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### By

## Hiroshi MIYAKE

#### With 13 Tables, 1 Text-figure and 14 Plates

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ABSTRACT: The present study is concerned with the so-called skarns designated after the rule applied to generally and the related minerals comprised in several ore deposits distributing in the Chūgoku District with respect to their geneses and in pursuit of so-called pyrometasomatic or contact effects. Inspection of the paragenetic relationships among, and identification of, the respective minerals have been fulfilled as elaborately as possible by means of an ordinary microscope and x-ray diffractometer in conjunction with heat treatments especially for some specimens, resulting in more precise interpretation other than various concepts given previously by many authors and furthermore in discovery of some rare minerals taking an important part in the subject under consideration.

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#### I. INTRODUCTION

The gangue minerals found occurring in, or in the vicinity of, the contact, contact-metasomatic or pyrometasomatic deposits designated prevailingly according to the antecedent definition are commonly represented by those composed of silicates, aluminosilicates and some others rich in alkaliearths, the assemblages of which have as a whole been called 'skarn' or 'tactite' all but regardless of their genetic processes. Besides, these skarns have hitherto merely for the reason of their apparent occurrence

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in Nature been classified into those derived respectively through reaction of the country rocks with certain sort of emanation and through recrystallization as a result of thermal effects on one hand and into those produced primarily or secondarily (furthermore endo- or exothermically) through mobilization of some substances on the other. These views together with some others are certainly considered to have placed emphasis upon metasomatism of the carbonate rocks through pneumatolytic action.

However, taking account of that so-called skarns happen to be associated either with some species of the ore minerals or without any of them and hence parallelism of the former to the latter in occurrence be not always establishable in every case, such an expression as the contact deposit or the like simply owing to coexistence of the minerals, though allied to skarns in property but different in source, with the ores in some geologic unit also seems by no account justifiable and should rather be restrictedly applied to in the case of such assemblages as are to more or less extent genetically related to ore formation.

In addition, it seems almost above suspicion that there are comparatively few formed at higher temperature literally in conjuction with the contact effects, for example, between granitic rocks and limestone; that is to say, an obscurity still remains in that, in spite of being grouped into those produced through pneumatolysis at relatively high temperature, so-called contact deposits are likely to bear no direct connection with the skarns comprised in the part distant from the related contactzone.

Detailed inspections for the mineral parageneses appearing in this sort of the deposits and for the experimental data of their artificial synthesis clearly point to at the same time that most of the skarns are possibly those formed at certain temperature lower than suggested previously in close connection with low-temperature mineralogenesis, no matter whether from a simple process or miscellaneously over-lapped sequences at certain stage prior or subsequent to that for ore genesis.

Important cues to clarifying the subjects alluded to above have been pursued in the present work concerning several ore deposits found at the Yamato mine in Yamaguchi Prefecture, the Tsumo mine in Shimane Prefecture, the Mihara, Sampō, Takinomaru, Motoyama, and Sano mines in Okayama Prefecture and the Mihara mine in Hiroshima Prefecture etc.

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## II. SKARNS AND THE RELATED MINERALS

It is in severe sense problematical to what extent the minerals in question may, even if recognized seemingly within, and in the vicinage of, the contact zone, be assuredly identified just with skarns or not, and why those allied or similar to themselves in characteristics are found occurring in various kinds of the country rocks even far from the contact zone. Accordingly, the minerals mentioned hereunder are those taken as a sort of skarns in such sense as has hitherto been used, as to which special cares have however been paid on their occurrence and genesis from the point of view other than ever given.

## A. Wollastonite

As was reported already by MIYAKE and AKATSUKA (1963), the host for the ore bodies composed mainly of chalcopyrite in the Yamato mine consists of limestone, chert, and slaty hornfels replaced abundantly by wollastonite in massive and veinand network-like occurrence. The most predominance of wollastonite in chert, no matter whether color of the latter may be grey or purely white, is especially to be noted while the very contacts between the two are nowhere transitional but clearly demarcated with one another (Plate 51, 4), as be the case with limestone. The massive bodies of wollastonite, a part of which in the Hokoku adit amount to more than 10m in diameter and have been worked for the raw materials of cermics on an industrial scale, are especially in limestone difficult to discriminate megascopically from the surroundings but in occurrence similar to in the cases of other country rocks. On the other hand, the veins and the networks of its veinlets are also found isolatedly far from, near by. and within the massive bodies (Plate 41, 1, 2, 3, and Plate 51, 6), suggesting their formation through different processes at somewhat different stages, while paragenetic relation to other later minerals may, as will be discoursed later, give a clue to elucidating their formation at another later stage.

Mineralogically, three normal modifications of calcium metasilicate, such as triclinic or pseudohexagonal pseudowollastonite ( $\alpha$ -type), monoclinic parawollastonite ( $\beta$ -type) and triclinic or pseudomoclinic wollastonite have been well known, of which the first was recognized merely in the pyrometamorphic rock (MCINTOCK, 1932) and in the synthetic materials but generally not in Nature and the latter two discovered at Crestmore by JOAN (1958) and MURDOCH (1961) are distinguishable in the respective spacings at 4.37Å and 3.73Å ( $\beta$ -type) and at 4.05Å and 3.40Å (common wollastonite) from each other on the basis of ASTM data. The specimens obtained from the Yamato, Mihara (Hiroshima) and Sampō mines have not been precisely identified with each of the above-mentioned while those from the Takinomaru mine alone have been surely taken as wollastonite.

There is another point to be clarified in detail. MIYAKE, AKATSUKA and UMEGAKI (1964) indicated the formation of wollastonite (difficult to be distinguished from para-wollastonite) as a sort of decomposition product through heating of prehnite at

1,000°C and that of pseudowollastonite through heating of apophyllite at the same temperature. This surely points to the stability of pseudowollastonite at certain temperature lower than 1,000°C but its presence has never been confirmable in the specimens provided for the present research although BUCKNER and Roy (1955) already demonstrated the inversion from low-temperature wollastonite to high-temperature pseudowollastonite at 1,120°C±10°C.

Various opinions have so far been made public as for genesis of wollastonite. KAKI-TANI (1962) experimentally revealed its formation through solid reactions between lime and silica to be txtremely confined within a narrow width between the two and reached a justification of his own view also in field observations concerning the STRINGHAM (1952) pointed out the mineral in question to be numerous localities. hydrothermally precipitated from alkaline solution at about 500°C and BARREL and DENNY (1961) also manifested its possibility similarly in hydrothermal condition at 125°~450°C. These data are considered in good harmany with low-temperature vein-like occurrence of wollastonite at least seemingly indifferent to, or without any reaction with, the surrounding host rocks, although the natural states composed-of complicated components are of course not always to be defined in mere dependence on temperature, and the precedents have placed a considerable emphasis on genesis through the metasomatic effects at rather high temperature (cf. KATO, 1913; 1937) or as a result of metasomatism of the pre-existing rocks through gaseous emanation derived from the related intrusive (cf. TILLEY, 1951). As far as the deposits under consideration are concerned, the occurrences of wollastonite surely are, because of no matter whether the host rocks may be deficient in calcium or not, in opposition to the metasomatic theory. In addition, the interstices of their prismatic crystals are often found filled with fluorite in the cases of the earlier ones (Plate 45, 1, 2) but not in those replacing garnet formed at later stage.

Remarkable is that the specimens sampled from the above-mentioned four deposits are yellowish brown to yellowish orange in flourescent color with radiation of 3,660Å in wavelength and become glittering for the ray with wavelength of 2,537Å. Details of other characteristics have already been shown in the previous work reported by the writer.

#### B. GARNET

The minerals in question are involved abundantly in the skarns appearing in all of the deposits alluded to above, the species and quantitative ratios of which, though different in each case, reveal their own characteristics, and are composed ordinarily of grossularite and andradite.

As for the relations of their chemical constituents to the cell constant  $a_0$ , refractive index and specific gravity, several sorts of the related diagrams have hitherto been proposed (cf. OMORI, 1953; SKINNER, 1956; WINCHELL, 1958), according to which the specimens from the Yamato mine are identified with those ranging from grossularite ( $a_0 = 11.87$  Å and S.G.=3.62) to andradite ( $a_0 = 12.05$  Å and S.G.=3.81) in

composition. Again after OMORI's view garnet found in pegmatite and mica-schist is believed to crystallize in trapezohedral form and that obtained from skarns in limestone is so in dodecahedral habit owing to the lower grade of the ionic bonding while in limestone subjected to the contact effects in higher grade the former associated with the latter are observable as a result of conspicuous addition of the thermal effects necessary for accelerating the ionic bondings. However, those with trapezohedral form have, though poor in the well-defined crystals, been discernible in none of the specimens sampled from the Yamato, Sampō and Mihara (Hiroshima) mines.

Both zonal structure and optical anomaly appearing often in garnet are now to be disputed. In relation to this regard, the specimens are classifiable into (a) anisotropic one with zoning, (b) isotropic one with zoning and (c) isotropic one without zoning on the basis of megascopic and microscopic differences in color, partial differences in optical feature, pressence of cracks between the respective parts and others. There are a profusion of examples indicating the marked difference of reddish browncolored inside (representing grossularite) from yellowish green-colored outside (representing andradite) in common cases and such a repeated zoning as has been ascertained in the specimens from the Sampo mine. Those revealing a division into six twin-sectors different from one another in optical property and zoning with different optical characteristics in arrangement parallel to the crystal planes surrounding a unit crystal and those characterized only with the latter feature are often recognizable. In addition, those classified into (a) are generally confirmed in a unit crystal or in its aggregates embraced in wollastonite and some others of the Yamato mine; those grouped into (b) are observed accompanying in part with a little amount of (a) in the skarn veins supposed to have been produced in certain condition different considerably from those for (c) because of the latter occurring as a continuous vein but not as isolated grains (Plate 42, 2, 3).

On the other hand, the optically abnormal characteristics are likely to appear restrictively in the cases of grossularite and andradie. OMORI (1953) ascribed them to the partial distortion caused by packing index in the inner structure and Kozu *et al.* (1940) reported their disappearance through heating at 1,225 °C for 10 min, whereas HEINRICH (1963) gave no allusion to genesis of the zoned anisotropic garnet, obtained from garnetiferous carbonate veins in serpentinite and almost similar to (a), and there still remains a sort of uncertainty in the former theories.

Be that as it may, the modes of occurrence of garnets appearing in the Yamato mine are classifiable into such types as: (1) that occurring merely in wollastonite but not in chert, slaty hornfels and limestone without any other accompanied minerals as an isolated simple crystal or small masses represented by grossularite or andradite, well-defined with (110) habit, characterized microscopically with the optically abnormal zoned structure, and related probably to genesis at certain stage nearly contemporaneous with formation of wollastonite not through the contact reactions but from a sort of mineralizer, (2) that appearing, for example, in the Shinsei adit

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as the large-scaled masses, about 10m in diameter, composed of the aggregates of the former type, megascopically different in color and composition in the inner and outer parts of a single grain (Plate 43, 1), (3) that occurring in wollastonite and representing a sort of zonation with wollastonite intercalating between the inner grossularite and the outer andradite (Plate 42, 4, 5, 6), (4) that belonging to the optically abnormal, brown-colored grossularite replaced gradually from the outer part by fine-grained, white-colored and seemingly waxy wollastonite replaced furthermore by bornite accompanying a subordinate amount of chalcopyrite, as is recognized merely in the skarns of the Fukuju deposit and rather resembling to calcite in occurrence (Plate 44, 1, 2, 3, 4), (5) that relating to yellow- to greencolored andradite with a zonation associating the narrow anisotropic zone in parallel alignment (Plate 43, 2), replaced by calcites, well-defined in some druses, bearing a genetic connection with some ore minerals such as chalcopyrite, sphalerite, galena and so on at a certain stage subsequent to alteration of hedenbergite and feldspars, the relics of which are in part observable, (6) that corresponding to the optically normal, brown-colored, ill-crystalline grossularite enclosed predominantly within the vesuvianite-garnet veins (relatively well-defined, brown- to greenish brown-colored vesuvianite, with zonal structure, replaced partly by grossularite in this case) (Plate 51, 2) or rarely within the garnet veins, microscopical to about 2cm in width, running irregularly across wollastonites and with a certain direction across limestone and accompanying wollastonite veins along its both side or the veins of granular diopside in limestone (Plate 51, 1), and (7) those subdivided into one composed of garnet-diopside-vesuvianite veins, simple in form and  $1 \sim 2$  cm in width, in massive wollastonite of the Fukuju deposit (Plate 53, 3, 4) and the other composed of garnetdiopside-vesuvianite-prehnite-feldspar veins, irregular in width and in extension, in wollastonite near the contact between wollastonite and limestone on the middle level of the Daido deposit (Plate 44, 5 and Plate 52, 5) (vesuvianite replacing granular garnet partly and being replaced by the later grossularite vein, and diopside being blue in the former vein but bluish grey in the latter because of being mingled with feldspar or prehnite), indicating a mineralogenic sequence of vesuvianite, through grossularite and to diopside in order.

