	2.1809 $\alpha_2$			4	117, 211	66.0	1.4154
42.7	2.1175	8	117, 211	66.2	1.4116 $\alpha_1$	11	228
43.8	2.0668	25	108, 204	66.6	1.4041 $\alpha_2$		
45.6	1.9893	2	125, 028	67.0	1.3967 $\alpha_1$	12	038, 228
47.0	1.9333	28	118, 214	67.4	1.3894 $\alpha_2$		

(Operator : J. Kitahara)

and the lattice constants obtained are  $a=4.595 \text{ \AA}$ ,  $b=5.495 \text{ \AA}$ ,  $c=18.423 \text{ \AA}$ . These values, as compared with those which have been reported, are found to be somewhat smaller in  $a$  and  $c$ , while a little larger in  $b$ .

The cell dimensions and space group (P 2cm) of prehnite were first determined by Gossner and Mussnug (1931). Nuffield (1943) on the basis of a detailed morphological, physical and chemical study confirmed the cell constant and indexed the x-ray powder pattern.

### Chemistry

Chemical analysis of the pure white prehnite specimen was performed by the writer. The result and its calculated atomic ratios on the basis of 24 (O, OH) are as denoted in Table 2.

Table 2. Chemical Composition of Prehnite from Hirose Mine

	Wt.%	Mols	Atom. Ratios		Ditto when 24 (O, OH)
SiO <sub>2</sub>	41.70	695	Si	695	5.788
TiO <sub>2</sub>	0.0	0	Ti	0	0
Al <sub>2</sub> O <sub>3</sub>	25.18	247	Al	494	{ 0.212
Fe <sub>2</sub> O <sub>3</sub>	0.45	3	Fe <sup>3</sup>	6	{ 3.902
FeO	0.32	4	Fe <sup>2</sup>	4	{ 0.050
MnO	0.04	1	Mn	1	{ 0.033
MgO	0.51	13	Mg	13	{ 0.006
CaO	26.76	478	Ca	478	{ 0.108
Na <sub>2</sub> O	0.23	4	Na	8	{ 3.981
K <sub>2</sub> O	0.18	2	K	4	{ 0.066
H <sub>2</sub> O <sub>+</sub>	4.32	240	H <sub>+</sub>	480	{ 0.033
H <sub>2</sub> O <sub>-</sub>	0.21		O	2882	{ 3.997
Total	99.90				

(Analyst : J. Kitahara)

As obviously seen in the analytical table, amounts of K<sub>2</sub>O, Na<sub>2</sub>O, MgO, MnO, and FeO+Fe<sub>2</sub>O<sub>3</sub> are all small, and, of these, the latter three are present, replacing Al<sub>2</sub>O<sub>3</sub>, and a small fraction of SiO<sub>2</sub> is partly replaced by Al<sub>2</sub>O<sub>3</sub>. Similar replacements seem to be present in the prehnite of this locality, and K<sub>2</sub>O, Na<sub>2</sub>O also replace some part of CaO. As compared with the chemical composition of prehnites of other localities, no marked difference is seen in the amount of each component. Though general formula of prehnite is shown by H<sub>2</sub>Ca<sub>2</sub>Al<sub>2</sub>(SiO<sub>4</sub>)<sub>3</sub>, the structural formula is given as (SiO<sub>3</sub>)<sub>3</sub>Ca<sub>2</sub>(AlOH). AlO<sub>2</sub>H or Si<sub>3</sub>AlO<sub>10</sub>(OH)<sub>2</sub>AlCaCa. The chemical formula of the prehnite from the mine is described as (OH)<sub>4.00</sub> (Na<sub>0.07</sub>, Ca<sub>3.98</sub>, K<sub>0.03</sub>)<sub>4.08</sub> (Al<sub>3.90</sub>, Fe<sup>3</sup><sub>0.05</sub>, Mg<sub>0.11</sub>, Ti<sub>0.00</sub>, Fe<sup>2</sup><sub>0.03</sub>, Mn<sub>0.01</sub>)<sub>4.10</sub> (Si<sub>5.79</sub>, Al<sub>0.21</sub>)<sub>6.00</sub>O<sub>20.00</sub>.

