American Mineralogist, Volume 67, pages 937–943, 1982

Experimental investigation of yugawaralite–wairakite equilibrium

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Abstract

Yugawaralite ($CaAl_2Si_6O_{16}{\cdot}4H_2O$) occurs most commonly in active geothermal areas and is associated with other zeolites including laumontite and wairakite. The stability field of yugawaralite was experimentally investigated using conventional hydrothermal apparatus over a $P–T$ range of 200–350° C and 0.25–2 kbar and run duration of 14 to 61 days. The reversal experiments indicate that the $P–T$ curve of the reaction yugawaralite = wairakite + $2SiO_2$ + $2H_2O$ passes through 225±5° C at 0.25 kbar, 223±5° C at 0.5 kbar, 238±5° C at 1 kbar and 245±5° C at 2 kbar P_{fluid}. The thermodynamic parameters of yugawaralite were calculated: $\Delta S_f = -2175.46\pm12.01$ J/mol-K, $\Delta G°_f = -8387.69\pm4.01$ kJ/mol, $\Delta H_f = -9036.19\pm4.01$ kJ/mol. The equilibrium curve, yugawaralite = laumontite + $2SiO_2$, was estimated by employing the thermodynamic data and the calculated slope of 31.3 bars/° C. Present and previous experiments define an invariant point where yugawaralite, wairakite and laumontite are stable. The invariant point was located at 234° C and 550 bars in experimental conditions where $P_{H_2O} = P_{\mathrm{total}}$. In geothermal systems where P_{H_2O} is about 1/3 P_{total}, the occurrence of yugawaralite is restricted to depths not much greater than 0.5 km. This result is consistent with its limited occurrence in active geothermal areas.

Introduction

Yugawaralite ($CaAl_2Si_6O_{16}{\cdot}4H_2O$) was first described by Sakurai and Hayashi from vein fillings near the Yugawara Hot Spring, Japan in 1952. This calcium zeolite has since been reported to occur often with other zeolites, including laumontite and wairakite as euhedral crystals filling cavities and veins in geothermal areas and as microcrystals in low-grade metamorphosed volcanic rocks. Yugawaralite of the first case includes the type locality; Heinabergsjokell, Iceland (see Barrer and Marshall, 1965); Onikobe (Seki and Okumura, 1968) and Shimoda, Japan (Sameshima, 1969); Chena Hot Springs areas, Alaska (Eberlein *et al.*, 1971); Osilo, Sardinia, Italy (Pongiluppi, 1977); Bombay, India (Wise, 1978) and Yellowstone National Park (Bargar and Beeson, 1981; Bargar *et al.*, 1981). These occurrences suggest that yugawaralite is formed at low pressures and temperatures and is related to geothermal activity. Yugawaralites from the Tanzawa Mountains (Seki *et al.*, 1969a) are believed to be the product of low-grade metamorphism, with an extremely high geothermal gradient.

$P–T$ stability relations of some calcium zeolites including laumontite, wairakite, and stilbite in the system $CaAl_2Si_2O_8–SiO_2–H_2O$ have been investigated (Liou, 1970 and 1971a, b); however the stability of yugawaralite and its relationship to those of other calcium zeolites has not been experimentally determined. Seki (1969) and Harada (1969) have suggested that the stability field of yugawaralite is intermediate between the stabilities of laumontite and wairakite with regard to temperature.

The purposes of the present study are (1) to experimentally determine the univariant equilibrium curve of the reaction yugawaralite = wairakite + $2SiO_2$ + $2H_2O$; (2) to calculate the thermodynamic parameters of yugawaralite; (3) to outline the $P–T$ stability relationships among yugawaralite, wairakite and laumontite, and (4) to correlate the experimental results with paragenesis of Ca-zeolites in geothermal systems. Chemographic relations of laumontite, yugawaralite, wairakite, quartz and some other calcium zeolites are plotted in the system $CaAl_2Si_2O_8–SiO_2–H_2O$ in Figure 1.

