

Studies on Oxyhornblende

By

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Part I. X-ray Studies on Oxyhornblendes

Abstract

With oxyhornblendes from Korea and Japan, rotation and Weissenberg photographs were taken, and it was found that these minerals were composed of two different lattices, one being the monoclinic amphibole lattice ($I2/m$), the other the monoclinic pyroxene lattice ($C2/c$). The proportion of the two lattices in these minerals are likely to vary from specimen to specimen. The two lattices are presumably grown submicroscopically side by side.

Introduction

T. Ito (1939, 1950) is the first to find that moonstone from Korea is composed of several, different feldspars. He showed that the mineral is built up from two monoclinic and two or three triclinic feldspars, the latter ones being twinned after the pericline law, and that of these one monoclinic feldspar is potash-rich and the remainders soda-rich. Soon after, ITO and SADANAGA (1952) described a microcline from Korea which is also built up from two different feldspars, both being triclinic, but one being potash-rich and the other soda-rich. They also reported in the same article that an anorthoclase from Korea is built up from two or three different feldspars, one of them being potash-rich and monoclinic, the remainders soda-rich and triclinic. MORIMOTO (1956) examined the monoclinic pyroxene found in andesite of Hakone Volcano, Japan, and reported that the mineral is built up from two different pyroxenes, one being pigeonite ($P2_1/c$) and the other augite ($C2/c$). Bown and Gay (1959) reported that a hornblende which is grown with augite is often oriented in such a way that its a, b, and c axial directions are parallel respectively with those of the augite.

Recently, the author has been studying amphiboles from the chemical and structural viewpoints. In the course of this study he, too, found a titaniferous oxyhornblende

(1963) and an oxyhornblende (1964) which are built up from two different lattices. In the following, the details of the study will be given.

Experimental

Specimens used in the present experiments were a titaniferous oxyhornblende from Ullung-do (Dagelet Island), Korea, and an oxyhornblende from Ikebukuro, Nagano Pref. Japan.

The specimens of a titaniferous oxyhornblende were kindly given to the author by Dr. A. HARUMOTO, and are 1~4 mm in length and 1~2 mm in diameter. The mineral makes up globular tough masses, 2~10 cm in diameter, together with alkaline basaltic matrix. The globular masses are contained in a rhyolitic lava and are considered to be a kind of volcanic ejecta.

Table 1. Chemical compositions and optical properties.

	titaniferous oxyhornblende *)	oxyhornblende
SiO ₂	39.20%	39.58% **)
Al ₂ O ₃	13.89	19.42
Fe ₂ O ₃	4.08	7.54
TiO ₂	6.53	3.04
FeO	7.33	0.57
MgO	11.96	14.92
MnO	0.11	0.11
Ca ₂ O	12.37	12.30
Na ₂ O	1.99	2.58
K ₂ O	1.45	0.20
H ₂ O+	0.87	0.00
H ₂ O—	0.25	0.06
Total	100.03	100.32
α	1.680	1.672 ***)
β	1.700	1.695
γ	1.709	1.719
2V	—75	—92 24'
X	pale greenish brown	light greenish brown
Y	reddish brown	greenish brown
Z	dark reddish brown	reddish brown

*) A. Harumoto (1933) Chikyū 19, 96. (written in Japanese)

**) Chemical composition. K. Tomita (1962) Memo. Coll. Sci., Univ. Kyoto. 27, 4, 549.

***) Optical properties. S. Kozu & B. Yoshiki (1927) Sci. Rep. Tohoku Imp. Univ. 3rd. 3, 107,

The specimens of an oxyhornblende were found abundantly as phenocrysts in hornblende andesite which is bitterly fragile. The crystals are usually elongated along the c-axis with well developed (110) and (010) faces. The surface of the crystals is always coated with a very thin, black or brown layer. The interior is reddish brown in colour.

These specimens are perfectly homogenous to the naked eye or under the microscope. The chemical compositions and optical properties of these specimens are shown in Table 1.

With the titaniferous oxyhornblende and oxyhornblende rotation and Weissenberg photographs about the c-axis were taken by using $\text{Cu-K}\alpha$ radiation. On the rotation photographs all the spots are arranged perfectly on the layer lines. (Fig 1).

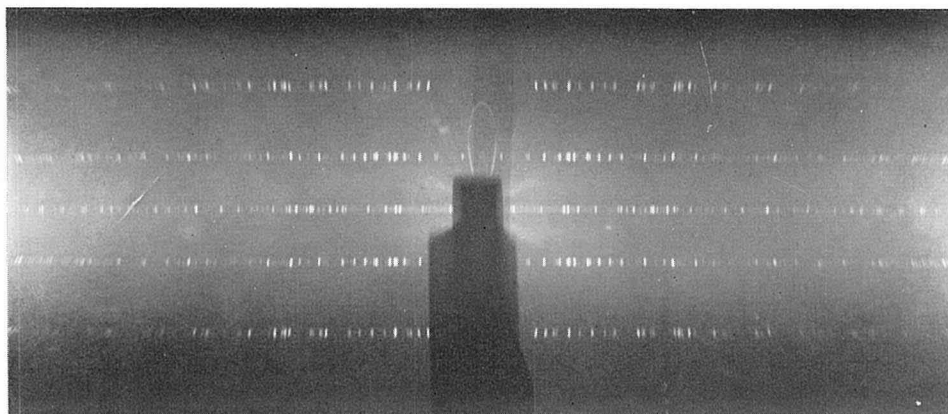


Fig. 1. A Rotation photograph of Ti-oxyhornblende about the c-axis.

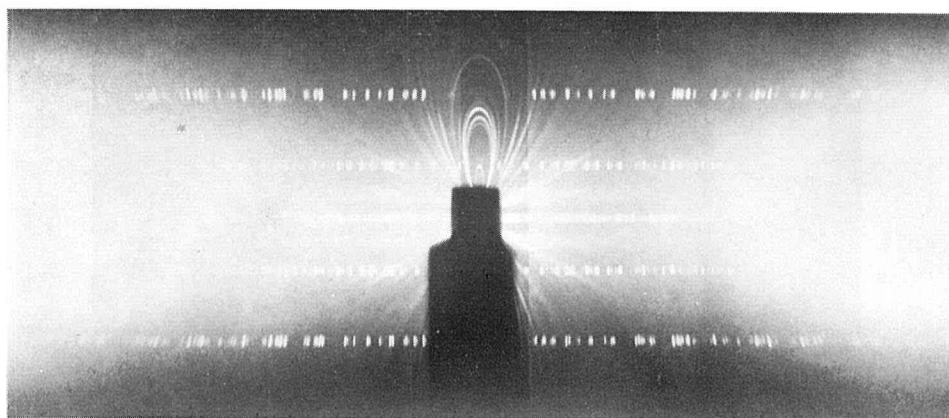


Fig. 1. B Rotation photograph of oxyhornblende about the c-axis.



Fig. 2. A Weissenberg photograph of Ti-oxyhornblende about the c-axis (zero level)



Fig. 2. B Weissenberg photograph of the oxyhornblende about the c-axis (zero level)

On the zero level Weissenberg photograph and the 1st and 2nd level equi-inclination Weissenberg photographs taken with Ti-oxyhornblende (Fig. 2 A), however, there are number of extra spots as compared with the corresponding photographs taken with tremolite. The same is the case with the oxyhornblende. (Fig 2 B).

The extra spots also constitute an another set of reciprocal lattice points.

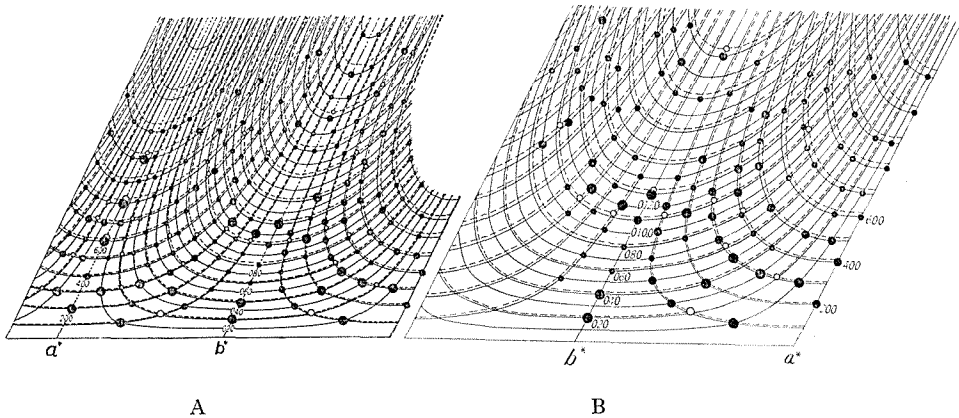


Fig. 3. A $h k 0$ Weissenberg diagram of titaniferous oxyhornblende from Korea.
Fig. 3. B $h k 0$ Weissenberg diagram of oxyhornblende from Japan.

In Fig. 4 are illustrated the zero level Weissenberg diagrams in which the solid circles denote one of the sets and the open circles the other set. In position and intensity the former is the same as that of tremolite and the latter is the same as that of diopside. It follows from these observations that these minerals are composed of two different

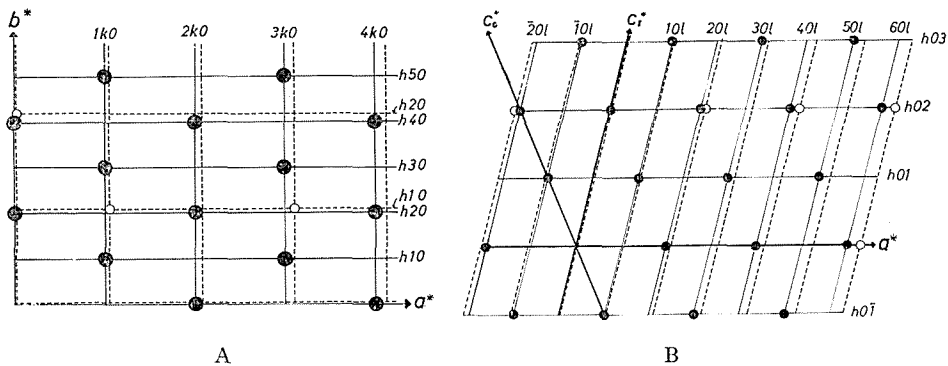


Fig. 4. A $h k 0$ Reciprocal net plane.
Fig. 4. B $h 0 l$ Reciprocal net plane.

lattices and the c-axis of the component lattices are parallel in direction and identical in length.

With the Ti-oxyhornblende rotation and Weissenberg photographs about the b-axis were next taken. In the rotation photograph all the spots are arranged perfectly on the layer lines. In the zero level Weissenberg and the 2nd level equi-inclination Weissenberg photographs there appear extra spots as compared respectively with the corresponding photographs taken with tremolite. In the 1st level Weissenberg photograph such extra spots are not observed. It follows from these fact the b-axis of the component lattices are parallel in direction, one being twice in length the other.

In Fig. 4 are shown the $(h k 0)$ and $(h 0 l)$ reciprocal net planes derived from the zero level Weissenberg photographs about the c-axis and b-axis respectively. Setting the reciprocal axes as a^* , b^* , c_1^* in Fig. 4, in one set of spots $h k l$ reflexions appear when $h+k+l=2n$, $h 0 l$ reflections when $h+l=2n$ and $0 k 0$ reflections when $k=2n$, hence the space group: $I_{2/m}$, while in the other set of spots $h k l$ reflections appear when $h+k=2n$, $h 0 l$ reflections when $h=2n$, $l=2n$, and $0 k 0$ reflection when $k=2n$, hence the space group: $C_{2/c}$. Setting the reciprocal axes as a^* , b^* , c_c^* in Fig. 4, in the former set $h k l$ reflections appear when $h+k=2n$, $h 0 l$ reflections when $h=2n$, and $0 k 0$ reflections when $k=2n$, hence the space group: $C_{2/m}$; while in the latter set $h k l$ reflections appear when $h+k+l=2n$, $h 0 l$ reflections when $h=2n$, $l=2n$ and $0 k 0$ reflections when $k=2n$, hence the space group: $I_{2/a}$.

Referring the axes a^* , b^* , c_1^* the lattice dimensions and the axial angles of the two lattices were calculated. Results are shown in Table 2.

Table 2. Lattice dimension.

