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WAIRAKITE-ANALCIME SOLID SOLUTION AS AN INDICATOR OF WATER PRESSURES IN LOW-GRADE METAMORPHISM

YOTARO SEKI*

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Preface

COOMBS (1960, 1961, 1970), LIOU (1970) and the present writer (SEKI, 1969 b, 1971) noted that wairakite is one of the most important minerals in rocks metamorphosed at relatively low pressures (probably lower than 2,000 bars) and at temperatures characteristic of the highest part of the zeolite facies and the pumpellyite-prehnite facies.

Complete solid solutions from $NaAlSi_2O_6 \cdot H_2O$ to $CaAl_2Si_4O_{12} \cdot 2H_2O$ stably associated with some of phases epidote, prehnite, laumontite, natrolite, chlorite, phengite, calcite, and quartz are found as devitrification products, as alteration products of plagioclase and as amygdaloidal minerals in low-grade metamorphosed Triassic volcanic rocks of Vancouver Island, Canada (SURDAM, 1966, 1967).

SEKI and ONUKI (1971) said that wairakite-analcime solid solutions must be stably formed at relatively higher pressure conditions than those which prevailed in active geothermal areas where almost pure wairakites are stably associated with analcime-quartz and/or albite.

Wairakite-analcime solid solution minerals have been found in weakly metamorphosed Miocene submarine volcanic rocks (the so-called Green Tuffs) of the highest-grade part of the laumontite-mixed layer chlorite zone and the lower-grade part of the pumpellyite-prehnite-chlorite zone of the south Tanzawa Mountains, central Japan (SEKI, HARAMURA, ONUKI, OKUMURA and HIRAGA, 1966; SEKI, 1969 b; SEKI, OKI, MATSUDA, MIKAMI and OKUMURA, 1969). Mineralogical characters of three of these wairakite-analcime series minerals have been described in some detail (SEKI and OKI, 1969; SEKI, 1971).

Recently the finding of wairakite-analcime solid solutions have also been reported from other districts of the same Miocene volcanic geosynclinal area as follows:

Yugami (SEKI, 1966)
Hanawa (SEKI, TAKEYASU, ONUKI and NAKAJIMA, 1968)
Nakatsu (SHIMAZU, TABUCHI and SUZUKI, 1968)
Motojuku (Geol. Survey of Japan, 1969)
Kawaji (SEKI and ONUKI, 1971)

Wairakites from Hanawa and Kawaji districts are very poor in sodium and are stably associated with epidote, albite, sericite, chlorite and quartz.

In Yugami, Nakatsu and Motojuku districts, minerals ranging widely in chemical composition from wairakite to analcime are found in association with some of the following minerals: epidote, prehnite, pumpellyite, chlorite, albite, laumontite and quartz.

In this paper the distribution and some mineralogical characters of wairakite-analcime series minerals found in the north Tanzawa Mountains will be described in some detail. By the mineralogical comparison of these minerals to those found in Yugami, south Tanzawa and Fujikawa districts, wairakite-analcime series as an indicator of P_{H_2O} in low-grade metamorphism will also be discussed (Figure 1).

Wairakite-analcime solid solutions in north Tanzawa Mountains

Geology and metamorphism of the northern side

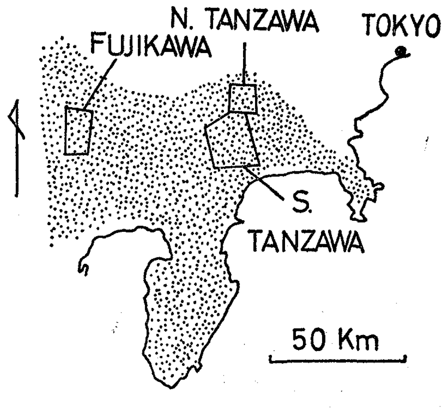

Fig. 1. Localities of north Tanzawa, south Tanzawa and Fujikawa districts in Miocene submarine volcanic geosynclinal area (dotted) of central Japan.

* Department of Foundation Engineering, Saitama University, Urawa, Japan.

