

ALTERATION OF WALL-ROCKS INTO WAIRAKITE-BEARING ASSEMBLAGES AT THE KAWAJI DAMSITE, CENTRAL JAPAN

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Introduction

Wairakite associated with albite, epidote, laumontite, chlorite and quartz has been found as a hydrothermal alteration product of granodiorite-porphyrite and Miocene submarine volcanic rocks of the Kawaji district, central Japan. The Kawaji district is located in Nikko National Park and is about 280 km north of Tokyo (Fig. 1). The modes of occurrence and some mineralogical properties of wairakite and associated minerals will be described in this paper.

Mr. H. YUGETA and Mr. Y. SASAKI of Dia Consultant Co. guided the senior author in the field. Dr. Y. OKI of Hot-Spring Research Institute kindly helped us in the separation of wairakite from laumontite. This manuscript was critically read by Professor Akiho MIYASHIRO of State University of New York at Albany. The present authors wish to express their sincere thanks to these friends.

Geology

Geology of the Kawaji district has been briefly studied by Iwao and IMAI (1951) and IMAI (1952). This district is mainly composed of the following three geologic units: Late Paleozoic slate and sandstone formation, granodiorite-porphyrite intrusive and Miocene dacite-andesite lava and tuff formation.

The exposure of Paleozoic rocks is limited to a small area to the east of Kawaji Spa. This is mostly composed of the alternation of black slate and hard sandstone intercalated with small amounts of limestone, chert and mafic volcanic materials. Same types of rocks can be commonly found as xenoliths within granodiorite-porphyrite intrusive mass as well as in the dacite-andesite volcanic formation.

Granodiorite-porphyrite is mostly medium-grained hornblende-biotite granodiorite mixed with small amounts of hornblende-biotite-porphyrite and biotite porphyry. These intrusive rocks have lots of angular or subangular xenoliths

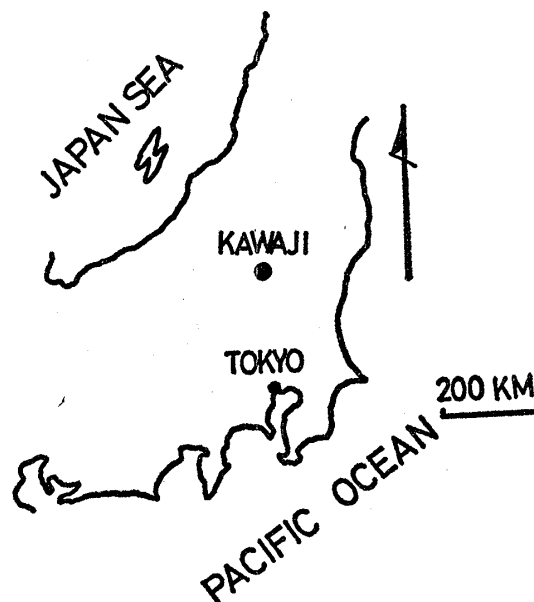


Fig. 1. Location of the Kawaji area, central Japan.

of Paleozoic slate and sandstone, diameter of which varies from 1.5 mm to several meters. These xenoliths were generally altered into biotite-bearing hornfels by thermal effect of the intrusive rocks. The age of intrusion of the granodiorite-porphyrite into Paleozoic formation is unknown, but is probably in Late Cretaceous.

The dacite-andesite lava and pyroclastic formation underlies the greatest area of this district. This formation is chiefly composed of brecciated lava, welded tuff and tuff breccia of dacitic-andesitic composition intercalated with small amounts of thin beds of slaty tuff. The finding of marine fossils in these slaty beds indicates that these volcanic rocks were extruded at the ocean bottom. A part of these volcanic rocks is intruded into the above-noted Paleozoic rocks and intrusive complex. Also at the north of Kawaji Spa, a sandstone bed having *Pectunnuculus* sp. and *Arca* sp. as fossils is intruded by a member of the dacite volcanic rocks. Hence, the geological age of the dacite-andesite lava and pyroclastic formation is believed to be Early or Middle Miocene (Iwao and IMAI, 1951).