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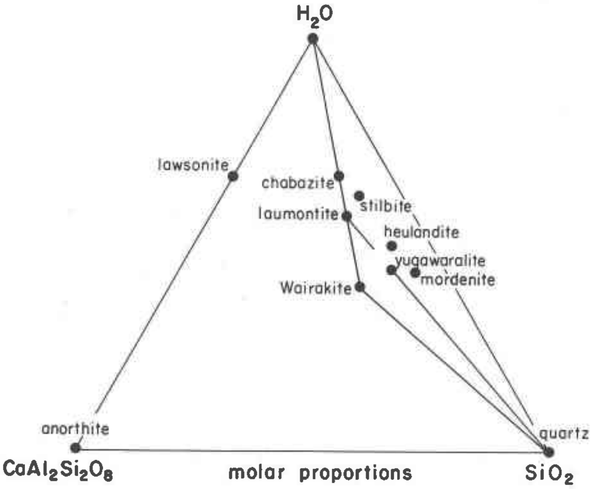

Fig. 1. Chemographic relations of some calcium zeolites and related minerals in the ternary system $CaAl_2Si_2O_8$–SiO_2–H_2O.

Experimental methods

Conventional cold seal hydrothermal apparatus was used in the present study; details of the experimental procedure are described by Liou (1971a). The starting materials include wairakite from Onikobe, yugawaralite from Yugawara, and laumontite from the Tanzawa Mountains, Japan. The compositions, optical and X-ray properties, and abbreviations of these zeolites are listed in Table 1. To ensure their purity, all of these minerals and quartz were ground and examined by X-ray diffraction. The charges for each experiment consisted of mixtures of yugawaralite as reactant and wairakite + quartz (with molar ratio of Wr/Qz[2] = 1/2) as products in subequal weight proportions and excess H_2O. Other charges had mixtures of laumontite and yugawaralite + wairakite + quartz in the weight ratio 1:2 with the ratios among yugawaralite, wairakite and quartz the same as above.

The charges were run from 30 to 61 days at temperatures ranging from 200° C to 350° C, and pressures from 250 bars to 2000 bars. The recorded temperatures are thought to be accurate to ±5° C and the pressures indicated by the gauges to be ±30 bars. All the charges were examined by microscope, X-ray diffraction and a scanning electron microscope with X-ray energy dispersive system.

Use of the above mineral mixtures as starting materials in the experiments avoided nucleation problems and enhanced the growth of seeded materials in their fields of stability. Phase relations were deduced by observing which assemblage grew at the expense of the other. The direction of reaction was established by comparing the relative intensities of main X-ray diffraction peaks (which were measured by area-ratio) of reactants and products before and after the experiment. When the mixture of yugawaralite + wairakite + quartz was used in the experiments, three intensity ratios, $Yu_{(140)}/Wr_{(400)}$, $Yu_{(111)}/Wr_{(211)}$, and $Yu_{(002)}/Wr_{(332)}$, were used. For the mixtures of laumontite + yugawaralite + wairakite + quartz, the intensities of only four peaks, $Lm_{(110)}$ and $Lm_{(002)}$, $Yu_{(111)}$ and $Wr_{(211)}$, were measured because the other X-ray diffraction peaks overlapped substantially.

Newton (1966) demonstrated that, where the extent of reaction is not large, the direction of reaction cannot be positively recognized by comparison of X-ray diffractometer charts. This difficulty was encountered in some experiments which are close to the equilibrium temperatures. In the present study, many runs with durations of 60 days and under closely similar P_{fluid}–T conditions were made and the results showed consistent reaction direction. Each isobaric equilibrium temperature for the reaction $Yu = Wr + Qz + H_2O$ was determined by at least 4 runs within a 50° C temperature interval.