The compositional discontinuity between grossularite and andradite in these cases has been alluded to by WHITE (1959) and structurally ascribed to the ionic radius of Ca<sup>++</sup> by OMORI (1953). Wollastonites seen within and around the garnet grains are considered pertaining to a sort of the products later than garnet while fluorites instead of wollastonite play the similar rôle in the case of the Sampō deposit (Plate 42, 1) and vesuvianites replace the inner core of garnet in the sketch illustrated by WHITTEN (1952). These relations are for the present deduced from that the unstable interval of solid solution between each end member of garnet might have been inferably replaced by the later minerals.

#### C. VESUVIANITE

Vesuvianites are, though always in minor amount, found involved in all of the deposits mentioned above. The radial growth of their slender prisms and their aggregates are often isolatedly embraced in wollastonite and fluorite while in the cases of their paragenesis with grossularite their prismatic crystals occur as the veins, accompanied with the former, running across limestone and wollastonite or along other skarn veins parallel or irregularly oblique to the walls.

As for their occurrence, BURNHAM (1959) reported their wide-spread but solitary distribution in limestone far from its contact zone with igneous rocks, WHITTEN (1952) illustrated the replacement of garnet with idocrase in his sketch, WATTERS (1958) pointed out both their stout-prismatic habit in the case of being included in the wollastonite zone and their shapeless crystals in the grossularite zone, and in spite of ECKERMAN's suggestion (1923), BARTH (1963) emphasized that microscopically there were no evidences pointing to instability of vesuvianite in skarns. On the other hand, WARREN and MODELL (1931) manifested an intimacy between grossularite and vesuvianite from the similarity in their values for  $a_0$  and  $c_0$ , McConnell (1939) confirmed a conspicuous interval between the two in his chemically obtained diagram and BURNHAM (1959) again deduced the producibility of grossularite, diopside and wollastonite through a simple addition of silica to idocrase. Anyhow, vesuvianites are accustomed to being replaced by the grossularite veins in most of the deposits mentioned in the present work exclusive of a part of the Yamato deposit revealing replacement of the latter with the former formed at the earlier stage and characterized by zoned structure.

Colors of vesuvianite are miscellaneously variable but seem to depend on its inner structure, being reflected on the special spacing and cell constant  $c_0$ , as are shown in Table 1.

Sample	Locality	Color	d (204, 440 <sup>+</sup> ) in Å	$c_0$ in Å	a <sub>0</sub> (Å) of paragenetic grossularite
1 (F 517)	Fukuju deposit (Yamato mine)	Greenish brown	2. 747	11.78	11.87
2 (F 517)	"	Red	2.753	11.81	_
3 (D 38)	Hōkoku adit (Yamato mine)	Brown	2. 751	11.79	_
4 (D 42)	"	Greenish brown	2. 751	11.79	11.86
5 (S 15)	Shinsei adit (Yamato mine)	Reddish yellow	2.756	11.78	
6 (M12)	Mihara mine (Okayama)	Tea-like	2.760	11.84	11.91
7 (S 14)	Sampō mine	Deep brown	2. 752	11.82	11.87

TABLE 1. RELATION BETWEEN VARIATION IN COLOR AND  $c_0$  of Vesuvianite and  $a_0$  of Paragenetic Grossularite.

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The values for  $c_0$  of vesuvianite in the table have been calculated form  $d_{(004)}$  and paragenesis of grossularite or increase of its  $a_0$  seems to influence to a slight extent but essentially in obscurity, while inspection of the related data in ASTM card (11-145) however indicates such a relation that the spacing for browish one is  $0.25 \sim 0.5\%$  larger in Å than that for yellow-colored.

#### D. Apophyllite

In the Yamato mine the apophyllite veins, 1mm in width on an overage, are found either traversing the assemblages of wollastonite, vesuvianite, and grossularite etc. and those of hedenbergite and quartz in the midst of the Fukuju deposit (Plate 45, 3 and Plate 51, 5) or running along the boundary of skarnized zone in the chert coming into contact with slender veins of wollastonite in the Hōkoku adit, being included in the small-scaled lodes,  $1\sim 2$ cm in width, composed chiefly of arsenopyrite in connection with the main deposits in this mine. In this case, apophyllites often occur in the interstices of arsenopyrite veins and of arsenopyrite crystals or as the veins along the former veins in chert and their own interstices are again filled with the later calcite (Plate 45, 4).

As a cue to deducing their formation temperature, those for this arsenopyrite together with other ones obtained in this mine have been determined on the basis of the theories related to the decrease in value of (131) spacing in connection with increase of S to As ratio (CLARK, 1960) as well as to correlatability of (131) spacing with the compositional variation (MORIMOTO and CLARK, 1961), the results being presented in Table 2.

Specimen	d <sub>(131)</sub> in Å	As %	Temperature of formation
a (D 32)	1.6335	33. 40	< 475°C
b (F 505)	1.6320	32.05	< 400°C
c (F 126)	1.6314	31.50	< 375°C
d (D 11)	1.6309	30.95	< 345°C

TABLE 2. FORMATION TEMPERATURE OF ARSENOPYRITE FROM THE YAMATO MINE

a: fine-grained crystals contained in the quartz vein including chalcopyrite worked on industrial scale in the Hökoku adit.

b: crystals occurring as masses in the arsenopyrite-bearing quartz vein in the Fukuju deposit.

c: those coexisting with the workable ores composed of chalcopyrite, sphalerite and galena associating calcite in the Fukuju deposit.

d: those obtained from the vein comprising apophyllite in the Hökoku adit.

Most significant is that in this deposit an intimate coexistence of apophyllite with arsenopyrite is restricted to in the case of the latter inferred as the lowest-temperature species, and furthermore worthy mentioning is the instability of apophyllite at the temperature higher than 300°C, as is deduced from the data of DTA and TGA for the specimens obtained from the coarse-grained and nearly pegmatitic granite

							Deadurat	abto!	1					
Ap	ophyll	ite	Specime	n from	Specime	n from	through	heating	W	ollasto	nite	Pseuc	lowoll	astonite
(AS:	ГМ 7-	170)	Yamate	o mine	Sampō	mine	of the spec	Sampo imen	(AS	5TM 10	0-487)	(AS	TM 1	0-486)
d(Å)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	hkl	d(Å)	I/I1	hkl
7.85	9	002			7.84	75								
7.76	9	101	7.80	100					7.7	40	200			
							7.66	11				ľ		
			6.34	13	6.38	11								
												5.87	10	100
												5.73	40	101
												5.10	20	102
4.534	20	103	4.54	65	4.54	39								
4.473	3	200			4.50	33						4.39	40	103
									4.05	10				•
3.943	100	004	3.94	91	3.950	39			•			3.97	10	005
3.886	6	211	3.88	43	3.892	36								
		•			3.857	21	3.826	26	3.83	80	400			
										•		3.75	20	104
3.606	2	104												
3.570	10	212	3.57	52	3. 577	42						3.58	$10_{B}$	
					3. 519	19	3.504	33	3.52	80	002			
							3.417	13	3.40	5		3.41	40	110
3.347	9	114			3.355	16								
					3. 323	22	3.306	41	3.31	80	202			
							3.226	40				3.20	100	112,105
3.168	5	220	3.16	30	3.160	57			3.16	5	111			
					3.099	18	3.082	27	3.09	30	202			
2.976	70	105	2.980	96	2.979	100	2.971	100	2.97	100	310	ĺ		
2.934	6	301	2.940	30	2.942	31								
2.812	3	314			2.816	9	2.803	26	2.80	. 10 <sub>B</sub>	311			
2.788	2	311			2. 799	9						2. 79	80	114
					2.722	4	2.715	8	2.72	10	402			
2.664	3	312	2.664	30	2.669	13						2.69	10	203
			2. 595											
					2. 557	7	2.554	8	2.55	30	112,600			
							2, 521	6				2.53	10	204
							2.474	21	2.47	60	112,402	2.46	60	116
							2.449	14			000			
							2.342	16	2.33	40	$510, 31\overline{2}$	2.35	20	205
					2.304	12	2.296	16	2.29	40	601, 203			
<u>.</u>			2.238	17										

1

TABLE 3. X-RAY DIFFRACTION DATA FOR APOPHYLLITE AT THE NATURAL STATE AND FOR ITS PRODUCT OBTAINED THROUGH HEATING AT 1000°C