	Ti-oxyhornblende		Oxyhornblende	
	$I_{2/m}$	$C_{2/c}$	$I_{2/m}$	$C_{2/c}$
a	$9.84 \pm 0.01 \text{ \AA}$	$9.63 \pm 0.01 \text{ \AA}$	$9.85 \pm 0.01 \text{ \AA}$	$9.75 \pm 0.01 \text{ \AA}$
b	$18.25 \pm 0.01 \text{ \AA}$	$8.96 \pm 0.01 \text{ \AA}$	$18.08 \pm 0.01 \text{ \AA}$	$8.88 \pm 0.01 \text{ \AA}$
c	$5.33 \pm 0.01 \text{ \AA}$	$5.33 \pm 0.01 \text{ \AA}$	$5.33 \pm 0.01 \text{ \AA}$	$5.33 \pm 0.01 \text{ \AA}$
β	$105^\circ 23'$	$105^\circ 23'$	$105^\circ 43'$	$105^\circ 43'$

Evidently, the lattice of which space group is $I_{2/m}$ is a monoclinic amphibole lattice, and that of which space group is $C_{2/a}$ a monoclinic pyroxene lattice.

Weissenberg photographs about the c-axis were taken with several other specimens of both Ti-oxyhornblende and oxyhornblende. In these photographs the spots brought by the monoclinic pyroxene are not always in coincidence with one another in number and in intensity. The ratio of two component minerals, therefore, varies from specimen to specimen. The monoclinic amphibole is, however, always more predominant than the other.

Consideration

The Ti-oxyhornblende and the oxyhornblende examined in the present investigation are built up from two component minerals, one being monoclinic amphibole, the other monoclinic pyroxene, and both being submicroscopic. The two component minerals are probably arranged side by side, and are associated in such a way that their three axis are parallel to each other respectively, if an *I*-lattice is chosen for the monoclinic amphibole and a *C*-lattice is for the monoclinic pyroxene.

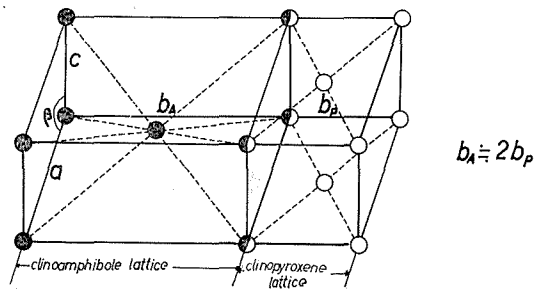


Fig. 5. Relationship between two component lattice.
(amphibole lattice $I2/m$, pyroxene lattice $C2/c$).

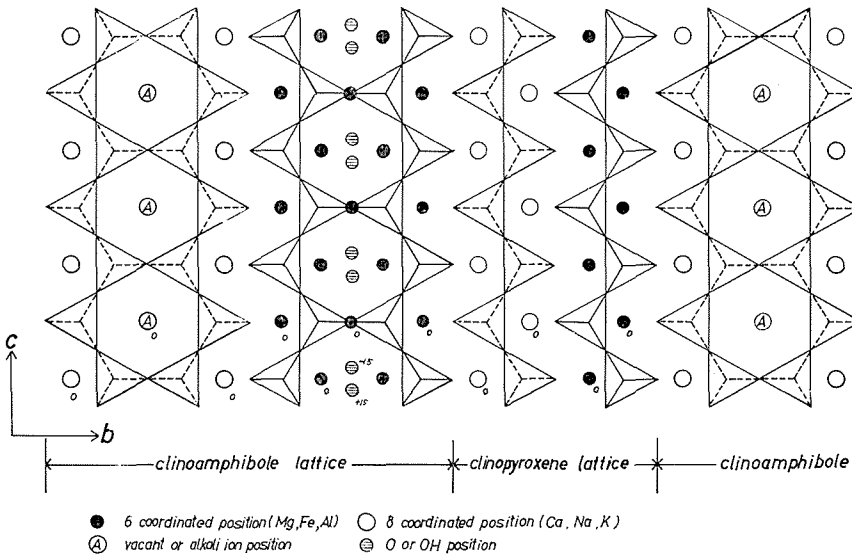


Fig. 6. (100) Projection showing silicon-oxygen tetrahedra of oxyhornblende composed of two different lattices.

Relationship between two component lattices are considered as illustrated in Fig. 5. The relationship of crystal structure between clinoamphibole (tremolite) and clinopyroxene (diopside) were investigated by B. E. Warren (1929). He showed that, when space groups of tremolite and diopside is chosen as $I2/m$, $C2/c$ respectively, the projection of tremolite structure on (010) are almost identical with that of diopside structure on (010) . It will be seen, therefore, that the two component minerals of Ti-oxyhornblende and the oxyhornblende may be connected on the ac plane $[(010)$ face]. In Fig. 6 is illustrated a diagrammatic view of the structure of the oxyhornblendes constructed by connecting clinoamphibole and clinopyroxene on the ac plane.

The chemical formula of the present oxyhornblende may be established from the results of chemical analyses in such a way that it will be in conformity with the general formula of amphibole. This is, however, not adequate. For, the present oxyhornblendes are built up from the two component minerals as said above. The author calculated the ratio of the two component minerals from the results of chemical analyses, paying attention to the water content, and obtained a result looked as if pyroxene were much more predominant than that estimated from the intensities in the X-ray photographs. In the amphibole composing the present Ti-oxyhornblende and oxyhornblende, therefore, some of its OH ions much be replaced by O atoms, electro-neutrality being balanced by additional elements.

References

- BOWEN, N. L. and E. PONSJAK, (1931): The role of water in tremolite, *Am. Jour. Sci.*, 22, 203.
 BOWN, M.C. and P. GAY, (1959): The identification of oriented inclusions in pyroxene crystals, *Am. Min.*, 44, 592.
 HARUMOTO, A. (1933): Studies on kaersutite and pigeonite contained in volcanic ejecta at Ullung-do, Chikyu, 19, 96. (Written in Japanese)
 ITO, T. (1939): The determination of lattice constants of triclinic crystals from one crystal setting- a special case, *Zeit. Krist.*, 100, 437.
 ———, (1950): X-ray studies on polymorphism, 122, Maruzen Co., Tokyo.
 ———. and R. SADANAGA, (1952): The lamellar structure of certain microcline and anorthoclase, *Acta cryst.*, 5, 441.
 MORIMOTO, N. (1956): The existence of monoclinic pyroxene with the space group C_{2h}^5 - $P2_1/c$, *Proc. Jap. Acad.*, 32, 750.
 TOMITA, K. (1962): Chemical and crystallographical studies on calciferous amphiboles, *Mem. Coll. Sci., Univ. Kyoto, Ser. B*, 28, 4, 549.
 UEDA, T. and K. TOMITA, (1963): Kaersutite composed of two different lattices, *Proc. Japan Acad.*, 39, 223.
 WARREN, B.E. (1930): The crystal structure of tremolite, *Zeit. Krist.*, 72B. 42.

Part 2. Phase Change of Calciferous Amphiboles by Heating

Abstract

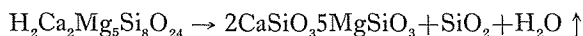
Some of the calciferous amphiboles were heated in air for 2 hours. Tremolite and actinolite heated at about 900°C and hornblende (tschermakite) heated at about 1000°C show that they are composed of two phases; one is the clinoamphibole phase, the other the clinopyroxene phase. The two phases are associated in the same way as in the case of the oxyhornblendes described in Part 1.

Introduction

KOZU, YOSHIKI and KANI (1927) made a thermo-optical investigation on some common hornblendes (calciferous amphibole). They observed that common hornblende was transformed to basaltic hornblende (oxyhornblende) at 750°C.

BARNES (1930) reported that common hornblende was transformed to basaltic hornblende by heating to about 800°C, and basaltic hornblende to original hornblende by heating to about 800°C in hydrogen gas. He thus showed that the transformation was reversible. He thought that it was caused by subtracting or adding hydrogens, the electrical neutrality being maintained by concomitant oxidation or reduction of iron respectively.

POSNJAK and BOWEN (1931) studied the dehydration with a tremolite from Ham Island, Alaska. They observed that the dehydration took place very slowly up to 850°C, but promptly between 900°C and 1000°C, losing about 70% combined water; and that dehydration between 900°C and 1000°C resulted in structural disintegration, that is a formation of pyroxene which belongs to a clinoenstatite-diopside series, and free silica. They thought that the change proceeded according to the following equation:



They also observed that the tremolite heated at 900°C for 24 hours gave an amphibole pattern together with a pyroxene pattern in an X-ray powder photograph.

WITTELS (1952) and VERMAAS (1952) observed by the D.T.A. that a tremolite shows an exothermic reaction between 615°C and 824°C, and a strong endothermic reaction between 930°C and 988°C, the former not showing any apparent change in the crystal structure, but the latter showing a disintegration of the structure. They presumed that the exothermic reaction might take place on account of the oxydation of the ferrous iron present in these minerals.

By the D.T.A. of actionlite, VERMAAS (1952) observed an endothermic reaction,

and showed that the reaction was accompanied by a loss of water and a formation of clinopyroxene and cristobalite. BELYANKIN and DONSKAYA (1939) obtained almost similar results.

KORZHINSKY (1955) carried out thermal studies with ten hornblende crystals from eastern Siberia, covering a wide range of composition. They showed two endothermic reaction between 900°C and 1030°C, and between 1240°C and 1300°C, the loss of weight being from 0.3% to 2.5%.

On the basis of thermal, chemical and X-ray investigation of fourteen amphiboles, OVCHINNIKOV et. al. (1955) showed that the constitutional water (about 50% of the total) was lost at 400°C~500°C, the lattice not being destroyed, but OH being replaced by O and Fe^{+2} by Fe^{+3} ; at 950°C~1100°C the remaining water was lost, the lattice being destroyed with a formation of clinoenstatite and hematite.

WITTELS (1952) examined the effect by heating with hastingsite and pargasite. He reported that hastingsite was disintegrated at 1090°C, and pargasite at 1100°C, and the product of the thermal reaction was a mixture of poliaugite, plasioclase and hematite.

Recently, the author (1963) found a titaniferous oxyhornblende and an oxyhornblende which are made up of two different phases, monoclinic amphibole and monoclinic pyroxene, both being submicroscopic. The two component minerals are probably arranged side by side, and are associated in such a way that their three axes are parallel to each other if an *I*-lattice is chosen for the monoclinic amphibole and a *C*-lattice is for the monoclinic pyroxene.

Taking into account the results hitherto obtained by many investigators, the author presumed that calciferous amphibole might change by heating into two phases, which may be associated in some crystallographic relationship. He therefore studied the thermal change of calciferous amphibole röntgenographically. In the following part, the details of the study will be given.

Experimental

1. Specimens:

Calciferous amphiboles used in the present investigation were picked out from the host rocks and agglomerate shown in Table 1. The specimens were crushed into grains, the size of which was from 100 mesh to 150 mesh. The crushed specimens were first separated from magnetic impurities, quartz and feldspars, using isodynamic separator, and then the specimens were purified under the microscope. The purified specimens were pulverized into powder in an agate mortar.

Single crystals of tremolite, actinolite and hornblende were also examined. They were 1~4 mm in length and 0.2~0.5 mm in diameter. Under the microscope no twinning or zonal structure was observed and they were perfectly homogeneous.

Table 1. Specimens.

sample	host rock	locality
tremolite	limestone of contact zone	Antung hsien, Liaotung, China
actinolite	tale shist	Sekigawa, Niihama-city, Ehime Pref.
hornblende (tschermakite)	agglomerate of volcanic ejecta	Gaspan, Parao Island
Ti-oxyhornblende (kaersutite)	agglomerate of volcanic ejecta	Ullung do (Dagelet Is.). Korea.
oxyhornblende	hornblende andesite	Ikebukuro, Fujimi, Suwa, Nagano Pref.

Table 2. Chemical compositions and optical properties.

	tremolite	actinolite	hornblende	oxyhornblende ²⁾	Ti-oxyhornblende ¹⁾
SiO ₂	58.54	56.74	41.07	39.58	39.20
Al ₂ O ₃	0.79	1.45	17.94	19.42	13.87
Fe ₂ O ₃	0.03	0.49	3.59	7.54	4.08
TiO ₂	0.04	tr.	0.18	3.04	6.53
FeO	0.11	3.96	7.79	0.57	7.33
MgO	23.60	21.51	13.37	14.92	11.96
MnO	tr.	0.20	0.14	0.11	0.11
CaO	12.75	11.76	12.05	12.30	12.37
Na ₂ O	0.59	0.75	1.83	2.58	1.99
K ₂ O	0.07	0.11	0.31	0.20	1.45
H ₂ O+	2.52	2.47	1.46	0.00	0.87
H ₂ O--	0.51	0.40	0.57	0.06	0.27
Total	99.19	99.84	100.33	100.32	100.03
Analyst	K. Tomita	K. Tomita	K. Tomita	K. Tomita	U. Ushizima
α	1.598	1.624	1.658	1.672	1.680
β	—	—	—	1.695	1.700
γ	1.635	1.642	1.683	1.719	1.709
γ - α	0.037	0.018	0.025	0.047	0.029
2V	-70° ³⁾	-65°	-76°	-92°24'	-75°
C $\wedge$ z	23°	17°	18°	—	—
X	—	very pale green	pale yellowish green	light green brown	brown pale green
Y	—	very pale yellowish green	pale brown-green	greenish brown	reddish brown
Z	—	pale green	dark yellowish green	reddish brown	dark reddish brown

1) cited from the report by A. Harumoto (1933) Chikyu, 19, 96.

