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# "Yugawaralite", A New Zeolite

By

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## Preface

While studying zeolites from Yugawara Hot Spring area, Kanagawa Prefecture, in 1930, SAKURAI found a zeolite which was quite different from Epistilbite, Desmine, Laumontite, Ptilolite, Chabazite etc. By the morphological study and chemical analysis, I assumed this to be a new unknown zeolite. Later in 1948, I was able from detailed study with HAYASHI's co-operation, to certify this be an unknown new mineral.

## Locality and Occurrence

This unknown new mineral is found in rocks near "Fudo-no-taki" Fall (Fig. 1 Atami 1/25.000 Ac) on the right side of the road to Okuyugawara Hot

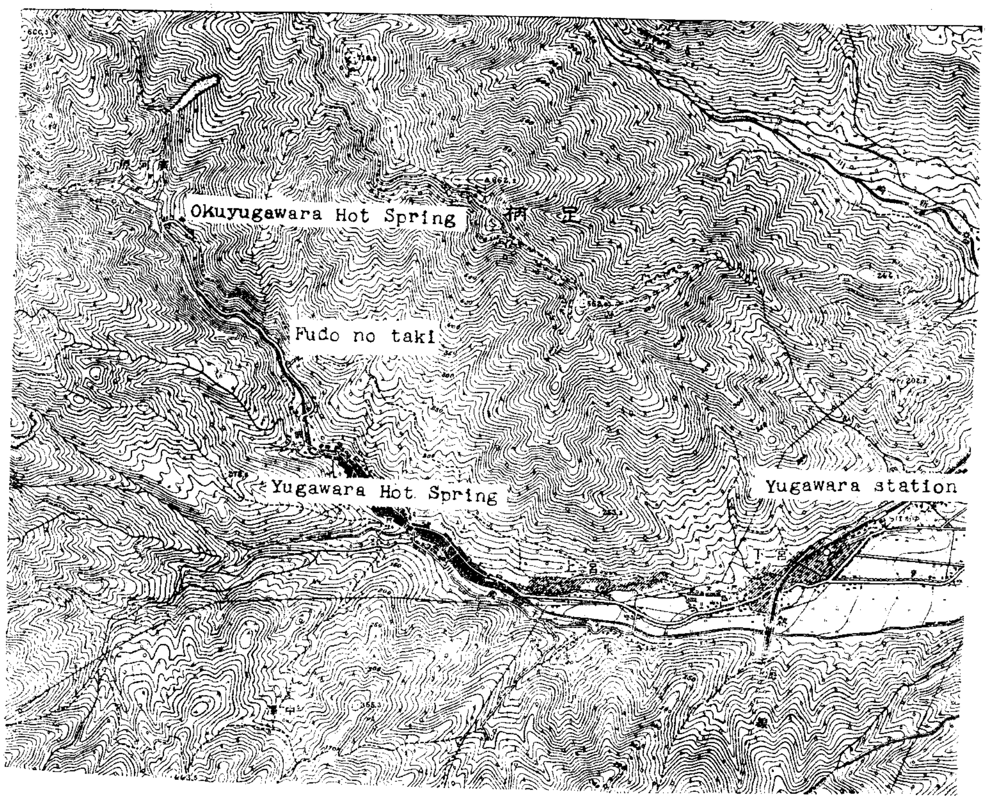


Fig. 1

Spring leading along the Chitose River, this place is about 300 m from Yugawara Hot Spring. Along the Chitose River, "Yugashima" series (lower miocene, tertiary) develops, and "Tenshozan" Andesite, develops over it. Yugawaralite occurs, forming net work, in the Andesite tuff of this Yugashima series, which, being metamorphosed very much by hot spring action, is grayish green near the outcrop. The net work vein is 5~15 mm wide and has many pretty crystals in its cavity. Co-existing mineral is Rock crystal, and a few amount of Laumontite, Chabazite, Ptilolite.

