Clinopyroxene Intermediate between Jadeite and Aegirine from Suberi-dani, Tokushima Prefecture, Japan

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Introduction

We have a number of chemical analyses of jadeite and of aegirine in the literature, but our knowledge on the solid-solution relation between the two pyroxenes is still very incomplete. It is not known whether or not there exists a complete series of solid-solution between the two pyroxenes. No reliable analyses of intermediate members are known (Fig. 2).

MIYASHIRO and BANNO (1958) stated that probably there exists a continuous series of solid-solution between them, and the presence of aegirine molecule in solid-solution enlarges the stability range of jadeitic pyroxenes toward lower pressures. However, they did not have sufficient data for the statement.

The writer found a clinopyroxene intermediate in composition between jadeite and aegirine in a rock from the valley of Suberi-dani, Katuura-mati, Katuura-gun, Tokushima Prefecture, Japan (33°53' N, 134°28' E). In this paper, the mode of occurrence and mineralogy of the clinopyroxene will be described in detail.

This pyroxene is associated with lawsonite, and the host-rock has a rather unusual bulk chemical composition. The significance and origin of the rock will be discussed briefly.

Mode of occurrence and petrographic description of the host-rock in Suberi-dani.

The host-rock containing the clinopyroxene occurs in association with lawsonite-glaucophane schists in the Paleozoic terrain (early-middle Permian) to the north of the Kurosegawa (Tectonic) Zone (Fig. 1.).

The Tectonic Zone contains Silurian Sediments along with some protoclasic granodiorites, amphibolites and biotite schists. A narrow area of Mesozoic sediments and serpentines are intercalated between the Paleozoic terrain and Kurosegawa (Tectonic) Zone. All the boundaries between the Tectonic Zone, the Mesozoic sediments, and the Paleozoic terrain are faults, into which serpentines intruded.

The Sanbagawa-Mikabu Metamorphic Zone is situated about 8 km. to the north of the locality of the host-rock. Between the locality and the Metamorphic Zone are exposed Paleozoic formations made up of almost non-metamorphic sediments. A part of the Paleozoic is covered by Cretaceous formations.

The host-rock under consideration contains lawsonite, which is a typical mineral of glaucophanitic metamorphism, and moreover, it is associated with

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Fig. 1. Geological sketch map of the Suberi-dani district, Tokushima Prefecture.

1. Mesozoic sediments (Jurassic-Cretaceous)
 2. Paleozoic sediments (late Carboniferous-middle Permian)
 3. Serpentine
 4. Basic metamorphic rock complex belonging to the Sanbagawa-Mikabu Terrain
 5. Kurosegawa Tectonic Zone proper.
- × Locality of the rock described in this paper.

The faults are shown by full lines and the boundaries of rocks by dotted lines. This figure is based on the geological map of Kenzan surveyed by HIRAYAMA, YAMASITA, SUYARI and NAKAGAWA (1956) with slight modifications by the present writer.

The host-rock of the clinopyroxene is intimately associated with serpentines and lawsonite-glaucophane schists. The glaucophane schists contain lawsonite, glaucophane, muscovite, calcite, chlorite, albite and some dusty sphene along with relict augite. The glaucophane is usually light in color with $b=Y$.

glaucophane schists. Then, it is reasonable to consider that the origin of the host-rock is related in some way to the glaucophanitic metamorphism of the Sanbagawa-Mikabu Zone.

The metamorphic complex containing the host-rock now under consideration forms a narrow belt extending ENE to WSW over a distance of more than 4 km., and about 30 m. in width.

Serpentines occur on the south of this belt. The northern limit of the belt is in contact with a weakly schistose stratum* by a strike fault. It is not clear whether progressive mineralogical changes exist or not between this belt and the Paleozoic stratum on the north. This belt is too narrow to be shown in the map.

Table. 1. Optic properties of the glaucophanic amphiboles in lawsonite-glaucophane schists and in albite-quartz veinlets cutting through the schists

I. Amphibole of the schists (glaucophane).

$b=Y$; optic plane parallel to (010).
 $c \wedge Z = 3^\circ - 7^\circ$
 $2V(-) = 33^\circ$ for yellow light. $r > v$.
 $\alpha = 1.643$, $\beta = \text{about } 1.658$, $\gamma = 1.661$,
 $\gamma - \alpha = 0.018$.
Pleochroism: X=very pale yellow, Y=pale violet, Z=pale blue. $Z > Y = X$.

II. Amphibole of albite-quartz veinlets (magnesian riebeckite).

$b=Z$; optic plane normal to (010).
 $c \wedge Y = 37^\circ$ for yellow light (dispersion strong).
 $2V(-) = 42^\circ$ $v > r$.
 $\alpha = 1.651$, $\beta = 1.662$, $\gamma = 1.672$, $\gamma - \alpha = 0.021$.
Pleochroism: Parallel to b =violet.
Practically parallel to c =blue.
Normal to the above two=
very pale yellow.
Absorption: violet > blue > very pale yellow.