2) optical data are cited from the report by S. Kozu and B. Yoshiki (1927) Sci. Rep. Tohoku Imp. Univ. 3, 143.

3) approximate value owing to the wave extinction.

Chemical compositions and optical properties of these specimens are shown in Table 2.

In Table 3. are shown the atomic ratios based on O=2400, which were derived from the results of chemical analyses. Al is divided into two groups, viz, 4-coordination (Al^{IV}) and 6-coordination (Al^{VI}). The chemical formulas of the Ti-oxyhornblende and oxyhornblende were not established, because these specimens are composed of two component minerals as mentioned in part I.

Table 3. Atomic ratio based on (O, OH)=2400.

	tremolite	actinolite	hornblende (tschermakite)	oxyhornblende	Ti- oxyhornblende
Si	792	784	599	552	586
Al^{IV}	8	16	201	248	214
Al^{VI}	4	8	108	70	30
Ti	0	0	2	32	74
Fe^{+3}	0	5	39	79	47
Fe^{+2}	0	46	95	7	92
Mn	0	2	2	2	2
Mg	477	443	291	310	267
Ca	181	174	189	183	198
Na	15	19	52	70	57
K	0	1	6	3	22
OH	227	227	142	0	86

The chemical formulas of these specimens run approximately as follows:

tremolite	$\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}(\text{OH})_2$
actinolite	$(\text{Ca}, \text{Na})_2(\text{Mg}, \text{Fe}^{+2})_5\text{Si}_8\text{O}_{22}(\text{OH})_2$
hornblende (tschermakite)	$(\text{Na}, \text{K})_{0.5}(\text{Ca}, \text{Na})_2(\text{Mg}, \text{Fe}^{+2})_4(\text{Fe}^{+3}, \text{Al}^{\text{VI}})\text{Si}_6\text{Al}^{\text{IV}}_2\text{O}_{22}(\text{OH})_2$

2. X-ray powder diffraction of powdered specimens heated at several temperatures:

Powdered specimens were heated in air at several temperatures. The temperature was raised up at the velocity of $5\sim 10^\circ\text{C}/\text{min.}$, up to a definite temperature, where the specimens were kept for 2 hours. (Table 4) Then, they were cooled rapidly by placing them on a metal surface. A specimen used for heating at a definite temperature was not used for heating at other temperatures.

X-ray powder patterns of the cooled specimens were taken by using diffractometer (NORELCO, North Am. Philp Co.). The results are shown in Fig. 1~5, Table 5~9.

Actinolite and hornblende (tschermakite) change in color by heating at 850°C from green to reddish brown.

The X-ray powder diffraction patterns of tremolite and actinolite heated at 850°C for 2 hours show no change, but the patterns of the specimens heated for 2 hours at

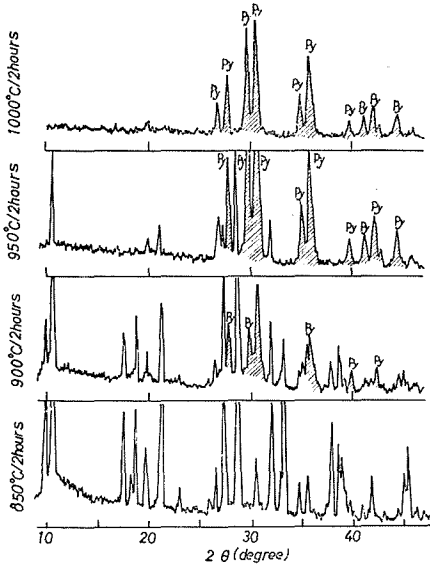


Fig. 1. X-ray powder diffraction patterns of tremolite, heated.

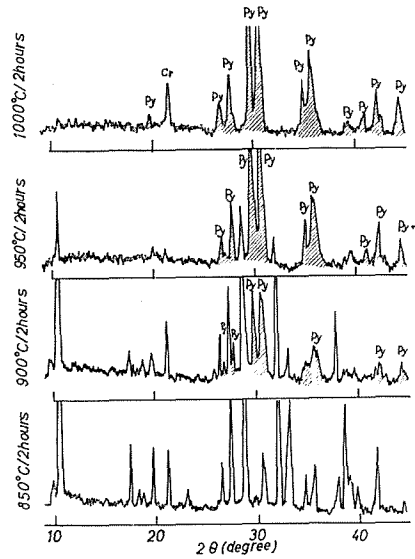


Fig. 2. X-ray diffraction patterns of actinolite, heated.

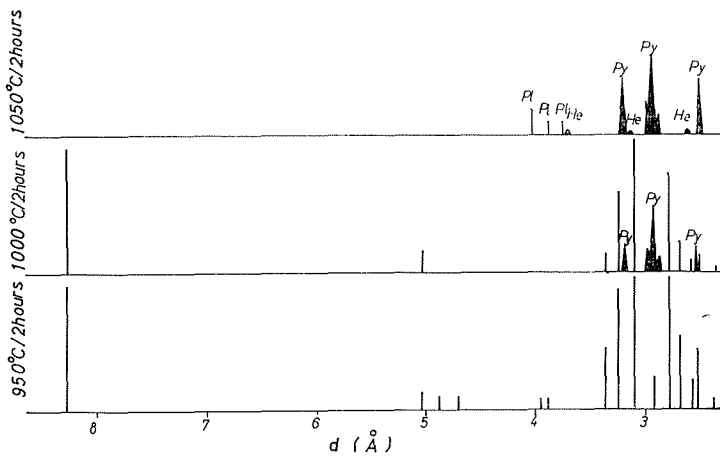


Fig. 3. X-ray diffraction patterns of hornblende, heated.

900°C and 950°C show a change to two phases, the clinoamphibole phase and the clinopyroxene phase. Further, the patterns show that the proportion of the two phase varies. The pyroxene phase is more abundant in the specimens heated at 950°C than

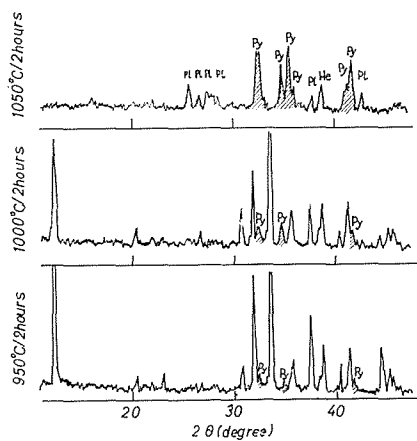


Fig. 4. X-ray diffraction patterns of oxyhornblende, heated.

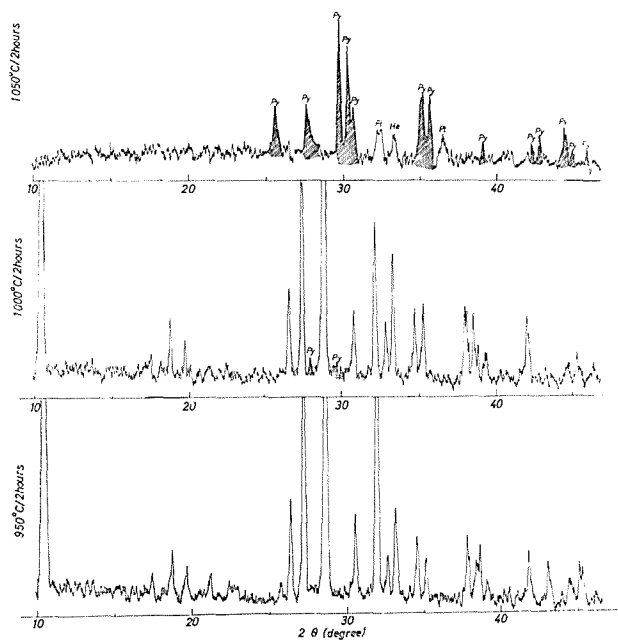


Fig. 5. X-ray diffraction patterns of Ti-oxyhornblende, heated.

in the specimens heated at 900°C. The X-ray diffraction patterns of tremolite and actinolite heated at 1000°C for 2 hours show that the original phases change to two phases, that is, clinopyroxene and cristobalite.

Table 4. Ignition Condition.

Specimen	temperature					duration
tremolite	850°C	900°C	950°C	1000°C		2 hours
actinolite	850°C	900°C	950°C	1000°C		2 hours
hornblende (tschermakite)	850°C	900°C	950°C	1000°C	1050°C	2 hours
oxyhornblende	850°C	900°C	950°C	1000°C	1050°C	2 hours
Ti-oxyhornblende (kaersutite)	850°C	900°C	950°C	1000°C	1050°C	2 hours

Table 5. x-ray power diffraction patterns of tremolite and that heated in air.

h	k	l	natural specimen			heated at 850°C for 2 hours		
			2θ (degree)	d (Å)	I/I ₀	2θ (degree)	d (Å)	I/I ₀
0	2	0	9.75	9.07	19	9.84	8.98	11
1	1	0	10.50	8.42	109	10.58	8.35	149
1	0	1	17.45	5.08	10	17.47	5.07	11
0	1	1	18.13	4.90	6	18.20	4.87	4
2	0	0	18.62	4.77	11	18.72	4.74	15
0	4	0	19.63	4.55	11	19.68	4.51	6
2	2	0	21.10	4.21	24	21.16	4.20	41
0	3	1	22.86	3.89	6	22.96	3.87	3
1	4	1	26.30	3.40	23	26.40	3.37	4
2	4	0	27.23	3.28	41	27.26	3.27	34
3	1	0	28.53	3.13	100	28.60	3.12	100
0	5	1	30.36	2.94	16	30.42	2.93	5
3	3	0	31.92	2.81	27	31.92	2.80	38
2	3	1	32.76	2.73	12	32.80	2.73	4
2	5	1	33.08	2.71	28	33.12	2.70	24
1	1	2	34.55	2.59	9	34.64	2.59	3
2	0	2	35.37	2.54	12	35.46	2.53	4
1	3	2	37.35	2.40	1	37.80	2.37	11
3	0	1	37.75	2.38	7	38.56	2.33	7
4	0	0						
3	2	1	38.50	2.34	14	38.80	2.32	5
4	2	0	39.15	2.30	5	39.20	2.30	2
2	7	1	41.74	2.16	14	41.80	2.16	3
3	3	2						
2	0	2	44.92	2.02	8	45.00	2.01	4
5	1	0	48.00	1.90	14	48.10	1.89	25
5	6	1	55.65	1.65	8	55.70	1.65	7
4	8	0	56.15	1.64	2	56.18	1.64	2
5	2	1	56.88	1.62	2	56.94	1.62	2
6	0	0	58.40	1.58	1	58.20	1.58	2
1	0	3						
1	2	3	59.43	1.56	2			
5	7	0	60.30	1.53	2	60.44	1.53	1
6	2	2						
2	8	2	61.70	1.50	3	61.70	1.50	2
2	12	0	64.75	1.44	18	64.84	1.44	8

Table 5. (continued)

Tremolite heated at 900°C for 2 hours.

2 θ (degree)	d (Å)	I/I ₀	h	k	l	note
9.76	9.05	69	0	2	0	reflection of clinoamphibole
10.46	8.45	9	1	1	0	"
17.44	5.08	8	1	0	$\bar{1}$	"
18.68	4.75	11	2	0	0	"
19.71	4.50	5	0	4	0	"
21.12	4.20	14	2	2	0	"
26.37	3.38	4	1	4	$\bar{1}$	"
27.26	3.27	38	2	4	0	"
27.76	3.21	10	2	2	0	reflection of clinopyroxene
28.60	3.12	100	3	1	0	reflection of clinoamphibole
29.78	2.99	10	2	2	$\bar{1}$	reflection of clinopyroxene
30.58	2.92	18	3	1	0	"
31.96	2.80	21	3	3	0	reflection of clinoamphibole
33.12	2.70	9	2	5	$\bar{1}$	"
34.60	2.59	4	1	1	$\bar{2}$	"
35.01	2.56	5	2	0	$\bar{2}$	reflection of clinopyroxene
35.46	2.53	8	2	0	$\bar{2}$	reflection of clinoamphibole
35.80	2.51	9	0	0	2	reflection of clinopyroxene
37.75	2.44	5	3	0	1	reflection of clinoamphibole
38.56	2.33	10	4	0	0	"
39.12	2.30	2	3	2	1	"
39.77	2.26	3	4	2	0	"
42.30	2.13	4	4	0	0	reflection of clinopyroxene
44.50	2.03	3	3	3	0	"
44.92	2.02	3	0	4	1	"
44.92	2.02	4	2	0	2	reflection of clinoamphibole
48.06	1.89	8	5	1	0	"
50.28	1.81	5	3	1	2	"
55.66	1.65	5	1	5	0	reflection of clinopyroxene
56.15	1.64	4	5	6	$\bar{1}$	reflection of clinoamphibole
56.64	1.62	4	4	8	0	"
58.16	1.58	4				reflection of clinopyroxene
58.16	1.58	17	6	0	0	"
61.71	1.50	5	1	0	3	reflection of clinoamphibole
64.80	1.44	4	2	8	2	"
66.56	1.40	3	2	12	0	"
66.56	1.40	3				reflection of clinopyroxene

Table 5. (Continued)

Tremolite heated at 950°C for 2 hours.