## Crystal

Monoclinic, flat crystal. Reaches to 5 mm (a-axis), 2 mm (b-axis) and 10 mm (c-axis), Crystal faces are as follows:

*a* (100), *b* (010), *c* (001), *m* (110), *l* (120), *u* (011), *p* ($\bar{1}$11).

*b* is considerably developed. The crystal is slightly longer along c-axis. Normally, the crystal consists of *b, m, a, c, u* and *p,* (Fig. 2) but sometimes, of *b, m, l, u* and *p.* (Fig. 3). Stereo projection is shown on Fig. 4. Often, two or more crystals are in parallel growth along (010). Irregular mass of many crystals are found in the cavity of the vein. Data measured by GOLDSCHMDT's double circle goniometer are on Table I.

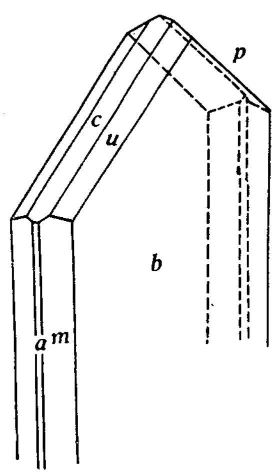


Fig. 2

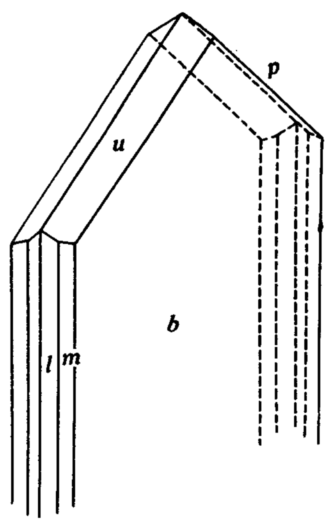


Fig. 3

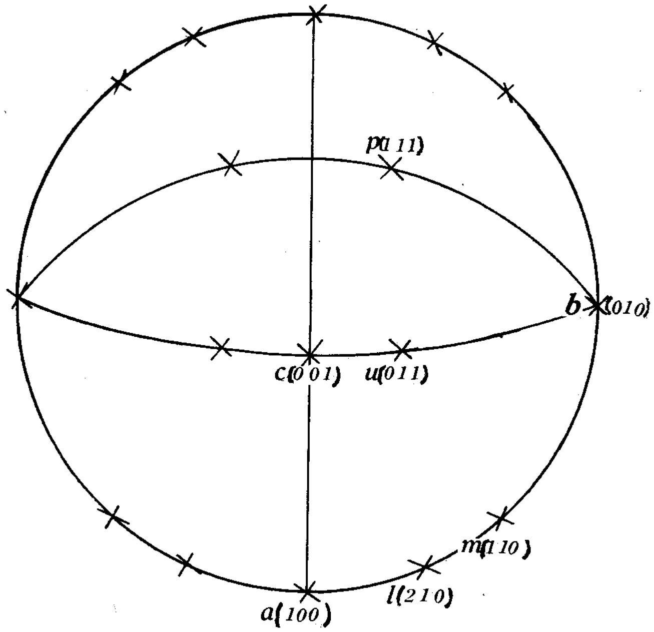


Fig. 4

Table I

| | Observed | | Calculated[1] | | Refrection[2] Strength | Size of[3] Face |
|---|---|---|---|---|---|---|
| | ρ | φ | ρ | φ | | |
| c (001) | 21°32′* | 89°56′* | 21°32′ | 90°00′ | A | M |
| b (010) | 90°00′ | 0°00′ | 90°00′ | 0°00′ | A | V. L |
| a (100) | 90°00′ | 90°00′ | 90°00′ | 90°00′ | A | S |
| m (110) | 90°00′ | 47°48′* | 90°00′ | 47°49½′ | B | M |
| l (210) | 90°00′ | 65°33′ | 90°00′ | 65°36′ | B | M |
| p ($\bar{1}$11) | 54°30′* | −59°25′* | 54°23′ | −59°20′ | A | M |
| u (011) | 39°30′ | 28°58′ | 39°10½′ | 29°05′ | A | M |

1) Calculated from marked * by formula.
2) A: strong. B: middle. C: weak.
3) V.L: very large, M: middle, S: small.