Fig. 2 shows the geological map of the area 1.5 km west of Kawaji Spa from where wairakite

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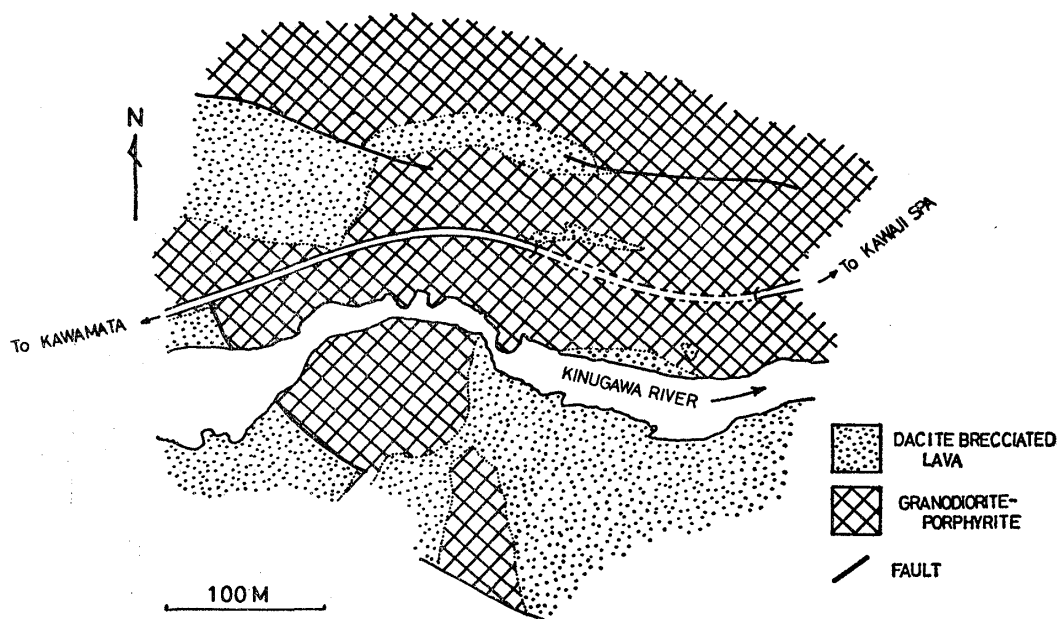


Fig. 2. Geological map of the Kawaji area, central Japan.

associated with epidote, laumontite, albite and quartz was found in the Miocene volcanic rocks and granodiorite-porphyrite complex. The Miocene volcanic rocks of this area are mostly brecciated lava, probably erupted out along the cracks within the granodiorite-porphyrite complex mass. Strike and dip of flow plane or brecciated plane observed in these volcanic rocks readily vary within a short distance of exposure. The volcanic rocks usually have a great deal of angular or subangular xenoliths of the surrounding granodiorite-porphyrite and also angular slaty xenoliths probably came from the Paleozoic formation of the basement of this area.

Hydrothermal alteration of granodiorite-porphyrite and Miocene volcanic rocks

All rocks represented in Fig. 2 have generally been sheared and altered under the hydrothermal conditions.

Hornblende and biotite which originally constituted the granodiorite-porphyrite have been partly or completely altered into chlorite or chlorite-sericite assemblage with a minor amount of sphene. Plagioclases were partly replaced along their cleavage planes or crystal margins by sericite, albite, epidote and wairakite. Many veins of the following mineral assemblages were formed along the sheared planes within these hydrothermally altered granodiorite-porphyrite:

calcite-wairakite-quartz,
wairakite-quartz,
wairakite-epidote-quartz,

wairakite-epidote-albite-quartz.

Among these veins, the occurrence of calcite-bearing one is very rare.