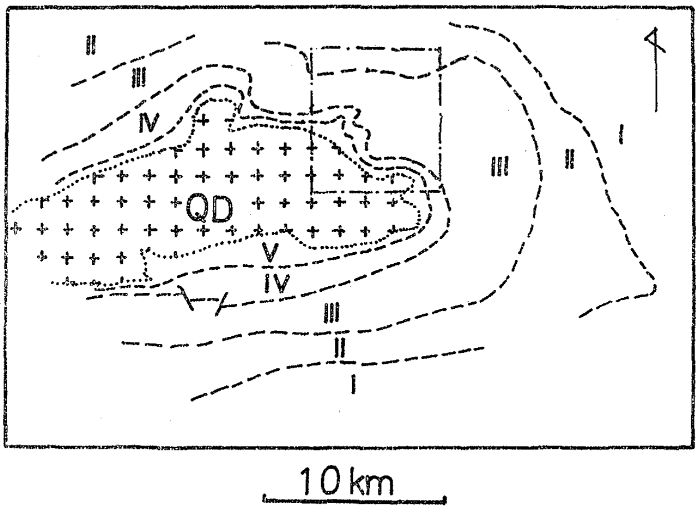

Fig. 2. Metamorphic zones developed surrounding a quartz diorite mass in the Tanzawa Mountains, central Japan.
QD : Quartz diorite mass
Ⅴ : Amphibolite or hornblende hornfels zone
Ⅳ : Actinolite greenschist or actinolite hornfels zone
Ⅲ : Pumpellyite-prehnite zone
Ⅱ : Laumontite zone
Ⅰ : Stilbite zone
The area marked by chain is described in this paper.

of the Tanzawa Mountains have been described in detail in other papers (SEKI, 1969 a: SEKI, OKI, ONUKI and ODAKA, 1971). The Miocene formation of this district is composed chiefly of a thick pile of basaltic, andesitic and dacitic submarine lavas and pyroclastics intercalated with small amounts of mudstone, sandstone and conglomerate.

These Miocene rocks suffered contact metamorphism related to the intrusion of a big mass of quartz diorite (Figure 2). The metamorphic aureole can be divided into the following four mineralogic zones: hornblende hornfels zone; actinolite hornfels zone; pumpellyite-prehnite zone; and laumontite zone (Figure 3). Sample localities on which metamorphic zone boundaries were based are not shown in this illustration.

In regard to the grade of metamorphism, these zones can be correlated to Zone V, Zone IV, Zone III and Zone II of the southern side of the same mass of quartz diorite in the south Tanzawa Mountains (SEKI, OKI, MATSUDA, MIKAMI and OKUMURA, 1969) (Figures 2 and 3).

Wairakite-analcime series minerals were found in the area of the actinolite hornfels zone, the pumpellyite-prehnite zone and the laumontite zone.

In the actinolite hornfels zone, wairakite-analcime solid solutions occur only in the following vein mineral assemblages: wairakite · analcime s.s. -quartz and wairakite · analcime s. s. -laumontite-quartz.

In the pumpellyite-prehnite zone and the laumontite zone, wairakite-analcime solid solutions were formed by the replacement of plagioclase phenocrysts and groundmass or glassy parts of original volcanic rocks. The association of metamorphic minerals of these wairakite · analcime-bearing rocks are summarized as shown in Table 1. Besides these occurrences, veins of the following mineral assemblages were also found in the pumpellyite-prehnite zone: wairakite · analcime s. s. -quartz, wairakite · analcime s. s. -natrolite, wairakite · analcime s. s. -laumontite-quartz, wairakite · analcime s. s. -stilbite-quartz.

Wairakite-analcime series minerals in the pumpellyite-prehnite zone and the laumontite zone must have been stably formed under the physical conditions of the pumpellyite-prehnite facies and the laumontite subfacies of the zeolite facies. Wairakite-analcime series minerals found as vein-forming minerals within rocks of the actinolite-hornfels zone may have been formed in the stage of retrogressive decrease of temperature after the metamorphism of the actinolite hornfels or greenschist facies.