2 θ (degree)	d (Å)	I/I ₀	h	k	l	note
10.54	8.39	56	1	1	0	reflection of clinoamphibole
19.90	4.45	7	0	4	0	"
21.10	4.21	14	2	2	0	"
26.78	3.33	22	0	2	1	reflection of clinopyroxene
27.24	3.27	17	2	4	0	reflection of clinoamphibole
27.78	3.21	53	2	2	0	reflection of clinopyroxene
28.60	3.12	100	3	1	0	reflection of clinoamphibole
29.78	2.99	83	2	2	$\bar{1}$	reflection of clinopyroxene
30.66	2.91	102	3	1	0	"
31.88	2.80	20	3	3	0	reflection of clinoamphibole
34.96	2.56	28	2	0	$\bar{2}$	reflection of clinopyroxene
35.87	2.50	66	0	0	2	reflection of clinopyroxene
39.76	2.27	11	4	0	0	"
41.28	2.19	14	0	2	2	"
42.30	2.13	24	3	3	0	"
42.80	2.11	6	3	3	$\bar{1}$	"
44.54	2.03	17	0	4	1	"
46.00	1.97	6	4	0	$\bar{2}$	"
48.04	1.89	7	2	4	$\bar{1}$	"
50.20	1.82	15	5	1	0	reflection of clinoamphibole
52.28	1.75	6	3	1	2	reflection of clinopyroxene
55.08	1.67	7	0	4	2	"
56.62	1.62	36				"
59.90	1.52	6				"
61.74	1.50	19	2	8	2	reflection of clinoamphibole
66.56	1.40	17				reflection of clinopyroxene

tremolite heated at 1000°C for 2 hours.			natural diopside from Ham Is.*		
2 θ (degree)	d (Å)	I/I ₀	d (Å)	I/I ₀	h k l
26.76	3.33	27	3.33	5	0 2 1
27.77	3.21	55	3.23	80	2 2 0
29.76	2.99	100	2.98	100	2 2 $\bar{1}$
30.64	2.92	122	2.94	70	3 1 0
35.00	2.56	35	2.56	10	2 0 2
35.83	2.50	71	2.53	40	0 2 2, 1 3 $\bar{1}$
39.80	2.26	10	2.29	10	1 1 $\bar{2}$, 2 2 1
41.32	2.18	12	2.18	5	4 0 0
42.26	2.14	20	2.12	20	0 2 2
42.84	2.11	6	2.10	30	3 3 $\bar{1}$
44.57	2.03	12	2.03	10	4 2 $\bar{1}$
46.13	1.97	4	2.002	30	0 4 1
52.28	1.75	4	1.959	5	4 0 $\bar{2}$
55.08	1.67	4	1.748	40	2 4 $\bar{1}$
56.64	1.63	35	1.668	5	0 4 2
			1.622	20	

Cu-K α radiation 35 kV-10 mA., time constant 4 sec, multiplier 1, scale factor 4, slit 1°-0.006'-1°, scanning speed 1°/min.,

* natural diopside from Ham Is., Alaska, Ref., Yoder, Am. J. Sci., 245-48, (1950).

space group of clinoamphibole: $I2/m$.

space group of clinopyroxene: $C2/c$.

Table 6. x-ray diffraction patterns of actinolite and that heated in air.

<i>h</i> <i>k</i> <i>l</i>	natural specimen			heated at 850°C for 2 hours		
	2θ (degree)	d (Å)	I/I ₀	2θ (degree)	d (Å)	I/I ₀
0 2 0	9.80	9.02	56	9.80	9.02	4
1 1 0	10.40	8.50	95	10.55	8.38	100
1 0 $\bar{1}$	17.36	5.10	33	17.46	5.07	12
0 1 1	18.16	4.88	24	18.25	4.86	3
2 0 0	18.65	4.80	21	18.70	4.74	3
0 4 0	19.65	4.52	65	19.72	4.50	11
2 2 0	21.12	4.20	80			
	22.92	3.87	16	23.00	3.86	4
1 4 $\bar{1}$	26.25	3.39	37	26.44	3.37	8
2 4 0	27.24	3.27	90	27.30	3.26	30
3 1 0	28.52	3.13	100	28.70	3.11	100
0 5 1	30.37	2.94	49	30.48	2.93	10
3 3 0	31.88	2.80	85	32.04	2.79	41
2 5 $\bar{1}$	33.04	2.71	60	33.24	2.69	24
1 1 $\bar{2}$	34.52	2.60	19	34.64	2.59	6
2 0 $\bar{2}$	35.34	2.54	33	35.50	2.53	9
4 0 0						
3 0 1	37.71	2.38	30	37.86	2.37	6
3 2 1	38.48	2.34	34	38.65	2.33	25
4 2 0	39.11	2.30	18	39.25	2.29	5
2 7 $\bar{1}$						
3 3 $\bar{2}$	41.68	2.17	28	41.48	2.18	2
2 0 2	44.91	2.02	26	45.07	2.01	10
5 1 0	48.06	1.89	26	48.34	1.88	10
5 3 0	50.28	1.81	27	50.50	1.81	12
5 0 1	54.38	1.68	9			
4 8 0	55.74	1.65	33	55.86	1.644	13
5 2 1	56.88	1.617	10	56.96	1.615	4
6 0 0						
1 0 3	58.34	1.580	24	58.42	1.578	7
6 2 $\bar{2}$						
5 7 0	60.40	1.531	10	60.56	1.527	3
2 8 2	61.66	1.502	16	61.86	1.498	3
2 12 0	64.89	1.435	49	65.10	1.431	24

actinolite heated at 900°C for 2 hours.

2θ (degree)	d (Å)	I/I ₀	<i>h</i> <i>k</i> <i>l</i>	note
10.48	8.43	77	1 1 0	reflection of clinoamphibole
17.46	5.07	3	1 0 $\bar{1}$	"
18.72	4.74	2	2 0 0	"
19.66	4.51	3	0 4 0	"
21.24	4.18	6	2 2 0	"

actinolite heated at 900°C for 2 hours (continued)

2θ (degree)	d (Å)	I/I ₀	h k l	note
26.42	3.37	4	1 4 $\bar{1}$	reflection of clinoamphibole
26.82	3.32	2	0 2 1	reflection of clinopyroxene
27.36	3.26	9	2 4 0	reflection of clinoamphibole
27.36	3.26	4	2 2 0	reflection of clinopyroxene
28.70	3.11	100	3 1 0	reflection of clinoamphibole
29.76	2.99	9	2 2 $\bar{1}$	reflection of clinopyroxene
30.58	2.92	8	3 1 0	"
32.06	2.89	26	3 3 0	reflection of clinoamphibole
33.16	2.70	3	2 5 $\bar{1}$	"
35.00	2.56	2	2 0 $\bar{2}$	reflection of clinopyroxene
35.84	2.50	3	0 0 2	"
37.86	2.37	7	4 0 0	reflection of clinoamphibole
39.70	2.27	1	4 2 0	"
41.82	2.16	1	2 7 $\bar{1}$	"
			3 3 $\bar{2}$	"
42.26	2.14	1	3 3 0	reflection of clinopyroxene
44.42	2.04	2	0 4 1	"
48.36	1.88	5	5 1 0	reflection of clinoamphibole
50.48	1.81	4	5 3 0	"
52.50	1.74	1	0 4 2	reflection of clinopyroxene
55.88	1.64	9	4 8 0	reflection of clinoamphibole
56.04	1.639	1		reflection of clinopyroxene
56.48	1.627	2		"
56.96	1.615	6	6 0 0	et al. reflection of clinoamphibole
65.04	1.432	3	2 12 0	"

actinolite heated at 950°C for 2 hours

2θ (degree)	d (Å)	I/I ₀	h k l	note
10.58	8.35	128	1 1 0	reflection of clinoamphibole
26.80	3.32	51	0 2 1	reflection of clinopyroxene
27.77	3.21	107	2 2 0	"
28.68	3.11	100	3 1 0	reflection of clinoamphibole
29.82	2.99	268	2 2 $\bar{1}$	reflection of clinopyroxene
30.66	2.91	241	3 1 0	"
31.98	2.80	41	3 3 0	reflection of clinoamphibole
35.04	2.56	45	2 0 $\bar{2}$	reflection of clinopyroxene
35.88	2.50	107	0 0 2	"
39.60	2.27	18	4 0 0	"
41.18	2.19	17	0 2 2	"
42.36	2.13	70	3 3 $\bar{1}$	"
44.54	2.03	41	0 4 1	"
48.30	1.88	14	5 1 0	reflection of clinoamphibole
52.34	1.75	14	0 4 2	reflection of clinopyroxene
56.70	1.62	54		"
60.12	1.54	18		"
65.04	1.43	14	2 12 0	reflection of clinoamphibole

Table 6. (continued)

actinolite heated at 1000°C for 2 hours			natural diopside from Ham Is.		
2θ (degree)	d (Å)	I/I ₀	d (Å)	I/I ₀	h k l
19.88	4.46	9	4.44	5	0 2 0
21.72*	4.09	28			
26.78	3.33	28	3.33	5	0 2 1
27.77	3.21	33	3.23	80	2 2 0
29.82	2.99	100	2.98	100	2 2 $\bar{1}$
30.66	2.91	63	2.94	70	3 1 0
35.02	2.56	29	2.56	10	2 0 2
35.82	2.50	46	2.53	40	0 0 2, 1 3 $\bar{1}$
41.20	2.19	6	2.18	5	1 1 2, 2 2 1
42.36	2.13	23	2.146	20	0 2 2
42.86	2.108	7	2.101	30	3 3 0
44.54	2.033	16	2.034	10	4 2 $\bar{1}$
52.32	1.747	4	1.748	40	0 4 1
56.70	1.622	25	1.622	20	0 4 2
61.74	1.501	5	1.520	10	

* reflection of cristobalite.

Cu-Kα radiation 35 kV-10 mA, time constant 4 sec, multiplier 1, scale factor 4, slit 1°-0.006'-1°, scanning speed 1°/min., space group of clin amphibole: $I2/m$. space group of clinopyroxene: $C2/c$.

Table 7. x-ray powder diffraction patterns of hornblende and that heated in air.

h k l	natural specimen			heated at 950°C for 2 hours		
	2θ (degree)	d (Å)	I/I ₀	2θ (degree)	d (Å)	I/I ₀
1 1 0	10.50	8.42	103	10.60	8.33	100
1 0 $\bar{1}$				17.44	5.08	3
0 1 1				18.14	4.88	3
2 0 0				18.84	4.71	3
0 4 0	19.60	4.53	11			11
1 4 $\bar{1}$	26.27	3.39	19	26.44	3.37	
2 4 0	27.20	3.28	28	27.40	3.25	24
3 1 0	28.50	3.13	100	28.81	3.10	100
0 5 1	30.35	2.94	17	30.58	2.92	6
3 3 0	31.85	2.81	32	32.18	2.78	36
2 3 1	32.60	2.75	17			
2 5 $\bar{1}$	33.00	2.71	24	33.22	2.69	16
1 1 2	34.52	2.60	13	34.64	2.59	7
2 0 $\bar{2}$	35.10	2.56	13	35.26	2.54	13
4 0 0	37.75	2.38	6	37.96	2.37	7
3 2 1	38.36	2.34	16	38.64	2.33	5
4 2 0	39.05	2.30	7	38.98	2.31	4
3 3 $\bar{2}$	41.76	2.16	18	41.94	2.15	8
2 0 2	45.00	2.01	10	45.20	2.00	6
5 1 0	48.05	1.89	4	48.50	1.88	3
5 3 0				50.66	1.80	6
5 6 $\bar{1}$	55.70	1.65	16	56.08	1.64	13
5 2 1	56.97	1.62	6			
1 0 3	58.15	1.59	10	58.34	1.58	13
2 8 2	61.65	1.50	11			
2 12 0	64.70	1.44	11	65.20	1.43	12

Cu-Kα radiation 35 kV-10 mA, time constant 4 sec., multiplier 1, scale factor 4, slit 1°-0.006'-1°, scanning speed 1°/min., space group of clin amphibole: $I2/m$.