The Miocene dacite-andesite volcanic rocks generally suffered much stronger alteration than the above-noted granodiorite-porphyrite. Hornblende and biotite were almost completely altered into chlorite with or without sericite and sphene. Plagioclase was also almost completely replaced by the assemblages of albite, epidote, sericite, quartz and wairakite. In these altered volcanic rocks many veins or lenticular pools of the following mineral assemblages are observed:

albite-epidote-quartz,
wairakite-laumontite-epidote-quartz,
epidote-laumontite-quartz,
epidote-wairakite-quartz,
laumontite-epidote,
wairakite-albite-laumontite-quartz,
wairakite-thomsonite-quartz,
laumontite-epidote-albite-quartz,
wairakite-quartz,
laumontite-epidote-wairakite,
laumontite-wairakite-quartz.

Besides these assemblages, veins of wairakite-calcite-quartz assemblage can rarely be found. Width of these veins varies from 0.2 mm to 23 cm.

As far as examined in thin sections under the microscope, no laumontite has been found in the altered granodiorite-porphyrite and the Miocene volcanic rocks themselves. The occurrence of laumontite is confined to veins developed along the shear planes or joints of these rocks. Probably

laumontite-bearing veins were formed at somewhat lower temperature conditions than those prevailed during the alteration of wall-rocks into wairakite-bearing assemblages.

Chemical compositions of wairakite, laumontite and epidote

Pure specimens of wairakite, laumontite and epidote were separated by means of heavy liquids from a wairakite-laumontite-epidote-quartz vein sample collected from an inspection gallery L-56 (560.01 m) for dam construction. Chemical compositions and atomic ratios calculated therefrom of these minerals are represented in Table 1.

The wairakite has only minor amounts of $\text{NaAlSi}_3\text{O}_8\text{-mH}_2\text{O}$ and $\text{NaAlSiO}_4\text{-nH}_2\text{O}$ components. X-ray powder patterns show that the analysed wairakite is a typical monoclinic calcium analogue of analcime (SEKI, 1966, 1968; LIU, 1970).

The density of the wairakite measured by suspension method is 2.274 ± 0.001 . Double refraction is clearly observed and the presence of fine-polysynthetic twins is a characteristic feature of the wairakite under the microscope of crossed-nicols.

The epidote has always idiomorphic crystal form. It is colorless in thin section and has pale straw brown color by the unaided eye. The epidote is rich in clinzoisite molecule as shown under Column B of Table 1.

The laumontite generally shows beautiful tabular crystal form of milky white color by the unaided eye. It is very rich in calcium and poor in sodium and potassium as shown under Column C of Table 1.

Other minerals

The indices of refraction of sodic plagioclase ($n_1=1.532\text{--}1.533$) associated with wairakite, laumontite, epidote, quartz and chlorite in altered granodiorite, dacitic volcanic rocks and veins indicate that the sodic plagioclase belongs to albite, probably having less than 10% anorthite.

Chlorite is pale green under the microscope and shows very low double refraction and optical negative character.

Stability fields of three kinds of wairakite-bearing mineral assemblages

SEKI, TAKEYASU, NAKAJIMA, and ONUKI (1968) have described the alteration of Miocene rhyolite-dacite tuff into wairakite-epidote-albite-chlorite-quartz and wairakite-laumontite-epidote-albite-chlorite-sericite-quartz assemblages from the Hanawa Mining district, north Japan. The present paper reveals that the same type of alteration of the Miocene volcanic rocks is not confined to the Hanawa district. In the Hanawa district, it was thought that the wairakite facies alteration may have been related to an geothermal activity in the Pliocene or Pleistocene time. The occurrence of wairakite in the Kawaji district may also be a "fossil" of a past violent local geothermal activity, probably related to the Pleistocene Nikko volcano. The existing gush of hot-spring in Kawaji Spa may be a trace of the geothermal activity.