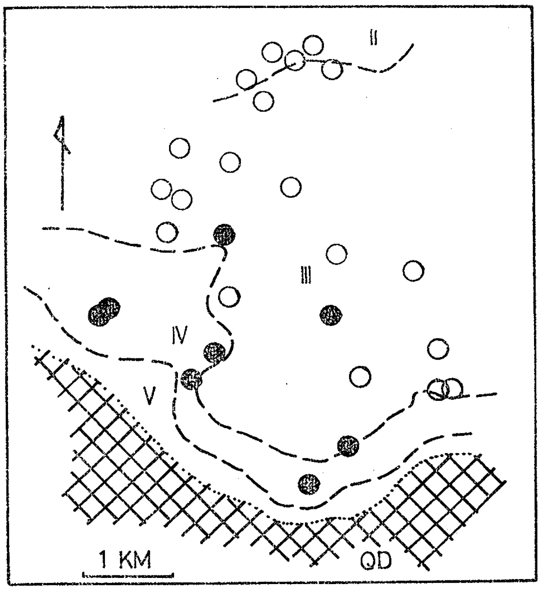

Fig. 3. Metamorphic zoning and localities from which wairakite-analcime solid solutions are found.

Solid circle : Wairakite-analcime series minerals occur in zeolite veins developed along joints and sheared surfaces.

Open circle : Wairakite-analcime series minerals occur as metamorphic minerals in rock themselves.

QD : Quartz diorite mass

Ⅴ : Hornblende hornfels zone

Ⅳ : Actinolite hornfels zone

Ⅲ : Pumpellyite prehnite zone

Ⅱ : Laumontite zone

Geologic basis is after SEKI, OKI, ONUKI and ODAKA (1971).

X-ray properties of wairakite-analcime solid solutions

The most characteristic differences between X-ray powder patterns of wairakite and those of analcime are the clear separation of 400 reflection from that of 004 reflection and the sharp appearance of the 200 reflection in wairakite (SEKI, 1968). In this paper the difference of $2\theta_{004}$ from $2\theta_{400}$ (in degree CuK α_1) and the intensity ratio of $I_{200}/(I_{200}+I_{211})$ will be represented simply by $\varDelta\ 2\theta$ and I ratio respectively.

The maximum $\varDelta\ 2\theta$ value caused by the smaller value of c dimension than a dimension in natural wairakite is 0.19. If the difference of c dimension from a dimension is very small, $\varDelta\ 2\theta$ can hardly be measured. Practically $\varDelta\ 2\theta$ less than 0.05 can not be detected precisely. It is expected that $\varDelta\ 2\theta$ larger than 0.05 can usually be measured with the error of ±0.01 or ±0.02.

I ratio may vary by different mounting methods to give different degrees of sample orientation. However, as observed by the present writer, the I ratio does not change significantly if the same diffractometer is used employing the same conditions (in this study the present writer used the diffractometer of TOSHIBA ADG-101 type, copper radiation at 15 mA, 30 kV, time constant 4, multiplier 1 and scale factor 2).

Figure 4 shows the relation of $\varDelta\ 2\theta$ and I ratio to Na/(Na+Ca) ratio of wairakite-analcime solid solution minerals. From this Figure it can be said that $\varDelta\ 2\theta$ and I ratio generally decrease with increasing analcime component.

SURDAM (1967) has also found in his microprobe study of wairakite from Vancouver Island that $\varDelta\ 2\theta$ regularly increases with increasing Ca as follows:

Ca (wt %)	2θ
2.0	0.03
4.0	0.05
6.0	0.13
8.0	0.17
8.5	0.18

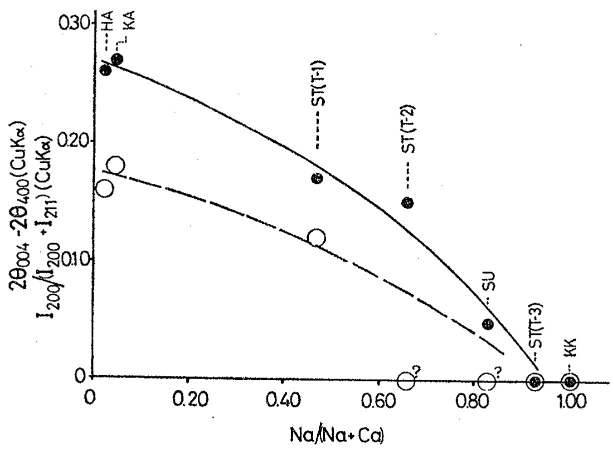

Fig. 4. Relations between Na/(Na+Ca) and $2\theta_{004}-2\theta_{400}$ (open circles) and Na/(Na+Ca) and $I_{200}/(I_{200}+I_{211})$ (closed circles) of wairakite-analcime solid solutions.