In order to obtain reproducible intensity ratios of diffraction peaks, samples for the X-ray mounts were prepared according to the same procedure and 2 to 5 X-ray diffraction patterns of each charge were taken under identical conditions (Cu*Kα* radiation, 35 kV accelerating potential, 16 mA emission current, ¼° 2*θ*/min scanning speed, 30 in./hr chart speed, intensity amplifier 1×10^2 and time constant 4). The areas of diffraction peaks were accurately measured by using a compensating polar planimeter. All run products were also examined by using a scanning electron microscope (SEM/EDAX) to detect the growth and dissolution of phases in order to verify the results obtained by X-ray diffraction.

Experimental results

The experimental results from a total of 35 runs using the mixture of yugawaralite + wairakite + quartz as starting material are listed in Table 2 and shown in Figure 2. The growth of wairakite at the expense of yugawaralite was readily detected for those experiments at temperatures greater than 250° C because 20–50% conversion occurred. However, the phase equilibration at temperatures lower than 250° C, especially *P*–*T* conditions close to

Table 1. Chemical compositions, optical and X-ray properties of natural wairakite, laumontite and yugawaralite used as starting materials

	Wairakite (Wr)*	Laumontite (Lm)**	Yugawaralite (Yu)***
SiO_2	54.91	50.99	59.29
TiO_2	0.01	0.02	tr.
Al_2O_3	22.75	21.87	17.43
Fe_2O_3	0.64	0.09	0.05
FeO	0.44	0.11	0.25
MnO	0.01	0.02	none
MgO	0.23	tr.	0.11
CaO	11.69	12.00	9.90
Na_2O	0.60	0.38	0.26
K_2O	0.31	0.10	tr.
H_2O^+	8.23	14.02	12.85
H_2O^-	0.19	0.29	0.10
Total	100.01	99.89	100.24
α	1.498±0.002	1.506±0.002	1.493
β		1.512±0.002	1.499
γ	1.502±0.002	1.516±0.002	1.501
a_o	13.65	14.737±0.004	6.73
b_o	13.66	13.006±0.002	13.95
c_o	13.56	7.550±0.003	10.03
β	90°20'	111.97±0.02	111°30'

*After Liou (1970)
**After Liou (1971a)
***After Seki and Okumura (1966)

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[2] Abbreviations: Wr—wairakite; Qz—quartz; Yu—yugawaralite; Lm—laumontite.

Table 2. Experimental results for the reaction: yugawaralite (Yu) = wairakite (Wr) + $2SiO_2$ + $2H_2O$

Run No.	Temperature °C	Duration days	Direction of reaction
	P_{H_2O} = 500 bars		
43	210	61	Yu ← Wr
39	220	30	Yu ← Wr
72	222	60	Yu ← Wr
50	230	60	Yu ← Wr
66	235	60	Yu → Wr
71	244	60	Yu → Wr
32	255	28	Yu → Wr
33	275	28	Yu → Wr
20	294	21	Yu → Wr
13	325	21	Yu → Wr
	P_{H_2O} = 1000 bars		
17	217	28	Yu ← Wr
74	222	60	Yu ← Wr
51	224	61	Yu ← Wr
73	228	60	Yu ← Wr
36	234	30	Yu ← Wr
61	235	60	Yu ⇆ Wr
45	241	31	Yu ⇆ Wr
62	246	60	Yu → Wr
35	250	30	Yu → Wr
23	256	28	Yu → Wr
18	275	28	Yu → Wr
15	300	28	Yu → Wr
21	325	14	Yu → Wr
	P_{H_2O} = 2000 bars		
52	221	59	Yu ← Wr
22	236	32	Yu ← Wr
42	238	32	Yu ← Wr
63	240	60	Yu ← Wr
65	250	58	Yu → Wr
64	260	60	Yu → Wr
19	264	30	Yu → Wr
37	274	30	Yu → Wr
38	287	31	Yu → Wr
14	301	28	Yu → Wr
16	333	28	Yu → Wr
7	348	31	Yu → Wr