## 2. Tourmaline (Dravite) from the Hirose Mine, Tottori Prefecture

### Occurrence

The dravite for which the following description will be made was found in a feldspar vein (see Fig. 3), penetrating the serpentinite body at a spot, about 330 m inward from the entry of the 34th level of this mine.



Fig. 3. Dravite (black) associated with plagioclase (white). Scale : 4/5  
34th level of Hirose Mine, Tottori Pref.

### Crystal Habit and Structure

The crystal consists of a parallel group of slender bodies, each 1–3 mm wide and 10 cm long, formed with  $(11\bar{2}0)$  and  $(10\bar{1}0)$ .

Belov and Belova (1949, 1950) gave the structure for dravite, with a 16.00 Å,  $c$  7.24 Å, assuming the formula unit  $\text{NaR}_3\text{B}_3\text{Si}_3(\text{O}, \text{OH})_{30}$ , while Ito and Sadanaga (1951) determined the structure of rubellite from Brazil, with a 16.0 Å,  $c$  7.17 Å, using the formula  $(\text{Na}, \text{Ca})(\text{Li}, \text{Al})_3(\text{Al}, \text{Fe}, \text{Mn})_6(\text{OH})_4(\text{BO}_3)_5\text{Si}_3\text{O}_{18}$ . The chief difference between the structure proposed by Donnay and Buerger and that of Ito and Sadanaga (1951) is

in the configuration of the  $\text{Si}_6\text{O}_{18}$  group. The structure described by Belov and Belova (1950) is similar in general to that of Donnay and Buerger (1950), but with some differences in the exact position of the atoms. Epprecht (1953) has discussed the structure and concludes that elbaite may have the structure proposed by Ito while dravite possesses the structure proposed by Donnay and Buerger : schorl might have an intermediate structure.

Epprecht (1953) carried out precision determinations of the lattice constants on tourmaline of known composition and demonstrated that dravite and elbaite are immisible with each other, but dravite and schorl, and schorl and elbaite make continuous solid solution in each series. Epprecht's extrapolated values for the three end-members are :

	a(Å)	c(Å)	c/a
Elbaite	15.842	7.009	0.4485
Schorl	16.032	7.149	0.4465
Dravite	15.942	7.224	0.4530

### Physical and Optical Properties

The dravite is black with vitreous luster. The mineral is 7 + on the Mohs scale of hardness, and gives a specific gravity of  $G_4^{15} = 3.034$ . The streak of tourmaline is generally colourless, but that of the mineral is pale blue grey.

The colour of tourmaline is extremely variable but in general terms it can be related to the composition in so far as the iron-bearing tourmalines are black, while the elbaites tend to light shades of blue, green or pink, and the dravites vary from dark brown to yellow and colourless. In the dravite-schorl series the decrease in colour with decreasing iron is similar to that shown by many ferromagnesian mineral series, and grades through brown and light brown for the low-iron members to almost completely colourless for the pure magnesium tourmaline.

Under the microscope, the mineral shows straight extinction and negative uniaxial character. Good cleavage is not observed in the mineral section. The pleochroism is Z=colourless, pale yellow and X=colourless : the interference colour is intermediate or high. The refractive indices are  $\epsilon = 1.6167$ ,  $\omega = 1.6380$ , and the double refraction is  $\omega - \epsilon = 0.0213$ .

The refractive indices, relating to the atomic number of 0.204 of  $(\text{Fe}^{2+} + \text{Fe}^{3+} + \text{Mn})$  which was computed from the atomic ratio 31 (O, OH, F) obtained from the chemical analysis that will be shown later, are  $\epsilon = 1.617$ ,  $\omega = 1.639$ , and double refraction is 0.022. Judging from these optical properties, the tourmaline of this mine is supposed to be dravite, as stated above.