HA : Wairakite from Hanawa district, Japan (Seki, Takeyasu, Onuki and Nakajima, 1968).

KA : Wairakite from Katayama geothermal area, Japan (Seki, Onuki, Okumura and Takashima, 1969).

ST : Wairakite-analcime series minerals from south Tanzawa Mountains, Japan (Seki and Oki, 1969).

SU : "Calcium analcime" from Sugashima, Japan (Harada and Sakurai, 1967).

KK : Analcime from Kasukabe, Japan (Hashimoto, 1968).

Figure 5 shows the relation between $\Delta\ 2\theta$ and I ratio of wairakite-analcime solid solution minerals separated from south Tanzawa, north Tanzawa, Hanawa, Onikobe, Nakatsu, Motojuku and Fujikawa districts. It is clear from this Figure (see also Fig. 4) that in the wairakite-analcime series, $\Delta\ 2\theta$ generally increases with increasing I ratio. The reasons why the relation between 2θ and I ratio of this series does not show any linear distribution but has moderately wide distribution are supposed to be (1) the previously noted error of measurements of 2θ and I ratio and (2) variations of physical characters in an end member analcime having wide chemical range from $NaAlSi_{1.5}O_5 \cdot mH_2O$ to $NaAlSi_3O_8 \cdot nH_2O$ (SEKI and OKI, 1969).

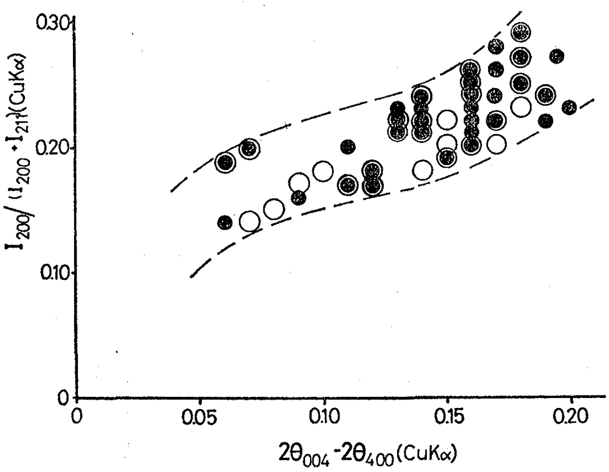

Fig. 5. Relation between $2\theta_{004}-2\theta_{400}$ and $I_{200}/(I_{200}+I_{211})$ of wairakite-analcime solid solution minerals.
Open circles: Wairakite-analcime from north Tanzawa, Japan.
Closed circles: Wairakite-analcime from other districts of Japan.
(Wairakite-analcime series minerals of $2\theta_{004}-2\theta_{400}<0.05$ are not shown in this diagram)

Wairakite-analcime solid solutions and their mineral associations

From Table 1 and Figure 6, it can be said that in north Tanzawa district wairakite-analcime solid solutions associated with some of other minerals such as epidote, pumpellyite, prehnite, albite, chlorite, calcite, laumontite and quartz have a wide range of chemical composition from almost pure wairakite to almost pure analcime.

In rocks of the pumpellyite-prehnite facies and the laumontite facies, the intensity of recrystallization is generally not great and relics of original minerals are preserved. Neither foliation nor schistosity was developed. In such low-grade metamorphosed rocks it is very hard to definitely determine the assemblages of *stably* associated metamorphic minerals.

However, in regard to the mineral assemblages of wairakite-analcime solid solutions in the Tanzawa Mountains, at least the following two distinct characters have been observed.

(1) None or only very small amounts of sodic plagioclase were formed in most rocks where analcime-rich minerals ($\Delta\ 2\theta<0.05$ and I ratio <0.10) are found. Especially in the laumontite

Table 1. Associated minerals, $2\theta_{004}-2\theta_{400}$, and $I_{200}/(I_{200}+I_{211})$ of wairakite-analcime solid solutions in the laumontite zone and the pumpellyite-prehnite zone of north Tanzawa district, central Japan.