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$															
	Ap	ophylli	tc	Specimer	ı from	Specimer	1 from	Product	obtained heating	w	ollasto	nite	Pscud	owoll	astonite
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	(AST	CM 7-1	70)	Yamato	mine	Sampō	mine	of the S	Sampō	(AS	TM IC	)-487)	(AS	TM	)-486)
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $						F		speci	men						
2.199       5       216       2.197       13       2.200       16       2.211       5       2.18       60 $\frac{607}{312}, 511$ 2.17       20       2112         2.157       3       202       2.159       11       2.172       9       1.18       1.18 <td< td=""><td>d(A)</td><td>I/I<sub>1</sub></td><td>hkl</td><td>d(Å)</td><td>I/I<sub>1</sub></td><td>d(Å)</td><td><math>I/I_1</math></td><td>d(Å)</td><td>I/I1</td><td>d(Å)</td><td>I/I<sub>1</sub></td><td>hkl</td><td>d(Å)</td><td>I/I<sub>1</sub></td><td>hkl</td></td<>	d(A)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>	d(Å)	$I/I_1$	d(Å)	I/I1	d(Å)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>	hkl
2.187       7       107       2.184       12       2.180       20       2.18       60       602, 511       2.17       20       2.17       20       2.17       20       2.17       20       2.17       20       2.17       20       2.17       20       2.17       20       2.12         2.103       10       315       2.108       2.08       5       403       2.10       20       2.13         2.004       4       2.010       4       2.018       7       2.018       5       403       2.10       20       213         2.004       4       2.021       7       2.018       15       1.98       20       602       10       214         2.004       4       2.021       7       2.018       1.91       20       602       10       214         1.930       33       1.935       34       1.936       6       1.91       20       600       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10       1.94       10 <td>2.199</td> <td>5</td> <td>216</td> <td>2. 197</td> <td>13</td> <td>2.200</td> <td>16</td> <td>2.211</td> <td>5</td> <td></td> <td></td> <td>_</td> <td>2.21</td> <td>20</td> <td>211</td>	2.199	5	216	2. 197	13	2.200	16	2.211	5			_	2.21	20	211
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	2.187	7	107			2.184	12	2.180	20	2:18	60	$\frac{602}{312}, 511$			
2.155       3       202       2.108       26       2.159       11       2.106       30       2.108       20       2.13       2.008       5       403       2.10       20       213         2.104       6       420       2.007       17       2.003       21       2.018       7       2.01       20       512       1.96       60       2.03       10       214         2.004       6       420       2.007       17       2.003       21       1.988       15       1.98       20       602       1.96       60       300,118         1.930       33       1.935       4       1.975       31       1.94       10       1.95						2.172	9					512,	2.17	20	212
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	2.155	3	202	1		2.159	11								
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	2.109	10	315	2. 108	26	2.106	30								
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	2.103	10	324							2.08	5	403	2.10	20	213
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						2.040	4						2.03	10	214
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		,				2.021	7	2.018	7	2.01	20	512			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	2,004	6	420	2.007	17	2.003	21								
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$								1.988	15	1.98	20	602			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$								1.975	31				1.96	80	300,118
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.930	33	1.935	4				1.94	10	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.906	6	1.916	5	1.91	20	800			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.878	4	1.878	6	1.88					
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$								1.849	5	1.86					
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.831	4	1.824	19	1.83	60		1.828	30	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$										1.80	5				
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.792	7			1.79	5				
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.771	7				1.786	7	1.789	6						
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.764	10				1.765	18	1.752	11	1.75	40		1.742	20	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.721	5				1.720	6	1.713	16	1.72	60		1.708	10	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.675	3		1.671	13	1.675	4	1.679	5				1.684	30	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.655	2				1.649	11								
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.620	2				1.620	4	1.615	7						
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	1.607	2				1.607	4	1.599	12	1.602	40		1.601	30	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.578	55		1.579	22	1.579	15								
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$						1.545	9	1.542	5						
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$						1.533	6	1.531	7	1.531	10		1.533	10 <sub>D</sub>	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.522	8											1.529	10	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	1.510	2						1.512	5	1.515	5				
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$						1.490	7					ĺ	1.493	60	
1.470       4       1.408       7       1.471       12         1.453       2       1.459       7       1.455       30         1.442       2       1.415       4       1.426       5         1.398       4       1.305       4       1.387       5         1.306       6       1.358       30								1.474	11	1.478	$20_B$				
1.453       2       1.459       7       1.455       30         1.442       2       1.415       4       1.426       5         1.398       4       1.305       4       1.387       5         1.305       4       1.358       30	1.470	4				1.408	7	1.471	12						
1.442       2         1.398       4         1.305       4         1.360       6	1.453	2				1.459	7			1.455	30				
1.398       4       1.415       4       1.426       5         1.305       4       1.387       5         1.305       4       1.358       30	1.442	.2													
1. 398     4     1. 305     4       1. 305     4     1. 358     30		· ·				1.415	4			1.426	5				
1.305     4       1.360     6       1.358     30	1.398	4								1.387	5				
1.360 6 1.358 30						1.305	4								
						1.360	6			1.358	30				

TABLE 3 (continued)

(Vorma, 1961).

In the Sampo mine apophyllites are observable not as the well-defined veins but as predominant impregnations appearing limitedly within a zone in such a skarnized part as is found distributing definitely between granite and limestone while the main ore-body situates in limestone or in slate revealing the irregular but sharp contact with ES strike and southward (in the upper part) or northward (in the lower part) plunge roughly along the boundary between the sedimentaries on the northern side and the granitic rocks on the southern side, and in the part where its wings composed almost purely of massive magnetite are squeezed the skarn veins become predominant, as are confirmable typically on No. 7 level in the main gallery. Notwithstanding the limited width of  $2\sim15$ cm between granite and limestone is there such a marked zonal disposition of the mineral assemblages as (Plate 52, 2, 3): (A) granite-(B-zone) prehnite and hendenbergite in association with wollastonite-(C-zone) grossularite and vesuvianite-(D-zone) wollastonite, apophyllite, and fluorite accompanied with prehnite-(E) limestone, of which C-zone, reddish brown in color, is almost linearly demarcated with B and D while the remainders are in irregular occurrence and in part deficient: going into details, B-zone is found thrusting out into, or running through, (A); hedenbergite is accustomed to traversing along, or into, C-zone on (A) side and vesuvianite, deep brown in color, to distributing on (E) side; wollastonite in D-zone bears a tendency coming into contact with (E); and any variations other than saccharoidal textures recognized in proper crystalline limestone are not affirmable even in (E) adjoining with these veins. Microscopically, apophyllite grains accompanying fluorite are present sporadically in wollastonite and in prehnite, filling the interstices in the crystals of the latters (Plate 46, 1, 2). The branches of these veins poking irregularly into limestone commonly involve wollastonite, and radite and hedenbergite accompanied with the later fluorite replacing andradite and associating, or replaced with, the ore minerals (Plate 42, 1 and Plate 52, 1).

It is of interest that x-ray diffraction for the final product derived from heating of apophyllite, dim brownish in color and translucent, taken out from D-zone, at 1,000°C for 6 hours surely results in emergence of pseudowollastonite and wollastonite (see Table 3). Save for the latter acquired similarly through heating of hydrogrossular, vesuvianite and prehnite at the same temperature, produceability of the former merely from apophyllite is specifically to be noted in relation to its native genesis.

In the Mihara mine (Hiroshima) the irregularly branching veins of skarns such as wollastonite outside and andradite inside accompanied with fluorite and some others are found thinning out in saccharoidal limestone (Plate 53, 5) and slate near their contact with granite. Parageneses of apophyllite with wollastonite are also common though the mutual relation, whether carlier or later, be difficult to discriminate excepting the special case. X-ray inspection for the specimens collected from some masses, corroded in shape, orange in color and more or less than 1cm in diameter, in such parts as are relatively abundant in apophyllite indicates the coexistence of

wollastonite, apophyllite and unknown crystals with intense diffraction at 14.8Å pointing probably to a sort of decomposition product after apophyllite. Another sort of pale greenish veins with repeated stripes of andradite, diopside and wollastonite etc. in the arrangement parallel to the walls,  $3\sim10$ cm in width, are found including apophyllite in amount far less than are observed at the ends of the conspicuously splitted veinlets.

#### E. PREHNITE

The minerals obtained from the Sampō mine are white in color, massive in form, observable megascopically in the skarn veins traversing along the contact zone between granite and limestone, and characterized microscopically by lamellar structure and undulatory extinction (Plate 46, 3, 4). Worthy mentioning is that those included in B-zone described already in the paragraph of apophyllite considerably replace the crystals of hedenbergite and those contained in the part of B-zone sticking into aplitic granite at the same time reveal replacement for the main components of the latter such as microcline and a small amount of plagioclase, both bearing no vestige of the replaced minerals and coexisting with wollastonite, fluorite and apophyllite though different from one another in formation stage.

Those comprised in the grossularite-vesuvianite vein penetrating irregularly across wollastonite on the middle level of the Daidō deposit in the Yamato mine are, because of their extremely minute size, mega- and microscopically difficult to be identifiable but those embraced in these veins or in other veins, bluish grey in color and irregular in form, running nearly parallel to the former and chiefly composed of diopside and feldspar are only röntgenometrically confirmable, both being surely formed at the later stage of not different but continued process of mineralization and thus replacing all of the earlier minerals.

Their occurrence in the similarly later veinlets coexisting with grossularite and some others in serpentinite has been described in the former report.

## F. SCAWTITE AND HILLEBRANDITE

(1) Scawtite. Its occurrence together with those of plazolite, cuspidine, bultfonteinite and hillebrandite are for the time being restrictedly assurable in the Mihara mine (Okayama).

The ore deposits in this mine are found embraced in the skarns formed in a part of the Paleozoic formation of limestone, schalstein and slate, producing chalcopyrite, cubanite, pyrrhotite, magnetite and so forth as the main ore minerals. Their largest mass, flattened in shape and 6m in maximum width, indicates a trend with strike of 70°W and dip of N30°~40° in such a scale as is continuously extended by about 130m along strike-side and down to about 250m along dip-side, wherein of due significance is that chalcopyrite is most predominant in the upper part, pyrrhotite in the middle part and magnetite in the lower part. Near or within the main

<u></u>	Scawt (ASTM 12	ite 2-777)	Scawtif the Miha (Okay	te from ara mine yama)	Product through scawtite Mihar	obtained heating of from the ra mine	Wollas (ASTM	stonite 10-487)	Calcio-o (ASTM	olivine 1-1029)
d(Å)	I/I1	hkl	d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	d(Å)	I/I1	d(Å)	I/I <sub>1</sub>
8.36	20	110	8.33	6				-		
7.93	20	?	7.62	6	7.63	9	7.7	40		
5.99	60	011,101	5.99	13						
					-				5.6	8
4.99	20	200	4.95	10						
4.52	60	130	4.52	26					4.32	24
4.19	60	121,220	4.18	21			4.05	10	4.05	6
3, 80	60	040	3.789	15	3.813	24	3.83	80	3.80	24
3, 55	60	211	3.535	22	3.500	22	3.52	80		
							3.40	5	3.38	9
					3.302	26	3.31	80		
			3.238	6			3.16	5		
3.21	60	141,112	3.200	28			3.09	30		
3.03	100	141,240	3.015	100	3.074	20	2.97	100	3.01	48
2.99	100	022,202	2.988	82	2.967	100				
2.96	40	231,321	2.954	32						
			2.884	4	2.864	11	2.80	10 <sub>B</sub>	2, 89	9
2.78	60	330,301+							2.74	100
			2.768	23	2.745	<b>5</b> 0	2.72	10		
					2.714	30			2.60	6
			2.594	3	2.612	13	2.55	30		
2.54	20	060,132+	2.540	13	2.550	7			2. 51	9
2.52	40	202	2.520	19						
2.49	60°	251,400+	2.485	34			2.47	60		
					2.469	. 17			2.44	6
			2.443	6	2.434	7				
2.39	20	222	2.390	4	2.399	6	2.33	40	2.32	6
2.33	20	161,251	2.327	6	2.343	15	2.29	40		
			2.287	7	2, 292	17				
					2.275	13			2.24	5
			2.252	7						
2.23	60	152	2.226	31						
2.21	40	103	2.206	16						
2.17	5	411	2.170	6	2.182	41	2.18	60	2.18	6
2.15	5	013	2.145	7						
2.12	20	170,152+	2.111	12						

TABLE 4. X-RAY DIFFRACTION DATA FOR SCAWTITE AT THE NATURAL STATE AND FOR ITS PRODUCT OPTAINED THROUGH HEATING AT 1000°C

# Hiroshi MIYAKE

	-	· · · · · · · · · · · · · · · · · · ·		I AB		<u> </u>				
(AS	Scawtit STM 12-	с -777)	Scawtite from the Mihara mine (Okayama)		Product through l scawtite Miharz	obtained neating of from the a mine	Wollas (ASTM	tonite 10-487)	Calcio- (ASTM	olivine 1-1029)
d(Å)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>
			2.090	7	2.085	6	2.08	5		<u> </u>
					2.042	7				
2.01	60	062,303+	2.008	13	2.020	15	2.01	20	2.02	6
					1.979	11	1.98	20		
					1.974	13				
					1.938	7				
			1.915	10	1.916	7	1.91	20	1.90	48
1.89	80		1.886	34	1.891	6				•
1.87	60				1.879	6	1.88	20		
			1.862	16			1.86	10		
					1.827	17	1.83	60	•	
1.81	40		1.801	7			1.80	5	1.80	32
					1.788	7	1.79	5		
1.777	40		1.772	15						
1.776	40		1.763	13						
					1.753	· 11	1.75	40	1.75	20
			1.743	6						
					1.713	18	1.72	60		
									1.68	20
					1.619	9			1.63	28
				۵ <b>۵</b>	1.600	15	1.602	40		
					1.581	7				
					1.547	9				
					1.542	9				
							1.531	10	1.53	8
							1.515	5	•	
									1.50	6
					1.472	9	1.478	$20_B$	1.47	9
					1.452	6	1.455	30		
					1.426	6			1.44	5
							1.426	5		
					1.360	6	1.387	5	1.41	5
							1.358	5		

TABLE 4 (continued)

ore-body are observed the dike-like occurrences of diorite, porphyrite and their intermediates formed prior to genesis of the skarns as well as of quartz-porphyry containing the fragments of deep-reddish grossularite while some small-scaled lodes named 'Jamon', 'Yataka' and 'Sempo' etc. are also found traversing near the main orc-body.