Table 7 (continued)

Hornblende heated at 1000°C for 2 hours

2θ (degree)	d (Å)	I/I ₀	h k l	note
12.40	8.28	140	1 1 0	reflection of clinoamphibole
20.42	5.04	3	1 0 $\bar{1}$	"
30.86	3.36	4	1 4 $\bar{1}$	"
31.98	3.25	19	2 4 0	"
32.50	3.20	5	2 2 0	reflection of clinopyroxene
33.74	3.08	100	3 1 0	reflection of clinoamphibole
34.86	2.99	6	2 2 $\bar{1}$	reflection of clinopyroxene
35.60	2.93	12	3 1 0	"
36.30	2.87	3	3 1 $\bar{1}$	"
37.64	2.77	23	3 3 0	reflection of clinoamphibole
38.76	2.69	7	2 5 $\bar{1}$	"
40.50	2.584	3	1 1 $\bar{2}$	"
41.36	2.54	7	0 0 2	reflection of clinopyroxene
44.50	2.36	2	4 0 0	reflection of clinoamphibole
45.30	2.32	2	3 2 1	"
49.20	2.15	3	3 3 $\bar{2}$	"
57.19	1.87	4	5 1 0	"
59.80	1.79	4	3 1 2	reflection of clinopyroxene
77.77	1.43	4	2 12 0	reflection of clinoamphibole

Co-K α radiation 30 kV-10 mA, time constant 4 sec., multiplier 1. scale factor 4. slit 1°-0.006'1°, scanning speed 1°/min. space group of clinopyroxene: C2/c.

Hornblende heated at 1050°C for 2 hours

2θ (degree)	d (Å)	I/I ₀	h k l	note
25.60	4.04	40		reflection of plagioclase
26.64	3.88	20		"
27.56	3.76	20		"
27.98	3.70	17		reflection of hematite
32.45	3.20	85	2 2 0	reflection of clinopyroxene
43.78	2.99	60	2 2 $\bar{1}$	"
35.48	2.94	100	3 1 0	"
35.89	2.89	30	3 1 $\bar{1}$	"
38.76	2.69	34		reflection of hematite
41.71	2.51	85	0 2 2 } 1 3 $\bar{1}$ }	reflection of clinopyroxene
42.70	2.46	14		reflection of plagioclase
48.88	21.6	14		reflection of clinopyroxene
49.64	2.13	18	0 2 2	"
50.88	2.08	15	3 3 0	"
52.27	2.03	14	4 2 $\bar{1}$	"
58.52	1.83	18	0 4 1	"
61.45	1.75	17	1 5 0	"
67.00	1.62	15	0 4 2	"
67.70	1.61	9		"
72.70	1.509	30		
74.70	1.474	20		reflection of hematite
78.50	1.413	15		

Co-K α radiation 30 kV-10 mA, time constant 4 sec., multiplier 1, scale factor 4, slit 1°-0.006'-1°, scanning speed 1°/min.

Table 8. x-ray powder diffraction patterns of oxyhornblende and that heated in air

<i>h</i>	<i>k</i>	<i>l</i>	natural specimen*			heated at 950°C for 2 hours		
			2 θ (degree)	<i>d</i> (Å)	I/I ₀	2 θ (degree)	<i>d</i> (Å)	I/I ₀
1	1	0	10.60	8.35	84	12.32	8.34	156
1	0	$\bar{1}$				20.38	5.06	5
0	4	0	19.68	4.51	9	22.98	4.49	6
1	4	$\bar{1}$	26.41	3.38	16	30.80	3.37	9
2	4	0	27.34	3.27	33	31.92	3.25	44
3	1	0	28.70	3.11	100	33.60	3.09	100
0	5	1	30.55	2.93	12	35.68	2.92	12
3	3	0	32.02	2.80	14	37.48	2.78	27
2	3	1	32.75	2.73	14	38.38	2.72	4
2	5	$\bar{1}$	33.14	2.71	28	38.72	2.70	16
1	1	$\bar{2}$	34.62	2.59	14	40.42	2.59	10
2	0	$\bar{2}$	35.20	2.55	13	41.26	2.54	16
4	0	0	37.90	2.37	5	44.34	2.37	16
3	2	1	38.56	2.33	9	45.12	2.33	8
4	2	0				45.50	2.31	4
3	3	$\bar{2}$	41.95	2.15	9	49.10	2.15	6
2	0	$\bar{2}$	45.17	2.01	10	52.20	2.03	6
5	1	0	48.43	1.88	5	56.94	1.87	2
5	3	0	50.60	1.80	7	59.60	1.80	3
4	8	0	56.05	1.64	10	66.16	1.64	10
5	2	1				66.60	1.63	3
2	8	2				73.16	1.50	35
2	12	0				77.44	1.43	4

* Cu-K α radiation

oxyhornblende heated at 1000°C for 2 hours

2 θ (degree)	<i>d</i> (Å)	I/I ₀	<i>h</i>	<i>k</i>	<i>l</i>	note
12.34	8.32	78	1	1	0	reflection of clinoamphibole
20.34	5.07	8	1	0	$\bar{1}$	"
26.68	3.88	6	0	3	1	"
30.78	3.37	17	1	4	$\bar{1}$	"
31.90	3.30	35	2	4	0	"
32.46	3.20	8	2	2	0	reflection of clinopyroxene
33.64	3.09	100	3	1	0	reflection of clinoamphibole
34.76	2.99	10	2	2	$\bar{1}$	reflection of clinopyroxene
35.68	2.92	17	0	5	1	reflection of clinoamphibole
37.50	2.78	20	3	3	0	"
38.48	2.71	8	2	3	1	"
38.72	2.70	20	2	5	$\bar{1}$	"
40.42	2.59	6	1	1	$\bar{2}$	"
41.26	2.54	21	2	0	$\bar{2}$	"
41.68	2.51	6	0	0	2	reflection of clinopyroxene
44.36	2.40	4	4	0	0	reflection of clinoamphibole
45.16	2.33	7	3	2	1	"
45.60	2.31	6	4	2	0	"
49.10	2.15	4	3	3	$\bar{2}$	"
52.96	2.01	10	2	0	2	"
59.64	1.80	5	5	3	0	"
66.16	1.64	8	4	8	0	"
77.46	1.43	18	2	12	0	"

oxyhornblende heated at 1050°C for 2 hours

2 θ (degree)	d (Å)	I/I ₀	h k l	note
25.60	4.04	27		reflection of plagioclase
26.40	3.92	16		"
27.50	3.76	19		"
28.46	3.64	14		"
32.46	3.20	74	2 2 0	reflection of clinopyroxene
34.80	2.99	57	2 2 $\bar{1}$	"
35.48	2.94	100	3 1 0	"
35.92	2.90	38	3 1 $\bar{1}$	"
37.78	2.76	14		reflection of plagioclase
38.72	2.70	30		reflection of hematite
41.19	2.543	30	2 0 $\bar{2}$	reflection of clinopyroxene
41.74	2.51	62	0 0 2	reflection of clinopyroxene
42.72	2.45	16		reflection of plagioclase
42.76	2.21	8	4 0 0	reflection of clinopyroxene
52.20	2.03	8	0 4 1	"
58.14	1.84	14	1 5 0	"
61.62	1.75	11	0 4 2	"
63.62	1.696	5	3 1 $\bar{3}$	"

Co-K α radiation, 30 kV-10 mA, time constant 4 sec., multiplier 1, scale factor 4, slit 1°-0.006'-1°, scanning speed 1°/min., space group of clinopyroxene: $I2/m$. space group of clinopyroxene: $C2/c$.

Table 9. x-ray powder diffraction patterns of kaersutite and that heated in air

h k l	natural specimen			heated at 950°C for 2 hours		
	2 θ (degree)	d (Å)	I/I ₀	2 θ (degree)	d (Å)	I/I ₀
1 1 0	10.50	8.42	69	10.60	8.34	132
1 0 $\bar{1}$				17.42	5.09	3
2 0 0				18.74	4.73	6
0 4 0				19.66	4.51	4
2 2 0				21.20	4.19	3
1 4 $\bar{1}$	26.30	3.39	13	26.41	3.37	11
2 4 0	27.15	3.29	32	27.30	3.26	47
3 1 0	28.54	3.13	100	28.70	3.11	100
0 5 1	30.33	2.95	11	30.56	2.92	10
3 3 0	31.80	2.81	21	32.00	2.79	65
2 3 1	32.46	2.75	10	32.68	2.74	5
2 5 $\bar{1}$	32.96	2.71	13	33.18	2.70	11
1 1 $\bar{2}$	34.43	2.61	11	34.58	2.59	7
2 0 $\bar{2}$	35.30	2.54	16	35.16	2.55	5
4 0 0	37.66	2.39	9	37.82	2.38	7
3 2 1	38.25	2.35	10	38.66	2.33	6
4 2 0				39.14	2.30	2
3 3 $\bar{2}$	41.66	2.17	8	41.86	2.16	6
2 0 2	44.88	2.02	7	45.12	2.01	4
5 1 0	48.00	1.90	6	48.26	1.88	5
5 3 0				50.50	1.81	12
4 8 0				55.98	1.64	8
5 2 1	56.80	1.62	7	56.26	1.63	4
6 0 0 } 1 0 3 }	58.06	1.59	9			
2 8 2				58.38	1.58	8
2 12 0	64.56	1.44	7	64.78	1.44	4

Table 9. (continued)

kaersutite heated at 1000°C for 2 hours

2θ (degree)	d (Å)	I/I_0	h k l	note
10.58	8.35	117	1 1 0	reflection of clinoamphibole
18.74	4.73	11	2 0 0	"
19.68	4.51	6	0 4 0	"
26.44	3.38	16	1 4 $\bar{1}$	"
27.28	3.27	45	2 4 0	"
27.68	3.22	2	2 2 0	reflection of clinopyroxene
28.74	3.10	100	3 1 0	reflection of clinoamphibole
29.80	2.99	1	2 2 $\bar{1}$	reflection of clinopyroxene
30.58	2.92	12	0 5 1	reflection of clinoamphibole
32.02	2.79	28	3 3 0	"
32.68	2.74	10	2 3 1	"
33.18	2.697	22	2 5 $\bar{1}$	"
34.56	2.59	12	1 1 $\bar{2}$	"
35.12	2.55	13	2 0 $\bar{2}$	"
37.86	2.37	12	4 0 0	"
38.40	2.34	11	3 2 1	"
39.14	2.30	4	4 2 0	"
41.88	2.15	11	3 3 $\bar{2}$	"
45.12	2.01	4	2 0 2	"
48.26	1.88	4	5 1 0	"
50.44	1.81	8	5 3 0	"
55.98	1.64	10	4 8 0	"
58.42	1.58	5	5 2 1	"
64.80	1.437	18	2 12 0	"

kaersutite heated at 1050°C for 2 hours

2θ (degree)	d (Å)	I/I_0	h k l	note
25.68	3.47	38	0 2 1	reflection of clinopyroxene
27.68	3.22	39	2 2 0	"
29.80	2.99	100	2 2 $\bar{1}$	reflection of clinopyroxene
30.35	2.94	80	3 1 0	"
30.78	2.90	37	3 1 $\bar{1}$	"
32.40	2.76	22		reflection of plagioclase
33.32	2.69	18		reflection of hematite ?
35.20	2.55	40	2 0 $\bar{2}$	reflection of clinopyroxene
35.70	2.51	44	0 0 3	reflection of clinopyroxene
36.52	2.46	18		reflection of plagioclase
39.12	2.30	11	4 0 0	reflection of clinopyroxene
42.36	2.13	11	3 3 0	"
42.90	2.11	18	4 2 $\bar{1}$	"
44.52	2.03	22	0 4 1	"
56.52	1.63	16		"
60.12	1.54	11		"

Cu-K α radiation, 35 kV-15 mA, time constant 4 sec., multiplier 1, scale factor 4, slit 1° -0.006'- 1° , scanning speed 1° /min., space group of clinoamphibole: $I2/m$. space group of clinopyroxene: $C2/c$.

The hornblende (tschermakite) heated for 2 hours at 850°C, 900°C and 950°C does not show any change, but that heated at 1000°C for 2 hours shows two phases, that is, clinopyroxene phase and clinoamphibole phase. The hornblende heated at 1050°C for 2 hours shows a structure collapse, producing clinopyroxene, plagioclase and hematite. The temperature at which the structure collapse of hornblende takes place is higher than that at which the structure collapse of actinolite and tremolite takes place, and the temperature range at which clinoamphibole and clinopyroxene phases coexist is narrower in the case of hornblende than in the case of tremolite and actinolite.