	Laumontite	Albite	Chlorite-clay	Epidote	Pumpellyite	Prehnite	Quartz	Calcite	$2\theta_{004}-2\theta_{400}$	$I_{200}/(I_{200}+I_{211})$
Laumontite zone			+				+	+	0.08−0.17	0.15−0.20
	+		+				+		<0.05−0.15	0.00−0.20
		+	+				+	+	0.15−0.19	0.22−0.24
	+	+	+			+	+		0.10−0.13	0.18−0.22
Pumpellyite-prehnite zone			+				+	+	0.18	0.23
		+	+	+				+	0.15−0.17	0.19−0.22
		+	+			+	+	+	0.11−0.14	0.17−0.18
		+	+		+		+	+	<0.05−0.09	0.00−0.17
			+				+		<0.05−0.14	0.00−0.18
		+	+	+			+		0.16−0.18	0.20−0.28
		+	+	+		+	+	+	0.14	0.22−0.24
		+	+				+		0.17−0.19	0.22−0.24
		+	+	+		+	+		0.14	0.22−0.24
		+	+		+	+	+		<0.05−0.12	0.00−0.17
		+	+	+	+		+		0.17	0.22−0.25
		+	+	+	+	+	+		0.14	0.23−0.24
	+	+	+			+	+	+	0.14	0.24

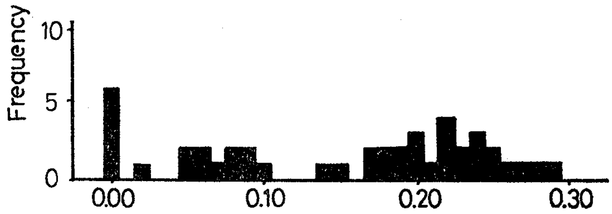

Fig. 6. Frequency distribution of $I_{200}/(I_{200}+I_{211})$ of wairakite-analcime solid solutions from north Tanzawa, central Japan.

facies area, rocks of analcime-clay-calcite-quartz and laumontite-analcime-clay-quartz assemblages do not have albite as a stable phase.

(2) Wairakite-analcime solid solutions associated with epidote (without prehnite) show $\Delta 2\theta$ higher than 0.15 and I ratio higher than 0.19. Wairakite-analcime series minerals associated with prehnite (without epidote), however, are lower than 0.13 in $\Delta 2\theta$ and are lower than 0.22 in I ratio. $\Delta 2\theta$ and I ratios of wairakite-analcime solid solutions associated with both of epidote and prehnite are 0.14 and 0.22–0.24 respectively (Figure 7). $\Delta 2\theta=0.14$ and I ratio$=0.22$ roughly correspond to the chemical composition of Na/(Na+Ca)=0.30 (Figure 4). Thus the mineral association of wairakite-analcime solid solutions with epidote, prehnite and pumpellyite in north Tanzawa district can be represented by (Al+Fe''')-Ca-Na diagram as shown in Figure 8.

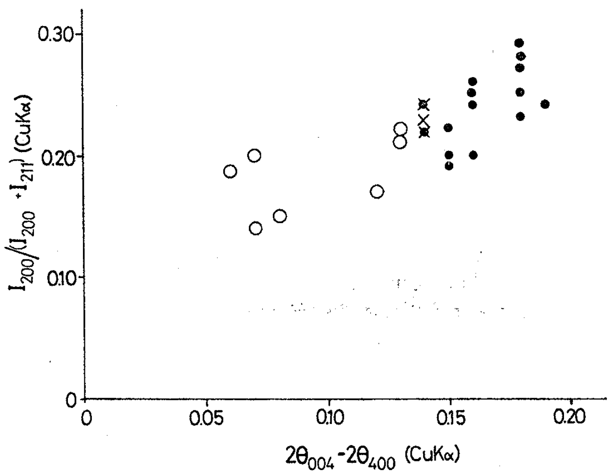

Fig. 7. Relation between $2\theta_{004}-2\theta_{400}$ and $I_{200}/(I_{200}+I_{211})$ of wairakite-analcime series minerals in epidote-bearing rocks (closed circles), prehnite-bearing rocks (open circles) and epidote-prehnite-bearing rocks (crosses) in north Tanzawa Mountains, central Japan. Wairakite-analcime series minerals of $2\theta_{004}-2\theta_{400}<0.05$ are not shown in this diagram.