On the surface *b* are often found streaks parallel to principal axis. From these data, axial ratio and axial angle are obtained as follows, which are different from those of other zeolites.

$$a : b : c = 0.9758 : 1 : 0.7129$$
$$\beta = 68°28'$$

## Physical Properties

Colorless transparent or white semi-transparent, strong vitreous lustre. On *b* has often iridescence. Streak: white. Cleavage: incomplete on *b*. Hardness: $4^1/_2$. Specific gravity: 2.201 (SAKURAI), 2.198 (HAYASHI).

## Optical Properties

Optically positive, optic axial plane: perpendicular to (010). 2v: 56°, 68°, 76°, 89° etc. (different by the measured specimens) Optically negative is not observed.

Refraction index: $\alpha=1.495$ $\beta=1.497$ $\gamma=1.504$ $\gamma-\alpha=0.009$

Shows straight extinction, elongation is negative.

Absorbtion: $\rho < \nu$

## Chemical Properties

Results of chemical analysis by Messrs. A. HAYASHI and N. SIMODA are shown on Table II (1, 2).

Table II

| | 1 | 2 |
|---|---|---|
| $SiO_2$ | 57.94% | 58.44% |
| $Al_2O_3$ | 17.65 | 17.31 |
| $Fe_2O_3$ | 0.35 | 0.36 |
| CaO | 9.79 | 9.75 |
| MgO | 0.86 | 0.42 |
| MnO | — | — |
| $Na_2O$ | 0.38 | 0.36 |
| $K_2O$ | 0.41 | 0.12 |
| $H_2O(+)$ | 10.70 | 10.55 |
| $H_2O(-)$ | 1.80 | 2.03 |
| | 99.88 | 99.34 |
| | by A. Hayashi (1948) | by N. Shimoda (1951) |

The following atomic ratio of the elements in unit cell of Yugawaralite is calculated from volume of unit cell, specific gravity, etc.

Si 19.60, Al 6.99, Fe 0.08, Mg 0.42, Ca 3.56, Na 0.24, K 0.16, $H_2O$ 14.11, O 54.00.

From these data is obtained molecular formula $Ca_4Al_7Si_{20}O_{54} \cdot 14H_2O$. Furthermore, this zeolite has a specific characteristic that is not attacked by cold or hot hydrochloric acid and does not produce gelatine, the latter is decided from non-coloration by methylene-blue.

Namely, Yugawaralite, like Mordenite and Ptilolite, belongs to zeolite group which in not attacked by hydrochloric acid.

## Study By X-Ray

The following lattice constant is determined from X-ray oscillation photograph around every axis. Camera radius: 3.41 mm, Anticathode: Mo ($MoK_\alpha$, $\lambda$=0.71Å)

$$a_0=13.26\text{Å},\qquad b_0=13.65\text{Å}\qquad c_0=9.73\text{Å}\qquad \beta=68°30'$$

Axial ratio from these data a: b: c=0.9715: 1: 0.7136 coincides very well with that from goniometric data.

The lines of Debye-Scherrer photograph of Yugawaralite do not coincide with those of Mordenite, Heulandite, Epistilbite and Desmine. Table III shows lattice distance and intensity I of these zeolites.

Table III

| Yugawaralite | | Mordenite | | Epistilbite | | Heulandite | | Desmine | |
|---|---|---|---|---|---|---|---|---|---|
| $CoK_\alpha$ D=2.865cm | | $CuK_\alpha$ D=5.50cm | | $FeK_\alpha$ D=2.865cm | | $CuK_\alpha$ D=5.019cm | | $CuK_\alpha$ D=2.865cm | |
| d (Å) | I | d (Å) | I | d (Å) | I | d (Å) | I | d (Å) | I |
| | | 10.00 | 8 | 7.65 | 8 | 9.73 | 1 | 5.82 | 5 |
| | | 8.26 | 1 | 6.11 | 10 | 8.18 | 8 | 5.09 | 6 |
| | | 7.26 | 8 | 5.25 | 10 | 4.89 | 1 | 4.57 | 6 |
| 6.48 | 6 | 6.41 | 6 | 4.53 | 9 | 4.45 | 2 | 4.03~4.22 | 10 |
| 5.69 | 10 | 5.48 | 5 | 3.78 | 8 | 3.88 | 10 | 3.83 | 6 |
| 5.23 | 5 | 4.95 | 8 | 3.59 | 2 | 3.25 | 1 | 3.45 | 6 |
| 4.71 | 9 | 4.48 | 10 | 3.50 | 2 | 3.07 | 4 | 3.04 | 10 |
| | | | | 3.18 | 3 | 2.92 | 10 | 2.79 | 4 |
| 4.27 | 4 | 4.30 | 6 | 2.96 | 4 | 2.77 | 5 | 2.61 | 4 |
| 3.90 | 4 | 4.14 | 5 | 2.77 | 6 | 2.68 | 4 | 2.36 | 3 |
| 3.72 | 10 | 3.74 | 10 | 2.60 | 2 | 2.37 | 2 | 2.08 | 5 |
| 3.52 | 3 | 3.51 | 10 | 2.31 | 4 | 1.94 | 6 | 1.84 | 3 |
| 3.20 | 2 | 3.33 | 5 | 2.17 | 5 | 1.77 | 5 | 1.68 | 2 |
| 3.07 | 10 | 3.13 | 5 | 2.11 | 2 | 1.71 | 1 | 1.60 | 6 |
| 2.91 | 4 | 2.98 | 3 | 2.02 | 2 | 1.64 | 1 | 1.56 | 5 |
| | | 2.72 | 4 | 1.92 | 1 | 1.48 | 3 | 1.46 | 3 |
| | | 2.48 | 3 | 1.85 | 4 | 1.35 | 2 | 1.37 | 2 |
| | | 2.21 | 2 | 1.79 | 2 | 1.27 | 2 | 1.31 | 5 |
| | | 2.05 | 2 | 1.71 | 1 | 1.23 | 1 | 1.24 | 5 |

We obtain n=1.04 by calculation from specific gravity (2.20), molecular formula (Ca, Mg, Na, K)$_{4.83}$ (Al, Fe)$_{7.07}$ Si$_{19.60}$ $O_{54}$·14.11 $H_2O$ (Table II, 1) and volume of unit cell which is obtained from the above mentioned axial length and axial angle of unit cell. Therefore the number n of molecules in unit cell of Yugawaralite is 1. Crystal constants and space groups of zeolites now known, are found in Table IV.

Table IV

| | Axial length. (unit Å) | | | Axial angle. | Space group. | | |
|---|---|---|---|---|---|---|---|
| | $a_0$ | $b_0$ | $c_0$ | | | | |
| Mordenite | 18.25 | 20.35 | 7.50 | —— | $D_{2h}^{17}$ or $C_{2v}^{12}$ | W. H. Taylor | (1938) |
| Heulandite. | 7.45 | 18 70 | 15.85 | 88°36′ | $C_{2h}^{3}$ | T. Wyart | (1933) |
| Epistilbite. | 14.90 | 17.87 | 10.32 | 89°20′ | | A. Hayashi | (1949) |
| Yugawaralite. | 13.26 | 13.65 | 9.73 | 68°28′ | | K. Sakurai<br>A. Hayashi | (1949) |
| Desmine. | 13.60 | 18.13 | 11.29 | 52°±½′ | $C_{2h}^{3}$ | T. Sekania<br>T. Wyart | (1937) |
| Harmotome. | 9.80 | 14.10 | 8.66 | 55°10′ | $C_{2h}^{2}$ or $C_{2}^{2}$ | T. Sekania<br>T. Wyart | (1937) |
| Phillipsite. | 10.00 | 14.25 | 8.62 | 54°20′ | $C_{2h}^{2}$ or $C_{2}^{2}$ | P. Chatelain<br>T. Wyart | (1938) |
| Gismondite. | 13.68 | 14.28 | 10.60 | —— | $D_{2h}^{19}$ or $D_{2h}^{22}$ | O. Kraus | (1939) |
| Stellerite. | 17.52 | 18.34 | 13.43 | —— | $V_{h}^{23}$, V | K. Takane | (1937) |
| Chabazite. | 13.75 | —— | 14.95 | Rhombohedral<br>a=9.37<br>α=94°34′ | $D_{3d}^{5}$ | T. Wyart | (1933) |
| Thomsonite. | 13.04 | 13.06 | 13.22 | —— | $C_{2v}^{10}$ | T. Wyart | (1933) |
| Gonnardite. | 13.35 | 13.35 | 6.65<br>(?) | —— | | M. H. Hey<br>F. A. Banister | (1932) |
| Natrolite. | 18.3 | 18.6 | 6.57 | —— | $C_{2v}^{19}$ | W. H. Taylor<br>C. A. Meek | (1933) |
| Mesolite. | 5.67 | 6.54 | 18.44 | 90°±20′ | $C_{2}^{3}$ or $C_{2v}^{19}$ | T. Wyart | (1933) |
| Scolesite. | 18.44 | 18.90 | 6.53 | —— | $C_{s}^{4}$ = $C_c$ | T. Wyart | (1933) |
| Edingtonite. | 9.59 | 9.59 | 6.53 | —— | $V_{d}^{3}$ | M. H. Hey<br>F. A. Banister | (1934) |
| Analcime. | 13.68 | —— | —— | —— | $O_{h}^{10}$ | W. Haritng | (1931) |

molecular formula
(Table II, I) and
ioned axial length
molecules in unit
ıps of zeolites now

| | |
|---|---|
| V. H. Taylor | (1938) |
| Γ. Wyart | (1933) |
| A. Hayashi | (1949) |
| C. Sakurai<br>A. Hayashi | (1949) |
| C. Sekania<br>C. Wyart | (1937) |
| C. Sekania<br>C. Wyart | (1937) |
| P. Chatelain<br>C. Wyart | (1938) |
| O. Kraus | (1939) |
| C. Takane | (1937) |
| Γ. Wyart | (1933) |
| Γ. Wyart | (1933) |
| M. H. Hey<br>F. A. Banister | (1932) |
| W. H. Taylor<br>C. A. Meek | (1933) |
| Γ. Wyart | (1933) |
| Γ. Wyart | (1933) |
| M. H. Hey<br>F. A. Banister | (1934) |
| W. Haritng | (1931) |



## Thermal Properties

Mr. M. Koidzumi gives thermal data about several zeolites as follows:

a). Curve of Heating Loss (Fig. 5). The curve of Yugawaralite (Fig. 5, A) is clearly different from that of Heulandite (Fig. 5, C), Deimine (Fig. 5, D) or Ptilolite (Fig. 5, E), and generally de-hydrates in four steps and is somewhat similar to that of Laumontite (Fig. 5, B). Namely, Yugawaralite loses rather rapidly more than 3 molecule of water till 100°C, then slowly 6 till 400°C, next rapidly 4 between 400°C and 450°C, later very slowly 1~1.5 molecule.