Wairakite-epidote-albite-chlorite-quartz (\pm calcite) is a characteristic mineral association in altered rocks of the Kawaji and Hanawa districts. The same mineral association has been reported to occur in a deeper part of drill holes in Wairakei active geothermal area, New Zealand (STEINER, 1968). The epidote-wairakite-albite-adularia-chlorite-quartz association occurs in

Table 1. Chemical compositions and physical properties of wairakite, epidote and laumontite occurred in Miocene pyroclastic rocks of Kawaji damsite area, central Japan.

	A	B	C
	Wairakite	Epidote	Laumontite
SiO ₂	54.16	37.96	51.01
TiO ₂	0.04	0.34	tr.
Al ₂ O ₃	23.59	27.59	21.62
Fe ₂ O ₃	0.39	7.06	0.47
FeO	n. d.	0.35	n. d.
MnO	tr.	0.31	0.01
MgO	tr.	0.02	tr.
CaO	13.01	23.56	11.13
Na ₂ O	0.19	0.04	0.06
K ₂ O	0.23	0.01	0.50
H ₂ O ⁺	8.25	2.11	11.88
H ₂ O ⁻	0.12	0.29	3.45
Total	99.98	99.64	100.13
O	6	12	48
Si	1.972	2.971	15.991
Al	{0.028} 2.000	{0.029} 3.000	{0.009} 16.000
Fe ^{III}	{0.984} 0.995	{2.526} 2.942	{7.974} 8.083
Ti	0.011	0.416	0.109
Ti	0.001	0.020	—
Fe ^{II}	—	0.023	—
Mn	—	0.021	0.001
Mg	—	0.002	—
Ca	0.507	1.975	3.738
Na	0.014	0.007	0.038
K	0.010	0.001	0.200
H ₂ O	1.001	0.551	16.022

Analysed by H. ONUKI

Density	2.274	3.32	n. d.
α	1.500	1.711	1.507
β	n. d.	n. d.	1.517
γ	1.505	1.720	1.520

deeper part of the Ohaki-Broadlands active geothermal area of New Zealand (BROWNE and ELLIS, 1970). The temperature condition under which these wairakite-epidote-albite-chlorite-quartz assemblages were stably formed is 250°C or higher. No laumontite is found in association with this mineral assemblage in Wairakei. Probably water pressures prevailed in deeper part of the Wairakei geothermal area is lower than those to form laumontite-wairakite-albite-association. In Wairakei, a zone characterized by the wairakite-epidote-albite-chlorite·clay-quartz assemblage passes to the mordenite-clay-quartz zone through the wairakite-chlorite·clay-quartz and laumontite-chlorite·clay-quartz zones with decreasing temperature (and with decreasing depth).*

Wairakite-analcime-chlorite·clay-quartz (\pm calcite) must be a characteristic mineral association of rocks in deeper part of Katayama geothermal area (SEKI, ONUKI, OKUMURA and TAKASHIMA, 1969) and in wairakite-chlorite·clay zone of the Wairakei area.** This mineral association must change in deeper parts of active geothermal area to the above-noted wairakite-epidote-albite-chlorite·clay-quartz association by

* The high concentrations of potassium, sodium and carbon-dioxide in rocks and in hydrothermal liquids would form the assemblages of K-feldspar, K-mica, Na-feldspar and calcite instead of the assemblages of wairakite, epidote and chlorite at the same range of temperature and pressure conditions (ZEN, 1961; ELLIS, 1967; MUFFLER and WHITE, 1969; BROWNE and ELLIS, 1970).