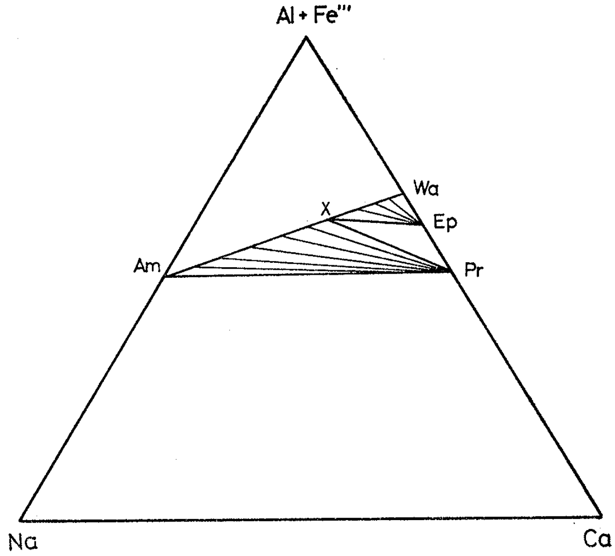

Fig. 8. (Al+Fe''')-Na-Ca diagram showing the associations of wairakite-analcime solid solution, epidote and prehnite in north Tanzawa Mountains, central Japan.
Wa : Wairakite
Am : Analcime
Ep : Epidote
Pr : Prehnite
X : Wairakite-analcime solid solution stably associated with epidote and prehnite

Wairakite-analcime solid solutions in the Fujikawa district

Wairakite-analcime series minerals occur in many weakly metamorphosed Miocene submarine volcanic rocks of the Fujikawa district, central Japan (Figure 1). The geology of district has been studies in detail by MATSUDA (1958, 1961). The present writer has recently also collected many samples from this district.

On the basis of detailed study of rock-specimens and thin sections, this district can be roughly divided into three zones representing a progressive metamorphism: (I) the mordenite zone, (II) laumontite zone and (III) pumpellyite-prehnite zone (Figure 9). Albite is stable in the last two zones. The occurrence of epidote is confined to rocks of Zone III and eastern half of Zone II. Narrow contact aureoles (less than 650 meters in map width) were developed along the diorite and diorite-porphyrite margins. The aureoles consist of hornblende hornfels, biotite hornfels and garnet skarn. The intrusive masses and their associated contact aureoles are confined within the pumpellyite-prehnite zone. Probably the thermal structure to make the zonal arrangement of the mordenite zone, laumontite zone and pumpellyite-prehnite zone in this district must have been determined mostly by the intrusion of diorite and diorite porphyrite.

Figure 9 also shows the distribution of wairakite-analcime solid solutions having larger values of $\Delta 2\theta$ than 0.05. Minerals very close to analcime end-member having smaller value of $\Delta 2\theta$ than 0.05 are distributed virtually over the entire district. This means that wairakite-analcime solid solutions in the lower grade part of the laumontite zone and in the mordenite zone have always smaller value of $\Delta 2\theta$ than 0.05 and are probably very rich in analcime composition.

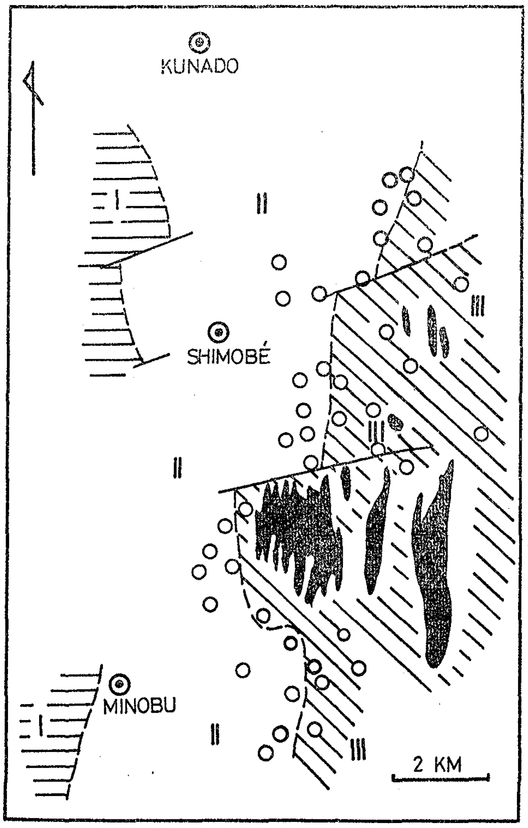

Fig. 9. Metamorphic zoning and distribution of wairakite-analcime solid solutions in low-grade metamorphosed Miocene submarine volcanic area of the Fujikawa district, central Japan.
I : Mordenite zone
II : Laumontite zone
III : Pumpellyite-prehnite zone
Circles indicate the localities from which wairakite-analcime solid solutions were found.
(Wairakite-analcime series minerals of $2\theta_{004}-2\theta_{400}<0.05$ are not shown in this map).
Black areas show quartz diorite and diorite-porphyrite intrusives.
Geologic basis is after MATSUDA (1958).