The oxyhornblende and Ti-oxyhornblende were also heated. Before heating, these specimens were composed of two phase, that is, clinoamphibole and clinopyroxene phases, but the latter phase was somewhat difficult to observe by an X-ray powder diffraction. Specimens heated both at 950°C and at 1000°C for 2 hours show a distinct coexistence of the two phases, clinopyroxene and clinoamphibole, the latter being more predominant. Specimens heated at 1050°C for 2 hours shows the disappearance of amphibole phase and the coexistence of pyroxene, plagioclase and hematite.

Specimens did not show any trace of melting by heating, so the reactions may have occurred in a solid state.

3. Thermo-gravimetric analysis:

The thermo-gravimetric analyses were carried out with tremolite, actinolite, hornblende (tschermakite) and Ti-oxyhornblende in order to find out the relationship between dehydration and phase change. The analyses were carried out by using recording thermobalance equipment (Rigaku Denki Co.).

Thermo-gravimetric curves obtained are shown in Fig. 6, and the results of thermo-gravimetric analyses are illustrated in Table 10.

Tremolite and actinolite show a continuous, gradual weight loss from room temperature up to about 900°C. In this range absorbed water may be driven off. At the temperatures from 900°C, the weight loss curves show an abrupt rise. In this range, a removal of the hydroxyl ions in the structures may take place and may be driven off as water molecules.

The dehydration temperature of the hornblende is higher than that of tremolite and actinolite as seen from the weight loss curves. The weight loss curve of the former shows an abrupt rise at the temperatures from 1050°C to 1150°C.

The weight loss curve of the Ti-oxyhornblende does not show an abrupt rise. This may be due to the fact that the Ti-oxyhornblende does not hold any hydroxyl ions, or holds only a little amount which is not visible with this equipment. In the Ti-oxyhornblende oxygen ions may have taken the place of the structural position of hydroxyl ion, that is, in this mineral the secession of hydrogens from hydroxyl ions may have already taken place.

Table 10. Loss of weight of calciferous amphiboles by heating progressively up to about 1100°C

a) tremolite, prismatic crystal

(sample weight, 1.904 g at start; heating velocity, 200°C/hr.)

temperature °C	loss of weight (mg)	percent of loss of weight	percent of total loss of weight
200	9.2	0.49	12.21
300	15.2	0.79	20.16
400	20.0	1.05	26.53
500	22.4	1.17	29.72
600	24.4	1.24	32.37
700	28.0	1.47	37.15
800	29.6	1.55	39.27
900	32.0	1.68	42.46
1000	48.8	2.56	64.75
1100	75.36	3.95	100.00

b) actinolite

(sample weight, 2.440 g at start; heating velocity, 200°C/hr.)

temperature °C	loss of weight (mg)	percent of loss of weight	percent of total loss of weight
100	3.2	0.13	4.56
200	7.2	0.29	10.27
300	10.1	0.41	14.27
400	14.1	0.58	20.11
500	15.7	0.64	22.39
600	17.3	0.71	24.67
700	20.0	0.82	28.53
800	21.4	0.88	30.53
900	24.8	1.02	35.37
930	25.3	1.04	36.09
1000	36.5	1.50	52.06
1100	68.6	2.81	97.86
1200	70.1	2.87	100.00

c) hornblende (tschermakite)

(sample weight, 1.860 g at start; heating velocity, 200°C/hr.)

temperature °C	loss of weight (mg)	percent of loss of weight	percent of total loss of weight
100	2.8	0.15	7.42
200	4.8	0.26	12.73
300	6.4	0.34	16.97
400	10.2	0.55	27.05
500	10.5	0.56	27.85
600	11.8	0.63	31.29
700	12.9	0.69	34.22
800	16.4	0.88	43.50
900	20.0	1.08	53.05
1000	22.0	1.18	58.35
1080	25.2	1.36	66.84
1100	29.6	1.59	78.51
1130	36.1	1.94	95.75
1200	37.7	2.03	100.00

d) Ti-oxyhornblende

(sample weight 1.652 g at start; heating velocity, 200°C/hr.)

temperature °C	loss of weight (mg)	percent of loss of weight	percent of total loss of weight
100	2.4	0.14	8.95
200	7.2	0.44	26.86
300	11.6	0.70	43.28
400	13.6	0.82	50.74
500	14.8	0.89	55.22
600	16.4	0.99	61.19
700	18.8	1.14	70.15
800	21.6	1.31	80.59
900	23.2	1.40	86.56
1000	24.0	1.45	89.55
1100	24.8	1.50	92.54
1200	26.8	1.62	100.00

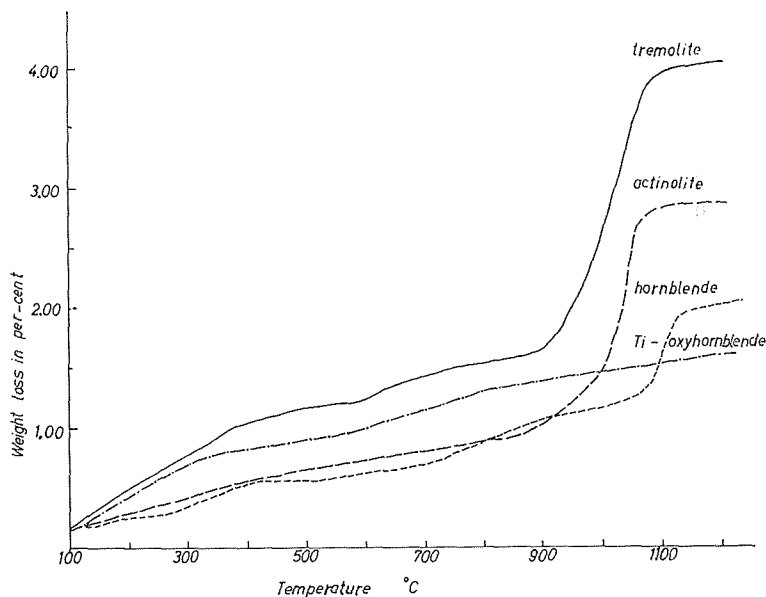


Fig. 6. Thermo-gravimetric curves of calciferous amphiboles.

In the tremolite and actinolite, the dehydration temperature coincides with the temperature of phase change, but in the hornblende it does not coincide; the dehydration temperature is higher than that of the phase change by about 50°C.

The loss of weight of tremolite, actinolite, hornblende and Ti-oxyhornblende nearly coincides with the weight of total water determined by the gravimetric chemical analysis.

4. X-ray analysis with heated single crystals:

X-ray powder patterns of the tremolite and actinolite heated between 900°C and 1000°C for 2 hours show that the clinopyroxene phase coexists with the clin amphibole phase. This is also the case with the hornblende heated between 1000°C and 1050°C for 2 hours. Therefore Weissenberg photographs were taken with single crystals of tremolite, actinolite and hornblende heated to see whether the clinopyroxene phase coexists with the amphibole phase in the single crystals as well, and if these two phases coexist, whether there is any crystallographic relationship between them.

Specimens used in the present experiment were 0.2~0.5 mm in diameter and 2~3 mm in length. They were quenched rapidly after heating.

Rotation and Weissenberg photographs of these specimens about the c-axis were taken by using Cu-K α radiation. In the rotation photographs all the spots are arranged perfectly on the layer lines. However, in the zero, 1st and 2nd level

Weissenberg photographs of the tremolite and actinolite heated at 900°C for 2 hours, there are always found an extra set of spots which, compared with the corresponding photographs of natural tremolite and actinolite, are not accountable on the basis of an ordinary lattice. (Fig. 7~10)

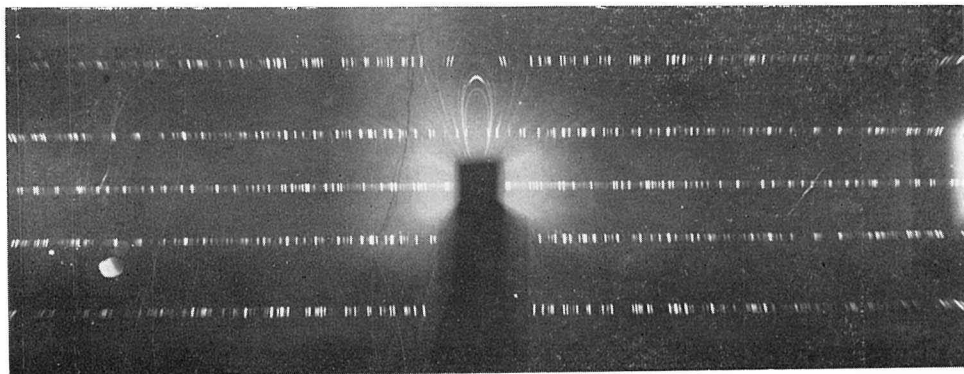


Fig. 7. (A) Rotation photograph of tremolite about c-axis.

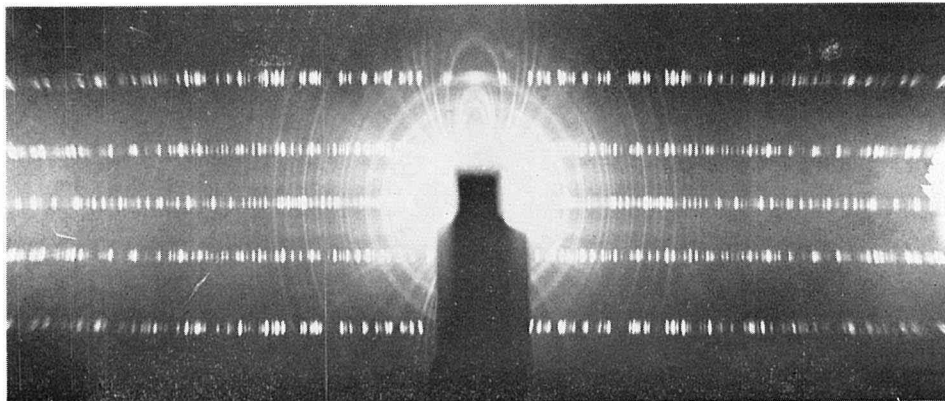


Fig. 7. (B) Rotation photograph of tremolite about c-axis, heated at 900°C for 2 hours.

When the lattice rows on the Weissenberg photographs are traced and observed closely, it is found that there are two sets of spots, of which one is the same as that of natural clin amphibole, the other is the same as that of diopside (Fig. 8 (C), Fig. 10 (C)).

It follows immediately from these observations that the tremolite and actinolite heated at 900°C for 2 hours are made up of two different lattices, e.g. clin amphibole and clinopyroxene lattices, $b^* c^*$ reciprocal net planes being overlapped and the direct

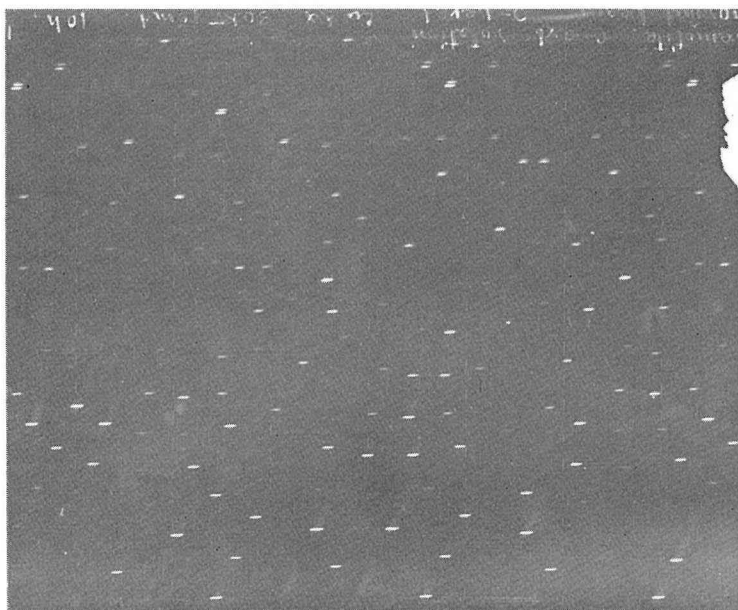


Fig. 8. (A) Weissenberg photograph of tremolite about c-axis.