The iron and manganese of tourmaline is seen to control the refractive index, while the birefringence also increases with rising iron content. The pleochroism is variable in intensity but in the iron-bearing tourmalines it is normally very strong. The iron-tourmalines typically show pleochroism from yellow or brown to pale yellow, or from blue or green to pale yellow or colourless : the magnesium tourmalines often show yellow to pale yellow pleochroism.

### X-Ray Powder Diffraction Pattern

Shimazu Recording X-Ray Diffractometer was used for X-ray diffraction experiment of the mineral. The conditions are as follows.

X-ray	35 kVp, 15 mA (Cu-K $\alpha$ radiation)
Full scale	1000 C. P. S
Scanning speed	1°/min.
Chart speed	10 mm/min.
Time constant	1.25 second
R. slit	0.8 mm
D. slit	4.0 mm
Filter	Ni

The X-ray powder data are as shown in Fig. 4 and in Table 3.

According to the expression of the powder diffraction file by Fink, the atom plane distance of tourmaline, d-value, for the maximum reflection intensity (1st rank) is 2.58, and the d-value of the tourmaline of this mine, 2.578, corresponding to it, is the 2nd rank of the reflection intensity, and the d-value of 2.04, shown for the tourmaline in the Fink's table for the 2nd rank of the intensity, corresponds to that of our tourmaline for the 3rd rank of the reflection intensity. D-value of 2.961 of our tourmaline for the maximum reflection (1st rank) corresponds to that in the Fink's table for the 8th rank of the

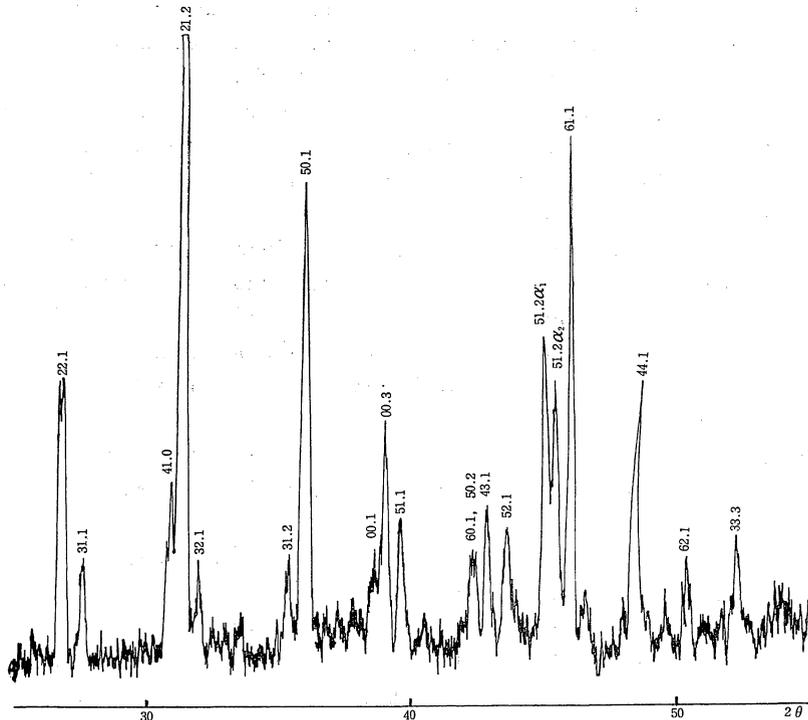


Fig. 4. X-ray diffraction pattern of tourmaline (dravite)  
from Hirose Mine, Tottori Pref.