Figure 10 indicates the relation between $\Delta 2\theta$ and I ratio of wairakite-analcime solid solution minerals separated from metamorphic rocks of the Fujikawa district. As in the case of north Tanzawa district, it is also clear from Figure 10 that wairakite-analcime solid solutions associated with prehnite are different from those associated with epidote in regard to the relation between $\Delta 2\theta$ and I ratio. $\Delta 2\theta$ and I ratio of a wairakite-analcime series mineral stably associated with both of epidote and prehnite (and pumpellyite, albite, chlorite and quartz) are 0.16 and 0.23 respectively. Probably the point X in Figure 8 shifts slightly toward Wa composition in the case of the Fujikawa district.

Wairakite-analcime solid solution as an indicator of water pressure conditions of low-grade metamorphism

SEKI and OKI (1969) have described a wairakite-

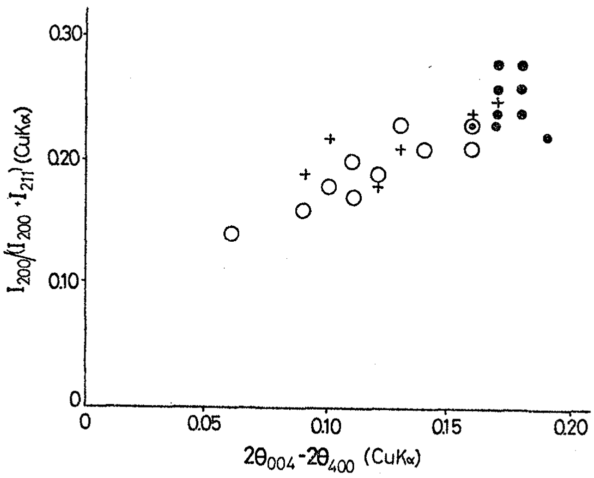

Fig. 10. Relation between $2\theta_{004}-2\theta_{400}$ and $I_{200}/(I_{200}+I_{211})$ of wairakite-analcime solid solutions in Fujikawa district, central Japan.
Closed circles : Wairakite-analcime in epidote-bearing rocks.
Open circles : Wairakite-analcime in prehnite-bearing rocks.
Circle with a dot : Wairakite-analcime in epidote-prehnite-bearing rock.
Crosses : Wairakite-analcime series minerals in rocks with neither epidote nor prehnite.
Wairakite-analcime solid solutions of $2\theta_{004}-2\theta_{400}<0.05$ are not shown in this diagram.

analcime series mineral (T-1) stably associated with prehnite and epidote from a weakly metamorphosed Miocene volcanic rock of south Tanzawa Mountains. $\Delta\ 2\theta$ and I ratio of this mineral are 0.12 and 0.17 respectively.

Seki (1966) also described a wairakite-analcime series mineral associated with prehnite and epidote in Miocene volcanic rock of the Yugami district $\Delta\ 2\theta$ and I ratio of this mineral are 0.20 and 0.28 respectively.

It has been shown in this paper that $\Delta\ 2\theta$ and I ratio decrease with decreasing wairakite composition. If this is true, point X in Figure 8 must shift toward Wa with the following order: south Tanzawa→ north Tanzawa→ Fujikawa→ Yugami (X→X′ in Figure 11).