** No analcime has been reported from rocks of the shallower part than the wairakite-epidote-albite-chlorite·clay zone of Wairakei and from rocks came from drill-holes of Katayama geothermal area. However, it is noticeable that wairakites separated by heavy liquids from Katayama rocks often show extraordinarily weak X-ray diffraction peaks of 200 and 004. Under the microscope a small amount of zeolite having low refractive index ($=1.48-1.49$) and very weak double refraction can be observed to be associated with wairakite with relatively high refractive index and characteristic polysynthetic twin in these Katayama rocks. It looks like analcime. Complete purification of wairakite from rocks having both of wairakite and analcime is practically impossible. It is also impossible to definitely determine the presence of a small amount of analcime by only X-ray powder pattern, because almost completely the X-ray powder peaks of analcime are concealed within those of wairakite. However, the above-noted evidences show that analcime must be stably present in some, not all, of wairakite-bearing rocks of the Katayama geothermal area.

increasing temperature. The transition temperature of these two mineral associations in 230—240°C at Wairakei. Maximum temperature measured in Katayama geothermal area in which neither epidote nor albite is associated with wairakite is 210°C. Both of these two mineral associations wairakite-epidote-albite-chlorite·clay-quartz and wairakite-analcime-chlorite·clay-quartz may belong to the wairakite facies defined by SEKI (1969). The transitional temperature between the wairakite facies and the higher grade facies has been assumed to be 200°C at pressure of 0.5 kb (SEKI, 1969). This temperature, however, may have to be shifted by almost 70—100°C to the higher temperature side as indicated by LIU (1970) and COOMBS (1970).

The Wairakite-analcime solid solution minerals are found forming altered volcanic rocks in the higher-grade part of Zone II (laumontite subfacies of the zeolite facies) and in Zone III (the pumpellyite-prehnite facies) of the Tanzawa Mountains, central Japan (SEKI and OKI, 1969; SEKI, OKI, ODAKA and MATSUDA, 1969). Wairakite-analcime solid solution minerals are stably associated with the following mineral assemblages: chlorite-quartz,

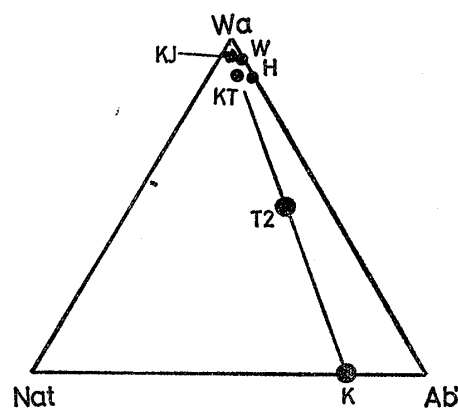
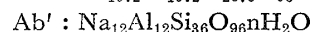
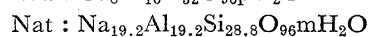
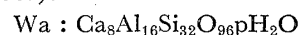


Fig. 3. Chemical relation between wairakites (Kj : Kawaji (this paper), W : Wairakei (STEINER, 1955), KT : Katayama (SEKI, ONUKI, OKUMURA and TAKASHIMA, 1969), H : Hanawa (SEKI, TAKEYASU, NAKAJIMA and ONUKI, 1968), Kasukabe analcime (K) (HASHIMOTO, 1964) and Tanzawa intermediate mineral (T-2) (SEKI and OKI 1969).



Calculation procedure of the Wa-Nat-Ab' ratio of wairakite-analcime solid solutions has been proposed by SEKI (1971).

chlorite-quartz-calcite,
 chlorite-albite-quartz,
 chlorite-albite-prehnite-laumontite-quartz,
 chlorite-albite-epidote-sericite-quartz,
 chlorite-albite-prehnite-epidote-pumpellyite-
 quartz,
 chlorite-prehnite-quartz-calcite,
 chlorite-prehnite,
 chlorite-prehnite-epidote-sericite-quartz,
 chlorite-prehnite-epidote-albite-sericite-quartz,
 chlorite-laumontite-quartz.