Table 3. X-Ray Powder Data for Dravite from Hirose Mine

2 $\theta$	d	I/I <sub>1</sub>	hk.l	2 $\theta$	d	I/I <sub>1</sub>	hk.l
10.5	8.4249	1	11.0	42.8	2.1127	3	41.5
11.8	7.4995	1	51.1	43.8	2.0668 $\alpha_1$	32	
12.85	6.8891	1	20.0	44.29	2.0450 $\alpha_2$	21	51.2
13.7	6.4634 $\alpha_1$	7		44.85	2.0209 $\alpha_1$	37	
13.9	6.4168 $\alpha_2$	9	10.1	45.4	1.9976 $\alpha_2$	3	61.1
17.92	4.9498	4	20.1	47.46	1.9156	24	44.1
19.3	4.5988 $\alpha_1$	4		48.4	1.8808	2	61.2
19.4	4.5753 $\alpha_2$	5	30.0	49.05	1.8571 $\alpha_1$	2	
20.2	4.3959	1	52.1	49.2	1.8519 $\alpha_2$	3	62.1
21.15	4.2006	10	21.1	49.35	1.8466	2	71.0
22.1	4.0220 $\alpha_1$	9		51.05	1.7889	5	33.3
22.35	3.9777 $\alpha_2$	21	22.0	54.1	1.6952	2	21.4
25.5	3.4930 $\alpha_1$	16		55.03	1.6685 $\alpha_1$	6	
25.65	3.4729 $\alpha_2$	17	22.1	55.1	1.6607 $\alpha_2$	12	60.3
26.3	3.3885 $\alpha_1$	10		55.9	1.6447	13	72.1
26.43	3.3722 $\alpha_2$	9	31.1	56.4	1.6313	1	63.1
29.6	3.0178 $\alpha_1$	4		57.6	1.6002 $\alpha_1$	23	
29.7	3.0079 $\alpha_2$	14	41.0	57.75	1.5964 $\alpha_2$	8	40.4
30.18	2.9612	100	21.2	58.3	1.5826	2	73.0
30.8	2.9028	5	32.1	58.92	1.5674	2	81.1
34.05	2.6330 $\alpha_1$	3		60.05	1.5407 $\alpha_1$	2	
34.15	2.6250 $\alpha_2$	5	31.2	60.2	1.5371 $\alpha_2$	15	64.1
34.8	2.5779	51	50.1	61.3	1.5144	21	50.4
37.4	2.4044	2	00.1	62.4	1.4881 $\alpha_1$	2	
37.6	2.3921 $\alpha_1$	3		62.55	1.4850 $\alpha_2$	3	33.4
37.8	2.3799 $\alpha_2$	29	00.3	62.7	1.4817	2	81.2
38.4	2.3441	12	51.1	63.67	1.4615 $\alpha_1$	19	51.4
41.05	2.1987 $\alpha_1$	7		63.8	1.4588 $\alpha_2$	22	
41.15	2.1936 $\alpha_2$	7	60.1	64.8	1.4387 $\alpha_1$	6	10.5
41.2	2.1910	7	50.2	65.05	1.4338 $\alpha_2$	6	
41.67	2.1664	10	43.1	65.88	1.4177 $\alpha_1$	5	20.5
42.42	2.1308 $\alpha_1$	12		66.05	1.4145 $\alpha_2$	18	
42.6	2.1222 $\alpha_2$	2	52.1				

(Operator : J. Kitahara)

reflection intensity. Thus, the atom plane distance coincides well with that of tourmaline, though the reflection intensity shown by the corresponding atom plane is different. D-value of 2.961 of our tourmaline for the maximum reflection intensity corresponds to that of 2.951 of the tourmaline of Kanakura Mine, Nagono Pref., which is for the 2nd rank of the reflection intensity. D-value of the tourmaline from Ishikawayama, Fukushima Pref. is 2.956, and it is for the 4th rank of the reflection intensity. In short, each atom plane distance of the tourmaline of Hirose Mine corresponds to each respective ones of the mineral from other localities, but the sequence of the x-ray reflection intensity for respective atom plane distance is not same.

The lattice constants of the mineral were obtained from the atom planes  $21\bar{3}2$  and  $50\bar{5}1$  with strong x-ray reflections. The d-values (atom plane distances) were measured accurately compared with the x-ray reflection position of standard quartz, and the lattice constants calculated from the d-values are  $a=15.944 \text{ \AA}$ ,  $c=7.192 \text{ \AA}$ , and the axial ratio is  $c/a=0.4511$ .

