

*The pumpellyite mineral series.*

By D. S. COOMBS, M.Sc., Ph.D., F.G.S.

Geological Department, University of Otago, New Zealand.

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## INTRODUCTION.

THE bluish-green epidote-like mineral, pumpellyite, was first described by Palache and Vassar (1925) from the copper-bearing amygdaloidal lavas of the Lake Superior region. It has since been reported from many parts of the world in rocks which have undergone low grades of metamorphism, especially types such as amygdaloidal spilites and diabases (Burbank, 1927; Waldmann, 1934; Tsuboi, 1936; Quitzow, 1935, 1936; Klein, 1939; Lacroix, 1942; Sæther, 1942; Zavaritsky, 1944; Korzhinsky, 1944; de Roever, 1947a, b; Kvasha, 1950) and in glaucophane-schists and even eclogites in which pumpellyite is often associated with lawsonite (Irving, Vonsen, and Gonyer, 1932; Quitzow, 1935, 1936; Lacroix, 1942; Switzer, 1945; de Roever, 1947a, b, 1950). W. P. de Roever (1947a, 1950) has suggested a pumpellyitic facies of metamorphism as a lower-grade equivalent of his lawsonite-glaucophane subfacies, characterized by high confining pressure, but subordinate shearing stress and thermal influence, and passing into un-metamorphosed rocks. Nevertheless, pumpellyite can certainly form under conditions of strong shearing stress accompanied by relatively low temperatures (Hutton, 1937; Amies, 1950).

Pumpellyite is widespread in southern New Zealand and has been described from the less highly metamorphosed rocks of the chlorite zone by Hutton (1937, 1940), Turner (1939), Amies (1950), and Reed (1950).

Professor C. E. Tilley has kindly allowed the writer to examine material collected by him from amygdaloidal pillow lavas at Howell's Point, Southland. The present writer has found that in the Taringatura district of Southland the mineral occurs with albite in the lower members of a thick series of tuffaceous greywackes which are quite unmetamorphosed apart from such changes as zeolitization of vitric tuffs and albitization of plagioclase. Some of this Taringatura pumpellyite has hitherto unrecorded optical properties.

In spite of the wide distribution of pumpellyite, knowledge of its crystallography has been very scanty. This is undoubtedly due to a tendency towards unusually poor crystal development. All observers have noticed elongation parallel to the *b* crystal axis (optical  $\beta$ ) and although Palache and Vassar thought that the mineral was probably orthorhombic, its monoclinic nature was correctly deduced by Burbank (1927). Several writers have reported a good cleavage in the zone of the *b*-axis and they have tentatively considered it as a basal cleavage. It appears from the present work that the basal plane of Irving, Vonsen, and Gonyer, and of Tsuboi (herein adopted) is not that of other workers such as Palache and Vassar, Burbank, Quitzow, Korzhinsky, and de Roever.

J. E. de Villiers (1941) made a careful study of the optical and crystallographic properties of a fairly well-crystallized mineral from Johannesburg which he considered to be pumpellyite, but the intercleavage angle ( $65-70^\circ$ ) does not agree with that found in the present study ( $82.4^\circ$ ), and optical properties of the Johannesburg mineral are notably discordant from those of all proven pumpellyites. It is clear that de Villiers was working with some other mineral, and his paper will not be referred to further.

Knowledge of the optical properties has been a little more detailed than that of the morphology of pumpellyite, but it is still far from complete. The orientation of the  $\alpha$  and  $\gamma$  optical directions with respect to the 'basal plane' as recorded by Vonsen and co-workers and by Tsuboi contrast sharply with that described by the other workers quoted above. One major discrepancy was recently removed when Nel, Strauss, and Wickman (1949) proved that the mineral noted by Söhngé (1945) as 'pumpellyite (?)' is a new species, lombaardite, with a distinctive X-ray powder pattern. In view of our rather limited knowledge of pumpellyite, an X-ray and optical study has been undertaken of all available material, and some new partial chemical analyses have been performed.