Fig. 8. (B) Weissenberg photograph of tremolite about c-axis, heated at 900°C for 2 hours. (zero level)

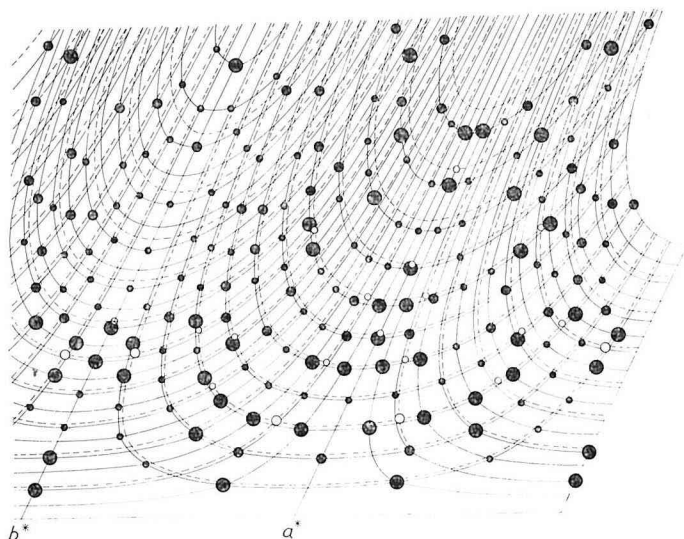


Fig. 8. (C) $hk0$ Weissenberg diagram of tremolite heated at 900°C for 2 hours. Solid circles denoted one set and open circles the other set of spots.

c-axes being similar in direction and identical in length. The space group of the former is $I2/m$, that of the latter $C2/c$. These are similar to the facts observed with the natural Ti-oxyhornblende from Korea and oxyhornblende from Japan (part I).

In the zero, 1st and 2nd level Weissenberg photographs of the hornblende heated, at 950°C for 3 hours, there is not found any "extra" spot when compared with the

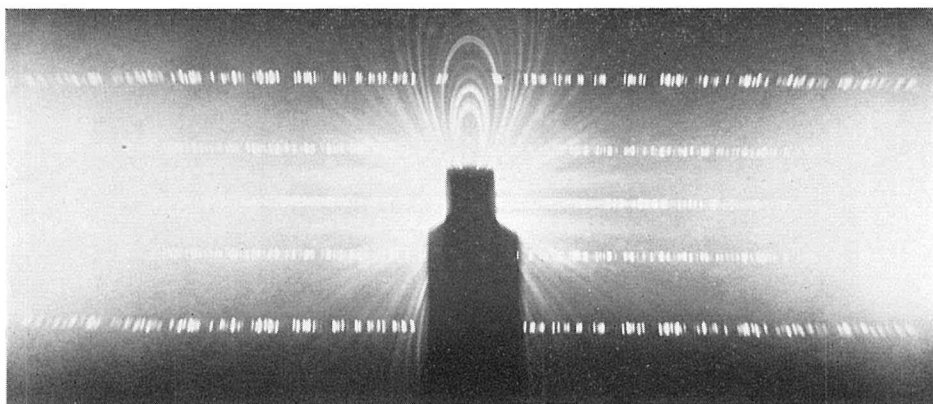


Fig. 9. (A) Rotation photograph of actinolite about c-axis.

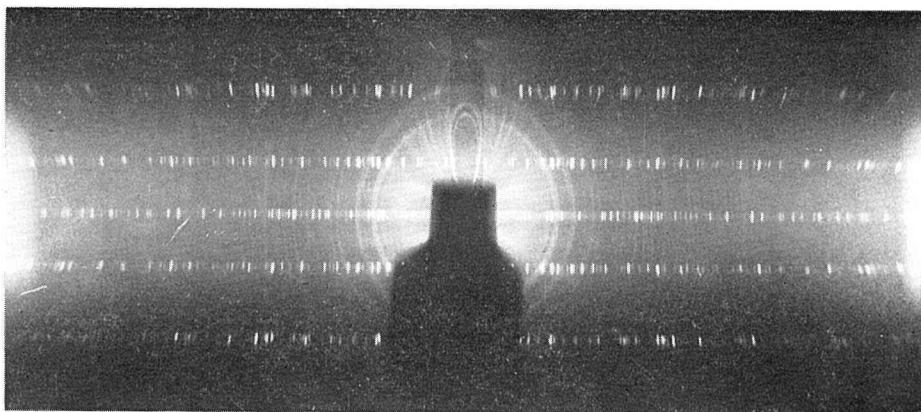


Fig. 9. (B) Rotation photograph of actinolite about c-axis heated at 900°C for 2 hours.

photographs of the natural hornblende. Therefore, although the hornblende changes to reddish brown in color, the crystal structure does not change. On the other hand, Weissenberg photographs of the hornblende, heated at 1020°C for 2 hours, about the c-axis show that the hornblende changes into the two phases stated above, associated in the same way, and clinopyroxene being predominant over the other. (Fig. 11, 12).



Fig. 10. (A) Weissenberg photograph of actinolite about c-axis. (zero level)

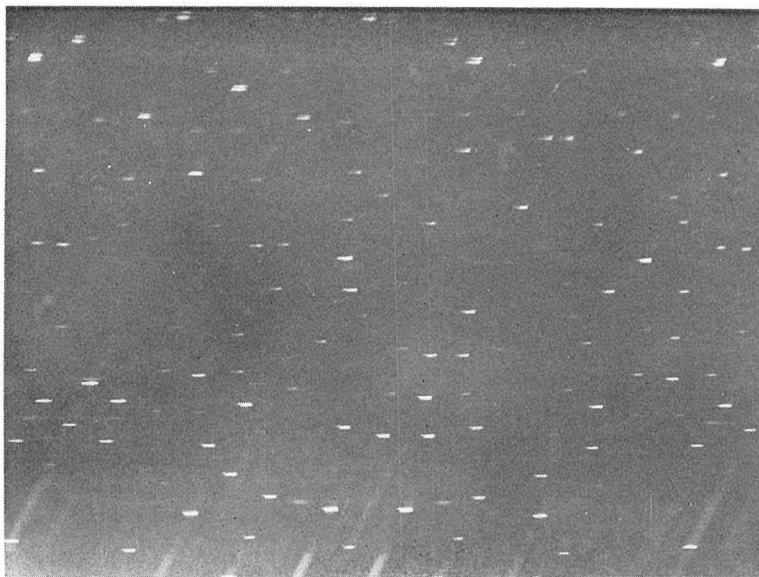


Fig. 10. (B) Weissenberg photograph of actinolite about c -axis, heated at 900°C for 2 hours. (zero level)

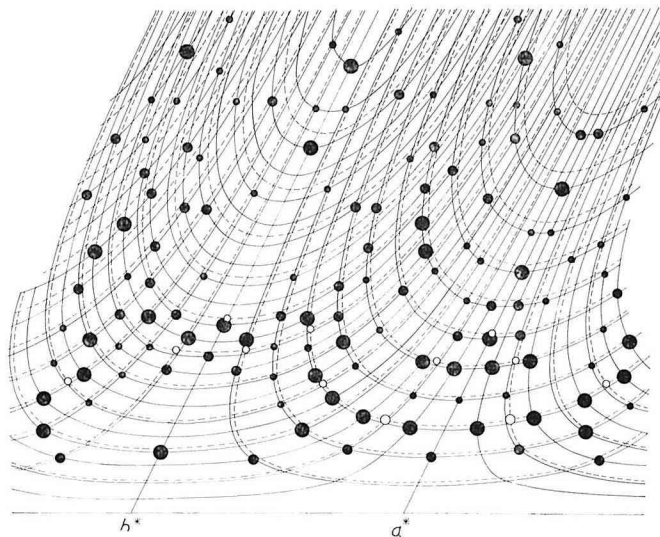


Fig. 10. (C) $hk0$ Weissenberg diagram of actinolite heated 900°C for 2 hours. Solid circles denote one set and open circles the other set of spots.

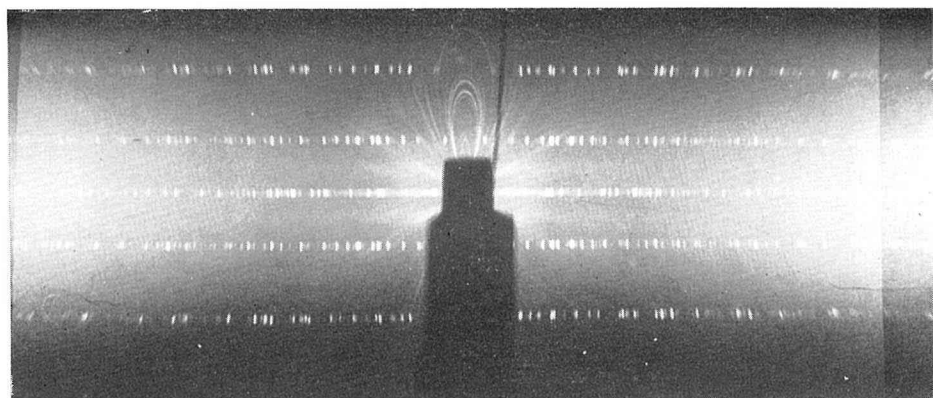


Fig. 11. (A) Rotation photograph of the hornblende (tschermakite) about c-axis.

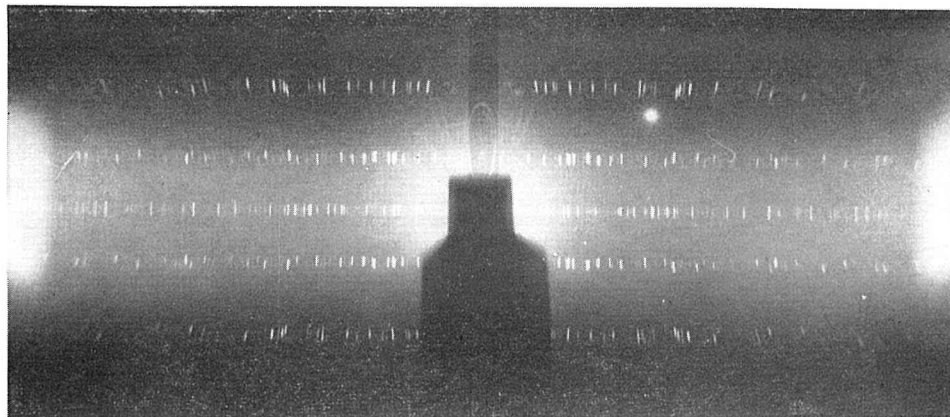


Fig. 11. (B) Rotation photograph of the hornblende about c-axis, heated at 950°C for 3 hours.

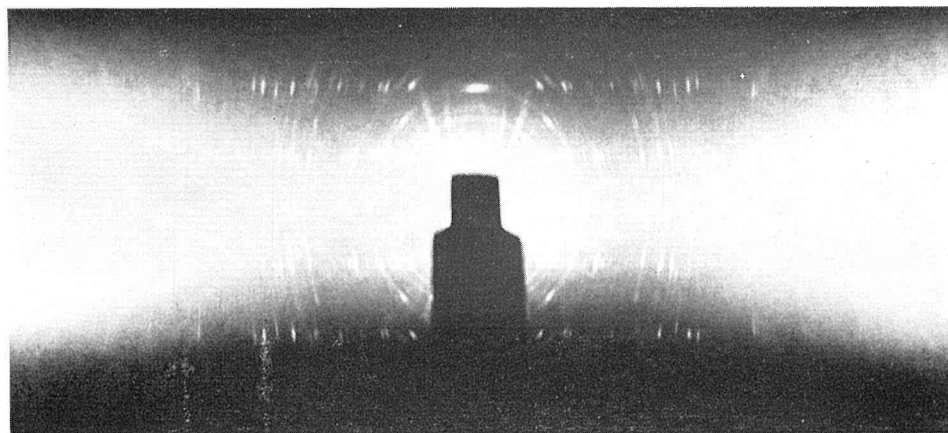


Fig. 11. (C) Rotation photograph of the hornblende about c-axis, heated at 1020°C for 2 hours.