Several kinds of the skarn assemblages characterized leadingly by garnet and hedenbergite in specific association with some hydrous minerals are recognizable inside and outside of the ore bodies. Remarkable is that the contact between limestone appearing in the northwestern part of No. 12 adit and porphyrite occurring in its southeastern part is flattened, well demarcated with each other and intercalates only the calcite veins, less than 1cm in width, but no trace of the metasomatic effects, whereas porphyrite is found replaced with garnet-vesuvianite skarns accompanied with hydrous calium silicates such as scawtite and others. The massive aggregates, about 2cm in diameter, are in part composed merely of deep-brownish vesuvianite in the core and yellow-brownish grossularite, and it seems a tendency that the greater their diameter, the more the amount of hydrous silicate become abundant (Plate 47, 1, 2, Plate 53, 1 and Fig. 1, a). In these cases a sort of crustal zonation is, though common but somewhat variable, markedly establishable as follows:

porphyrite—yellowish pink-colored crust—yellowish brown-colored crust of grossularite and vesuvianite—yellowish white-colored crust of plazolite—white to greyish white-colored crust of scawtite

These examples are discernible on the eastern side of No. 10 adit in the main ore-body and on the southern side in the eastern part of No. 2 adit of the Yataka lode, being in their neighborhood accompanied with chalcopyrite and magnetite without any relation to the skarns in apparent occurrence in further association with the veinlets of deep-reddish brown-colored grossularite in the former adit and calcite veins with width of  $5 \sim 10$ cm in the latter and No. 12 adit, penetrating across porphyrite.

Scawtite is white, greyish white or pale fleshy in color, flinty and conchoidal in fracture, effervescent through reaction with cold solution of diluted hydrochloric acid though not so much as in the case of limestone, and microscopically composed of aggregates of fine grains including the patches of calcite in some case (Plate 47, 3) and cutting by the later veinlets of its own in other cases. X-ray diffraction data for the very mineral hand-picked megascopically as the most pure specimen are arranged with those for the heated product, which will be referred to later, and other resembling minerals in Table 4.

Various views as to genesis of the mineral concerned have already been proposed. TILLEY (1938) regarded it as a pseudomorph after spurrite, and MARSON (1956) did so as a primary mineral formed directly through such a low-temperature metasomatism of limestone as produces larnite and gehlenite in connection with certain intrusion or as a sort of the secondary mineral formed after wollastonite through hydration of wollastonite-calcite rock but at the same time reached another conclusion that extraordinarily high water-pressure caused by emanation from the intrusive or

obtained from high content of water in limestone might have been more suitable for formation of scawtite than for that of anhydrous wollastonite. In relation to this, the patches of calcite recognized in massive scawtite, as stated above, may prove an intimate affinity between the two but whether the former might be derived from the pre-existed limestone or not is of uncertainty. Moreover, the presences of calcite veins cutting through massive scawtite and of the veins composed simply of scawtite are properly believed to offer a cue for justifying the genesis of calcite through certain process almost contemporaneous with that of scawtite while the former is hardly ascribed to capturing of limestone in porphyrite because of their well-defined contact without any trace of reaction. Occurrence of the isolated scawtite-veins may also give disproof against the view suggesting their formation as the pseudomorphs after wollastonite and detailed inspection surely points to none of wollastonite in every type of scawtite though a very few amount of the former are merely röntgenometrically discernible with grossularite and vesuvianite in the skarn zone coming into contact with porphyrite.

McCONNELL indicated the reduction of birefringence in the product obtained through heating of Ballycraigy scawtite at 850°C to be due to decarbonation and its similarity to wollastonite in x-ray diffraction patterns. For comparison, the specimen, fleshy white in color, sampled in the Mihara mine has also been subjected to heat treatment for each duration of 6 hours with respective increase of  $100^{\circ}$ C at  $100^{\circ} \sim 1,000^{\circ}$ C, and after allowed to cooling in the air at the room temperature, the product has been röntgenometrically analysed. The results obtained exhibit that at 700°C the spacings for scawtite partially decrease their intensity and the new ones appear, at 800°C all of the forms completely disappear and the remainders increase in number, at 900°C numbers of the remainders increase a little more in their intensity, and at 1,000°C the spacings are not different in position and intensity from those at 900°C. As are shown in Table 4, it has become evident that the final products acquired through heating reached a good agreement with wollastonite and calcio-olivine after an assumed reaction:

 $6\text{CaO} \cdot 6\text{SiO}_2 \cdot 2\text{H}_2\text{O} \cdot \text{CaCO}_3 \rightarrow 5(\text{CaO} \cdot \text{SiO}_2) + (2\text{CaO} \cdot \text{SiO}_2) + 2\text{H}_2\text{O} + \text{CO}_2$ scawtite wollastonite calcio-olivine

The spacings for wollastonite produced through heat treatment of scawtite are, though more or less different from those for the natural one and impossible to be identified with those for parawollastonite, remarkably allied to those for the synthetic one given by TUTTLE and HARKER (1957). The spacings other than those for wollastonite are in accord with those for calcio-olivine corresponding to  $\tau$ -Ca<sub>2</sub>SiO<sub>4</sub> which is obtainable through slow cooling of the product formed through heating of the solid substance with the same compositions for 30min. at 1,525°C down to the room temperature but convertible into  $\alpha'$ -modification represented by native skannonite combined dimorphically with larnite ( $\beta$ -modification) occurring often in the skarn zone at the temperature higher than 820°C. After all, it results in that scawtite becomes unstable at this temperature and decomposes into wollastonite and others.

Another occurrence of scawtite has been recognized in the Daidō deposit of the Yamato mine, wherein the mineral in question is contained in the greenish grey part intervening between wollastonite and malachite zone pertaining to the first ore-body (Daiichi kōtai) composed of the secondarily enriched ores such as cuprite, malachite and so on, filling the cavities and fissures of limestone and wollastonite. Though megascopically difficult but only röntgenometrically possible to discriminate it from wollastonite, scawtite in this case is believed as an alteration product from wollastonite.

(2) Hillebrandite. Earthy veins, white in color, soft in property and  $3\sim 4mm$  in width, are found cutting almost linearly through the masses of scawtite, occurring in a close relation to the latter in the case of the Mihara mine. X-ray diffraction data for the specimens taken out from these veins surely indicate the content of hillebrandite and calcite, as are given in Table 5.

Hilleb from the mine (O	randite Mihara kayama)		Hillebrandite (ASTM 9-51)	······································	Cal (ASTM	cite 5-0586)
d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>
8.21	9	8.2	50	200		
6.73	5	6.7	20	201		• •
5.93	. 5	5.8	40	002		
4.80	20	4.76	90	202		
4.08	10 <sup>.</sup>	4.06	50	400		1
3.860	15				3.86	12
3.558	7	3. 52	50	120		
3.361	23	3.33	90	121		
3.034	100	3.02	80	122	3.235	100
2.933	36	2.92	100	321		
2.837	20	2.82	80	403,204		
2,782	19	2.76	80	600		
2.761						
2.722	4					
2.699	4	2.70	40			
		2.67	40	322,501		
2.649	7	2.63	50	123		
2.496	16				2.495	14
2.476	7	2.45	· 40	520		
2.394	11	2.37	80	521		
2.284	23				2.285	18
2.250	10	2.26	70	205		
		2.23	70	603,124+		
		2.10	10	324		
2.094	20				2.095	18
2.070	8	2.06	70	800		

TABLE 5. X-RAY DIFFRACTION DATA FOR HILLEBRANDITE.

#### Hiroshi MIYAKE

#### G. PLAZOLITE

That an identification of plazolite or hydrogrossular is not easy because of confusion with ordinary grossularite has been reported in the preceding work. In the Mihara mine, however, presence of the very mineral is precisely ascertainable in certain zone of the white-colored, massive materials including a plenty of minute grains of brown- to yellow-colored garnets,  $3\sim10$ cm in width, running between the whitecolored, central part of scawtite and the yellow brown-colored part, rich in garnet and vesuvianite, coming into contact with porphyrite (Plate 47, 4). X-ray diffraction data for the specimens sampled from this zone and treated with diluted hydrochloric acid evidently suggest the coexistence of hydrossular, vesuvianite and calcite, as are presented in Table 6.

Hydrogrossular in this case is of  $a_0 = 12.16$ Å and hence quite similar to plazolite given first by PAEST (1937). Referring to YODER's theory (1950) as to the relation of the unit-cell to the composition, its content of water is estimated  $2.5 \sim 3.0$  mol.

MASON (1957), after YODER's view, deduced its formation at 500°C from the research of the specimens obtained from the contact between limestone and dike rock, SCOTT (1951) ascribed its genesis to repeated precipitation from hydrothermal solution on the basis of presence of hydrogrossular replacing the phenocrysts of olivine in basalt and of appearance of concentrically colored layers with different refractive indices in hydrogrossular, BARRER and DENNY (1961) showed the results for synthetic minerals belonging to a series between grossularite and hydrous garnet in the experiments with gels of 3CaO·Al<sub>2</sub>O<sub>3</sub>·nSiO<sub>2</sub>aq at 200°~450°C, according to a part of which the ratio of SiO<sub>2</sub> to H<sub>2</sub>O in the product relates to the refractive indices of the raw materials and to heating temperature, the values for ao are variable in the range of  $12.6 \sim 11.89$ Å, and a gap without such compositions as correspond to 0.51<Si<1.81 per residual 3CaO·Al<sub>2</sub>O<sub>3</sub> is present between high-silica and low-silica fields, being correlative to the solubility diagram given hypothetically by YODER, who on the other hand pointed out the decomposition of grossularite into gehlenite, wollastonite and anorthite at the temperature lower than 1080°C, although the present writer however confirmed impossibility of this decomposition below 1.000°C. As for dehydration of hydrogrossular, BELYANKIN and PETROV (1941) recognized either the endotherms at  $650^\circ \sim 690^\circ C$ , connected with dehydration by Smith (1952) through decrepitation method or the exotherms at 870°C and 940°C. and SKINNER (1956) combined the endotherm revealed by hibschite (3CaO·Al<sub>2</sub>O<sub>3</sub>. 2SiO<sub>2</sub>·2H<sub>2</sub>O) at 650°C with its destruction and dehydration, while FRANKEL (1959) denied the decomposition of hydrogrossular up to 1,100°C and BARREN and DENNY manifested no variation in natural grossularite and synthetic hydrogrossular with ignition at 1,000°C. As a result, dehydration of hydrogrossular seems, though still of vagueness to more or less extent, to be influenced in temperature with its compositional variation.

In the light of these results, special cautions have been paid to heat treatment of hydrogrossular through such procedures as were similar to in the case of scawtite.

Plazoli the Mih (Okay	te from ara mine yama)	Plazo	lite (Hydrogr (ASTM 3-080	rossular) )1)	Idoo (ASTM	crase [11-145]
d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	hkl	d(Å)	I/I <sub>1</sub>
6. 10 5. 95 4. 96 4. 90	9 6 12 7	4.96	40	211	11. 0 5. 90 4. 69	5 10 5
4.31	7				4.03	20
3. 485 3. 241 3. 042 2. 995 2. 954 2. 751	4 18 42 19 19 37	3. 25 3. 03	40 80	321 400	3. 469 3. 244 3. 054 2. 999 2. 946 2. 752	20 10 20 5 40
2. 718 2. 595 2. 479	100 30 25	2.71 2.60 2.48	100 20 60	420 332 422	2. 593	80
2. 438 2. 383 2. 327 2. 305	21 4 4	2. 38	60	510,431	2. 452 2. 332	50 10 <sub>B</sub>
2. 217 2. 182 2. 161	34 6 6	2. 21	80	521	2. 194	10
2. 148	36	2.14	80	611 532	2.122 1.997	30 10 <sub>B</sub>
1.926	4	1.92	20	620	1.960	5
1.909 1.884 1.799 1.751 1.719	4 4 12 4	1. 76	50	444	1.882 1.793 1.762	10 5 20
1.686	19 12	1.68	80	640	1.679	5 30
1.623 1.606	42	1.62	100	642	1.621	60
1.558 1.544 1.519 1.493 1.478	4 4 12 6 4	1.52	50	800	1.568 1.556 1.539 1.525 1.495 1.476	5 10 5 10 5
1.360 1.343	13 4 10	1.36	60	840	1. 374 1. 345	5 10
1. 324	10	1.32	40	842		- 2 
1. 294	7	1.29	40	664	1. 297 1. 280 1. 264	20 10 10 <sub>B</sub>

TABLE 6. X-RAY DIFFRACTION DATA FOR PLAZOLITE.

Taking account of the specimens mixed with calcite, though removed through washing with diluted hydrochloric acid, and unavoidably with vesuvianite, the latter only has been preliminarily subjected to the treatment in a quite similar condition. Either the deep-brownish one obtained from the Yamato mine or the greenish one

Product obtai heating of from the Mi (Okaya	ned through plazolite hara mine ama)	Gehl (ASTM	enite [ 9-216)	Wolla (ASTM	stonite 10-487)	Gross (ASTM	ularite 3-0826)
d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	d(Å)	I/I <sub>1</sub>	d(Å)	I/I1
5. 44 5. 07 4. 23 3. 835 3. 697	8 7 5 6 20	5. 46 5. 08 4. 22 3. 67	5 5 3 20	3. 83	80		
3. 509 3. 433 3. 305 3. 182	7 7 8 7	3. 43	1	3. 52 3. 31 3. 16	80 80 5		
3.000 2.975 2.841	24 31 100	3.07 2.85	25 100	3.09 2.97	30 100	2.96	80
2.715 2.656	7 19	2. 72	5	2.80	10 <sub>B</sub> 10	2.65	100
2.529 2.471 2.429	6 4 16	2. 535 2. 435	7 20	2. 33	60	2. 53 2. 44	20 60
2. 394 2. 330 2. 285	18 5 14	2. 404		2. 33 2. 29	40 40	2. 33	50
2. 183 2. 170	6 5	2.135		2.18	60	2.16	60
2.039 2.034	11	2. 126 2. 043	1 15	2.08	5	2. 10	20
1.963 1.920	5	1.967 1.924	1	2.01 1.98	20 20	1.92	70
1.863 1.847	5	1.864	5	1.91 1.88 1.86	20 20 10	1.52	10
1.831 1.810	68	1.812	9	1.83 1.80 1.79	60 5 5		
1.748 1.713 1.645	28 11 5	1. 754 1. 723	35 5	1.75	40 60	1.71	60 80
1.623 1.605 1.599	5 5 5	1.632 1.616	1 3	1.602	40		
1.585 1.541 1.512 1.509	7 4 6	1.519	15	1.531 1.515	10 5	1.58	90 50
1.471	4	1.475	1	1.478	20	1.45	50
1.429	5	1.437	5	1.426	5		
1.372	8	1.375	9	1.507			

Table 7. X-ray Diffraction Data for the Product Obtained through Heating of Plazolite at 1000  $^\circ\mathrm{C}$ 

from the Sano mine indicate no difference at 900°C and complete decomposition at 1,000°C through such an assumed reaction as:

 $6\text{CaO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2 \rightarrow 2\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2 + 3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{SiO}_2 + \text{CaO} \cdot \text{SiO}_2$ 

vesuvianite gehlenite grossularite wollastonite X-ray diffraction data clearly justify this assumption. In the case of plazolite, the spacings for vesuvianite begin to be weaken at 800°C and vanish at 1,000°C, while those for plazolite reveal no changes up to 600°C, partially begin to be weaken at 700°C, decrease in such intensity as half of the initial at 800°C, and then perfectly disappear but those for gehlenite and grossularite newly emerge at 900°C, and those for the former markedly set off their intensity, those for the latter is invariable and those for wollastonite newly appear at 1,000°C (see Table 7). At this step it is difficult to determine whether emergence of gehlenite and wollastonite may be related to decomposition of plazolite or to that of vesuvianite contained in a small amount, but comparison of the spacings for each product from plazolite with those obtained preliminarily for vesuvianite yields a cue to such a possibility that plazolite also thermally decomposes into gehlenite, grossularite and wollastonite as follows:

 $\begin{array}{ccc} 3 \left( 3 CaO \cdot Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O \right) \rightarrow 2 \left( 2 CaO \cdot Al_2O_3 \cdot SiO_2 \right) + 3 CaO \cdot Al_2O_3 \cdot 3SiO_2 \\ & \text{plazolite} & \text{gehlenite} & \text{grossularite} \\ & + CaO \cdot SiO_2 + 6H_2O + CaO \\ & \text{wollastonite} & ? \end{array}$ 

In spite of the fact that decomposition of plazolite begins at 700°C and is accomplished at 900°C, its  $a_0$  displays a diminution at 600°C compared with that at the room temperature while all other spacings reveal no variation in intensity, implying an essential variability of this mineral at certain temperature lower than properly expected. To be added to here is that the decomposition temperature of vesuvianite in the case of coexisting with plazolite is lower than that in the case of being solitarily present. It is however questionable whether a view as to the catalytic effect through the coexisting mineral, elucidated by ROUTSALA (1963) concerning the other mineral, may hold, just as it is, in this case or not. In relation to this, that the products obtained through heating of plazolite are, as shown above, somewhat deviated from such an equilibrium suggested by YODER at 750°C and ordinary pressure as hydrogrossular  $\rightarrow$  gehlenite + wollastonite + anorthite may be concerned with accentuated formation of grossularite in the place of anorthite due to the presence of vesuvianite though in a little amount.

# H. CUSPIDINE AND BULTFONTEINITE

(1) Cuspidine. The mineral, white in color and opaque, is found occurring in scawtite included in the skarns traversing or replacing porphyrite in the Mihara mine (Okayama), being well distinguishable from scawtite in the cases of its veinlets, less than 5mm in width, but not so in the cases of its massive aggregates, less than 5cm in diameter, varying occasionally into scawtite (Plate 48, 1, 2 and Plate 43, 4). In the part within 10cm from porphyrite, massive cuspidine is complicatedly mingled

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with greyish scawtite, pale greenish vesuvianite, brownish garnet and calcite, from the greenish brown-colored part in which vesuvianite and bultfontenite have been discerned röntgenometrically.

Cuspidin the Miha (Okay	e from ra mine ama)	Synthetic (ASTM	cuspidin <del>e</del> 11-75)	Cuspidir the Miha (Okay	ne from ara mine vama)	Synthetic (ASTM	cuspidine 11-75)	
d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	
7.36	12	7.36	5	2.050	17	2.047	7	
7.11	7	7.11	3	2.014	28	2.016	17	
5.27	9	5.27	3	1.989	14	1.995	5	
5.14	5	5.13	5 <sub>D</sub>			1.985	5	
		4.62	3 <sub>D</sub>	1.962	5	1.959	1	
4.57	5	4.55	3 <sub>D</sub>	1.949	7			
		4.23	3	1.912	5			
3.681	12	3.68	7	1.882	24	1.884	25	
		3.42	5			1.832	7	
		3. 361	20	1.822	26	1.821	11	
3.254	55	3.257	7	1.810	17	1.809	7	
3.064	100	3.064	100	1.780	5			
2.936	30	2.944	17	1.770	8			
		2.930	15	1.729	21			
2.898	58	2.898	17	1.684	9			
2.869	97	2.871	40	1.655	4			
		2.819	5 <sub>D</sub>	1.621	5			
		2. 568	5	1.605	7			
2.551	17	2.550	7	1.598	8			
2.522	8	2. 520	1	1.576	8			
2.488	12	2.493	9	1.534	4			
2.450	10	2.452	5	1.487	9			
		2.307	9	1.467	4			
2.293	18	2. 292	15	1.447	5			
2.153	4	2.159	3	1.440	7			
2.086	8	2.089	3	1.410	4			
2.058	17	2.063	9	1.407	4			

TABLE 8. X-RAY DIFFRACTION DATA FOR CUSPIDINE.

X-ray diffraction patterns for cuspidine are in better accordance with those obtained through hydrothermal treatment by Van VALKENBURG and RYNDERS (1958) rather than the specimen from Monte Somma, Italy (see Table 8). Moreover, no phase changes are recognized up to 1,405°C, melting point of cuspidine, on DTA curve given by them. The results for heat treatment concerning the specimens sampled from the veinlets in scawtite in the Mihara mine are: at 800°C the spacings of cuspidine get to dwindle and the new ones bring forth, at 900°C the former completely disappear and those of alite, a sort of synthetic minerals in clinker, and lime become

evidently visible, and at 1,000°C the remained more and more increase in intensity while the spacings other than those of cuspidine and alite appeared at 800°C are utterly lost at 900°C and the products corresponding to these spacing are for the present impossible to be clarified to sufficient extent, as are given in Table 9. As a

Product obta heating of from the M (Okay	ined through Cuspidine Iihara min <del>e</del> yama)	A (ASTM	lite I 9-352)	L (ASTM	ime [ 4-0777)
d(Å)	I/I <sub>1</sub>	d(Å)	I/I1	d(Å)	I/I <sub>1</sub>
4.62 3.810 3.781	7 7 7 7	5. 901 3. 862 3. 510	40 30 20		· · · · · · · · · · · · · · · · · · ·
3. 365 3. 223 3. 169	6 7 6	3. 346 3. 227	20 10		
3. 035 2. 868 2. 772	9 21 100	3. 022 2. 957 2. 891 2. 818 2. 776	80 60 30 10 100	2. 778	34
2. 732 2. 602 2. 531 2. 434	69 37 9 13	2. 730 2. 670 2. 602 2. 549 2. 449	80 10 100 10 30		
2. 398 2. 274	31 22	2. 326 2. 304 2. 277 2. 234	60 50 20	2.405	100
2. 185 2. 159 2. 125 2. 087 2. 087	44 15 6 7	2. 185 2. 159 2. 125	100 100 30		
2.080 2.041 2.018 1.979	9 12 19 22	2.083 2.045 2.011 1.979 1.940 1.926	40 20 10 60 <sub>B</sub> 70 60		
1.910 1.907 1.889	7 9 12	1.900 1.863	20 <sub>B</sub> 10		
1.839 1.817 1.796	4 4 10	1.825 1.797	60 <sub>B</sub> 30		
1. 721	7	1.771 1.752	90 80		
1. 622	10	1.642 1.632 1.623	20 80 60	1.701	45

Table 9. X-ray Diffraction Data for the Product Obtained through Heating of Cuspidine at  $1000\,^\circ\text{C}$ 

result cuspidine is believed stable at the temperature lower than 800°C and its decomposition process may run as:

,

$$2(3CaO \cdot 2SiO_2 \cdot CaF_2) \rightarrow 2(3CaO \cdot SiO_2) + 2CaO + SiF_4 + SiO_2$$
  
cuspidine alite lime ?