One of these wairakite-analcime solid solutions (T-2) has the following chemical composition (SEKI and OKI, 1969):

$\text{Na}_{7.50}\text{K}_{0.13}\text{Ca}_{3.89}\text{Mg}_{0.38}\text{Al}_{14.79}\text{Si}_{32.87}\text{O}_{96}17.44\text{H}_2\text{O}$.
 This specimen T-2 has an intermediate composition between an analcime reported from Kasukabe by HASHIMOTO (1964) and wairakites (Fig. 3). It must be noted that the intermediate minerals T-2 has larger amount of water than analcimes and wairakites. Wairakites reported from Wairakei (STEINER, 1955), Hanawa (SEKI, TAKEYASU, NAKAJIMA and ONUKI, 1968), Tanzawa (SEKI, OKI, MATSUDA, MIKAMI and OKUMURA, 1969), KATAYAMA (SEKI, ONUKI, OKUMURA and TAKASHIMA, 1969) and KAWAJI (this paper) and Kasukabe analcime have 16.0 H₂O per O=96 calculated on the anhydrous basis. Consequently, it can be said that T-2 specimen is almost equal in che-

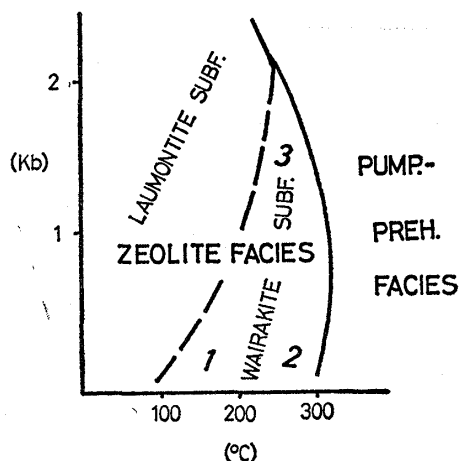


Fig. 4. Possible stability fields of three wairakite-bearing mineral assemblages ($P_{\text{solid}} > P_{\text{water}}$).

- 1 : Wairakite-analcime-chlorite-clay mixed layer mineral-quartz assemblage.
 - 2 : Wairakite-epidote-albite-chlorite-quartz assemblage.
 - 3 : Wairakite-analcime solid solution-epidote-chlorite-quartz assemblage.
- (Facies and subfacies boundaries modified from COOMBS (1969) and SEKI (1969)).

mical composition to the mixture of 50% Kasukabe analcime and 50% wairakite plus water. It is probable to think that under constant temperature and solid pressure conditions wairakite plus analcime react to form a one phase of intermediate chemical composition of these two phases with increasing water pressure. Thus, *Wairakite-analcime solid solution mineral-epidote-chlorite-quartz* (\pm calcite, laumontite, albite, prehnite and pumpellyite) must be the characteristic mineral association in rocks suffered the metamorphism of the physical conditions near the triple field (point) between the zeolite facies (laumontite subfacies), the pumpellyite-prehnite facies and the wairakite facies (or the triple field between the laumontite subfacies and the wairakite subfacies of the zeolite facies and the prehnite-pumpellyite facies (COOMBS, 1969)). As shown in Fig. 4.

References

- BROWNE, P. R. L. and A. J. ELLIS (1970), The Ohaki-Broadlands hydrothermal area, New Zealand: Mineralogy and related geochemistry. *Amer. Journ. Sci.*, vol. 269, p. 97-131.
- COOMBS, D. S. (1969), Present status of the zeolite facies. *Advance in Chemistry, Amer. Chem. Society*, p. 556-566.
- (1970), On the mineral facies of spilitic rocks and their genesis (in press).
- ELLIS, A. (1967), The chemistry of some explored hydrothermal systems, in Barnes, H. L., ed., *Geochemistry of Hydrothermal Ore Deposits: New York, Holt, Rinehart and Winston*, p. 465-512.
- HASHIMOTO, M. (1964), Analcime-veins in mudstone, 2,703 m beneath the surface of Kasukabe, Saitama Prefecture, Japan. *Jour. Geol. Soc. Japan*, vol. 70, p. 226-228.
- IMAI, I. (1952), Geology of the Ikari dams site (in Japanese). *Bull. Geol. Surv. Japan*, vol. 3, p. 387-390.
- IWAO, S. and IMAI, I. (1951). 1/75,000 Geological map sheet (Shiobara) and its explanatory text (in Japanese with English abstract). *Geol. Surv. Japan*.
- LIU, J. G. (1970), P-T stabilities of laumontite, wairakite, lawsonite and related minerals in the system $\text{CaO-Al}_2\text{O}_3-2\text{SiO}_2-\text{H}_2\text{O}$. *Amer. Min.* (in press).
- MUFFLER, L. J. P. and D. E. WHITE (1969), Active metamorphism of Upper Cenozoic sediments in the Salton Sea geothermal field and the Salton Trough, Southeastern California. *Bull. Geol. Soc. Amer.*, vol. 80, p. 157-182.
- SEKI, Y. (1966), Wairakite in Japan (I) (II), *Journ. Assoc. Japan. Min. Petr. Economic Geol.*, vol. 55, p. 254-261 and vol. 56, p. 30-39.
- (1968), Synthesized wairakites: their difference from natural wairakites. *Jour. Geol. Soc. Japan*, vol. 74, p. 457-458.
- (1969), Facies series in low-grade metamorphism. *Jour. Geol. Soc. Japan*, vol. 75, p. 255-266.
- (1971), Some physical properties of analcime-

- wairakite solid solutions. *Jour. Geol. Soc. Japan*, vol. 77, p. 1—8.
- , and Y. OKI (1969), Wairakite-analcime solid solutions from low-grade metamorphic rocks of the Tanzawa Mountains, central Japan. *Min. Jour.* vol. 6, p. 36—45.
- , OKI, Y., MATSUDA, T., MIKAMI, K. and OKUMURA, K. (1969), Metamorphism in the Tanzawa Mountains, central Japan. *Journ. Assoc. Japan. Min. Petr. Economic Geol.*, vol. 61, p. 1—25, 50—75.
- , Y. OKI, J. ODAKA and T. MATSUDA (1969), Geological research for hot spring resources in the Kannokawa area, Tsukui-machi, Tanzawa Mountains (in Japanese with English abstract). *Bull. Hot-Spring Res. Inst. Kanagawa prefecture*, p. 1—28.
- , ONUKI, H., OKUMURA, K. and TAKASHIMA, I. (1969), Zeolite distribution in the Katayama geothermal area, Onikobe, Japan. *Jap. Jour. Geol. Geogr.*, vol. 40, p. 63—79.
- , TAKEYASU, T., NAKAJIMA, M. and ONUKI, H. (1968), Wairakite from Hanawa Mine, northern Japan. *Jour. Assoc. Japan. Min. Petr. Economic Geol.*, vol. 59, p. 236—246.
- STEINER, A. (1955), Wairakite, the calcium analogue of analcime, a new zeolite mineral. *Min. Mag.*, vol. 30, p. 691—698.
- (1968), Clay minerals in hydrothermally altered rocks at Wairakei, New Zealand. *Clays and Clay Minerals*, vol. 16, p. 193—213.
- ZEN, E-an, (1961), The zeolite facies: an interpretation. *Amer. Journ. Sci.*, vol. 259, p. 401—409.

栃木県川治ダム予定地のワイラカイト相変質

関 陽 太 郎・大 貫 仁

(要 旨)

川治ダム予定地付近に発達する花崗閃緑岩—玢岩と中新世火山岩類は、ひろくワイラカイト、曹長石、エピソード、緑泥石および石英より成る鉱物組合せによってひろく変質している。それらの岩石の中には、ワイラカイ

ト、ローモンタイト、曹長石、エピソード、石英より成る脈が発達する。ワイラカイト、ワイラカイト・アナルサイム固溶体を含む変成相には3つの亜相を認めることができる。