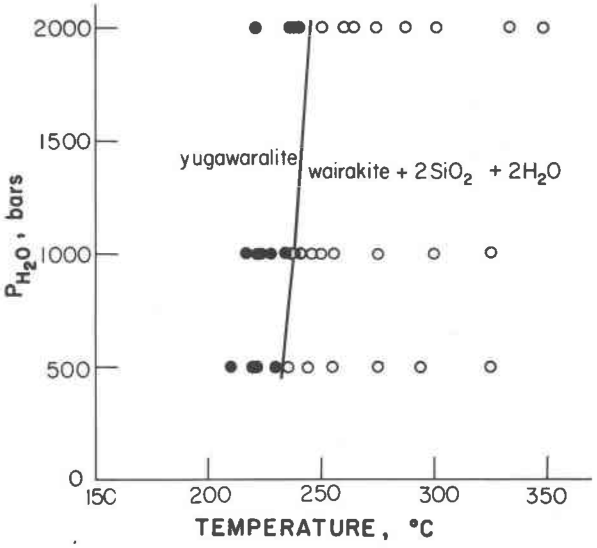

Fig. 2. P_{fluid}–T curve of the reaction yugawaralite = wairakite + $2SiO_2$ + $2H_2O$, experimentally determined by using the mixture of yugawaralite + wairakite + quartz (run data listed in Table 1).

equilibrium, is extremely difficult to detect because of the sluggishness of the reaction. Only about 5–20% change took place even in experiments of 2 months duration. In the present study many experiments under closely similar temperature conditions were made and examination of dissolution and growth of wairakite by using SEM was found to be very useful for determining reaction direction. In the reaction products from runs within the wairakite field (Fig. 2), especially at pressures higher than 1000 bars, freshly grown, well formed wairakite crystals with trapezohedron-cubic faces were found. In the products run within the yugawaralite field, poorly crystallized yugawaralite and subrounded wairakite occurred. No new phases were detected by either X-ray diffraction or SEM observation.

A combination of the criteria of intensity ratios of X-ray peaks and SEM observations was used to locate the equilibrium curve of the reaction as

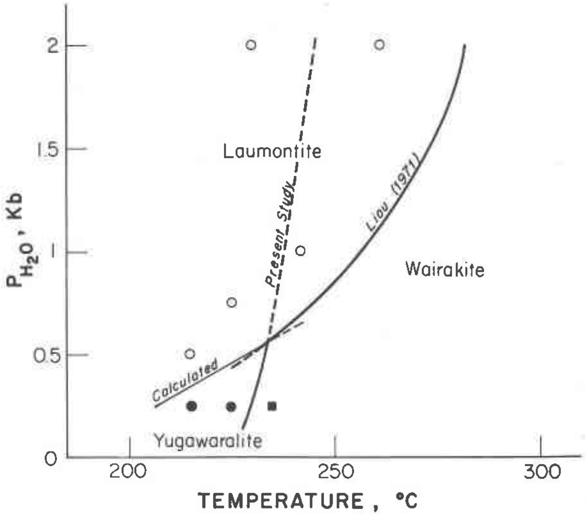

Fig. 3. P_{fluid}–T stability diagram for yugawaralite, laumontite, and wairakite in the presence of excess quartz and fluid. Open circles (laumontite), solid circles (yugawaralite) and solid squares (wairakite) show the strongest tendency of growth among the three zeolites in the four-mineral mixture experiments. The equilibrium P–T curve of the reaction yugawaralite = wairakite + $2SiO_2$ + $2H_2O$ is from the present experimental determination. The equilibrium dehydration curve of laumontite is from Liou (1971a); the P–T curve of the reaction, yugawaralite = laumontite + $2SiO_2$ is calculated.

passing through 233±5° C at 500 bars, 238±5° C at 1000 bars and 245±5° C at 2000 bars. The steep slope of the *P–T* curve suggests that the reaction is relatively insensitive to pressure in the range 0.5–2.0 kbar.