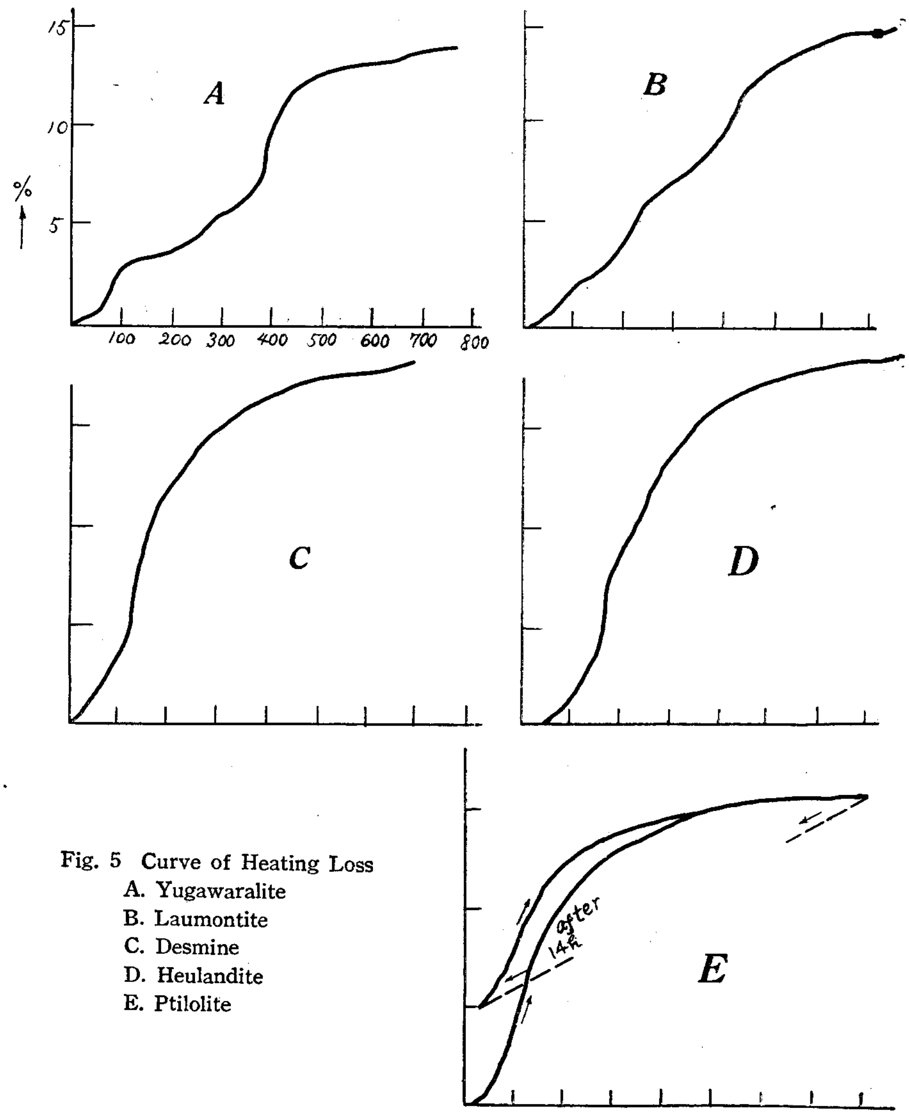


Fig. 5 Curve of Heating Loss
A. Yugawaralite
B. Laumontite
C. Desmine
D. Heulandite
E. Ptilolite

b). Curve of Differential Thermal Analysis (Fig. 6). This curve of Yugawaralite (Fig. 6, C) is very characteristic and quite different from that of other zeolites.

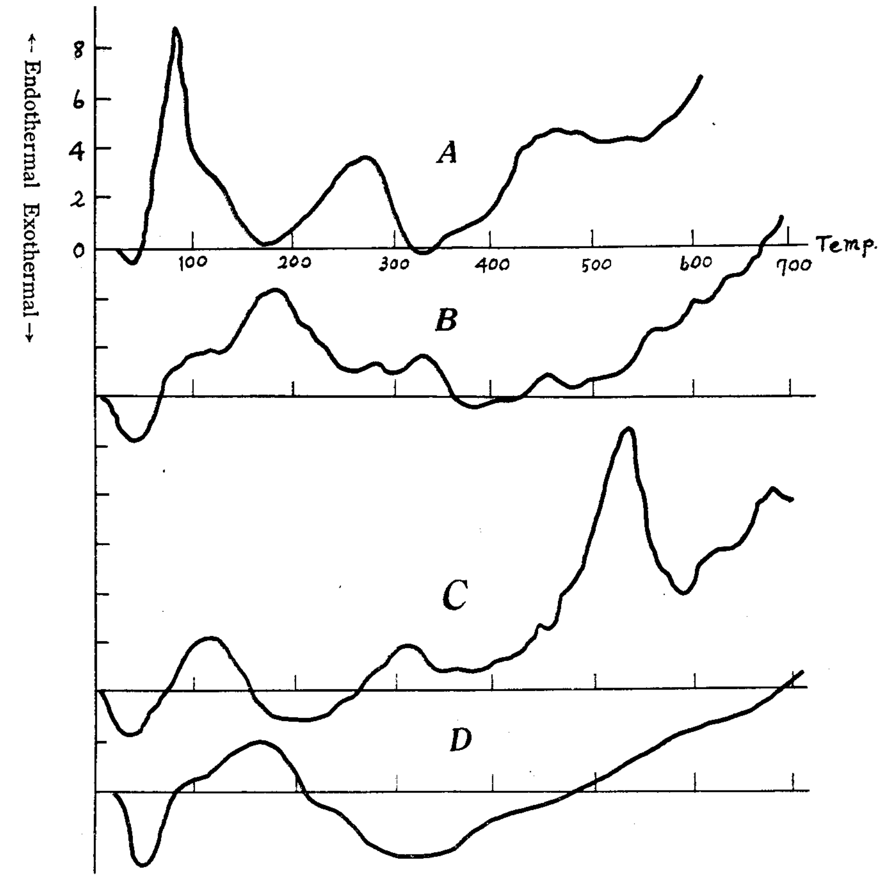


Fig. 6 Curve of Differential Thermal Analysis.
A Laumontite, B Desmine, C Yugawaralite,
D Ptilolite.

## Summary

1). Morphological: Monoclinic. a : b : c=0.9748 : 1 : 0.7129 $\beta$=68°28′. Crystal faces: *c* (001), *b* (001), *a* (100), *m* (110), *l* (120), *u* (011), *p* ($\bar{1}$11). *b* (010) is large. Flat.

2). Physical: Transparent, colorless or semitransparent, white. Strong vitreous lustre. Cleavage: imperfect on (010). Hardness: 4.5 Specific gravity: 2.20.

3). Optical: Optically positive, Optic axial plane ⊥ (010), 2V: 56°~89°
$\alpha$=1.495, $\beta$=1.497, $\gamma$=1.504

4). Chemical: $Ca_4Al_7Si_{20}O_{54} \cdot 14H_2O$. Not attacked by HCl.

5). X-ray: Lattice constant $a_0=13.26$, $b_0=13.69$, $c_0=9.73$, $\beta=68°28'$. Unit cell contains 1 molecule.
Debye-Scherrer photograph does not coincide with that of other zeolites.

6). Thermal: Yugawaralite dehyrates in 4 steps like Laumontite. Curve of differential thermal analysis is characteristic. Necessary to pay attention to endothermal reaction at 540°.

## Conclusion

By the data above mentioned, we can not identify this mineral with the other zeolites, and we may consider this to be a new zeolite. We wish to it "Yugawaralite" after its locality.

Finally, the authors hope to express their hearly thanks to Dr. T. Ito for his long kind guidance, and also to Messrs. E. Sorida, M. Koidzumi, N. Shimoda, T. Yokota, K. Ishioka, T. Kawai and K. Nagashima.