

## LOWER-GRADE STABILITY LIMIT OF EPIDOTE IN THE LIGHT OF NATURAL OCCURRENCES

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### Introduction

Epidote is a mineral commonly observed in a variety of metamorphic rocks of the greenschist facies, epidote amphibolite facies and pumpellyite-prehnite facies. It also occurs in skarns which are calcareous rocks metamorphosed under relatively high temperature conditions by the intrusion of salic magmas.

The higher-grade stability limit of epidote has been discussed by many authors. Epidote which is stable over almost the whole range of P-T fields of the greenschist facies and/or epidote amphibolite facies must break down in the higher-grade part of the amphibolite facies, the granulite facies and the pyroxene hornfels facies to form chiefly calcic plagioclase, Ca-garnet, calcic pyroxene and tschermakite component of calcic amphibole. Mineral equilibria including epidote, anorthite, Ca-pyroxene and Ca-garnet was theoretically and experimentally studied by KRETZ (1963), NITSCH and WINKLER (1965), STRENS (1965), HOLDAWAY (1966a and 1966b) and KEITH, *et al.*, (1968). These studies are mostly related to the high-temperature stability limit of epidote.

For the last two decades, the petrochemical study of areas of the lower-grade metamorphic facies than the greenschist facies has rapidly progressed. In these low-grade facies areas, the first appearance of epidote or epidote-bearing assemblage has often been used as an indicator of definite metamorphic grade. In this paper, the present writer will examine the lower thermal stability limit of epidote mostly on the basis of natural occurrence of this mineral.

### Review of stability of epidote in low-grade metamorphism

The present writer has classified metamorphic facies series into five types according to the pressure prevailed during the metamorphism (SEKI, 1969). In this chapter the epidote occurrence in low-grade metamorphic facies will also be reviewed chiefly on the basis of the different pressure type

metamorphism.

(A) Epidote in the Franciscan metamorphic terrane, California (Highest-pressure type facies series)

No epidote occurs in "in situ" blueschist facies metamorphic rocks including metamorphosed mafic volcanic rocks of Diablo Range and Goat Mountain areas in the Franciscan metamorphic belt (ERNST, 1965, 1971; ERNST and SEKI, 1967; ERNST *et al.*, 1970; SEKI *et al.*, 1969). In these rocks pumpellyite and lawsonite associated with albite, jadeitic pyroxene, chlorite, phengite, glaucophane, quartz, calcite (and aragonite) and sphene are commonly observed. Petrographical observations coupled with oxygen isotopic geothermometry and experimental phase equilibrium data indicate that these metamorphic rocks must have been formed at relatively low-temperature (150-300°C) and high pressure conditions (5-8 kb) (ERNST *et al.*, 1970).

The blueschist facies metamorphic area in the Franciscan belt passes with decreasing grade of metamorphism toward the zone characterized by the occurrence of laumontite-bearing mineral assemblages through an intermediate zone characterized by the occurrence of pumpellyite (neither prehnite nor epidote) in metamorphosed mafic volcanic rocks (BLAKE, 1969; ERNST *et al.*, 1970; COLEMAN and LEE, 1963; COLEMAN and LANPHERE, 1971; BLAKE *et al.*, 1967, 1969; BAILEY *et al.*, 1964). No epidote can be found as a metamorphic mineral in any type of metamorphic rocks of the laumontite zone.

In coarse-grained glaucophane-bearing rocks which occur as tectonic blocks intimately related with serpentinite intrusions, epidote occurs in association with glaucophane, phengite, garnet, omphacite and rutile. These tectonic blocks were called "high-grade blueschists" (COLEMAN and LEE, 1963; TAYLOR and COLEMAN, 1968; COLEMAN and LANPHERE, 1971). Some of these "high-grade blueschists" display the effects of retrogressive alterations related to later deformation into the assemblage of "low-grade in situ blueschists"; in this assemblage glaucophane,

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lawsonite, pumpellyite and chlorite are stable but garnet and epidote are replaced by a combination of lawsonite, pumpellyite and chlorite. Probably in high-pressure type metamorphic facies series observed in Franciscan metamorphic belt of California, epidote is stable in the high temperature field of the blueschist facies conditions which passes to the eclogite facies or epidote amphibolite facies with increasing grade of metamorphism.