Integrating the present results with a previous study on laumontite–wairakite equilibrium in the $CaAl_2Si_2O_8$–SiO_2–H_2O system by Liou (1971a), the portion of the present wairakite–yugawaralite curve above 500 bars is metastable in the field of laumontite. Apparently, the three Ca-zeolites, yugawaralite, wairakite and laumontite have identical CaAl formula contents; hence, in the system with excess SiO_2 and H_2O, they define an invariant point. The intersection of the two experimentally determined univariant curves for reactions Yu = Wr + $2SiO_2$ + $2H_2O$ and Lm = Wr + $2H_2O$ locate the invariant point at about 234° C and 550 bars. Radiating from the invariant point is another reaction, Yu = Lm + $2SiO_2$. The phase relationships shown in Figure 3 are also consistent with those proposed by Harada (1969).

In order to experimentally outline the invariant relationship, seven experiments with mixtures of laumontite + yugawaralite + wairakite + quartz were carried out. The data are listed in Table 3 and are plotted in Figure 3. Based on the reconnaissance experiments and results described in the previous sections, it was concluded that yugawaralite is stable at lower temperatures relative to wairakite and at lower pressures relative to laumontite. The stability field of yugawaralite is bounded by the reaction Yu = Wr + Qz + H_2O at high temperature and by the reaction Yu = Lm + Qz at high pressure.

Discussion

Calculation of thermodynamic data for yugawaralite

The thermodynamic parameters of yugawaralite were calculated from the experimentally determined reversed *P–T* brackets using the method of Fisher and Zen (1971) and Zen (1972). All data used in the calculations are listed in Table 4.

Table 3. Experimental results with the mixture of laumontite (Lm), yugawaralite (Yu), wairakite (Wr) and quartz (Qz)

Run No.	Temp. °C	P_{H_2O} bars	Duration days	Changes in Run Products			Tendency of mineral crystallization
				Lm/Yu	Wr/Lm	Yu/Wr	
84	230	2000	31	+	–	+	Lm > Yu > Wr
83	261	2000	31	+	–	–	Lm > Wr > Yu
86	242	1000	30	+	–	–	Lm > Wr > Yu
85	225	750	30	+	–	+	Lm > Yu > Wr
88	215	500	30	+	–	+	Lm > Yu > Wr
87	225	250	30	–	±	+	Yu > Wr > Lm
91	235	250	30	–	+	±	Wr > Yu > Lm

For the reaction, Yu = Wr + Qz + H_2O, the *P–T* brackets are: (1) 233±5° C, 500 bars; (2) 238±5° C, 1000 bars; (3) 245±5° C, 2000 bars. The combination of points 1, 3 and 2, 3 were used in the calculation. The calculated thermodynamic parameters of yugawaralite are:

$$\Delta S^{\circ}_{f(298,1)} = -2175.46\pm12.01 \text{ J/mol-K},$$

$$\Delta G^{\circ}_{f(298,1)} = -8387.69\pm4.01 \text{ kJ/mol};$$

$$\Delta H^{\circ}_{f(298,1)} = -9036.19\pm4.01 \text{ kJ/mol}.$$

Estimation of the P–T curve of the reaction yugawaralite = laumontite + $2SiO_2$

Using the data listed in Table 4, the slope of the equilibrium curve for Yu = Lm + $2SiO_2$ was estimated according to the Clapeyron equation. The calculated slope is 31.3 bars/° C, assuming that variation of volume and entropy changes with temperature for the solid–solid reaction is negligible. From the intersection of these experimentally determined univariant lines for Lm = Wr + $2H_2O$ and Yu = Wr + 2Qz + $2H_2O$, the *P–T* line for the solid–solid reaction Yu = Lm + 2Qz was drawn according to the calculated slope. The result is shown in Figure 3. The calculated phase relations among yugawaralite, wairakite and laumontite in the presence of excess quartz and fluid are consistent with the reconnaissance experiments using the three Ca-zeolites as starting materials described in the previous section. Figure 3 indicates that yugawaralite is restricted in its stability to temperatures lower than 233° C and pressures lower than 550 bars (about 1.5 km in depth) at the experimental conditions of $P_{H_2O} = P_{total}$.