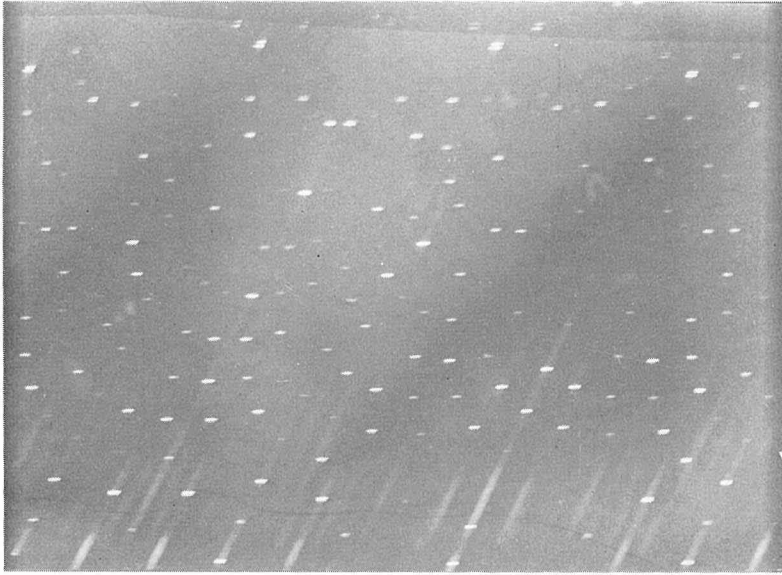


Fig. 12. (A) Weissenberg photograph of the hornblende about c-axis. (zero level)

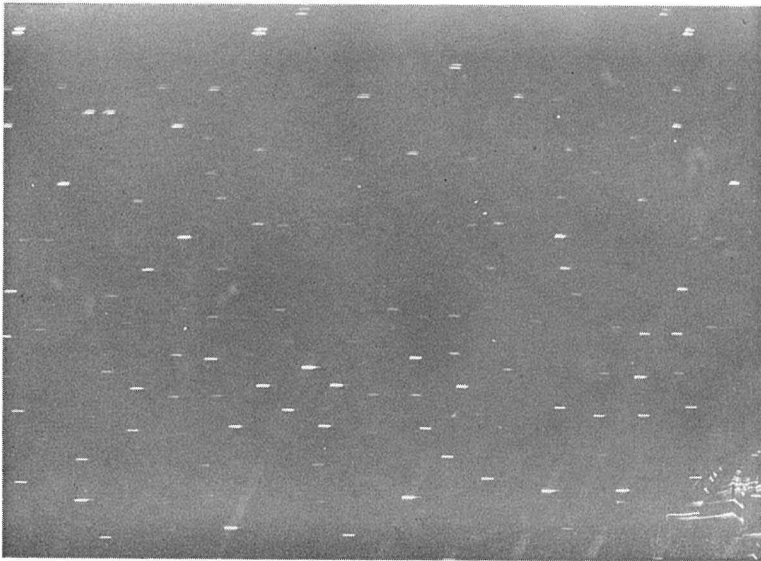


Fig. 12. (B) Weissenberg photograph of the hornblende about c-axis, heated at 950°C for 2 hours. (zero level)



Fig. 12. (C) Weissenberg photograph of the hornblende about c-axis, heated at 1020°C for 2 hours. (zero level)

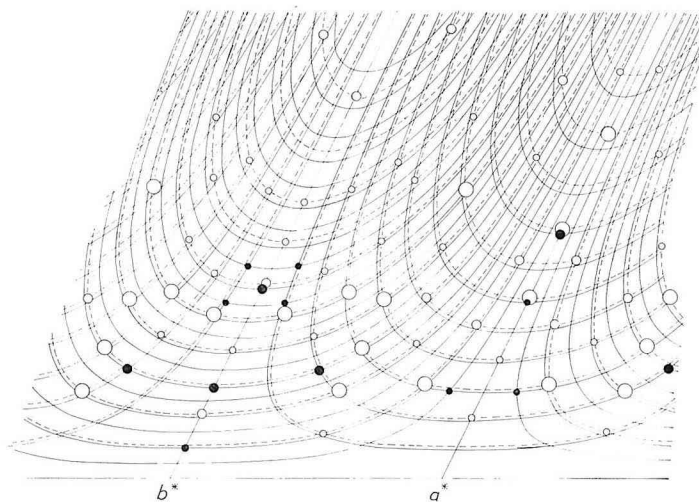


Fig. 12. (D) $hk0$ Weissenberg diagram of the hornblende heated at 1020°C for 2 hours. Solid circles denote one set and open circles the other set of spots.

Table 11. Unit cell dimensions of the amphiboles examined.

(A) tremolite from China

	natural specimen (space group I 2/m)	heated at 900°C for 2 hours	
		amphibole lattice (space group I 2/m)	pyroxene lattice (space group C 2/c)
a	9.97 Å	9.82 Å	9.57 Å
b	18.12	18.18	8.96
c	5.29	5.32	5.32
β	106°25'	105°22'	105°22'
a sin β	9.56 Å	9.56 Å	9.31 Å

(B) actinolite from Japan

	natural specimen (space group I 2/m)	heated at 900°C for 2 hours	
		amphibole lattice (space group I 2/m)	pyroxene lattice (space group C 2/c)
a	—	9.94 Å	9.77 Å
b	18.12 Å	18.08	8.93
c	5.30	5.30	5.30
β	—	108°18'	108°18'
a sin β	9.52 Å	9.44 Å	9.27 Å

(c) hornblende (tschermakite) from Parao Is.

	natural specimen (space group I 2/m)	heated at 1020°C for 2 hours	
		amphibole lattice (space group I 2/m)	pyroxene lattice (space group C 2/c)
a	10.01 Å	9.96 Å	9.56 Å
b	18.25	18.02	8.98
c	5.32	5.34	5.34
β	107°14'	99°42'	99°42'
a sin β	9.56 Å	9.81 Å	9.43 Å

In Table 11 are shown the cell dimensions of the natural tremolite, actinolite and hornblende (tschermakite), and of the phases produced by heating.

Consideration

The temperatures at which calciferous amphiboles, used in the present investigation, the phase changes and dehydration take place are diagrammatically illustrated in Fig. 13. The temperatures of the phase changes vary with their chemical composition; the more the Al substitutes for Si, the higher the temperatures of the phase changes are likely to become.

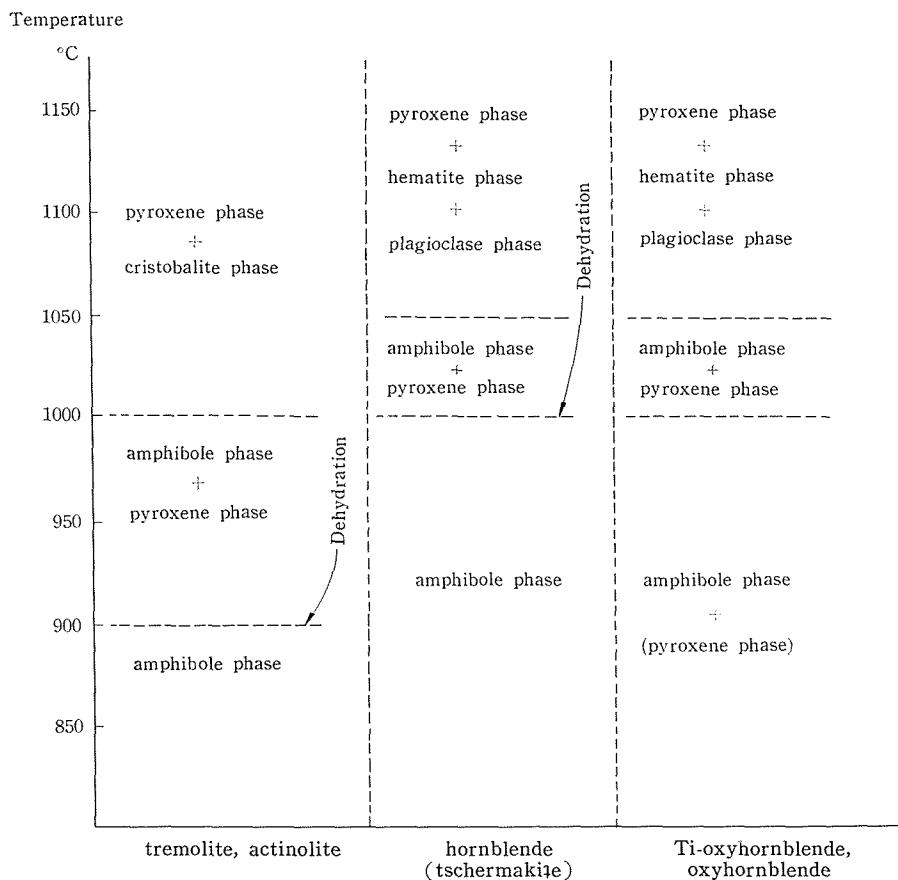
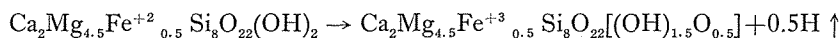
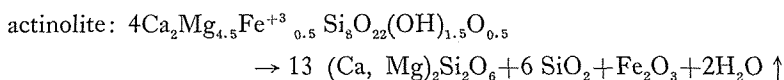
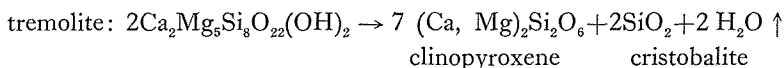


Fig. 13. The relationship between structural change and the temperature for amphiboles. Specimens were heated one by one for 2 hours at the temperature shown on the left side.

Actinolite changes in colour by heating at about 850°C from green to reddish brown, the lattice not being destroyed. The change may be brought about by an oxydation of ferrous irons to ferric state, electroneutrality being balanced by the simultaneous, partial loss of hydrogens from the hydroxyl ions. The chemical change may run as follows:

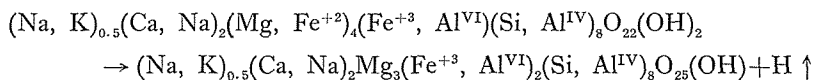


The phase change of tremolite and actinolite to clinopyroxene takes place at about 900°C and simultaneously dehydration may take place. Before the phase change is completed, there exist two phases, e.g. clinoamphibole and clinopyroxene phases, crystallographic axes being in common to both. At about 1000°C the product changes to a mixture composed of clinopyroxene and cristobalite. The change may run as follows:



Hornblende (tschermakite) changes in colour by heating at about 850°C from green to reddish brown, as in the case of actinolite, not being accompanied by the lattice destruction.

The chemical change may be the same as in the case of actinolite and may run as follows:



The phase change of hornblende to clinopyroxene takes place at about 1000°C. Before the phase change is completed, there exist the two phases mentioned above, and arranged in the same way. The dehydration of hornblende may take place at nearly the same temperature; although in the present investigation the temperature was somewhat higher than that of the phase change. The hornblende changes to a mixture of clinopyroxene, plagioclase and hematite at about 1050°C.

The clinopyroxene phase of Ti-oxyhornblende and oxyhornblende increase by heating at about 1000°C and change to a mixture of clinopyroxene, plagioclase and hematite at about 1050°C. Dehydration, however, does not take place anywhere. This may be due to the fact that in the clinoamphibole phase of these minerals a great part of the OH positions in the structure has already been occupied by O ions and a great part of the Fe^{+2} positions by Fe^{+3} ions.

In the present investigation the author observed that two types of phase changes of calciferous amphiboles to a mixture of clinopyroxene and cristobalite or a mixture of clinopyroxene, plagioclase and hematite, take place by heating. The one, through two steps; first, to an oxyhornblende which retains original structure, and secondly, to an oxyhornblende composed two phases, the crystallographic axes of which are common to both. The phase change of actinolite and hornblende progresses in the way mentioned above. The other, through one step, omitting the first step mentioned above. The phase change of tremolite goes on in this way.

The author is certain that natural oxyhornblende is a product of some calciferous amphiboles heated and oxydized in nature, and there may be two kinds, one which still retains amphibole phase, and the other which is composed of amphibole and pyroxene phases.

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References

- BARNES, v.E. (1930): Changes in hornblende at 800°C, *Am. Min.*, 15,393.
BELYANKIN, D.S. and E.V. DONSKAYA, (1939): Thermo-optical investigation of actinolite, *Bull. Acad. Sci. U.S.S.R., Ser. Geol.*, 1, 95, (M.A. 8-19)
KORZHINSKY, A.F. (1955): Thermal analysis of certain hornblende from eastern Siberia, *Acad. Sci. U.S.S.R.*, 266 (M.A. 13-396).
KOZU, S. and B. YOSHIKI, (1927): The dissociation temperature of brown hornblende and its rapid expansion at this temperature, *Sci. Rep. Tohoku Imp. Univ., Ser. 3*, 2, 107.
KOZU, S. and K. KANI, (1927): Note on the study of the transformation of common hornblende into basaltic hornblende 750°C, *Sci. Rep. Tohoku Impr. Univ., ser. 3*, 2, 143.
OVCHINNIKOV, L.N., A.S. SHUR, and EN.T. ELKINA, (1955): Thermo-analytical study of the amphiboles of certain skarn zones of Urals, *Acad. Sci. U.S.S.R.* 250 (M. A. 13-397).
POSNJAK, E. and N.L. BOWEN, (1931): The role of water in tremolite, *Am. Jour. Sci.*, 22, 203.
TOMITA, K. (1964): The existence of oxyhornblende built up from two different lattices, *Memo. Coll. Sci. Univ. Kyoto, Ser. B*, 30, 4.

- UEDA, T. and K. TOMITA, (1964): Titaniferous oxyhornblende built up from two different lattices, *Min. Jour.*, 4, 85.
- VERMAAS, F.H.S. (1952): The amphibole asbestos of South Africa, *Trans. Geol. Soc. South Africa*, 55, 1.
- WITTELS, M. (1952): Structural disintegration of some amphiboles, *Am. Min.*, 37, 28.
- WITTELS, M. (1951): Structural transformations in amphiboles, *Am. Min.*, 36, 751.