The present data seem in opposition to the suggestion given by VAN VALKENBURG and RYNDERS for convertibility into calcio-olivine through heating of synthetic cuspidine, though different in property from the native one, for 12 hours at 1,200°C in connection with that, as has been stated already, calcio-olivine should be stable at the temperature lower than 820°C and convertible into  $\alpha'$ -modification above this

Bultfonteinite from the Mihara mine (Okayama)		I (/	Bultfonteinite ASTM 8-223)	Hibschite (Hydrogarnet) (ASTM 4-0723)		
d(Å)	I/I <sub>1</sub> ·	d(Å)	I/I <sub>1</sub>	hkl	d(Å)	I/I <sub>1</sub>
8. 16 7. 10 6. 58 5. 65 5. 47	50 18 21 18 21	8. 12 6. 51	60 20 20	010 110 200		
4. 30 4. 08 3. 986 3. 901 3. 829	14 28 18 18 18	4. 06 3. 95	40 5	111 201	4.89	5
3. 641 3. 583 3. 507 3. 456 3. 335	18 25 53 54 14	3. 636 3. 60 3. 50 3. 44 3. 33 3. 26	5 5 40 40 5 5	211 211 211 021 220 120		
3. 032 2. 925 2. 889 2. 823 2. 776 2. 755 2. 738	39 93 100 61 57 89 50	2. 92 2. 88 2. 825 2. 77 2. 743 2. 72	60 60 20 40 30	121 221 221 221 221 102	3. 00	80
2.667		2.72	20 5	102	2.68	100
2.604 2.495 2.483	32 18 18	2.001	Ŭ		2. 57	20
2.461	25				2.46	50
2.353	18	2.36 2.333	5 5		2.36	40
2. 276	25	2. 272	30 <sup>-</sup>		2. 19	50
2. 184 2. 119 2. 037	25 36 28	2. 118 2. 037	40 50		2. 12	5
1.985 1.932	39 100	1.988 1.953 1.93	20 5 100		1.95	60
1.892	32	1.912 1.887	5 5			

TABLE 10. X-RAY DIFFRACTION DATA FOR BULTFONTEINITE.

#### temperatue.

(2) Bultfonteinite. The mineral appearing similarly in such skarns as contain cuspidine and some others in the Mihara mine is that presented in Table 10, and its occurrence as well as diffraction patterns are in good alliance to the specimen found associated with scawtite and afwillite etc. in the contact zone at Crestmore, California (MURDOCH, 1955), whereas in spite of its resembrance in chemical composition to cuspidine not so much distinct intimacy is confirmable anywhere in occurrence and no allusion to this mineral has been given even by TILLEY (1951) elucidating predominance of cuspidine in the boron- and fluorine-bearing skarns recognized in the vicinage of granite.

#### I. Feldspar

Feldspar divided into two types in occurrence have been collected from the Daidō deposit of the Yamato mine. One found in calcite associating the ores such as chalcopyrite, molybdenite and scheelite is euhedral, platy, about 3mm in width, dark green to black in color on the surface of its crystal, though with the green-colored inner part and the white-colored core, and easily discriminated from the greyish ores (Plate 53, 2). X-ray diffraction patterns for the white-colored specimen are, as are presented in Table 11, quite similar to those identified with yellow-colored ferriferous orthoclase from Madagascal by COOMBS (1954), according to whom colors of these specimens depend on the content of Fe to a certain extent. In those for the blackcolored crusts of the specimen, weakening of the spacings other than that at 3.23Å and strengthening of those for chlorite at 14.8Å and 7.2Å are conspicuously observ-Microscopical examination for this part clarifies the presence of green-colored able. materials composed probably of chlorite produced through decomposition but yields no proof for its identity with microcline in spite of appearance of the spacing at 2.903Å (Plate 49, 1, 2). The other, fine in grain-size and hard to be discriminated even with the microscope, is likely to occur as veins in the skarns in the same deposit, less in amount than the coexisting diopside, and röntgenometrically composed of plagioclase and potash feldspar, as has been alluded to in the case of garnet (7).

The allied occurrence of feldspars is surely recognizable in the part intervening between the mass of hedenbergite-wollastonite skarn and the dark purplish, biotitehornfels near granite, coarse in grain-size, rich in potash feldspar and combined continuously with its aplitic facies mentioned already, in the eastern part of No. 10 level in the main adit of the Sampō mine, and hence a zoned arrangement of the massive skarn from the core to the outer side is as follows (Plate 50, 1 and Plate 53, 3): (a) wollastonite, irregular but occasionally amygdaloidal in form and mostly 1~ 3cm in diameter, injecting into the interstices of calcite crystals—(b) hedenbergite, mostly 1~3mm in width—(c) feldspars, commonly pale pink in color, 2~5mm in width or extremely small in grain-size not discernible even with microscope and röntgenometrically composed of plagioclase and potash feldspar—(d) hornfels,

wherein wollastonite is in part deficient but feldspars are not so. Moreover,

Ferriferous orth	noclase	Ferriferous orthoclase from Madagascr				
from the Yamat	o mine	(three samples)				
ط(هُ)	T/T		1 7/7			
u(A)	1/11	d(A)	1/11	hki		
6.6	8					
6.51	11					
5, 87	11					
4.23	41	4.23	40	. 201		
3.94	16	3.95	25	111		
3.78	52	3, 78	70	130		
3.68	5					
3.62	13	3.62	20	131		
3.54	7					
3.47	36	3.47	30	112		
3.32	75	3.32	70	220		
3, 29	52	3.29	40	202		
3. 23	100	3.24	100	040,202		
2.996	39	2.997	50	131		
2.903	26					
2.769	15	2.769	10	$13\overline{2}$		
2.607	13	2.606	10	312		
2.581	21	2.579	20	241		
2.552	-10	2.550	10	112		
2. 526	8					
2. 518	8					
2.489	5	•				
2. 423	7					
2. 387	8					
2. 320	5					
2.169	21	2, 168	30	060		
2.130	8					
2.120	7					
2.072	5					
2.051	5					
2.007	7					
1.971	8					
1,928	10					
1.851	7					
1.796	23					
1.775	5					
1.630	5					
1.568	5					
1, 499	8					
1.449	7					
1.436	7					

TABLE 11. X-RAY DIFFRACTION DATA FOR FERRIFEROUS ORTHOCLASE.

1.1.1

hedenbergite remained from the replacement by feldspar formed at the later stage is enclosed in the latter and in wollastonite formed almost contempoaneously or a little later than its own while it is uncertain whether calcite embraced in wollastonite may be genetically combined with limestone contained originarily in hornfels or with mineralization of the related skarns but fluorite in wollastonite may point to a close relationship between them in the course of the similar mineralogenesis.

It thus results in that the crust of hedenbergite at the earlier stage and the core of wollastonite at the later stage might have been deposited only as the irregular masses because of compact hornfels without any cracks suitable for formation of the veins and then feldspars as the veins filling the boundaries between the massive skarns and the surrounding hornfels. Referring to the view offered by TAKEUCHI and YAMAOKA (1964) emphasizing the presence of the white-colored microcline rock in their discussion on difference of the breccia skarn from the banded skarn, occurring in the Ömine mine, Iwate Prefecture, that the skarns observed in the Sampō mine seem to bear some resembrance but are not quite similar to in irregularly massive occurrence and in no content of garnet still remains to be prusued.

In the Maruyama adit of the Tsumo mine in Shimane Prefecture a plenty of the feldspar veinlets, faint pink in color, about 5mm in width, composed of plagioclase and potash feldspar, and bearing a few amount of hedenbergite but without any trace of wollastonite, are found disposed parallel in the black-colored hornfels.

In the case of No. 11 level in the main adit of the Mihara mine (Okayama), the feldspar veins, fleshy white in color,  $1 \sim 5$ cm in width, and with content of calcite, hornblende, quartz, laumontite vein and epidote along the former, are observed cutting across the black-colored hornblende-bearing skarns replaced partially by magnetite (Plate 49, 3, 4). Laumontites in this case are pale pink in color, individually prismatic in form, and in radial arrangement as a whole, occurring as veins, less than 3cm in width, penetrating the sporadically disposed grains, about 1cm in maximum diameter, of chalcopyrite stained with purplish tint and remained from replacement by feldspars at a certain stage later than that of ore formation, as be also the case with those, a little reddish white in color, traversing haphazardly through slate in No. 10 level of the main adit in the Sampō mine.

#### J. UNDETERMINED MINERALS

Two species have, though in paragenetic relation to other sorts of skarns, been not yet precisely determinable. One recognized in hedenbergite, containing small grains of lievrite coming into contact with wollastonite in the central part of the Fukuju deposit in the Yamato mine is most commonly granular, about 0.2mm in grain-size, scattered along a definite direction, wine-purple in color, characterized with pleochroism varying from colorless to violet, and cut respectively by the apophyllite veinlets and by the calcite veinlets (Plate 50, 2). The data of x-ray diffraction are shown in Table 12, revealing some alliance to those of lombardite or graftonite. The other observed in hedenbergite accompanied with a few amount of grossularite

Unknown mineral from the Yamato mine		Lambardite (ASTM 11-127)		Graftonite (ASTM 6-0406)		Correlative
d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	d(Å)	I/I <sub>1</sub>	to
9.26 8.70 8.01 7.72 7.20 6.68 6.53 5.86 4.68 4.51	27 20 50 20 17 20 50 17 23 17					Hedenbergite
4. 14 3. 94 3. 76	17 17 17	9 51	50	4. 31	30	
3. 342 3. 251	43 17 77	3. 51 3. 22	30 30	3, 14	90	-
2. 979 2. 899 2. 869 2. 811	100 73 50 23	2. 90 2. 85	100 50	3. 01 2. 97 2. 90 2. 86	40 40 100 40	Hedenbergite
2.695 2.617 2.574 2.538	53 40 20 40	2.61	30	2.71	80	
2. 401 2. 315 2. 276 2. 226	17 17 17 17	2. 34 2. 48 2. 40 2. 29	10 20 20	2. 46 2. 41 2. 32 2. 27	10 30 10 10	
2. 173	43	2.15	20			
2. 117 2. 096	30 27	2.12	20	2. 13 2. 08 2. 05 1. 972 1. 927	10 10 10 10 20	
1.898	13	1.87	30	1.886 1.850	10 20	
1.790	13 20			1.774 1.769	10 20	
1.759	20			1.735 1.715	10 10	
1.629 1.556	30 13	1.62 1.58 1.551	50 20 10			

TABLE 12. X-RAY DIFFRACTION DATA FOR THE UNKNOWN MINERAL FROM THE YAMATO MINE.

in limestone occurs as veinlets, white to pale yellowish orange and in part faint pink in color, irregularly  $2\sim 5$ mm in width and elongated fibrously almost perpendicular to the walls of hedenbergite. In these veinlets hedenbergite is observed getting close the former and limestone, or near the very veinlets some sulfide ores



FIG. 1, a. Skarns replacing porphyrite, obtained from No. 12 level of the main adit in the Mihara mine (Okayama). Cf. Plate 53, 1 and 4.

- a: porphyrite.
- b: reddish brown-colored garnet zone.
- c: white-colored scawtite-plazolite zone.

d: calcite vein.

b. Unknown mineral in hedenbergite, obtained from the main adit of the Takinomaru mine. Cf. Plate 50, 3.

- a: green-colored hedenbergite.
- b: greyish brown-colored limestone.
- c: deep brown-colored sphalerite.
- d: white-colored unknown mineral.

composed mainly of small grains of sphalerite are found distributing narrowly within a zone,  $1\sim3mm$  in width (Plate 41, 3 and Fig. 1, b). The data of x-ray diffraction for the specimens, from which a scarce quantity of calcite filling the interstices among their fibrous crystals be removed through treatment with diluted hydrochloric acid, are presented in Table 13.

# III. CONSIDERATION ON CONTACT EFFECT

Assuming the reactions of some emanation derived from certain intrusives upon the pre-existed sedimentaries confined especially to carbonate rocks, pyrometasomatic

Unknown mineral from the Takinomaru mine		Correlative	Unknown mineral from the Takinomaru mine		Correlative
d(Å)	I/I <sub>1</sub>	to	d(Å)	I/I1	to
7.44	14		2. 241	38	
6.84	12		2.200	.8	
5.81	10		2.151	12	
5, 59	10		2. 241	38	
4.82	24		2.200	8	
4.46	10		2. 151	12	
4.25	26	quartz	2.113	20	
4.13	8		2.112	22	
4.03	10		2.095	14	
3.712	28		2.086	8	
3, 660	12		2.029	8	
3.629	8		1.975	8	
3. 526	8		1.927	12	
3.420	50		1.913	40	
3.341	100	quartz	1.888	8	none
3. 230	56		1.856	8	
3.128	50		1.847	8	
3.004	42		1.818	16	
2.950	10		1.777	10	
2,889	52		1.745	8	
2.710	10		1.710	20	
2.687	10		1.671	16	
2.643	16		1.662	16	
2.628	12		1.628	· 16	
2. 483	18		1.611	8	
2.458	20		1.589	8	
2.415	16		1.563	10	
2.368	10		1.552	10	
2. 334	8		1, 539	10	
2. 181	20				

TABLE 13. X-RAY DIFFRACTION DATA FOR THE UNKNOWN MINERAL FROM THE TAKINOMARU MINE.

or contact effects have so far generally been discoursed in conjunction with the genesis of so-called skarns, notwithstanding that the related effects are nowhere or very scarcely confirmable in the places embracing the skarns designated after the preceding rule of nomination or the related minerals are likely to appear in such parts and occurrence as are believed to be in no relation to the effects in question.

Concerning the ore deposit of the Yamato mine (previously called the Ofuku mine), on the basis of such experiences that the regular arrangements of skarn minerals are considered to have been combined with simultaneous formation through

diffusion and there might have also generally been many chances for the similar dispositions in the cases of ore skarns, though formed not always contemporaneously but different respectively in crystallization sequence, WATANABE (1956) suggested such a relation as:

limestone | wollastonite, hedenbergite | andradite | "granite" Cu-sulfides

KATō (1913) also in his diagrammatic sketch showing the relations among various minerals in the Ofuku deposit gave the arrangement: calcite-wollastonite (containing vesuvianite)-garnet-pyroxene-chalcopyrite (penetrating all others). However, inasmuch as (1) no evidences for the contact of granite with limestone are obtainable within, or in the neighborhood of, the deposit concerned, (2) an extremely small-scaled skarn veinlet (Plate 52, 4),  $2\sim3$ cm in total width and with zonation of (a) granodiorite—(b) carthy calcite, containing diopside, potash feldspar, plagioclase and micas etc., derived probably from replacement of granodiorite—(c) grain-sized aggregates of green-colored diopside and reddish orange-colored grossularite,  $2\sim$ 5mm in width—(d) reddish orange-colored grossularite, less than 1cm in width—(e) yellowish green-colored andradite, 3~5mm in width-(f) granular limestone, intervenes between crystalline limestone and granodiorite distributing at the western brim of the Ofuku plateau about 2km east from the deposit in contrast with occurrence of considerably large masses of the skarns rather near the ore-bodies, (3) predominance of wollastonite is rather common in chert and not so much in limestone while no trace of hedenbergite and wollastonite are observable even in the very contact between the intrusive and limestone, (4) the country rock of the ore deposit is, though deduced only from the surrounding walls of the cave excavated at all, composed mainly of chert and subordinately of limestone without any trace of contact with the intrusive, and (5) such a simple and well-defined zoning of the skarns as were shown by some authors are nowhere discernible, every ordinary concept established previously for this deposit becomes to be hardly applicable.

In the case of the Takinomaru mine, the skarn zone appearing in the contact between granite and limestone is  $30 \sim 50$  cm in width and far larger in scale than that seen at the western edge of Ofuku plateau. Such an arrangement as (Plate 48, and Plate 51, 3): granite—grossularite—andradite—limestone, of which the order of the second and the third is quite similar to in the case of Ofuku plateau, is general in this case but the definite boundaries between granite and skarn zone cannot be drawn and furthermore the relations of the two are often found repeated like the complicated stripes, as be also the case with Mihara fluorite deposit (Hiroshima) (Plate 48, 4). Although, as far as this type of skarns is concerned, a concept of endomorphic skarn believed a secondary derivative concerning replacement of limestone by hightemperature fluid seems to be possible, of contrary significance is that the large-scaled masses of wollastonite and hedenbergite etc. are found occurring in limestone utterly indifferent to the situation of granite and more detailed observation of the contacts between granite and skarn zone surely indicates the garnet veinlets to traverse the

granite and to replace the mafics at first and orthoclase, though hardly altered, at last.

The skarn zones appearing in the Sampō mine are, as has been referred to already, deemed not as those produced primarily, so to speak, through the reaction of granitic intrusion upon limestone but essentially as the aggregates of veins ascending along the weak zone suitable for passing of certain sort of mineralyser.

A part of the skarn veins observed in No. 12 level of the main adit in the Mihara mine (Okayama) are independently disposed  $2\sim3m$  distant from the linear contact between porphyrite and limestone and none of the skarns are recognizable within the other contact with dioritic rock combined probably with extension of porphyrite in the eastern part of No. 2 level in the Yataka adit.

After all, any evidences supporting the leading opinions as to genesis of so-called skarns through the effects of the intrusive emanation on the lime-rich rock, even though considered in common sense to be suitable for chemical reaction, at higher temperature have regrettably not been acquired in such modes of occurrence that all of the related minerals are always found as veins or veinlets and the contacts under consideration are occasionally irregular but well-demarcated in no direct relation to the presence of the former while the fact is that formation of the ores and so-called skarns are ascertainable not only in the lime-rich rock but also in other kind of the sedimentaries or rarely even in the intrusives of their own.

In addition, there still remain several questions with respect to the mineral species composing the skarns as well as to their definition now that almost all of the mineral concerned are not of so-called pyrometasomatic or contact product derived from reaction at higher temperature, even if proved synthetically at the solid state in the simple system with a very few component but of hydrothermal one precipitated in various scales of the fissures from low-temperature mineralyser in view of their intimate parageneses with considerably low-temperature hydrothermal minerals, and furthermore considered to have almost always been subjected to hydrothermal alteration concealing the earlier states to more or less extent.

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# EXPLANATION FOR PLATE XLI

(All figures  $\times$  30)

Fig. 1. Wollastonite vein traversing chert, obtained from the Hökoku adit of the Yamato mine in Yamaguchi Prefecture. (only with lower nicol)

Ct: white-colored chert.

W: wollastonite.

- FIG. 2. The part same as that in FIG. 1. (with crossed nicols)
- Fig. 3. Wollastonite vein penetrating limestone, obtained from the Daidō deposit of the Yamato mine. (only with lower nicol)

FIG. 4. The part same as that in FIG. 3. (with crossed nicols)
Pl. XLI





Pl. XLI



#### EXPLANATION FOR PLATE XLII

(Figs. 1, 2, 3 and 6,  $\times 30$ ; 4 and 5,  $\times 3$ )

Fig. 1. Vein-like occurrence of garnet obtained from the main adit of the Sampō mine in Okayama Prefecture. (only with lower nicol) Cf. plate 52, Fig. 1.

Garnet intervening between the crust and the core is replaced by fluorite in direction parallel to the crystal plane almost without optical anomaly.

G: the part rich in garnet including andradite.

Fl: fluorite.

FIG. 2. An end of the vein similar to that shown in FIG. 1. (only with lower nicol) Lt: white-colored, crystalline limestone.

G: garnet.

FIG. 3. The part same as that in FIG. 2. (with crossed nicols)

Fig. 4. Massive garnet obtained from the Fukuju deposit of the Yamato mine. (only with lower nicol) Cf. Figs. 5 and 6.

Zonal structure is conspicuous and the part intercalating between the crust and the core is replaced by wollastonite.

W: wollastonite.

G: garnet.

FIG. 5. The part same as that in FIG. 4. (with crossed nicols)

FIG. 6. Massive garnet obtained from the Fukuju deposit. (only with lower nicol) Cf. FIGS. 4 and 5.

The part included similarly in FIGS. 4 and 5 illustrates wollastonite filling the intervals between the core predominant in grossularite and the crust abundant in andradite as well as among the interstices of respective crystals of garnets with optical anomaly.

G: garnet.

W: wollastonite.



PL. XLII



Pl. XLII



#### EXPLANATION FOR PLATE XLIII (All figures × 30)

FIG. 1. Massive garnet obtained from the Daidō deposit of the Yamato mine. (with crossed nicols)

The specimen is characterized with optical anomaly, fan-shaped along the crystal planes.

G<sub>1</sub>: the part abundant in grossularite.

G<sub>2</sub>: the part abundant in andradite.

FIG. 2. Massive garnet obtained from the Fukuju deposit of the Yamato mine. (with crossed nicols)

The specimen reveals no fan-shaped twinning but slightly the zonal structure.

FIG. 3. Garnet vein obtained from the Fukuju deposit. (only with lower nicol) The specimen, typical as vein, discloses no traces of crystallographical form as well as of optical anomaly and replacement of vesuvianite with garnet, the latter two being traversed by diopside vein.

FIG. 4. The part same as that in FIG. 3. (with crossed nicols)



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### Pl. XLIII

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#### EXPLANATION FOR PLATE XLIV

(FIGS. 1 and 2,  $\times$ 3; FIGS. 3, 4 and 5,  $\times$ 30)

FIG. 1. Garnet replaced by wollastonite, obtained from the Fukuju deposit of the Yamato mine. (only with lower nicol) Cf. FIGS. 3 and 4.

Garnet and later wollastonite are again replaced by chalcopyrite and bornite.

G: reddish colored grossularite.

W: wollastonite.

- C-P: chalcopyrite.
  - B: bornite.

FIG. 2. The same part as that in FIG. 1. (with crossed nicols)

FIGS. 3 & 4. Garnet replaced by wollastonite and sulfide ores, obtained from the Fukuju deposit. (only with lower nicol)

The specimen same as that in FIGS. 1 and 2 indicates the replacement of garnet by wollastonite in FIG. 4 to be higher in grade than that in FIG. 3.

FIG. 5. Vesuvianite replaced by garnet, obtained from the Daidō deposit of the Yamato mine. (with crossed nicols) Cf. Plate 52, FIG. 5.

Vesuvianite is replaced by grossularite vein and futhermore by diopside, feldspar and prehnite.

V: vesuvianite.

G: grossularite without any optical anomaly.

D: diopside.

F: potash feldspar and plagioclase.

Pr: prehnite.

Pl. XLIV



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Pl. XLIV



#### EXPLANATION FOR PLATE XLV

#### (All figures $\times 30$ )

- FIG. 1. Diopside vein running acrose the mass of wollastonite, obtained from the Hökoku deposit of the Yamato mine. (only with lower nicol)
  - W: wollastonite.
  - D: diopside.
- F10. 2. Fluorite filling the interstices of wollastonite, obtained from the Hökoku deposit. (with crossed nicol)
  - W: wollastonite.
  - Fl: fluorite.

FIG. 3. Apophyllite obtained from the Fukuju deposit. (with crossed nicols) Cf. Plate 51, FIG. 5. Ap: apophyllite.

- V: vesuvianite.
- G: grossularite.

W: wollastonite replacing the former two.

FIG. 4. Apophyllite vein obtained from the Hökoku deposit. (with crossed nicols)

- Ct: white-colored chert including the veinlets of wollastonite.
- A-P: arsenopyrite vein running across chert.

AP: apophyllite vein cutting the former or surrounding the fragment of the former.

C: calcite vein traversing the former.



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## Pl. XLV



#### EXPLANATION FOR PLATE XLVI

#### (All figures $\times$ 30)

Fig. 1. Apophyllite and wollastonite comprised in skarn vein running along the contact between granite and limestone in the main adit of the Sampō mine in Okayama Prefecture. (only with lower nicol) Cf. Plate 52, Figs. 2 and 3.

Ap: apophyllite.

W: wollastonite accompanying an extremely small amount of fluorite.

FIG. 2. The part same as that in FIG. 1. (with crossed nicols)

FIG. 3. Prehnite and hedenbergite embraced in the skarn vein similar to that in FIG. 1. (only with lower nicol) Cf. Plate 52, FIGS. 2 and 3.

H: hedenbergite.

Pr: prehnite replacing the former.

FIG. 4. The part same as that in FIG. 4. (with crossed nicols)

Pl. XLVI



Pl. XLVI



#### EXPLANATION FOR PLATE XLVII

#### (All figures $\times$ 30)

FIG. 1. Porphyrite obtained from No. 12 level of the main adit in the Mihara mine in Okayama Prefecture. (with crossed nicols) Cf. FIG. 2, Plate 53, FIG. 1 and Plate 54 FIG. 1.

P: plagioclase.

Py: pyroxene.

FIG. 2. Contact between porphyrite and skarn replacing the former, obtained from the same spot in the Miharamine. (only with lower nicol) Cf. FIG. 1 Plate 53, FIG. 1 and Plate 54, FIG. 2.

Pt: porphyrite.

W: wollastonite.

FIG. 3. Scawtite obtained from the same spot in the Mihara mine. (with crossed nicols) Cf. Plate 53, FIG. 4.

S: scawtite.

C: calcite remained from replacement by the former.

Cu: cuspidine veinlet replacing scawtite.

FIG. 4. Plazolite (hydrogrossular) obtained from the same spot in the Mihara mine. (with crossed nicols) Cf. Plate 53, FIG. 1, and Plate 54, FIG. 1.

Hy: translucent, white-colored hydrogrossular.

Hy': transparent hydrogrossular.

C: calcite.



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Pl. XLVII



#### EXPLANATION FOR PLATE XLVIII

#### (All figures $\times$ 30)

FIG. 1. Veins of cuspidine and bultfonteinite obtained from No. 12 level of the main adit in the Mihara mine in Okayama Prefecture. (only with lower nicol) Cf. Plate 54, Fig. 1.

The veins are almost parallel to calcite vein in scawtite. Bultfonteinite vein is found cutting cuspidine vein.

Cu: cuspidine.

Bu: bultfonteinite.

C: calcite.

FIG. 2. The part same as that in FIG. 1. (with crossed nicols)

FIG. 3. Garnet in granite, obtained from the main adit of the Takinomaru mine. (with crossed nicols) Cf. Plate 11, FIG. 3.

Garnet vein replaces a part of granite, without any mafics, subjected to carbonatization as whole.

P: plagioclase.

O: orthoclase.

C: mainly adradite.

FIG. 4. Repeated Precipitation of skarns found in the fluorite deposit of the Mihara mine in Hiroshima Prefecture. (only with lower nicol)

Alternated stripes of colorless part composed of garnet, diopside and fluorite, colored part composed of actinolitic amphibole, and magnetite etc. are considered not as the zoned skarns but as the products derived from repeated precipitation of mineralyser.

Pl. XLVIII



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#### EXPLANATION FOR PLATE XLIX

#### (All figures $\times 30$ )

FIG. 1. Ferriferous orthoclase obtained from the Daido deposit of the Yamato mine. (only with lower nicol) Cf. Plate 53, FIG. 2.

O: frrriferous orthoclase.

Ch: chlorite replacing the former.

C: calcite.

Q: quartz.

C-P: chalcopyrite replacing orthoclase and chlorite.

Sc: scheelite.

FIG. 2. The part same as that in FIG. 1. (with crossed nicols)

FIG. 3. Feldspar vein obtained from No. 11 level of the main adit in the Mihara mine (Okayama). (only with lower nicol)

F: plagioclase.

A: black-colored hornblende cut by the veinlet of the former.

FIG. 4. Feldspar vein obtained from the same level in the Mihara mine.

F: plagioclase.

- E: epidote, pale green in color, appearing near the boundary between feldspar and laumontite.
- L: laumontite vein comprised in feldspar vein.



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# EXPLANATION FOR PLATE L (All figures $\times$ 30)

- FIG. 1. Skarns included in slaty hornfels, obtained from the main adit of the Sampō mine. (only with lower nicol) Cf. Plate 53, FIG. 3.
  - W: massive wollastonite containing fluorite in a little amount and occasionally calcite in the core.
  - H: hedenbergite replaced remarkably by feldspar.
  - F: patash feldspar and plagioclase.
  - Hf: hornfels.
- FIG. 2. Unknown mineral in hedenbergite, obtained from the Fukuju adit of the Yamato mine. (only with lower nicol)
  - H: hedenbergite.
  - U: unknown mineral.

Q: quartz.

FIG. 3. Unknown mineral in hedenbergite, obtained from the main adit of the Takinomaru mine. (only with lower nicol)

- H: hedenbergite.
- Z: sphalerite.
- U: unknown mineral.



PL. L



Pl. L

#### EXPLANATION FOR PLATE LI

- FIG. 1. Garnet-vesuvianite vein in limestone, obtained from the Hökoku adit of the Yamato mine.
  - Lt: crystalline limestone comprising a profusion of the garnet-vesuvianite veinlets.
  - G+V: vein composed of browish grossularite and greenish brown-colored, prismatic vesuvianite.
    - W: wollastonite arranged along the both side of the former.
    - C: calcite filling the interstices of vesuvianite crystals.
- FIG. 2. Garnet-vesuvianite vein in wollastonite, obtained from the Hökoku adit.
  - G+V: vein composed of brownish grossularite and brownish vesuvianite.
    - W: massive wollastonite.
- FIG. 3. Skarn vein in granite, obtained from the main adit of the Takinomaru mine. Cf. Plate 48, FIG. 3. Veins of garnet and calcite, penetrating granite, are furthermore surrounded by their veinlets, composing a replaced zone with a gradual variation into granite as if it were a sort of zoned skarn.
  - Gt: granite.
  - G: mainly green-colored andradite.
  - C: calcite cutting across granite.
- Fig. 4. Boundary between chert and massive wollastonite, obtained from the Hökoku adit of the Yamato mine. The boundary is markedly well demarcated.
  - Ct: greyish chert.
  - W: massive wollastonite.
  - D: greyish green-colored diopside.

FIG. 5. Apophyllite vein obtained from the Fukuju deposit of the Yamato mine. Cf. Plate 45. FIG. 3.

G+V: reddish orange-colored grossularite and honey-yellow vesuvianite.

W: wollastonite traversing across the former.

- Ap: apophyllite cutting across all of the skarns.
- FIG. 6. Wollastonite vein in chert, obtained from the Hökoku adit of the Yamato mine.
  - Ct: greyish chert.
    - W: wollastonite.



Pl. LI

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Pl. LI

#### EXPLANATION FOR PLATE LII

- FIG. 1. Skarn vein in limestone, obtained from the main adit of the Sampō mine. Cf. Plate 42, FIGS. 1, 2 and 3.
  - Lt: white-colored crystalline limestone.
  - G: garnet.
  - W: wollastonite.
  - H: hedenbergite.
  - C-P: chalcopyrite enclosed in garnet.

FIGS. 2 & 3. Skarn vein in the contact between limestone and granite, obtained from the main adit of the Sampō mine. Cf. Plate 46, FIGS. 1, 2, 3, and 4.

- Gt: granite.
- Pr+H: prehnite, hedenbergite and a little amount of wollastonite.
  - G: reddish brown-colored grossularite and a little amount of vesuvianite.
  - V: brownish vesuvianite and a little amount of grossularite.
  - Ap: translucent, pale brownish apophyllite comprising a little amount of wollastonite.
  - W: wollastonite including a little amount of fluorite and apophyllite.
  - Lt: white-colored, crystalline limestone.
- FIG. 4. Skarn vein in the contact between limestone and granodiorite, obtained from the Ofuku plateau, Miné City, Yamaguchi Prefecture.
  - Gt: granodiorite.
  - C: calcite.
  - G: reddish orange-colored grossularite and greenish andradite.
  - D: diopside.
  - Lt: white-colored, crystalline limestone.

FIG. 5. Veins of feldspar and prehnite in wollastonite, obtained from the Daidō deposit of the Yamato mine. Cf. Plate 44, FIG. 5.

Feldspar, diopside and prehnite contemporaneously replace wollastonite, grossularite and vesuvianite etc.

W: wollastonite.

- G+V: brown-colored grossularite-vesuvianite vein.
  - D: bluish grey-colored diopside replacing the former.
  - F: greyish feldspar-diopside vein replacing grossularite and vesuvianite.
- D+W+Pr: grey-colored diopside-wollastonite-prehnite vein replacing wollastonite.

PL. LII



Pl. LII



#### EXPLANATION FOR PLATE LIII

- FIG. 1. Skarn vein replacing porphyrite, obtained from No. 12 level of the main adit in the Mihara mine (Okayama). Cf. Plate 47, Figs. 1, 2, and 4, and Plate 54, FIG. 1.
  - Pt: porphyrite.
  - m: dark pinkish part composed of grossularite and wollastonite.
  - n: yellowish part composed of andradite, cuspidine and wollastonite.
  - o: purple- to reddish brown-colored part composed of vesuvianite and cuspidine.
  - p: opaque, white-colored part composed of plazolite and calcite.
  - q: translucent part similar to the former.
- FIG. 2. Ferriferous orthoclase obtained from the Daidō deposit of the Yamato mine. Cf. Plate 49, FIGS. 1 aud 2.
  - O: ferriferous orthoclase with black-colored surface.
  - C: calcite.
  - Sc: scheelite with greyish fracture.
- FIG. 3. Skarns in slaty hornfels, obtained from the main adit of the Sampō mine. Cf. Plate 50, FIG. 1.
  - Hf: dark purplish hornfels.
  - F: pale pinkish potash-feldspar and plagioclase.
  - H: deep green-colored hedenbergite.
  - W: wollastonite including a little amount of calcite and fluorite.
  - W': wollastonite vein.
- FIG. 4. Skarns replacing porphyrite, obtained from No. 12 level of the main adit in the Mihara mine (Okayama). Cf. Plate 7, FIG. 3 and Plate 54, FIG. 1.

Pt: porphyrite remained from replacement.

- G+V: yellowish brown- to yellowish green-colored part composed of garnet and vesu-vianite.
  - Cu: opaque, white-colored cuspidine replacing scawtite.
  - Bu: transparent vein of bultfonteinite.
- FIG. 5. Wollastonite vein in limestone, obtained from the Mihara mine (Hiroshima).
  - Lt: crystalline limestone.
  - W: wollastonite vein including a little amount of fluorite and a part rich in apophyllite.
  - G+D: yellowish green-colored vein of garnet and diopside contained in wollastonite and limestone.


Pl. LIII





Pl. LIII