## DETERMINATION OF CELL-SIZE AND SPACE-GROUP.

Through the courtesy of Dr. F. A. Bannister a small fragment of a specimen in the British Museum (B.M. 1926,226), was made available for study. It was originally forwarded to the Museum by Professor Palache and comes from his type locality in the Lake Superior region, Calumet and Hecla mine, Houghton County, Michigan. The fragment consists of an aggregate of small epidote crystals on matrix, some of the epidote carrying a bluish-green dusting of tiny bladed needles of pumpellyite. The specimen had been treated with dilute hydrochloric acid to remove calcite. Microscopic examination proved that the pumpellyite needles are sub-parallel and distorted growths, and they are heavily striated parallel to the length. Several of the more homogeneous-looking grains were set up for oscillation photographs about their long axes. All gave streaks along the powder rings, but one lath-shaped grain,  $0.27 \times 0.09 \times 0.04$  mm., gave two or three maxima in each streak sufficiently well defined to allow interpretation. This crystal was used for zero-layer normal-beam and first and second layer-line equi-inclination Weissenberg photographs with the crystal oscillating about the  $b$ -axis (fig. 1). Exposures of up to 90 hours were required using filtered  $\text{Cu-K}\alpha$  radiation. Although lattice planes are represented in the photographs by long streaks, maxima are well defined and the streaks themselves are very narrow, allowing accurate measurement of  $\xi$  values, both for  $K\alpha_1$  and  $K\alpha_2$  reflections. Between the maxima the streaks are continuous, indicating twisting of lattice planes about the  $b$ -axis.

The cell-edge  $b$  was estimated from  $\text{Cu-K}\alpha$  and  $\text{Cu-K}\beta$  layer-lines in oscillation photographs as 5.95 and 5.97 Å, respectively. Using the provisional value so obtained together with values of  $a$ ,  $c$ , and  $\beta$  obtained as below, lattice spacings were calculated for planes giving strong spots on the Weissenberg photographs. In this way many of the lines on a 19-cm. powder photograph were successfully indexed. Systematic discrepancies between observed and calculated powder line spacings of the forms  $(02l)$ ,  $(h2l)$  led to the refined value  $b$  5.94 Å. The dimensions  $a$ ,  $c$ , and  $\beta$  were determined from high-angle reflections in the zero-level Weissenberg photographs, with the probable limits of error listed in the tabulation below. The unit cell chosen is  $A$ -face-centred,  $\beta$  97.6°. This choice of cell emphasizes relationships with the epidote group and also with lawsonite. It accords with the choice of basal plane by Irving, Vonsen, and Gonyer, and by Tsuboi, but not with that of the other

workers, which becomes  $\{100\}$ . The wave-lengths used in the calculations were  $\text{Cu-K}\alpha = 1.540 \text{ \AA}$ .,  $\text{Cu-K}\beta = 1.544 \text{ \AA}$ . The Calumet pumpellyite cell is as follows:

$$a \ 8.81 \pm 0.02, \ b \ 5.94 \pm 0.01, \ c \ 19.14 \pm 0.02 \text{ \AA}., \ \beta = 97.6^\circ \pm 0.2^\circ.$$

Some difficulty in the interpretation of the oscillation photographs was caused by the presence of streaked spots falling irregularly between

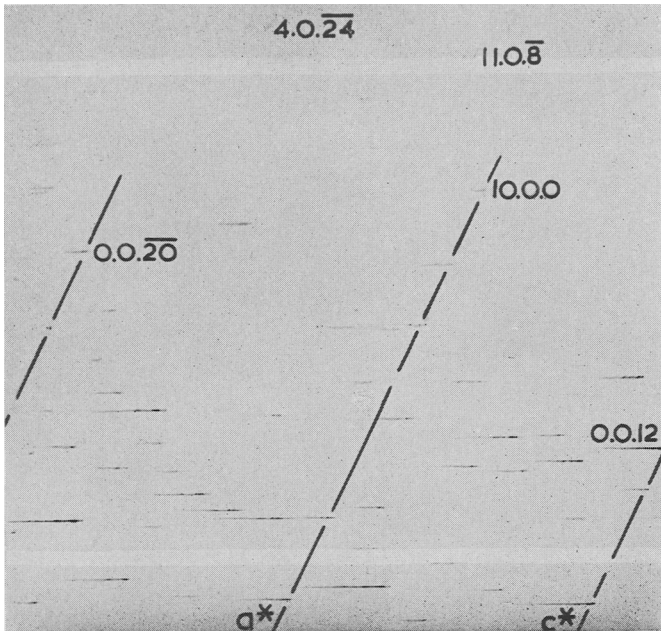


FIG. 1. Zero-layer, normal-beam Weissenberg photograph of pumpellyite from Calumet, Michigan, with the crystal oscillated about the  $b$ -axis. The streaking of reflections is due to twisting of the crystals about  $b$ .  $\text{Cu-K}\alpha$  radiation, Ni filter. Camera diameter 6 cm.

the layer-lines. After careful indexing, it was found that all the anomalous streaks could be satisfactorily accounted for as being due to specified strongly reflecting lattice planes which were in a position to reflect in disoriented parts of the crystal, but not in that part of the crystal responsible for the main maximum in each streak.

From a study of some hundreds of reflections of the form  $h0l$ ,  $h1l$ ,  $h2l$ , it was found that the condition for reflection from a plane  $hkl$  is that  $k+l = 2n$ . The structure must therefore belong to one of the

space-groups  $A2$ ,  $Am$ , or  $A2/m$ . No piezoelectric effect was detected in crystals of pumpellyite from Hicks Ranch, Sonoma County, California, by use of the Giebe and Scheibe tester, and no pyroelectric effect was detected when crystals were placed on aluminium foil and immersed in liquid air. Hence it is likely that the mineral is holosymmetric with the space-group  $A2/m$ .