Stability of epidote in similar type of metamorphic facies series in the Alps, Corsica, Calabria and New Caledonia can be summarized as follows:

1. The stable association of epidote with pumpellyite, prehnite, laumontite, albite and sericite in low-grade metamorphic rocks of the Alps has been reported by MARTINI and VUAGNAT (1965).
2. In Corsica, BROUWER and EGELER (1952) reported the absence or rare occurrence of epidote in the high-pressure metamorphic area where the glaucophane-jadeite-lawsonite assemblage is common. In contrast, epidote is an important rock-forming mineral in the area where glaucophane (or crossite) is stable but neither jadeite nor lawsonite was formed.
3. HOFFMAN (1970) also noted the absence of epidote in highpressure regional metamorphic rocks of north Calabria, Italy where the grade of metamorphism ranges from the pumpellyite facies to the blueschist facies with increasing temperature.
4. No epidote was formed in the lower-grade part, but is stably found in rocks of higher-grade part of the blueschist facies area of New Caledonia (COLEMAN, 1967; BROTHERS, 1970).

(B) Stability of epidote in the Kanto Mountains and Shikoku of the Sanbagawa metamorphic terrane, Japan (High-pressure type facies series)

Epidote is one of the most common minerals in low-grade metamorphic rocks of Sanbagawa metamorphic belt, Japan. In the Kanto Mountains and Shikoku which are the most thoroughly studied areas in this belt, epidote was stably formed in the actinolite-greenschist facies, the transitional facies between the greenschist and the blueschist facies and the pumpellyite-actinolite facies. (BANNO, 1964; ERNST, 1964; ERNST and SEKI, 1967; ERNST *et al.*, 1970; HASHIMOTO and KASHIMA, 1970; IWASAKI, 1963; MIYASHIRO and SEKI, 1958; SEKI, 1958, 1969). The pumpellyite-actinolite facies area generally passes with decreasing grade of metamorphism to the pumpellyite facies area in which neither actinolite nor prehnite was formed as metamorphic minerals in mafic volcanic rocks. The pumpellyite facies area probably passes to the laumontite subfacies area of the zeolite facies. Epidote disappears or

becomes to be very rare in the lower-grade part of the pumpellyite facies area.

(C) Stability of epidote in central Kii Peninsula of the Sanbagawa metamorphic terrane, Japan (Intermediate pressure type facies series)

Sanbagawa metamorphic area of the central Kii Peninsula can be divided into the following zones with increasing metamorphic grade (SEKI, ONUKI *et al.*, 1971):

laumontite zone  
pumpellyite-prehnite zone  
pumpellyite-actinolite zone  
actinolite-greenschist zone.

The wide development of the pumpellyite-prehnite facies area is a characteristic of Sanbagawa metamorphism of this area. No sodic amphibole, lawsonite and jadeitic pyroxene has been found in any metamorphic grade of this area. Analyses of the metamorphic paragenesis indicate that the regional metamorphism of central Kii Peninsula must have occurred under lower pressure conditions than those which prevailed in the Kanto Mountains and in Shikoku. Epidote is common in metamorphic mafic volcanic rocks of the pumpellyite-prehnite zone, the pumpellyite-actinolite zone and the actinolite-greenschist zone. Epidote seems to disappear in the laumontite zone.

In regard to the metamorphic facies series, metamorphic conditions in the following six areas seem to be very close to those which prevailed during the Sanbagawa metamorphism of the central Kii Peninsula.

1. Low-grade metamorphic rocks of the Katsuyama area, Japan were studied in detail by HASHIMOTO (1968). On the basis of the analyses of metamorphic mineral assemblages in pelitic, psammitic and mafic volcanic rocks, this area can be regionally divided into an actinolite-greenschist zone, a pumpellyite-actinolite zone and a pumpellyite-prehnite zone. Sodic amphiboles (mostly crossite and riebeckite) were formed in some metamorphic rocks of the pumpellyite-actinolite zone. Epidote is a widespread metamorphic mineral in all of above-noted zones, although its occurrence in the pumpellyite-prehnite zone is less common than in the two higher-grade zones.

2. In the New Zealand Geosyncline belt, epidote was stably formed in the pumpellyite-prehnite facies which passes into the zeolite facies and the greenschist facies area with decreasing and increasing grades of regional metamorphism respectively (HUTTON, 1940; COOMBS, 1960, 1961; COOMBS *et al.*, 1959; LANDIS and COOMBS, 1967). Epidote is also clearly the characteristic Ca-Al silicate of the greenschist facies and is probably stable in the pumpellyite-prehnite facies, but

was supposed to be of detrital origin in the zeolite facies.

3. PACKAM and CROOK (1960), CROOK (1960a, 1960b, 1961) and CHAPPELL (1968) also reported the stable occurrence of epidote in the pumpellyite-prehnite facies and the greenschist facies, but rare occurrence of epidote in the zeolite facies of Tamworth Trough, Australia.

4. Epidote may have been stably formed in the higher-grade part of the zeolite facies of east-central Puerto Rico which passes to the pumpellyite-prehnite facies with increasing metamorphic grade (OTALORA, 1964). Epidote is common in metamorphic rocks of the pumpellyite-prehnite facies.

5. BROWN and THAYAR (1963) noted the common occurrence of epidote in Triassic and Jurassic eugeosynclinal deposits of Oregon metamorphosed under pumpellyite-prehnite facies conditions.

6. SMITH (1968, 1969) reported the common occurrence of epidote in altered felsic or intermediate volcanic rocks developed in his albite-quartz zone and prehnite zone of the Tasman Geosynclinal area, Australia. Smith said that these two zones may represent a lower-grade part than the pumpellyite-prehnite zone which is distinguished from his albite-quartz zone and prehnite zone by the occurrence of both pumpellyite and prehnite associated with epidote in altered mafic volcanic rocks. Probably Smith's albite-quartz zone and prehnite zone represent the lower-grade part of the pumpellyite-prehnite facies. The absence of pumpellyite in rocks of these two zones may be, at least partly, due to the character of bulk chemical compositions of original rocks, not mafic but felsic or intermediate.

(D) Stability of epidote in Tanzawa Mountains, central Japan (Low-pressure type facies series)

Thick pile of Miocene mafic-intermediate pyroclastic rocks of the Tanzawa Mountains, Japan, were metamorphosed by the intrusion of a big mass of quartz diorite. The metamorphic area can be divided into the following five zones in the order of increasing grade of metamorphism (SEKI, OKI *et al.*, 1969; SEKI, OKI, *et al.*, 1971; SEKI, OKI *et al.*, 1972):

Clinoptilolite (stilbite) zone  
Laumontite zone  
Pumpellyite-prehnite zone  
Actinolite-greenschist zone  
Amphibolite zone

There is no zone characterized by the pumpellyite-actinolite assemblage.

Epidote is stable in rocks of the actinolite-

greenschist zone and the pumpellyite-prehnite zone. Epidote also occurs, though very rarely, in some of hematite-bearing mafic metamorphic rocks of the higher-grade part of the laumontite zone. In most of the zeolite facies area of the Tanzawa Mountains, mineral assemblages including prehnite and/or calcium zeolites (laumontite or stilbite) evidently formed instead of epidote-bearing mineral assemblages.

The pressure conditions which prevailed during the metamorphism of the Tanzawa Mountains were inferred to be 2 kb or lower (SEKI, 1969, 1971; SEKI and ONUKI, 1971).

(E) Stability of epidote in geothermal areas (Lowest-pressure type facies series)

Rocks in deeper parts of active geothermal areas are now being altered under relatively low pressure conditions. Recent development of the petrological and geological studies of cores from bore-holes drilled in these geothermal areas has given us many invaluable informations on the stability of epidote in metamorphism.

STEINER (1966, 1968, 1970) reported the occurrence of epidote in cores from the deepest parts of two bore holes of the Wairakei geothermal area, New Zealand. The intermediate-felsic volcanic rocks of this area can be divided into the following three zones by the assemblages of metamorphic minerals from the surface to depth:

mordenite-montmorillonite zone  
-----70 m 120°C  
laumontite-chlorite zone  
-----250 m 170-200°C  
wairakite-chlorite zone

Epidote associated with wairakite, chlorite, albite and adularia occurs commonly in altered volcanic rocks of the deeper part of the last zone. The depths and temperatures of the cores from which epidote appears first are 400-500 m and 230°C respectively. In the Wairakei geothermal area, epidote was probably stable at higher pressures than about 100 bars and higher temperatures than 230°C.

Local alterations of Miocene pyroclastics and their associated rocks into wairakite-albite-epidote-chlorite-white mica-quartz assemblages have been reported from the Hanawa Mine district (SEKI, TAKEYASU *et al.*, 1968) and the Kawaji district (SEKI and ONUKI, 1971), Japan. Geological and petrological evidence indicates that these epidote-bearing mineral assemblages must have been formed in the deeper (higher temperature) part of the past geothermal areas.

Epidote appears as a hydrothermal alteration product in bore-hole cores of the Reykjanes thermal area, Iceland (BJÖRNSSON *et al.*, 1970). Depths

and temperatures measured in cores where the first appearance of epidote was identified are as follows:

Hole number	Depth (m)	Temperature (°C)
4	550	200
3	720	250
8	1200	260

Epidote was stably associated with quartz, albite, prehnite, calcite, zeolites and some other minerals at greater depths.

SIGVALDASON (1959, 1963) briefly described the hydrothermal alterations of volcanic rocks into epidote (and clinozoisite)-and pumpellyite-bearing mineral assemblages at the Hveragerdi and Reykjavik geothermal areas, Iceland. The shallowest depths and lowest temperature conditions for the formation of epidote in these areas are as follows:

Hveragerdi	Reykjavik
450 m 255°C	750 m 130°C

The occurrence of pumpellyite and clinozoisite in such low temperature conditions as 120–140°C indicates that observed mineral occurrence and the metamorphic transformations in Reykjavik area may reflect the previous higher temperatures, not the present (pre-drilling) temperature environments.

KEITH *et al.*, (1963) reported the formation of epidote in the deeper part (1310–1500 m, 320°C) of the Salton Sea geothermal area, California. Epidote is associated with quartz, chlorite, K-feldspar, albite and, rarely, with tremolite. MUFFLER and WHITE (1969) also described the hydrothermal alteration of Pliocene and Quarternary sediments into epidote-bearing mineral assemblages (chlorite-epidote-albite-

Table 1. Stability ranges of epidote in various types of metamorphic facies series.

LM	B S		E A	Franciscan metamorphic belt.....Highest pressure type
	←			
PM	P A		G S *	Sanbagawa metamorphic belt (Kanto Mountains and Shikoku) .....High pressure type
←				
LM	P P	P A	G S	Sanbagawa metamorphic belt (Central Kii), New Zealand Geosynclinal belt, Sangun metamorphic belt (Katsuyama) .....Intermediate pressure type
	←			
LM	P P		G S	Tanzawa Mountains area.....Low pressure type
	←			
LM	W A			Wairakei geothermal area.....Lowest pressure type
	←			

Abbreviations:	LM	Laumontite subfacies	} Zeolite facies
	WA	Wairakite subfacies	
	P P	Pumpellyite-prehnite facies	
	PM	Pumpellyite facies	
	P A	Pumpellyite-actinolite facies	
	G S	Greenschist facies	
	B S	Blueshist facies	
	E A	Epidote amphibolite facies	

\* Intermediate facies between the actinolite greenschist facies and the blueschist facies

K-feldspar-quartz-pyrite) in deeper parts of the same geothermal area. The epidote-bearing associations occur at 1100 m or deeper and 300°C or higher.

In other geothermal areas such as Onikobe (SEKI, ONUKI, *et al.*, 1969), Atosanupuri (SEKI, 1970), Matsukawa (SUMI, 1968), Panzhetka (RUSINOV, 1965) and Steamboat Springs (SIGVALDSON and WHITE, 1961; SCHOEN and WHITE, 1965, 1967; WHITE 1968; SIGVALDSON, 1962), no epidote has been found as a hydrothermal alteration product of rocks, although the occurrence of other kinds of minerals such as prehnite, wairakite, laumontite and mordenite are not rare. Maximum temperatures and their depth of these geothermal areas are as follows:

Onikobe	270 m	210°C
Atosanupuri	200 m	150°C
Matsukawa	390 m	230°C
Panzhetka	800 m	200°C
Steamboat Spring	200 m	170°C

WHITE and SIGVALDSON (1963) referred to the paper by NABOKO and PIYP (1961) on the occurrence of epidotes in the Panzhetka geothermal area at 750 m depth and 140°C conditions. Later RUSINOV (1965), however, made it clear that the epidotes in this geothermal area are not newly formed ones at these depth and temperature but are relict clastic crystals.

Probably, the low-temperature stability limit of epidote in such low-pressure conditions as appropriate to geothermal areas is about 230°C.

#### Summarized low-temperature stability limit of epidote

SEKI (1969) classified the facies series of low-grade metamorphism into five types: highest pressure type, high pressure type, intermediate pressure type, low pressure type and lowest pressure type. Table 1 shows the stability ranges of epidote in type areas of these five facies series of low-grade metamorphism. The low-temperature limit of the stability field of epidote can be plotted in a reasonable temperature-pressure diagram of low-grade metamorphic facies as shown by Figure 1.

Chemical relations between epidote and wairakite, laumontite, pumpellyite and prehnite are simplified by the following four equations:

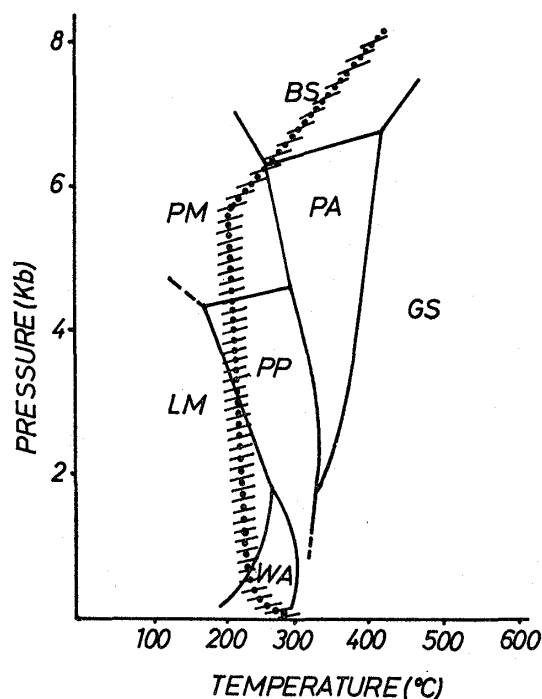
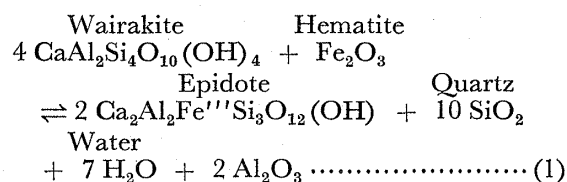


Fig. 1. Probable stability limit of epidote in low-grade metamorphism.

P-T fields of metamorphic facies came from SEKI (1969) partly modified by COOMBS (1969).

LM: Laumontite subfacies } Zeolite facies  
WA: Wairakite subfacies }

PP: Pumpellyite-prehnite facies

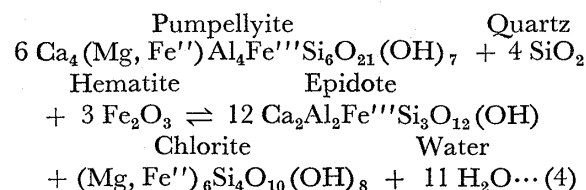
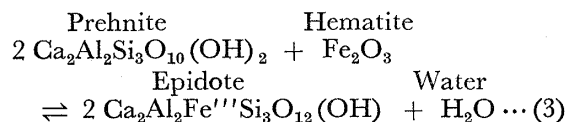
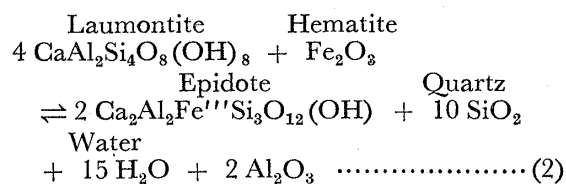
PM: Pumpellyite facies

PA: Pumpellyite-actinolite facies

BS: Blueschist facies

GS: Greenschist facies

Lower stability limit of epidote inferred from its natural modes of occurrence is represented by a hatched dot line.



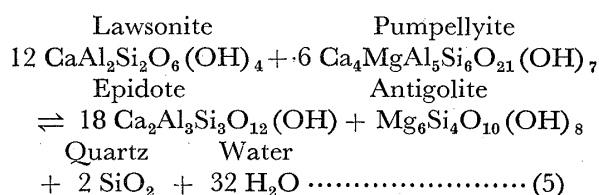
The left-hand sides of these equations show the

lower-temperature decomposition products of epidote.  $\text{Al}_2\text{O}_3$  and some  $\text{SiO}_2$  and  $\text{H}_2\text{O}$  in the right-hand sides of equations (1) and (2) may be used to form chlorite, prehnite or some other minerals associated with epidote.

Equation (1) represents the chemical relation of the first appearance of epidote at depth in active geothermal areas, characterized by low-solid pressure conditions. The first appearance of epidote in the high-grade part of the laumontite subfacies of the zeolite facies area and the lower-grade part of the pumpellyite-prehnite facies area can be represented by equations (2) and (3). Equation (4) may indicate the first appearance of epidote at lower-grade part of the pumpellyite-prehnite zone and the pumpellyite zone.

All chemical reactions from left-hand side to right-hand side in equations (1), (2), (3) and (4) are dehydration reactions. Thus, if water pressure is equal to solid pressure, transition curves for these equations must be of positive slope in temperature-pressure diagram. However, in natural metamorphic environments where water pressures are lower than solid pressures, the negative solid volume change which occurs during these chemical reactions at constant  $P_{\text{fluid}}$  gives much greater effects to the slope and shape of transition curves than those given by the hydration-dehydration relation. The decrease of solid volume accompanied with the formation of epidote by equations (1) and (2) (25–30%) is far larger than those which occur when epidote is formed by equations (3) and (4) (4–13%). Consequently, it is probable that, in temperature-solid pressure diagram (water pressure  $\ll$  solid pressure), the low-temperature stability limit of epidote at low pressure conditions such as those which prevailed at the shallow depth appropriate to geothermal areas must form a negative P-T curve but become almost parallel to the pressure axis with increasing pressure as shown by Figure 1.

In the lower temperature portion of the blueschist facies metamorphism characterized by very high pressure conditions, epidote is not stable but both lawsonite and pumpellyite appear as stable metamorphic minerals associated with glaucophane and/or jadeitic pyroxene. The chemical relation between epidote, lawsonite and pumpellyite can be represented as follows:



Pumpellyite formed under high pressure conditions is generally poor in  $\text{Fe}^{3+}$  content (SEKI, 1961; ERNST *et al.*, 1970; JOLLY, 1971). Epidote formed within the high-grade P-T part of the blueschist facies is also generally poor in  $\text{Fe}^{3+}$  content (COLEMAN and LEE, 1963; LEE, COLEMAN, BASTRON and SMITH, 1966). Antigolite molecule may be used to form glaucophane by the combination with albite molecule. It must be noted that the chemical reaction to form lawsonite and pumpellyite from epidote-chlorite-quartz assemblage by equation (5) is accompanied with the addition of a great amount of water and a great decrease in solid volume (13.5% decrease). So, the low temperature stability limit of epidote under very high pressure ( $\sim P_{\text{H}_2\text{O}}$ ) conditions such as those which prevailed during the blueschist facies metamorphism must form a curve of positive slope in temperature-pressure diagram.

The stability limit of epidote shown in Figure 1 by the present writer is still rather speculative.

It is probable that epidote may have been formed metastably at some low temperature regimes, because epidote is not very reactive at low temperature and low pressure conditions.

The effect of increasing  $\text{Fe}^{3+}$  content to the stability field of epidote and the influence of oxygen fugacity on both epidote stability and composition have been theoretically and experimentally shown by STRENS (1965), HOLDAWAY (1966a, 1966b) and LIOU (1972, personal communication). MIYASHIRO and SEKI (1958) have already reported that in the Kanto Mountains of the Sanbagawa belt the average Al content of epidote generally decreases with decreasing grade of metamorphism from the actinolite-greenschist facies to the pumpellyite facies. Probably the stability field of  $\text{Fe}^{3+}$ -rich epidote extends toward lower temperatures than that of Al-rich epidote as suggested by STRENS (1965). Epidote does not occur generally in "in situ" metamorphic rocks of the Franciscan belt. At South Fork Mountains, however, some mafic metavolcanics contain epidote whereas associated metagraywackes carry lawsonite (ERNST, 1971; ERNST, 1972, personal communication). Possibly the  $\text{Fe}^{3+}/\text{Fe}^{2+}$  ratio (i.e., rock bulk composition plus oxygen fugacity) determines the occurrence of epidote as well as pressure and temperature. It is also probable to think that in equation (5) the oxidation conditions favor epidote in which Al can readily be replaced by  $\text{Fe}^{3+}$  and more reducing conditions promote crystallization of pumpellyite and lawsonite in which no or minor iron can enter under constant temperatures and pressures.

The composition of individual phases associated

with epidote as well as rock bulk composition must also be important to decide the stability of epidote. The chemical potential of  $\text{CO}_2$  on the lower stability limit of epidote could not be neglected. In this aspect, the review of epidote occurrence in the calc-silicate environments such as skarn formation may be important.

Experimental studies of such chemical reactions as represented by equations (1), (2), (3), (4) and (5) at various conditions of oxygen partial pressure, various ratios of water pressure/solid pressure and  $\mu\text{H}_2\text{O}/\mu\text{CO}_2$  should also be undertaken, although the rate of reactions must be very slow.

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### 低変成度におけるエピドートの安定領域

関 陽 太 郎

(要 旨)

比較的高温度 (おそらく 600°C 以上) の条件の下ではエピドートは輝石・ザクロ石・灰長石などの組合せに分解する。このようなエピドートの高変成度での安定領域については、実験的にも、理論的にも、また天然の産状からもかなり多くの研究がすでにおこなわれた。しかし、低変成度でのエピドートの安定領域 (エピドートの安定領域の下限) についての知識はまだはなはたとぼしい。最近までに集積した岩石学的データをまとめると、エピドートの安定領域の下限は、第1図の点線で示され

たようなものであるらしい。この線より低温になると、エピドートは (1), (2), (3), (4), (5) であらわされるような化学反応で、沸石、プレーナイト、パンペリー石、ローソン石などに分解してしまうのであろう。

#### 地 名

Hanawa	花輪	Matsukawa	松川
Katsuyama	勝山	Onikobe	鬼首
Kawaji	川治	Tanzawa	丹沢