It should be pointed out that the equilibrium Yu = Lm + 2Qz is a solid–solid reaction, hence its *P–T* locations are independent of fluid composition and of P_{H_2O}/P_{total}. In contrast, the two experimentally determined dehydration reactions shown in Figure 3 are very sensitive to variation in P_{H_2O}/P_{total} ratio. The P_{fluid}/P_{total} ratio in geothermal systems has been suggested to be about 0.3 (Coombs *et al.*, 1959). The phase relationships among the three Ca-zeolites under such conditions of $P_{H_2O} = 0.3\ P_{total}$ were estimated using their calculated thermodynamic parameters and are shown in Figure 4. The invariant point shifts toward lower temperatures and lower pressures along the *P–T* line of Yu = Lm

Table 4. Thermodynamic parameters of some Ca-zeolites and quartz

Mineral	Laumontite	Wairakite	Quartz	Yugawaralite
Formula	$CaAl_2Si_4O_{12}\cdot 4H_2O$	$CaAl_2Si_4O_{12}\cdot 2H_2O$	SiO_2	$CaAl_2Si_6O_{16}\cdot 4H_2O$
ΔG^o_f (kJ/mol)	-6685.22	-6185.45	-856.65	-8387.69
ΔH^o_f (kJ/mol)	-7237.11	-6612.01	-911.08	-9036.19
ΔS^*_f (J/mol·K)	-1851.05	-1430.69	-182.58	-2175.46
V^o (J/bar·mol)	20.7638	18.6951	2.2697	26.6033
Source	Helgeson et al. (1978)	Helgeson et al. (1978)	Helgeson et al. (1978)	present study

*Calculated from ΔG^o_f and ΔH^o_f

+ Qz with decreasing P_{H_2O}/P_{total} ratio. At the $P_{H_2O} = 0.3\ P_{total}$, the invariant point is estimated to be about 172° C and 230 bars P_{total}. Apparently, in geothermal systems where the fluid phase contains abundant other components in addition to H_2O, and where P_{fluid}/P_{total} ratios may be approximately 0.3, yugawaralite stability would be considerably restricted to smaller *P–T* fields and to lower temperatures.

Geological applications

The experimental results, which have located the stability field of yugawaralite at low temperatures and pressures, are consistent with its occurrences in geothermal fields and low-grade metamorphic terrains. Seki and Okumura (1968) described yugawaralite from the Onikobe geothermal area at depths between those corresponding to the laumontite zone (110.00–139.00 m) and the wairakite zone (deeper than 141.20 m) and at 110° C. The yugawaralite is associated with laumontite, wairakite, quartz, pyrite and chloritic minerals in an altered dacite tuff. The results of temperature measurements in drill holes at Yellowstone National Park (Bargar and Beeson, 1981; Bargar *et al.*, 1981) indicate that yugawaralite in drill hole Y-2 is restricted to a narrow zone of 149–152.2 m at temperatures of about 200° C whereas in drill hole Y-3, it is formed in a zone of about 73 m in depth and 170° C. These direct temperature measurements in both Yellowstone and Onikobe geothermal systems are plotted in Figure 4 and are compatible with the present data.