* According to YAMASHITA (oral communication), the northern half of the stratum is composed of weakly schistose pelitic rocks and the southern half is composed of sandstones which are thought to be of Upper Permian and apparently free from metamorphism.

However, the writer rarely noticed glaucophane-like amphibole with darker color and $b=Z$ in some albite-quartz veinlets cutting through the schists. It may be magnesioriebeckite. Two of the principal axes of the absorption ellipsoid of this amphibole show a large departure from the axis of the indicatrix (Table 1). The glaucophane schists often contain calcite pools.

The host-rock is massive, compact and dirty green in color. Under the microscope, larger prismatic crystals of clinopyroxene are embedded in the matrix composed of idiomorphic lawsonites. No other minerals were noticed. The clinopyroxene is distinctly pleochroic. In some cases, crystals of the clinopyroxene are in radial arrangement. Albite veinlets form a network within the rock mass.

Mineralogical data on the clinopyroxene

The clinopyroxene was separated from the host-rock by means of Clerici solution. The sample obtained for chemical analysis is grass-green in color. It was analysed by Hiroshi HARAMURA. The analytical data and the calculated atomic ratios are given in Table 2.

Table. 2. Chemical analysis, atomic ratios and molecular composition of clinopyroxene from Suberi-dani.

Wt. %		Atomic ratios (O=6)		Molecular composition	
SiO ₂	52.41	Si	1.96	} 2.00	Aegirine 0.43
Al ₂ O ₃	9.47	Al	0.42		Jadeite 0.36
TiO ₂	1.56	Ti	0.04	} 1.06	Diopside 0.17
Fe ₂ O ₃	15.54	Fe ⁺³	0.44		Hedenbergite 0.04
FeO	1.43	Fe ⁺²	0.04		
MnO	0.12	Mn	0.00		
MgO	2.96	Mg	0.16	} 0.92	1.00
CaO	5.06	Ca	0.20		
Na ₂ O	9.97	Na	0.72		
K ₂ O	0.10	K	0.00		
H ₂ O ⁻	0.08				
H ₂ O ⁺	1.10				
P ₂ O ₅	0.06				
Total	99.86				

The table shows also that the clinopyroxene is composed of 43% aegirine, 36% jadeite, 17% diopside and 4% hedenbergite, according to the following *method of calculation*. Atomic ratios for O=6 are calculated from chemical analysis. When the value for Si is smaller than 2, the deficiency is filled up with the equivalent value of Al. First, jadeite and aegirine molecules are calculated by allotting Na to Al and Fe^{'''} respectively. When Fe^{'''}+Al is in excess of Na, the Na is allotted to Fe^{'''} and Al in the ratio of Fe^{'''}/Al present, and the excess of Fe^{'''}+Al is disregarded. When Na is in excess of Fe^{'''}+Al, the excess is disregarded. Then, diopside and hedenbergite molecules are calculated by allotting Ca to Mg and Fe^{''} respectively. When Mg+Fe^{''} is in excess of Ca, the Ca is allotted to Mg and Fe^{''} in the ratio of Mg/Fe^{''} present, and the excess is disregarded. When Ca is in excess of Mg+Fe^{''}, the excess is disregarded. At last, the molecular percentages of jadeite, aegirine and diopside + hedenbergite are calculated so that the sum is equal to 100.

Table. 3. X-ray diffraction data of the clinopyroxene from Suberi-dani (CuK α radiation for the upper 8 peaks and CuK α_1 radiation for the rest).

$C 2/c (C^{6_{2h}})$ hkl	d (Å)	I
110	6.32	3
020	4.37	3
220	3.164	2
221	2.964	8
310}	2.880	10
311}		
202}	2.668	3
131}		
112}		
221	2.460	3
400	2.283	1
040	2.190	2
331	2.108	2
041	2.007	1
402	2.016	1
150	1.726	1
421	1.722	1
531	1.601	1

Note: Quartz powder was used as an internal standard.

of aegirine. A similar relation holds as regards the composite peak of (310) and (311).

The associated lawsonite has the following optical properties: elongation (−), colorless, $\alpha=1.664\pm0.002$, $\gamma=1.686\pm0.002$, $\gamma-\alpha=0.022$, $2V(+)=80^\circ-86^\circ$. These optical data are in agreement with those of lawsonite in the literature. The identification was confirmed by the X-ray powder method.

Comparison with the existing data of alkalic clinopyroxenes

About 75 chemical analyses of clinopyroxene rich in Na, Al, and/or Fe were collected from the literature. Their molecular compositions were calculated according to the method outlined before. The result of the calculation is plotted in Figs. 2 and 3.

It is clear from the figures that there is a continuous series of solid-solution between diopside (hedenbergite) and jadeite and also between diopside (hedenbergite) and aegirine. The existence of the continuous series between diopside and aegirine was experimentally proved by OSTROVSKY (1946) and YAGI (1958). There exist, how-

Optical properties of the clinopyroxene are as follows. Pleochroism: X=pale green, Y=yellow, Z=pale brown; $b=Y$, optical plane//010, $c\wedge X=5^\circ$. The elongation is negative in slender prisms in thin sections. $2V(-)=74^\circ-80^\circ$, dispersion strong $r>v$. $X>Y>Z$. $\alpha=1.726\pm0.003$, $\gamma=1.750\pm0.003$, $\gamma-\alpha=0.024\pm0.003$, (all for yellow light).

The X-ray diffraction data of the clinopyroxene are shown in table 3. Peaks were provisionally indexed on the basis of a monoclinic unit cell with the following dimensions: $a_0=9.56$ Å, $b_0=8.77$ Å, $c_0=5.24$ Å, $\beta=72^\circ13'$. Here, the space group is assumed to be $C2/c(C^{6_{2h}})$. Peaks with 2θ larger than 60° were not observed. The peak of (221) of this clinopyroxene is situated halfway between the positions of the corresponding peak of jadeite and

Fig. 2. The molecular compositions of alkalic clinopyroxenes

Jad: $NaAlSi_2O_6$ Aeg: $NaFe^{III}Si_2O_6$

Diop. + Hed: $CaMgSi_2O_6 + CaFe^{II}Si_2O_6$

The symbols represent a classification of the host-rocks.

● Igneous rocks ○ Metamorphic rocks
⊙ Clinopyroxene described in this paper

ever, a large apparently vacant field between jadeite and aegirine. The clinopyroxene described in this paper is unique in composition, falling in the middle of the vacant field.

It follows that the vacant field is not real and jadeite and aegirine may form a continuous series of solid-solution.

The range of physical conditions under which jadeite is formed is very limited. On the other hand, the chemical conditions under which aegirine is formed are realized rather rarely on the earth. Then, probably the conditions for the formation of clinopyroxenes intermediate between jadeite and aegirine are fulfilled only rarely. Probably this is the reason why intermediate clinopyroxenes were not so far found.

Fig. 3. Composition field of alkalic clinopyroxenes. The encircled dot shows the clinopyroxene described in this paper.

Origin of the rock

Under some conditions, aegirine would react with lawsonite to form albite, calcite and hematite as shown by the following equation;

The mineral assemblage of the left-hand side would occur extremely rarely if ever while that of the right-hand side is rather common in metamorphic rocks. Then, this reaction is considered to be displaced toward the right under ordinary conditions of metamorphism.

The pyroxene under investigation, however, is not aegirine end member, but contains 43 mol. per cents of aegirine and 35 mol. per cents of jadeite. Then, the above consideration must be somewhat altered. If the pyroxene associated with lawsonite is not aegirine end member but a solid solution containing other molecules (e. g. jadeite molecule), the stability of the clinopyroxene-lawsonite assemblage will increase.

Moreover, the above reaction depends on the partial pressure of H_2O and CO_2 . Lower CO_2 pressure and higher H_2O pressure would have contributed to the increase of the stability of the left-hand side of the equation.

The rock under consideration is composed exclusively of lawsonite and aegirine-jadeite solid solution. The lawsonite is chemically equivalent to anorthite plus water, while, jadeite has a composition of albite plus nepheline. Accordingly, the bulk chemical composition of the rock must be undersaturated with silica. The bulk composition of the rock is calculated from the composition of the clinopyroxene and the estimated mode, neglecting minor components (Table 4).

The C. I. P. W. norm was calculated from the bulk composition, with the following result: ab 44.4, ne 3.3, an 30.2, hm 6.5, di 10.0, wo 5.6.

Table 4. Calculation of the bulk composition of the rock from the mode.

	Clinopyroxene	Lawsonite	Total (wt. %)
SiO ₂	30.21	17.19	47.40
Al ₂ O ₃	5.46	14.62	20.08
Fe ₂ O ₃	8.95		8.95
CaO	2.92	8.03	10.95
MgO	1.71		1.71
Na ₂ O	5.75		5.75
H ₂ O		5.16	5.16
Total	55.00	45.00	100.00

Thus, the chemical composition of the rock is unusual and may resemble those of some alkalic rocks. Since rocks with such unusual chemical composition are very rare in the earth's crust, one may consider that probably the rock was formed not by isochemical metamorphism, but by material migration during metamorphism.

Perhaps, such unique chemical composition may have originated from absorption of silica by the surrounding serpentine. The stability of jadeite increases under environments undersaturated with silica. Thus, such environment poor in silica may have promoted the formation of the mineral assemblage of this rock.

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徳島県迂り谷産の Jadeite と Aegirine の中間の成分をもつ輝石

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(摘 要)

Jadeite-diopside(hedenbergite) 間および aegirine-diopside(hedenbergite) 間には、それぞれ連続固溶体系列があることが知られている。ところが、jadeite と aegirine との中間の成分を持つ輝石は、今までに報告されていない。

この論文では、そのような輝石が徳島県勝浦町迂り谷で、いわゆる黒瀬川構造帯北縁の弱変成結晶片岩に伴つて見出されたことを報告し、jadeite と aegirine との間にも、連続固溶体系列があることを推定した。

その輝石は、ここでは clinopyroxene-lawsonite rock として、蛇紋岩に伴つて産出している。その岩石の成因を簡単に論じた。