When X and X′ in Figure 11 represent the chemical compositions of wairakite-analcime series to be

$$CaAl_2Si_4O_{12}\cdot 2H_2O+NaAlAi_2O_6\cdot H_2O$$

and

$$2CaAl_2Si_4O_{12}\cdot 2H_2O+NaAlSi_2O_6\cdot H_2O$$

respectively, the following chemical equation is obtained

3 Epidote + Prehnite
$Ca_2Al_3Si_3O_{12}(OH)$ $Ca_2Al_2Si_3O_{10}(OH)_2$

+ 2 Wairakite-analcime ss
$CaAl_2Si_4O_{12}\cdot 2H_2O+NaAlSi_2O_6\cdot H_2O$

+5 Quartz + 4 Water ⇄ Epidote
SiO_2 H_2O $Ca_2Al_3Si_3O_{12}(OH)$

+ 2 Prehnite
$Ca_2Al_2Si_3O_{10}(OH)_2$

+ 2 Wairakite-analcime ss
$2CaAl_2Si_4O_{12}\cdot 2H_2O+NaAlSi_2O_6\cdot H_2O$

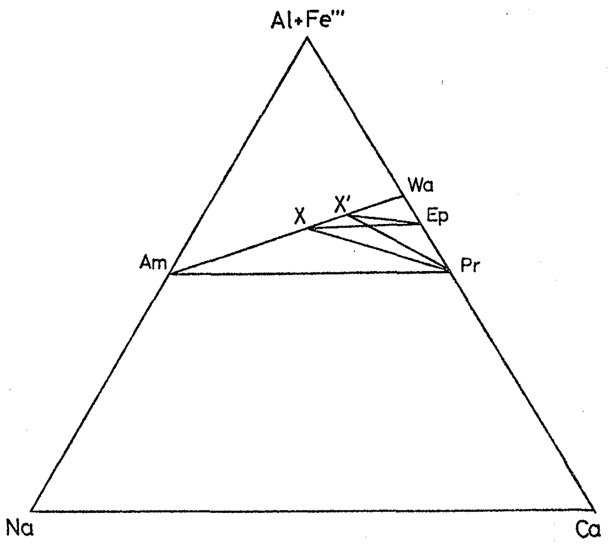

Fig. 11. (Al+Fe''')-Na-Ca diagram showing the shift of the composition of wairakite-analcime solid solutions associated with both of epidote and prehnite by increasing water pressure (X→X′).
Wa : Wairakite
Am : Analcime
Ep : Epidote
Pr : Prehnite

When the composition of X in Figure 8 further shifts toward the right and reaches to pure wairakite composition, the following chemical equation is obtained between prehnite + wairakite and epidote:

2 Epidote + 5 Quartz + 4 Water
$Ca_2Al_3Si_3O_{12}(OH)$ SiO_2 H_2O

⇄ Prehnite + 2 Wairakite
$Ca_2Al_2Si_3O_{10}(OH)_2$ $CaAl_2Si_4O_{12}\cdot 2H_2O$

Quartz is associated with wairakite-analcime solid solutions in most rocks which have been treated by the present writer.

The chemical reactions which represented by the above-noted two equations must have been displaced towards the right with addition of water under constant temperature and pressure conditions.

All of the wairakite · analcime-bearing rocks of Tanzawa Mountains, Fujikawa and Yugami districts have been found within relatively narrow areas of the ｢pumpellyite-prehnite facies and higher-grade parts of the zeolite facies. It seems improbable that the temperature conditions to form these wairakite · analcime series mineral-bearing rocks were significantly different in these districts. Probably one of the most important factors to shift the position of X in Figure 8, towards the wairakite composition or to analcime composition was water pressure or chemical potential of water which prevailed during the low-grade metamorphism of these districts. It is very probable that the wairakite-analcime ratio of wairakite-analcime solid solution stably associated with both of prehnite and epidote can be used as an indicator of water pressure condition (or chemical potential of water) to form low-grade metamorphic rocks, if these rocks were recrystallized at almost same low temperature condition.

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ワイラカイト・アナルサイム固溶体と，変成作用の水圧

関　陽太郎

(要　旨)

丹沢山地南部，同山地北部，富士川中流地域，福井県湯上地域の低変成度のグリーンタフの中には，ワイラカイト・アナルサイム固溶体がかなり広く見出される．この固溶体鉱物のX線による検討をおこなった結果，エピドートおよびプレーナイトと共存するこの固溶体のワイラカイト成分が上記の地域ごとにかなりちがっていることがわかった．もしこれらの地域の変成作用の温度がほぼ同じであるとしたら，このようなワイラカイト成分の差は，変成作用の時の水蒸気圧の差を反映しているらしい．