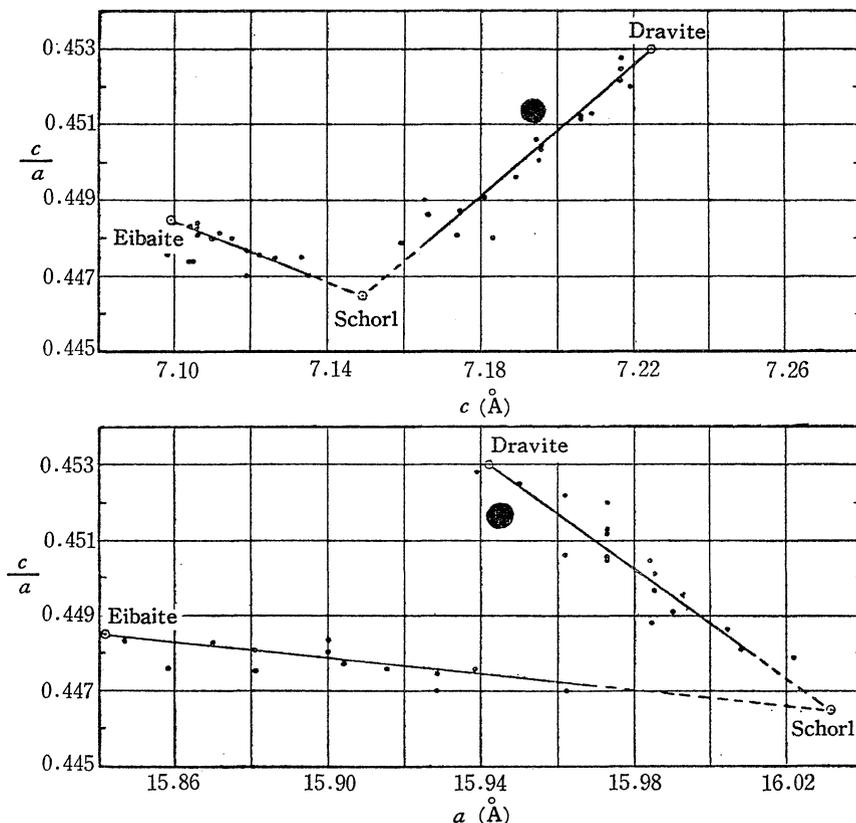


Fig. 5. Tourmaline axial ratios and cell dimensions. Great full circle : Dravite from Hirose Mine, Tottori Pref.

The lattice constants of dravite, schorl and elbaite which have been reported, are  $a=15.94-15.98$ ,  $c=7.19-7.23$ ;  $a=15.93-16.03$ ,  $c=7.12-7.19$ ; and  $a=15.98-15.93$ ,  $c=7.10-7.13 \text{ \AA}$  respectively. From the aspect of the lattice constants, the mineral from the Hirose Mine is considered to be dravite among the three species.

The tourmaline from the mine is plotted in the limit of dravite group on the diagrams (Fig. 5) showing tourmaline axial ratios and cell dimensions.

### Chemistry

The chemistry of tourmaline is complex and until recently its basic formula was very uncertain. Recent structural determinations make it reasonably certain that the formula is either  $\text{NaR}_3\text{B}_3\text{Si}_3(\text{O}, \text{OH})_{30}$  (Belov and Belova, 1950) or more probably  $\text{NaR}_3\text{Al}_3\text{B}_3\text{Si}_3$

$O_{27}(OH)_4$  (Donnay and Buerger, 1950 : Ito and Sadanaga, 1951). R can be predominantly  $Fe^{2+}$  as in schorl, Mg as in dravite, or (Al+Li) as in the elbaite series. There is no substitution of Al for Si in the tourmalines : in the hydroxyl position part of the (OH) is often replaced by F, particularly in the elbaite series. There appears to be a complete range of composition between the magnesian and iron rich members, and between the iron-rich and the lithium tourmalines, though intermediate members in the latter range are less common. It seems to be an immiscibility gap between elbaite and dravite.