#### RELATION OF X-RAY CELL TO MORPHOLOGY.

The best cleavage and the face parallel to which the crystals are flattened is (100) according to the above choice of unit cell. There is a less perfect cleavage parallel to (001). Due to curved growth, the available

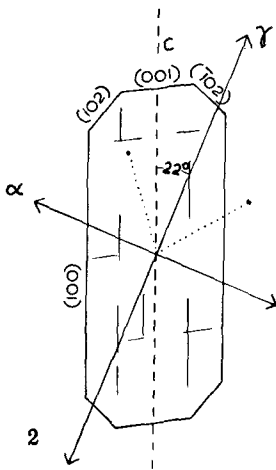


FIG. 2. Optical orientation of pumpellyite from Calumet, Michigan.

crystals are quite unsuitable for accurate optical goniometric measurement, and what appear to be single faces are found to give diffuse reflections over a range of as much as  $15^\circ$ . Nevertheless, reflections at approximately  $40\text{--}45^\circ$  from (100) are prominent, and some crystals or cleavage fragments give reflections at about  $80^\circ$  from (100). These reflections have been correlated with the faces (102),  $(\bar{1}02)$ , and (001). The face  $(\bar{1}01)$  has also been recognized. The angles which important faces should make with (100) have been calculated as follows: (102)  $39.1^\circ$ ,  $(\bar{1}02)$   $46.1^\circ$ , (001)  $82.4^\circ$ , (101)  $23.3^\circ$ ,  $(\bar{1}01)$   $25.9^\circ$ .

The optical orientation is illustrated in fig. 2. In both Calumet pumpellyite and in Californian pumpellyite,  $\gamma$  lies in the acute angle between the cleavages. In the former case the extinction angle  $\gamma:c$  is about  $22^\circ$  and in the latter case about  $4^\circ$ . The optic axial plane is parallel to (010).

#### TWINNING.

Pumpellyite may be twinned on (001) as reported by Irving, Vonsen, and Gonyer, and further described below. In addition, twinning sometimes occurs on (100) (fig. 3). Twinning on two planes approximately at right angles to each other has been observed by de Roever (1947a).

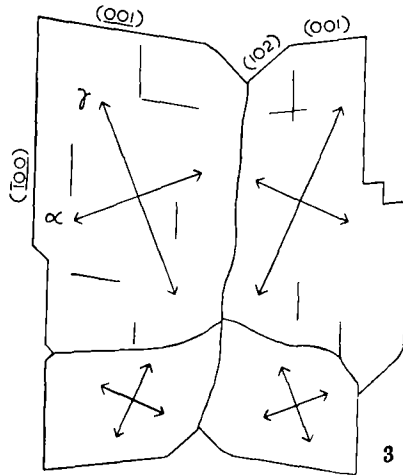


FIG. 3. Cruciform twin of pumpellyite from Calumet, Michigan.

#### RELATED MINERALS.

Palache and Vassar, Quitzow, also Irving, Vonsen, and Gonyer, have all pointed out similarities between pumpellyite and the epidote minerals. The members of both groups are prismatic in habit with elongation parallel to the symmetry axis, and they have generally similar refractive indices and a tendency towards strong dispersion. Pale varieties of pumpellyite are distinguished from zoisite by inclined extinction in (010) sections, and from both zoisite and clinozoisite by having distinctly lower refractive indices. A characteristic bluish-green  $\beta$  absorption tint and positive sign serve to distinguish typical ferrous pumpellyite from epidote, and the refractive indices and birefringence tend to be lower, although iron-rich pumpellyite (strongly coloured) has higher indices and birefringence than an iron-poor member of the epidote series. Such optically negative pumpellyites as have so far been recognized are brownish in colour, but may be difficult to identify without careful study. The interaxial and intercleavage angle is  $82.4^\circ$  for pumpellyite as against  $64^\circ 36'$  for epidote. Chemical similarities and differences have been summed up by Irving, Vonsen, and Gonyer in the suggested formulae:

pumpellyite:  $\text{Ca}_4\text{R}_6\text{Si}_6\text{O}_{23}(\text{OH})_3 \cdot 2\text{H}_2\text{O}$ , where

$\text{R}_6 = (\text{Al}, \text{Fe}''')_5(\text{Fe}'', \text{Mn}, \text{Mg})$ ;

zoisite:  $\text{Ca}_4\text{Al}_6\text{Si}_6\text{O}_{24}(\text{OH})_2$ .

Quitow (1936) has reported parallel growths of pumpellyite with epidote in rocks from Braunau, Kellerwald, Germany. It is no surprise to find that two cell-edges of pumpellyite,  $a$  and  $b$ , have dimensions similar to those of epidote.

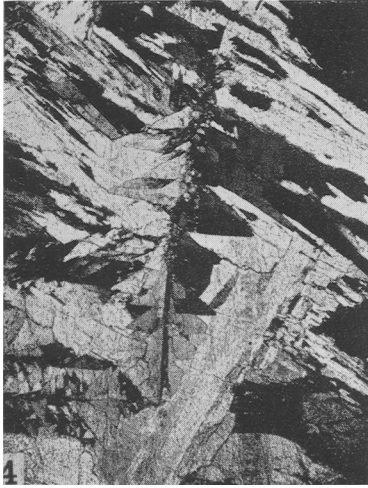


FIG. 4.

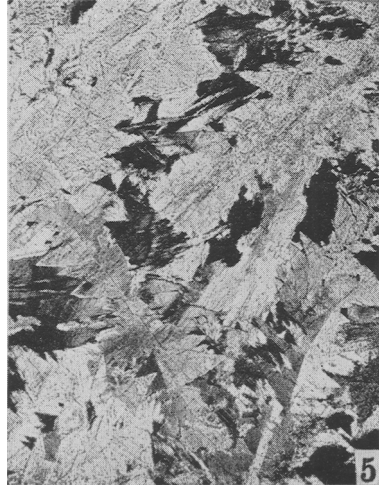


FIG. 5.

FIGS. 4 and 5. 'Oak-leaf' twinned intergrowths of pumpellyite and lawsonite from Sonoma County, California. In fig. 4, note lamina of lawsonite (in extinction) along the composition plane of the twinned aggregate in (010) section in the lower centre field. Much of this photomicrograph is occupied by sub-prismatic sections of the intergrowths. A cruciform arrangement will be seen in fig. 5 (lower left). Crossed nicols.  $\times 22$ .

Irving, Vonsen, and Gonyer described twinned crystals of pumpellyite shaped like oak leaves, with the twin plane (001) down the centre. Their locality was Mill Creek, California, where the pumpellyite, an iron-poor variety, was associated with lawsonite. During examination of pumpellyite-lawsonite-glaucophane-rocks from two other Californian localities, Hicks Ranch and Porter Creek, half a mile west of Heldsburg, both in Sonoma County, the present writer has observed similar twins. In most cases there is a thin lamina of intergrown lawsonite along the composition plane, separating the two halves of the twin. Oriented lawsonite also occurs between cusps of pumpellyite (figs. 4 and 5). In these intergrowths  $\alpha$  of the lawsonite is parallel to  $\beta = b$  of the pumpellyite, and  $\beta$  of the lawsonite is parallel to the  $a$  crystallographic axis

of pumpellyite, with which  $\alpha$  makes a small angle,  $12^\circ$ . This latter fact was confirmed by a special oscillation photograph.

On the basis of the lawsonite cell as described by Wickman (1947), we find a close correspondence between the lengths of the cell-edges along the shared (001) plane. Thus:

$$a (= \beta) \text{ of lawsonite, } 8.88 \text{ kX, is parallel to } a \text{ of pumpellyite, } 8.81 \text{ \AA.}$$

$$b (= \alpha) \quad ,, \quad 5.75 \text{ kX,} \quad ,, \quad b (= \beta) \quad ,, \quad 5.94 \text{ \AA.}$$

According to Wickman, the lawsonite structure consists of chains of octahedral groups of O and OH atoms surrounding Al and alined parallel to the  $b$ -axis and the principal cleavages. The chains are linked by  $\text{Si}_2\text{O}_7$  groups and by calcium atoms. Water molecules occupy the interstices. According to the second hypothesis of Ito (1950), the epidote structure also contains chains of (O,OH) and Al atoms parallel to the  $b$ -axis, but there are also independent (Al,Fe) $\text{O}_6$  octahedra. Linkage is effected both by  $\text{Si}_2\text{O}_7$  and by  $\text{SiO}_4$  groups. In view of the crystallographic similarities it seems probable that pumpellyite may have a basically similar type of structure to these two minerals. Cell dimensions of epidote, zoisite, lawsonite, pumpellyite, and lombaardite are listed in table I.

TABLE I. Cell constants of epidote and related minerals.

	1.	2.	3.	4.	5.
	Epidote.	Zoisite.	Lawsonite.	Pumpellyite.	Lombaardite.
$a$ ... ..	8.96 Å.	16.19 Å.	8.88 kX	8.81 Å.	16.10 kX
$b$ ... ..	5.63	5.64	5.75	5.94	5.62
$c$ ... ..	10.20	10.13	13.30	19.14	18.20
$\beta$ ... ..	$115^\circ 24'$	$90^\circ$	$90^\circ$	$97.6^\circ$	$115\frac{1}{2}^\circ$
	$P2_1/m$	$Pnma$	$C222_1$	$A2/m$ or $A2, Am$	$Pm, P2,$ or $P2/m$

1, Ito, 1947. 2, Ito, 1950. 3, Wickman, 1947. 4, This paper. 5, Nel, Strauss, and Wickman, 1949.

#### POWDER PHOTOGRAPH DATA.

Powder data are presented in table II for the pumpellyite sample from which the crystal used in the Weissenberg work was selected (Calumet, Houghton Co., Michigan) and for a sample from Hicks Ranch, Sonoma County, California. The photographs used for the measurements were taken with filtered  $\text{Cu-K}\alpha$  radiation on a 19-cm. camera. Spacings for the Californian mineral show a slight but distinct shrinkage of about 0.1 to 0.2 % relative to those of Calumet pumpellyite. According to fig. 8, the refractive index of this Calumet sample ( $\beta$  1.706)



TABLE II. X-ray powder data for pumpellyite.

(19-cm. diameter camera, Cu- $K\alpha$  radiation,  $\lambda = 1.5418 \text{ \AA}$ . Relative intensities estimated by eye.)

<i>hkl</i> .	Calumet, Michigan.		Hicks Ranch, California.	
	<i>d</i> (Å.).	Int.	<i>d</i> (Å.).	Int.
10 $\bar{2}$	6.9	< 1	6.8	< 1
102	6.1	< 1	6.0	< 1
004	4.75	1	—	—
111	4.66	1	4.67	2
200, 10 $\bar{4}$	4.38	2	4.39	1
20 $\bar{2}$	4.18	< 1	4.18	< 1
104	3.96	< 1	—	—
202	3.79	5	3.79	3
20 $\bar{4}$	3.45	1	3.45	1
11 $\bar{5}$ , 10 $\bar{6}$	3.11	< 1	3.11	< 1
204	3.03	2	3.03	2
020	2.96	1	2.95	1
115, 300, 30 $\bar{2}$	2.90	10	2.90	10
022	—	—	2.80	< 1
206	2.74	5	2.73	5
302	2.69	< 1	—	—
311	2.64	3	2.63	5
024	2.51	2	2.51	4
11 $\bar{7}$ , 2.0.10	2.452	4	2.445	5
222	2.330	2	2.325	3
	2.282	< 1	2.282	1
20 $\bar{8}$	2.210	3	2.205	5
40 $\bar{2}$	2.190	2		
026	2.163	< 1	2.158	1
	2.118	1	2.112	1
40 $\bar{4}$	2.094	1	2.088	< 1
402	2.072	1	2.069	1
	2.006	1	2.003	2
	1.920	1	1.917	1
028	1.853	1	1.847	2
	1.739	< 1	1.739	< 1
	1.718	< 1	1.720	< 1
	1.698	2	1.695	2
2.0.10, 1.1. $\bar{1}\bar{1}$	1.664	2	1.661	2
	1.638	< 1	1.636	< 1
424	1.597	3	1.594	4
0.0.12	1.581	2	1.578	2
2.2. $\bar{1}\bar{0}$	1.556	1	1.552	3
408	1.513	2	1.512	1
	1.48-1.49	2	1.488	2
	—	—	1.476	3
	1.450	1	1.447	2
	1.414	< 1	1.412	< 1

[continued on p. 122]

TABLE II (*cont.*)

1.394	< 1	1.398	1
1.369	1	1.367	1
1.344	< 1	1.340	< 1
1.317	< 1	1.314	< 1
1.301	1	1.299	2
1.263	< 1	1.260	< 1
1.249	1	1.247	1
—	—	1.229	1
1.210	< 1	1.208	< 1
1.170	< 1	1.170	< 1
—	—	1.113	< 1
1.101	< 1	1.099	< 1
1.090	< 1	1.089	< 1
1.082	< 1	1.079	< 1
1.072	< 1	1.070	< 1
0.986	< 1	0.985	< 1
0.953	< 1	0.953	< 1
0.912	< 1	0.911	< 1
—	—	0.831	< 1

indicates an iron content of about 8.2 % total iron as  $\text{Fe}_2\text{O}_3$ . A partial analysis of the Hicks Ranch mineral showed 3.7 % total iron as  $\text{Fe}_2\text{O}_3$  including 2.7 % FeO, or about 2 % of the theoretical ferric iron end-member. Allowing an average contribution of 3 % FeO as  $\text{Fe}_2\text{O}_3$ , calculation shows that the Calumet pumpellyite contains about 12 % of the ferric iron end-member. The observed slight changes in lattice spacings as  $\text{Fe}''$  replaces Al are in the expected direction.

#### CHEMICAL COMPOSITION.

Calculations from the analyses of four pumpellyites (table III), lead to figures closely approximating 56 oxygen atoms (including OH,  $\text{H}_2\text{O}$ ) per unit cell of 993 cubic Ångströms. Fifty-six oxygen atoms correspond to two formula units as proposed by Irving, Vonsen, and Gonyer (1932), namely:  $\text{Ca}_4\text{R}_6\text{Si}_6\text{O}_{23}(\text{OH})_3 \cdot 2\text{H}_2\text{O}$ , where  $R = \text{Al, Fe, Mg, Mn}$  (and Ti); and  $(\text{Ti} + \text{Al} + \text{Fe}'') : (\text{Mg} + \text{Fe}'' + \text{Mn}) = 5 : 1$ . Atomic proportions have therefore been calculated on the basis of  $26(\text{O}, \text{OH})$  per half unit cell, any excess or deficiency being placed in the  $\text{H}_2\text{O}$  group. The results show good agreement with Irving's formula. The earlier suggestion of Palache and Vassar (1925),  $6\text{CaO} \cdot 3\text{Al}_2\text{O}_3 \cdot 7\text{SiO}_2 \cdot 4\text{H}_2\text{O}$ , can be discarded. Five analyses of synonymous varieties and impure specimens are set out in table IV (p. 124), but only one of these warrants the calculation of a formula.

TABLE III. Chemical analyses of pumpellyite with corresponding atomic proportions.

	A.	B.	C.	D.
SiO <sub>2</sub> ...	37.18	38.01	37.63	37.53
TiO <sub>2</sub> ...	—	0.21	0.41	0.14
Al <sub>2</sub> O <sub>3</sub> ...	23.50	25.88	27.14	25.39
Fe <sub>2</sub> O <sub>3</sub> ...	5.29	1.11	—	1.74
FeO ...	2.09	2.90	3.25	2.95
MnO ...	0.13	0.17	1.03	0.13
MgO ...	3.18	1.81	1.47	2.65
CaO ...	23.08	22.70	21.49	22.56
Na <sub>2</sub> O ...	0.19	0.46	0.46	0.58
K <sub>2</sub> O ...	trace		0.08	nil
H <sub>2</sub> O+ ...	6.28	6.64	7.27	6.37
H <sub>2</sub> O- ...	0.06		0.12	0.09
P <sub>2</sub> O <sub>5</sub> ...	—	—	—	trace
	100.97	99.89	100.35	100.13
Sp. gr. ...	3.2	3.18	3.22	3.2
O,OH,H <sub>2</sub> O per unit cell as determined for A.	55.5	55.9	57.1	56.0

Atomic proportions on basis of 26(O,OH) per half unit cell.

Si ...	5.94	6.07	6.02	6.00
Ti ...	—	0.02	0.05	0.02
Al ...	4.42	4.88	5.12	4.78
Fe <sup>III</sup> ...	0.64	0.13	—	0.21
Fe <sup>II</sup> ...	0.28	0.39	0.43	0.39
Mn ...	0.02	0.02	0.14	0.02
Mg ...	0.76	0.43	0.35	0.63
Ca ...	3.95	3.88	3.68	3.86
Na ...	0.06	0.14	0.14	0.18
K ...	—	—	0.02	—
OH- ...	3.00	3.00	3.00	3.00
H <sub>2</sub> O ...	1.88	2.04	2.44	1.94

- A. Calumet, Michigan. Palache and Vassar, 1925. H. E. Vassar, anal.
- B. Mill Creek, California. Irving, Vonsen, and Gonyer, 1932. F. A. Gonyer, anal. (pale green pumpellyite).
- C. Skaggs, California. Irving, Vonsen, and Gonyer, 1932. F. A. Gonyer, anal. (pale brownish pumpellyite).
- D. Asahine, Japan. Tsuboi, 1936. S. Tanaka, anal.

TABLE IV. Inferior analyses of pumpellyite, and analyses of impure material.

	E.	F.	G.	H.	I.
SiO <sub>2</sub> ...	37.41	36.99	35.94	39.44	42.00
Al <sub>2</sub> O <sub>3</sub> ...	24.62	25.49	19.41	28.33	23.05
Fe <sub>2</sub> O <sub>3</sub> ...	2.21	6.48	6.80		2.64
FeO ...	1.81	—	4.54		3.74
MnO ...	—	—	—	—	0.12
MgO ...	3.46	—	2.48	3.20	1.67
CaO ...	22.20	19.90	22.77	22.21	21.04
Na <sub>2</sub> O ...	0.32	3.70	trace	0.93	—
K <sub>2</sub> O ...	—	0.40	—	—	—
H <sub>2</sub> O + } ...	7.72	7.22	8.40	6.58	5.62
H <sub>2</sub> O - }					0.12
	99.75	100.18	100.34	100.69	100.00

Sp. gr. — 3.180 3.11 3.229 (3.2)

Si ...	6.05	E. <i>Chlorastrolite</i> , Isle Royale, Michigan. Hawes, 1875.
Al ...	4.69	G. W. Hawes, 1900, anal.
Fe <sup>m</sup> ...	0.27	F. <i>Chlorastrolite</i> , Isle Royale, Michigan. Whitney, 1847. J. D. Whitney, anal.
Fe <sup>n</sup> ...	0.24	
Mg ...	0.83	G. <i>Zonochlorite</i> , Neepigon Bay, Lake Superior. Hawes, 1875. G. W. Hawes, anal.
Ca ...	3.84	
Na ...	0.10	H. <i>Lotrite</i> , Lotru Valley, Carpathians. G. Munteanu-Murgoci, 1900, anal.
OH— ...	3.00	I. Impure pumpellyite, Levikha, Ural Mountains. 'Corrected' by Kvasha to allow for quartz and sphene. Epidote is also present. The correction for quartz is plainly insufficient. $\beta$ 1.697–1.702; $\gamma$ — $\alpha$ 0.012; 2V <sub><math>\gamma</math></sub> (blue) 51°, 2V <sub><math>\gamma</math></sub> (red) 35°. Kvasha, 1950. T. L. Pokrovskaya, anal.
H <sub>2</sub> O ...	2.61	

#### THERMAL ANALYSIS.

Preliminary dehydration experiments on pumpellyite from Asahine, Japan, were carried out by Tsuboi (1936). A differential thermal analysis of Asahine pumpellyite by Dr. R. J. McLaughlin (fig. 6) shows no significant reaction until temperatures are reached of 740° and 810° C., where there are two strong overlapping endothermic peaks, followed by an exothermic peak at 910°. This evidence supports the suggested formula to the extent that it indicates two types of water bonding. The water is lost at a rather higher temperature than in the case of lawsonite, for which Dr. McLaughlin found endothermic peaks at 630° and 760°. Corresponding figures due to Norin (1942) are 675° and 805°. Kvasha (1950) found endothermic peaks for pumpellyite at about 700° and 800° followed by an exothermic peak at 850°.

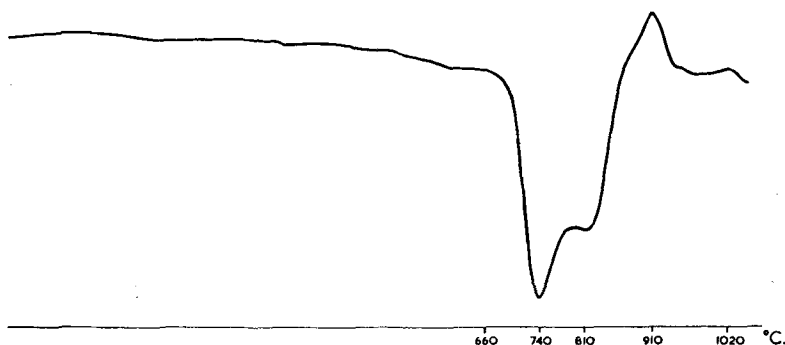


Fig. 6. Differential thermal analysis curve for pumpellyite.

#### OPTICAL STUDY

Due to the tendency for crystals to lie flat on (100) it is almost impossible to make satisfactory measurements of the  $\alpha$  and  $\gamma$  indices on powders. Hence, wherever possible, indices have been determined on single crystals mounted on the stage goniometer with the  $b$  ( $=\beta$ ) axis parallel to the axis of rotation. With this arrangement, the optic axial angle as well as all three indices can be measured on the same part of a single grain. For accurate results it is usually essential to use very narrow splinters in order to minimize the effects of zoning and distorted crystal growth. Under favourable conditions the accuracy actually obtained is estimated as  $\pm 0.001$  for indices and  $\pm 2^\circ$  for  $2V$  ( $\text{Na}_D$  light). Under less favourable conditions these errors may be more than doubled. The refractive index  $\beta$  can be determined fairly satisfactorily on loose crystals too small to be mounted on the stage goniometer. Due to the strong dispersion of the optic axes, sections parallel to the length of the crystals show anomalous interference tints, brownish for sections containing  $\beta$  and  $\gamma$ , and bluish for sections containing  $\alpha$  and  $\beta$ . Significant data are listed below, all measurements being for  $\text{Na}_D$  light.

*Calumet, Michigan* (B.M. 1926, 226, used for unit-cell determination).— Dark lustrous green crystals showing distinct zoning. Indices averaged from six crystals are considered representative of the sample as a whole.

	Average.	Extremes.	Pleochroism.
$\alpha$ ...	1.698	1.695–1.702	almost colourless
$\beta$ ...	1.706	1.703–1.709	deep bluish-green
$\gamma$ ...	1.720	1.717–1.722	almost colourless
$\gamma-\alpha$ ...	0.022	—	—
$2V_\gamma$ ...	$80^\circ$	$75^\circ-85^\circ$	
Dispersion $r < v$ strong.		$\gamma:c$ (in acute angle $\beta$ ) = 20–25°.	

The birefringence, 0.022, agrees fairly closely with the figures of 0.018–0.020 reported by Burbank (1927), Korzhinsky (1944), and Zavaritsky (1944) for comparable deeply coloured pumpellyites.

*Calumet, Michigan* (H 90919).—This material, a specimen of which has been provided by Professor C. Frondel, is that from which Palache and Vassar's original analysis sample was selected. Most of the crystals are strongly zoned and great variation in colour is apparent from very pale to very dark bluish-green or even slightly brownish bottle-green. The darker regions have higher indices than the lighter, stronger dispersion, and larger optic axial angles. In zoned crystals, dark-green zones usually surround lighter cores. Data for two representative zoned crystals are given below, and data for other crystals from the same sample are incorporated in fig. 9 (p. 131).

		Crystal no. 1.	Crystal no. 2.
$\alpha$	...	1.676–1.678	1.687–1.698
$\beta$	...	1.677–1.679	1.691–1.707
$\gamma$	...	1.690–1.693	1.706–1.720
$\gamma - \alpha$	...	0.014–0.015	0.019–0.022
$2V_{\gamma}$	...	29°–33°	57°–76°
Dispersion	$r < v$ distinct		$r < v$ very strong

In one small aggregate of pumpellyite needles from this specimen, it was found that although  $\beta$  varied from 1.694 to 1.710, for the majority of grains it varied within narrow limits, 1.701–1.702, which gives a fair average for the particular small aggregate as a whole. After crushing and centrifuging in bromoform to separate any feldspar and quartz, total iron was estimated as  $\text{Fe}_2\text{O}_3$  by the *o*-phenanthroline colorimetric method, with the result 7.6 %  $\text{Fe}_2\text{O}_3$ , including 2.1 % FeO. This compares closely with Palache and Vassar's figures of 5.29 %  $\text{Fe}_2\text{O}_3$  + 2.09 % FeO (7.61 % total iron as  $\text{Fe}_2\text{O}_3$ ). Two less homogeneous hand-picked dark-green samples with an estimated average  $\beta$  of 1.700, treated in the same way, gave 7.0 % and 7.1 % of total iron as  $\text{Fe}_2\text{O}_3$ . The iron-poor nature of the almost colourless Calumet pumpellyite was confirmed in a qualitative way by a colorimetric determination of 3.4 % total iron oxides on a small, impure, and rather strongly zoned sample.

*Hicks Ranch, Sonoma County, California*.—Veins of light green pumpellyite traverse pumpellyite-lawsonite-glaucophane-rock. Pumpellyite occurs in the veins as needles several millimetres long flattened on (100). Colorimetric determinations showed 2.7 % FeO, 0.7 %  $\text{Fe}_2\text{O}_3$  (3.7 % total iron as  $\text{Fe}_2\text{O}_3$ ), and 0.26 % MnO. The optical properties

correspond closely with those given by Irving, Vonsen, and Gonyer (1932) for iron-poor Californian pumpellyite.

	Hicks Ranch.	Mill Creek (Irving et al. 1932).	Pleochroism.
$\alpha$ ...	1.681	1.679	colourless
$\beta$ ...	1.682	1.680	very pale green
$\gamma$ ...	1.695	1.692	colourless
$\gamma - \alpha$ ...	0.014	0.013	
$2V_\gamma$ ...	39°	40°	
$\alpha : (001)$ ...	12°	12°	
Dispersion	$r < v$ (distinct)	$r < v$	

Thin sections from Hicks Ranch show the twinned pumpellyite-lawsonite intergrowths already referred to (figs. 4, 5). The extinction

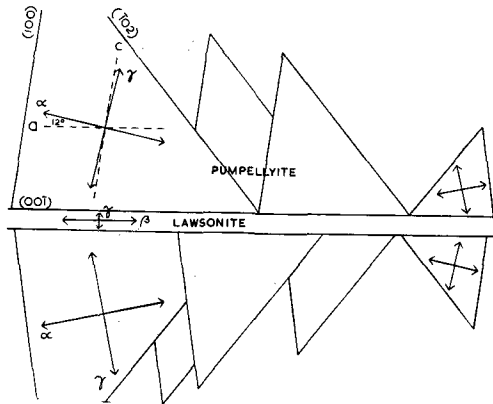


FIG. 7. