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Author(s)	WAKISAKA, Hiroshi; NISHIDO, Hirotsugu; YOSHIASA, Akira; KITAGAWA, Ryuji; TAKENO, Setsuo
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Zeolites in the Granitic Rocks of Chugoku District, Southwest Japan

By

Hiroshi WAKISAKA, Hirotsugu NISHIDO, Akira YOSHIASA,
Ryuji KITAGAWA and Setsuo TAKENO

with 3 Tables and 8 Figures

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Abstract :Ca-zeolites such as laumontite, stilbite and chabazite were found in the granitic rocks of both magnetite and ilmenite series of late Cretaceous to Tertiary age developed widely in the Chugoku district. These zeolites occur (1) in veins of various scale, (2) replacing plagioclase of the host granitic rocks, and (3) in pegmatitic druse. Mineralogy were investigated by X-ray powder diffraction method, the optical microscope, the scanning electron microscopy and the electron microprobe.

Based on the mode of occurrence and the associated minerals, laumontite and stilbite in veins and replacing plagioclases are concluded to be formed from hydrothermal solutions filling open fractures which have been formed under the paleo-stress field after the solidification stage of the host granitic rocks. Probable formation condition of laumontite and stilbite was estimated as follows; 200-250°C(temperature), 0.5-1.0 Kb(P total) and 20-40 bar(max P_{CO}).

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I. Introduction

Zeolites commonly occur in sedimentary and metamorphic rocks as well as in cavities and veins in volcanic rocks (Gottardi and Galli, 1985). Occurrence of zeolites in granitic rocks are relatively rare (e.g., Ishii, 1952; Raade, 1969; and Stepiewiez, 1978). However, zeolites are frequently found in granitic rocks developed in the Chugoku district. No detailed research on zeolite formation in granitic rocks has been published up to the present.

Zeolites occur in vein, as replacing plagioclase and even in pegmatitic druse. The most common occurrence is in vein developed in the granitic rocks and the mode of occurrence of the vein is quite similar to that of clay veins reported in detail by Kitagawa(1986). Although zeolites were already reported as the minor

constituent minerals of the clay vein (Kitagawa et al., 1982; Kitagawa,

1985), mineralogical description of these zeolites were too brief and no genetical consideration was presented. In this paper, mineralogy of zeolites found in the granitic rocks developed in the Chugoku district will be described and their formation condition will also be discussed.

II. Samples

Zeolites were collected from the following eight localities. (1)Kudani, Northwestern part of Hiroshima city, (2)Fukutomi-cho, Kamo-gun, Hiroshima Prefecture, (3)Mihara city and its suburbs, (4)Shikawa, northwestern part of Fukuyama city, (5)Yakake-cho, Oda-gun, Okayama Prefecture, (6)Miyori, Kiyone-mura, Tsukubo-gun, Okayama Prefecture and (7)Nabeyama and Igi,

Mitoya-cho, Iishi-gun, Shimane Prefecture. (8)Komaki, Yokota-cho, Nita-gun, Shimane Prefecture. Among the above 8 localities, sericite and halloysite deposits are developed in the two host granitic rocks at Nabeyama(7) and Komaki(8), respectively. In each locality, zeolites were collected from several to about 10 points.

As is seen in Fig.1, all localities are in the granitic rocks of Chugoku district from late Cretaceous to Tertiary age. According to Ishihara(1977) and Ishihara et al.(1980), only localities (7) and (8) belong to the magnetite series whose K-Ar ages are about 48 Ma, and the other belong to the ilmenite series of late Cretaceous age. Therefore, zeolites occur throughout the Chugoku district regardless of the rock type of the granite. All of the granitic rocks are coarse to medium grained biotite granite.

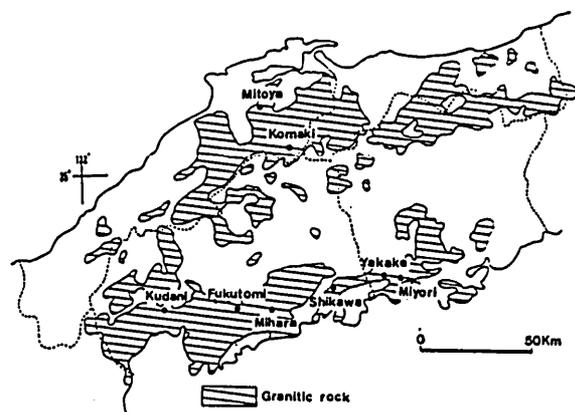


Figure 1. Locality map of sampling points of zeolite in the granitic rocks of Chugoku district.

III. Occurrence and mineral description

Occurrence of zeolites in the granitic rocks of Chugoku district can be classified into the following three types; (A) veins, (B) replacing plagioclase, and (C) pegmatitic druse. Types (A) and (B) are found in the localities (1), (2), (3), (4) and (7). At locality (8), only (A) type is recognized. Type (C) is found at the location of (5) and (6).

Zeolites and coexisting minerals were first identified by X-ray powder diffraction method and laumontite, stilbite and chabazite were confirmed. Some typical X-ray powder diffraction patterns of zeolites are shown in Fig.2. These X-ray powder diffraction patterns agree well with those of natural zeolites reported previously (Lapham, 1963; Tomita et al., 1979; Aumento, 1966; and Gude and Sheppard, 1966). Smectite, calcite, apophyllite, epidote, chlorite, quartz and feldspar

were identified as the coexisting minerals. Mineral paragenesis of zeolites are summarized in Table 1. In Table 1, mineral species of zeolite in the granitic rocks are very restricted and laumontite is the most common. Moreover, coexisting minerals of zeolite are also restricted. Quartz and/or smectite and calcite are the most common associated minerals of zeolite. In the following, three types of occurrence of zeolite mentioned above will be described.

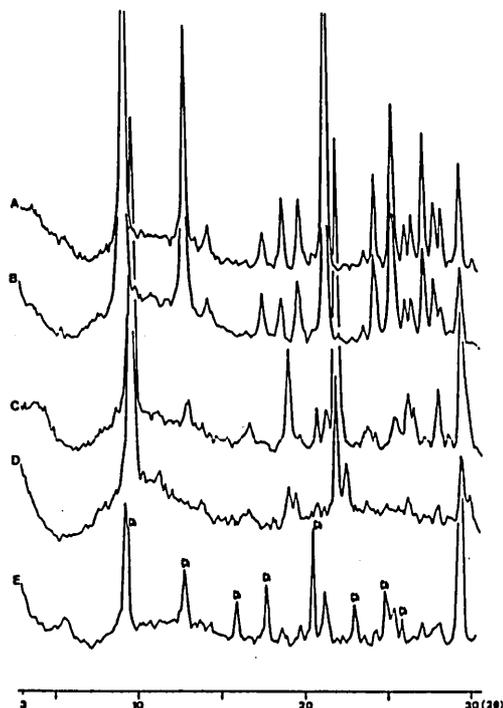


Figure 2. X-ray powder diffraction patterns of zeolites. A: laumontite in vein. B: laumontite in pegmatite druse. E: chabazite(ch) coexisting with laumontite in vein.

1. Veins

Zeolites are generally found in vertical veins developed in the granitic rocks. Occasionally zeolites are also found in horizontal veins. The width of veins varies from about one millimeter to several centimeters. Detailed field observation of the veins reveals the following 4 types of occurrences; (1) filling vertical (rarely almost horizontal) fractures of 1 millimeter to several centimeters in width. The fractures extend even to dike rocks intruded in the granitic rock (Fig.3 A), (2) in fault clays whose width is several to some tens centimeters, and also along slickenside (Fig.3 B), (3) filling the interstices of both sides of dike rock such as porphyry (Fig.3 C), and (4) as small network vein or veinlets developed only in small area

of about several tens centimeter. This type of occurrence is recognized only in Mitoya district, where small veinlets are developed densely as it looks almost one vein (Fig.3 D). These mode of occurrences are schematically shown in Fig. 3. It should be noted that the mode of occurrence of zeolite is almost similar to those of clay veins described in detail by Kitagawa(1986).

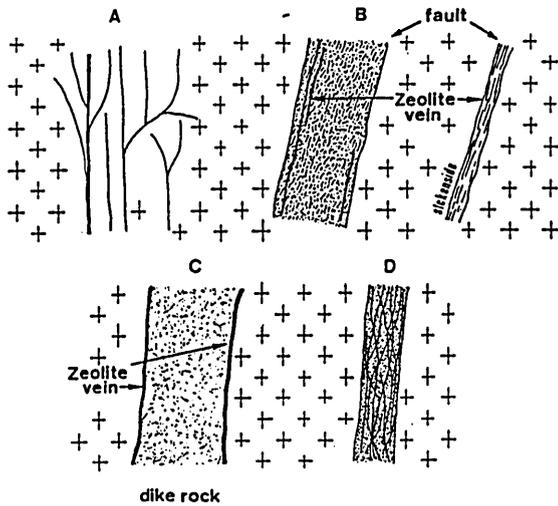


Figure 3. Schematic illustration of zeolite veins.

Mineral species of zeolite observed in veins are laumontite, stilbite and chabazite. Among these, laumontite is found in all types of veins, whereas stilbite and chabazite are found in only (4) type. In Table 1, mineral paragenesis of zeolite in the granitic rocks are summarized. As is evident in the Table, the assemblage is characterized by common association with smectite and/or calcite. The mineral assemblage found in vein is rather simple than that in pegmatite. Laumontite is the most common zeolite. It is pink in color which changes to white when laumontite associates with stilbite, chabazite and/or smectite. When only stilbite is present, the color of the vein is grayish white to white. Zeolites are, in general, aggregates of very fine crystals. In rare case, subhedral to euhedral crystals of laumontite of about several millimeters are observed under scanning electron microscope. Crystal surface of laumontite is in general smooth. In Shikawa (locality 4), a continuous development of laumontite veins of (1) type can be pursued more than 200m in vertical direction until the overlain mudstone of Paleozoic is reached.

The orientation of the strikes of the veins was systematically investigated in three localities where the veins are developed significantly, i.e., in Kudani, Shikawa and Mitoya. In Fig.4, orientation of the veins in each district is characterized by

several preferred directions. Main direction of zeolite veins in each region is almost same to the azimuth of maximum principal stress axis deduced from the orientation analysis of clay veins in each region (Kitagawa and Nishido, 1994).

2. Replacing plagioclase

Zeolite replacing plagioclase of the host granitic rocks was recognized along type (1) zeolite vein. Mineral species of zeolite of this type is restricted to laumontite and the mode of occurrence is characteristic as shown in Fig.5. In the Fig.5, plagioclase within several centimeters from a vein

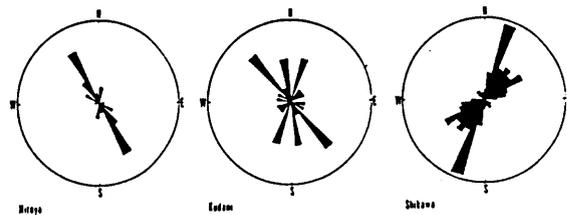


Figure 4. Histogram of strike of zeolite and smectite veins in Mitoya, Kudani and Shikawa districts.

is altered to laumontite. The color of the altered part, i.e., laumontite, is characteristically pinkish. Neither stilbite nor chabazite were found as replacing plagioclase (Table 1).

		V	R	P			V	R	P	
Kudani	lau	○	○		Mitoya	lau	○	○		
	lau-qtz	○				lau-qtz	○			
	lau-sm-qtz	○				lau-sm-qtz	○			
Fukutomi	lau	○	○		Komaki	lau	○			
	lau-sm-qtz	○				Yakake	lau-sti-cal-ap-qtz			○
	lau-sti-qtz	○					lau-qtz-fp			○
	sti-qtz	○			lau-epi				○	
Mihara	cal	○			Miyori	lau-chl			○	
	lau	○	○			lau-epi-qtz-fp			○	
	lau-qtz	○					lau-qtz-fp			○
	lau-sm-qtz	○								
Shikawa	lau-sti-qtz	○			lau:laumontite sti:stilbite cha:chabazite qtz:quartz cal:calcite epi:epidote chl:chlorite sm:smectite	ap:apophyllite fp:feldspar V :In vein R :Replacement product of plagioclase P :In pegmatite				
	lau-cal	○								
	lau-sm-cal	○								
	lau-sti-qtz	○								
	lau-cha	○								
		○								
		○								

Table 1. Mineral paragenesis of zeolites

3. Pegmatitic druse

Zeolites in pegmatitic druse are laumontite and stilbite. Laumontite is the most common and the association with small amounts of stilbite is also

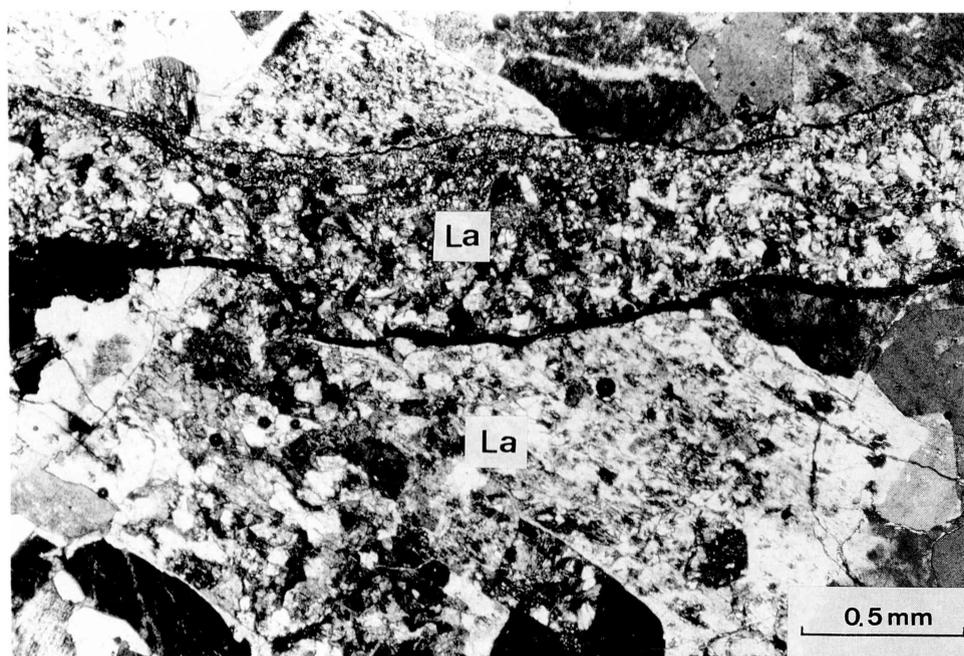


Figure 5. Microphotograph illustrating the mode of occurrence of laumontite. Note that laumontite vein cut host plagioclase and most part of the plagioclase is replaced by laumontite (see, text).

	1	2	3	4	5	6	7	8
SiO ₂	52.53	52.49	51.06	52.93	52.11	52.04	52.70	52.42
TiO ₂	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.00
Al ₂ O ₃	21.71	22.16	22.23	21.80	22.09	21.76	22.10	21.70
FeO*	0.01	0.06	0.02	0.05	0.01	0.00	0.04	0.07
MnO	0.01	0.03	0.02	0.04	0.00	0.00	0.01	0.02
MgO	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.01
CaO	11.83	12.21	12.35	11.62	11.64	11.23	11.77	11.26
Na ₂ O	0.08	0.05	0.02	0.16	0.20	0.13	0.16	0.12
K ₂ O	0.44	0.58	0.15	0.87	0.64	1.08	0.96	1.23
Si	16.11	15.97	15.83	16.12	16.00	16.07	16.02	16.10
Ti	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al	7.85	7.95	8.12	7.82	7.99	7.92	7.92	7.86
Fe*	0.00	0.02	0.01	0.01	0.00	0.00	0.01	0.02
Mn	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.00
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ca	3.89	3.98	4.10	3.79	3.83	3.74	3.83	3.71
Na	0.05	0.03	0.01	0.09	0.12	0.08	0.09	0.07
K	0.17	0.23	0.06	0.34	0.25	0.43	0.37	0.48
B.E.X	-1.8	-3.1	-1.8	-2.2	-0.3	-0.7	-2.4	-1.2

	9	10	11	12	13
SiO ₂	51.14	52.64	52.48	52.09	52.22
TiO ₂	0.00	0.00	0.00	0.00	0.00
Al ₂ O ₃	22.31	21.79	21.44	22.32	22.64
FeO*	0.00	0.00	0.00	0.00	0.00
MnO	0.01	0.01	0.01	0.01	0.02
MgO	0.00	0.00	0.00	0.00	0.00
CaO	12.45	12.03	11.86	12.63	12.40
Na ₂ O	0.10	0.13	0.17	0.11	0.19
K ₂ O	0.52	0.73	0.78	0.36	0.40
Si	15.78	16.06	16.12	15.87	15.84
Ti	0.00	0.00	0.00	0.00	0.00
Al	8.12	7.84	7.76	8.02	8.10
Fe*	0.00	0.00	0.00	0.00	0.00
Mn	0.00	0.00	0.00	0.00	0.00
Mg	0.00	0.00	0.00	0.00	0.00
Ca	4.12	3.93	3.90	4.13	4.03
Na	0.06	0.08	0.10	0.06	0.11
K	0.20	0.28	0.31	0.14	0.15
B.E.X	-4.5	-4.7	-5.5	-5.1	-2.7

*Total Fe

- 1,2,3 :In vein, Mitoya, Shimane Prefecture.
 4 :Replacement product of plagioclase, Mitoya, Shimane Prefecture.
 5,6,7 :In vein, Shikawa, Hiroshima Prefecture.
 8 :In vein, Fukutomi, Hiroshima Prefecture.
 9,10,11,12,13:In pegmatite, Yakake, Okayama Prefecture.

Table 2. Chemical composition and formulae on the basis of 48 oxygen atoms of laumontites.

	1	2	3	4	5
SiO ₂	61.59	59.58	60.93	61.17	54.08
TiO ₂	0.02	0.01	0.00	0.00	0.00
Al ₂ O ₃	16.75	18.12	17.49	18.27	17.19
FeO*	0.04	0.03	0.00	0.00	0.00
MnO	0.02	0.03	0.01	0.03	0.00
MgO	0.01	0.00	0.00	0.00	0.00
CaO	8.99	8.72	9.06	9.02	8.52
Na ₂ O	0.38	0.62	0.78	0.98	0.68
K ₂ O	0.09	0.12	0.06	0.08	0.03
Si	27.22	26.60	26.87	26.64	26.25
Ti	0.01	0.00	0.00	0.00	0.00
Al	8.73	9.53	9.09	9.38	9.83
Fe*	0.02	0.01	0.00	0.00	0.00
Mn	0.01	0.01	0.00	0.01	0.00
Mg	0.01	0.00	0.00	0.00	0.00
Ca	4.26	4.17	4.28	4.21	4.43
Na	0.32	0.54	0.66	0.83	0.64
K	0.05	0.07	0.03	0.05	0.02
B.E.X	-1.7	6.5	-1.8	0.9	3.3

*Total Fe

- 1,2 :In vein, Fukutomi, Hiroshima Prefecture.
 3,4,5:In pegmatite, Yakake, Okayama Prefecture.

Table 3. Chemical composition and formulae on the basis of 72 oxygen atoms of stilbitites.

recognized. These zeolites together with associated minerals occur in pegmatitic druse of relatively small scale, less than 1m. This type of zeolites are euhedral crystals and the color of laumontite and stilbite are pink and pale brown, respectively. The largest euhedral crystals of laumontite and stilbite are 3 to 4cm in length and 2 to 3mm in width, respectively. In Yakake and Miyori, zeolites were found only in pegmatitic druse and neither zeolite veins nor replacement zeolite were confirmed.

IV Chemical composition

Chemical composition (except H₂O) of zeolites were analyzed using JEOL electron microprobe (ACMA-733II, 15KV) on aggregates and single crystals. The results of chemical analysis and atomic formulae obtained on the basis of 48 or 72 oxygen atoms are shown in Tables 2 and 3. The balance error (B.E.%) in the tables, Passaglia, 1970) is also presented. Chemical compositions of laumontite and stilbite are almost constant regardless of localities and occurrences (Tables 2 and 3). The ratio of Na+K to Ca for laumontites and stilbites are shown in Fig.6 together with the previous results (Coombs, 1952; Deer et al., 1963 and Passaglia and Galli, 1978). Laumontite and stilbite of the present study are characteristically fall on near Ca end region (Fig.6). Laumontites related to granitic rocks in the literatures (open circles and stars in Fig.6) are almost coincide with the present result, i.e., Ca-rich composition. Concerning high alkali laumontites (open triangles), re-examination of the composition will be required since low possibility of alkali solubility is suggested considering the crystal structure (Kawahara, 1988). The present results support the Kawahara's suggestion.