As shown in Figure 4, the paragenetic depth sequence of Ca-zeolites in a geothermal system is highly dependent on imposed thermal gradient, P_{H_2O}/P_{total} ratio and other factors including solution compositions (*e.g.*, Giggenbach, 1981). In a geothermal system with relatively high geothermal gradient and high P_{H_2O}/P_{total} ratio, yugawaralite may be stable and the depth sequence could be mordenite→laumontite→yugawaralite→wairakite. On the other hand, in the regions with lower geothermal gradient and lower P_{H_2O}/P_{total} ratio, yugawaralite is not stable and zonation of Ca-zeolite could be mor-

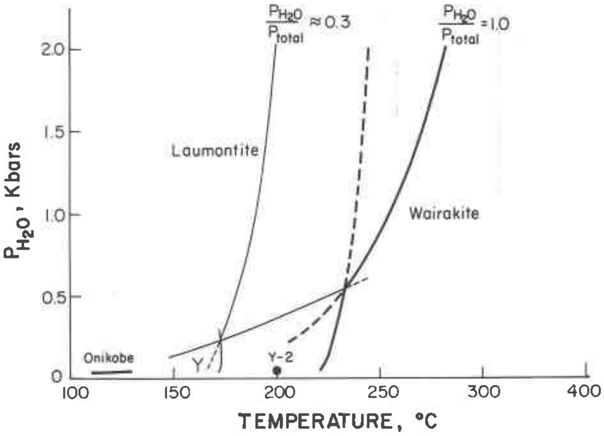

Fig. 4. P_{fluid}–*T* diagram showing the stability relations among yugawaralite (Y), laumontite, and wairakite in the presence of excess quartz and fluid at $P_{H_2O}/P_{Total} = 1.0$ and 0.3. The recorded temperatures and depths for yugawaralite occurrences in Onikobe (Seki *et al.*, 1969) and in Y-2 hole of Yellowstone (Bargar and Beeson, 1981) geothermal systems are also plotted for comparison.

denite→laumontite→wairakite. Because many geologic, solution and hydrologic conditions may control both P_{H_2O}/P_{total} ratio and geothermal gradient, different depth zonations of Ca-zeolite may occur even in a single geothermal system. Such variations have been recorded in the Onikobe geothermal area by Seki *et al.*, (1969b).

The yugawaralite in Alaska occurs about 14 miles from Chena Hot Springs and was suggested to have been deposited under conditions of low fluid pressure of 150–350 bars and temperature ranges of 200° to 300° C (Eberlein *et al.*, 1971). Although there is no past or present thermal spring activity at the yugawaralite locality, the yugawaralite is associated with stellerite, stilbite and laumontite. The suggested temperatures may be 100° C too high in the light of the presently determined stability of yugawaralite and that of stilbite (Liou, 1971b).

The occurrence of yugawaralite in the thermally metamorphosed aureoles of the Tanzawa Mountains (Seki *et al.*, 1969a) merits comment. The geothermal gradient for the hydrothermal formation of stilbite, laumontite and wairakite has been suggested to be high (Seki *et al.*, 1969a). Both wairakite and yugawaralite occur together with chlorite + prehnite + pumpellyite in some metamorphosed basaltic and andesitic rocks. Within these rocks the assemblages yugawaralite–chlorite–quartz, yugawaralite–laumontite–calcite–quartz, and yugawaralite–wairakite–calcite have been recorded in veins, cavities or druses. The close association of these three calcium zeolites together with quartz–chlorite suggests that they may have crystallized at *P–T* conditions very close to those of the invariant point. On the other hand, these zeolites may have formed at different times, and hence under different *P–T* conditions. Moreover, wairakites from the Tanzawa Mountains contain substantial amounts of Na (>1 wt.%) (Seki and Oki, 1969), hence, Na-bearing wairakite and yugawaralite may be stable within the yugawaralite field.

The present results provide a very restricted *P–T* stability field for yugawaralite which accounts for its restricted occurrence only at depths shallower than 500 m in geothermal systems. The presence of yugawaralite in hydrothermally altered rocks can potentially be used as a geobarometer.

Acknowledgments

This study was conducted during the senior author's 2-year advanced training at Stanford University, supported by the government of the PRC. The experimental study was supported by NSF grant EAR 79-09138 and DOE FC07-80ID-12148. We wish to thank Drs. R. C. Newton, Y. Seki and Mary Keskinen for discussion and review of the manuscript.

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Manuscript received, January 5, 1982;
accepted for publication, May 3, 1982.