Table 4. Chemical Composition of Tourmaline from Hirose Mine

	Wt.%	Mols	Atom. Ratios		Ditto when (O, OH, F) = 31
SiO <sub>2</sub>	36.19	603	Si	603	5.847
TiO <sub>2</sub>	0.29	4	Ti	4	0.039
B <sub>2</sub> O <sub>3</sub>	10.80	155	B	310	3.006
Al <sub>2</sub> O <sub>3</sub>	31.72	311	Al	622	6.031
Fe <sub>2</sub> O <sub>3</sub>	1.09	7	Fe <sup>3</sup>	14	0.136
FeO	0.49	7	Fe <sup>2</sup>	7	0.068
MnO	0.0	0	Mn	0	0.0
MgO	13.00	325	Mg	325	3.151
CaO	0.42	8	Ca	8	0.078
Na <sub>2</sub> O	2.25	36	Na	72	0.698
K <sub>2</sub> O	0.16	2	K	4	0.039
F	0.10	5	F	5	0.048
H <sub>2</sub> O <sup>+</sup>	3.34	186	H	372	OH 3.607
			O	3011	
	99.85				
O=F	0.04				
Total	99.81				

(Analyst : J. Kitahara)

The chemical analysis of the sample together with the atomic ratios on the basis of 31 (O, OH, F) obtained from the chemical analysis is given in Table 4.

From the table 4, it is ascertained that the mineral is dravite which iron content (FeO + Fe<sub>2</sub>O<sub>3</sub>) is poor and magnesium content is rich. As described above, the mineral is tourmaline as regard to its x-ray properties, but optically, it is supposed to be dravite. Judging from the result of chemical analysis, it is certainly dravite. Amount of FeO + Fe<sub>2</sub>O<sub>3</sub> shown by the analysis is as small as 1.58%, which indicates the mineral to be nearly an end member of dravite, and this amount may be near to the minimum responsible for the black colour of the mineral, and, really, the projected point of the dravite of this mine, shown in Fig. 5 is found near the boundary of black and brown varieties. Besides the small amount of total iron content, the surpassed amount of the trivalent iron upon the bivalent in their coexistence may be regarded as the cause of the deepening colour and blackish tint, in comparison with the variety containing the iron of only one valency.

For the atomic ratio of Si, determined from the chemical analysis as 18, that of Fe computed is 0.63. Some sample is brownish black in colour with atomic ratio of 0.2, and

the other one is black with that of 0.9, while some other sample is light brown with the atomic ratio of 0.4. Thus, it seems that the Fe atomic ratio of some 0.63, as that of the present tourmaline, is indicative of a composition near to the converting point of the colour of the mineral from brown to black. To sum up, as the deciding factor for the colouring of tourmaline, its iron content is to be pointed out, and, further, the ratio of the bivalent to the trivalent iron also seems to play an important part for the nature of the colour.

### Origin

The pneumatolytically injected plagioclase is extremely rich in  $Al_2O_3$ . By the influence of the solution the rock has recrystallized and some change of material has occurred. Some MgO from the peridotite or serpentinite may have been added, just as  $SiO_2$  has been resorbed from the pneumatolytic or succeeding solution. The formation of tourmaline is evidently favoured by the presence of rock rich in MgO. The dravite is found in metamorphic or metasomatic assemblages. The tourmaline has crystallized later than the plagioclase. The photo. Fig. 3, shows typical vein or concretion of chiefly plagioclase and tourmaline. The black part is thus almost wholly aggregates of tourmaline, usually with petty plagioclase between. Plagioclase and tourmaline generally occur together, the quartz mostly as separate vein. The tourmaline concretion may be up to 5 cm in width. The boron have been metasomatically introduced; it seems most likely that granitic magma was the initial source of the boron, the Mg/Fe ratio in the tourmaline being controlled by the mineral or rock in which the tourmaline is developed.

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### References

References were omitted from the paper.

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