In the crystal structure of stilbite, on the other hand, Ca ion is commonly substituted by alkali ions (i.e., Harada, 1962; Passaglia and Galli, 1978). Stilbite in the granitic rocks is characterized by high Ca content corresponding almost to that of stellerite. In Fig.7, dotted line and broken line show the stilbite-stellerite boundary and D(Ca+Mg)=4 line, respectively proposed by Passaglia and Galli (1978). The present stilbite contains much Ca than those examined by Passaglia and Galli (1978). Although point 1 in Fig.7 situates in the center of stellerite, precise examination of the X-ray powder pattern of the specimen by the method described by Passaglia and Galli (1978) clearly indicates that the mineral is stilbite.

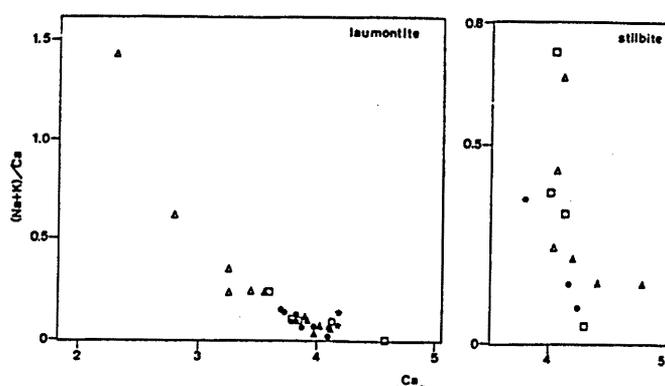


Figure 6. Na+K/Ca ratio of laumontites and stilbites in granitic rocks of Chugoku district and previous analyses. ●: in veins. ▲: in pegmatite druse, □: Deer et al. (1963). △: Gottaldi and Galli (1985). ○: in granitic rocks, Stepiewicz (1978). ☆: in granitic rocks, Coombs (1952) No. 13, 14. *: in granitic rocks, Passaglia and Galli (1978).

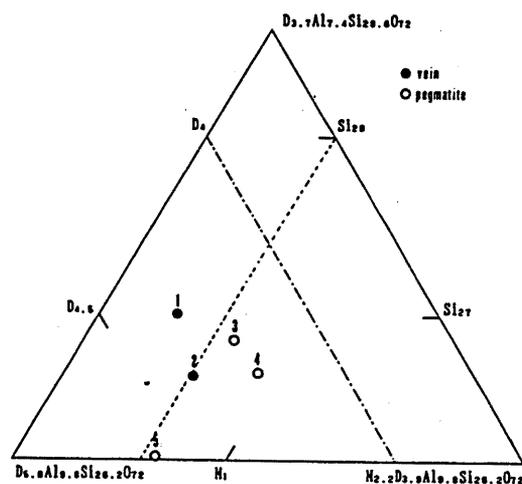


Figure 7. Chemical composition of stilbite in the granitic rocks of chugoku district in the diagram proposed by Passaglia and Galli (1978). D=Ca+Mg. M=Na+K. Numbers are correspond to those of Table 3.

V Discussion

Although zeolites are formed in various geological conditions, occurrence of laumontite, stilbite and chabazite are restricted to (1) diagenesis, (2) hydrothermal solution including percolating groundwater in volcanic rocks (Iijima, 1980) and (3) in low grade metamorphic rocks (Liou, et al., 1987).

Laumontite and stilbite found in veins were most probably formed from certain hydrothermal solution filling

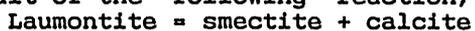
open fractures which were formed under the regional stress field subsequent to the solidification stage of the respective host granitic rocks. The main direction of strike of the zeolite veins in some regions examined in Fig.4 are almost similar to azimuth of the principle stress axes of clay veins developed in the same regions (Kitagawa and Nishido, 1994).

These zeolites are intimately associated with smectite and calcite. The clay minerals in the Komaki deposit (mainly composed of halloysite) is believed to be formed by hydrothermal solution related to the granitic activity (Kitagawa and Kameoka, 1986), though Sudo and Takagi (1993) inferred that the deposit was formed during weathering. Laumontite veins clearly cut the host clay deposit. Therefore, laumontite was probably formed at the latest stage of the hydrothermal activity, if this deposit was formed by hydrothermal activity.

Formation process of zeolites in the pegmatitic druse is rather problematical. P-T condition of zeolite formation in pegmatitic druse remains problematical. No veins connecting to the pegmatitic druse were confirmed during field observation. In addition, intimate association of the zeolite with epidote, chlorite and calcite suggests that the zeolites were formed later than the pegmatitic stage, i.e. hydrothermal origin. Similarity of chemical composition of zeolites in veins and in pegmatitic druse is suggestive.

Concerning the formation condition of Ca-zeolites, experimental result of Cho et al.(1987) and Liou et al.(1987) are available. Phase boundaries of Ca-zeolites in Fig.8 are taken from the two literatures in the system of $\text{CaAl}_2\text{Si}_2\text{O}_8\text{-SiO}_2\text{-H}_2\text{O}$. Upper temperature limit can be estimated from the absence of yugawaralite and wairakite. Pressure range can be estimated also from the absence of yugawaralite(lower limit) and heulandite(upper limit). The estimated temperature and pressure range is reasonably accepted for the case of zeolites in the granitic rocks of Chugoku district based on the facts of the associated clay minerals and also of the mode of occurrence, i.e. open cracks filling.

Coexistence of smectite and calcite with laumontite may be the result of the following reaction;



According to Ivanov and Gurevich(1975), this reaction takes place at about 200-250°C, 0.5-1.0 Kb(P total) and 20-40 bar(P_{CO_2}). Coexistence of laumontite, stilbite and quartz may probably produced by the breakdown of heulandite(Cho et al., 1987). Absence of heulandite may give significant information on the formation condition of the present zeolites, especially on the range of

pressure and temperature.

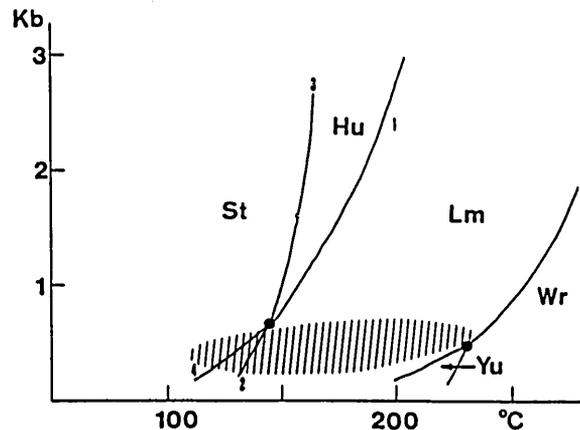


Figure 8. Possible formation condition of zeolites in the granitic rocks. Phase boundaries are taken from Cho et al.(1987).

Such conditions described above can be expected in certain stage related to the post granitic activities. Chemical composition of zeolites in the granitic rocks is characterized by high Ca content. Replacement of plagioclase by laumontite requires Ca addition. Common association of smectite with zeolite needs also extra Ca. In addition to these facts, absence of Na-zeolite such as analcime is indicative to the chemical nature of the related hydrothermal solution. Occurrence of zeolite veins in the granitic rocks are quite similar to those of clay veins(Kitagawa, 1986). The fact suggests that at relatively early stage of the post granitic activities, hydrothermal solution differentiate into two types, one is related to zeolite and the other to clay veins. The differentiation probably occurred due to mainly the change of P_{CO_2} and temperature. Present results suggest the common occurrence of zeolites in the granitic rocks of Chugoku district. However, reasonable explanations on the characteristics of chemical compositions of laumontite and stilbite, and the source of high Ca content in the hydrothermal solution in the later stage of the post granitic activities are remained to be solved.

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- Hiroshi WAKISAKA : Hiroshima city Ebayama museum of meteorology. Eba-Minami, 1-40-1, Naka-ku, Hiroshima, 730 Japan.
- Hirotsugu NISHIDO : Hiruzen Research Institute, Okayama University of Science, Okayama, 717-06 Japan.
- Akira YOSHIASA : Department of Earth and Space Science, Faculty of Science Osaka University. Machikaneyama, Toyonaka, 560 Japan.
- Ryuji KITAGAWA and Setsuo TAKENO : Department of Earth and Planetary Sciences, Faculty of Science, Hiroshima University, Kagamiyama, Higashihiroshima, 724 Japan.

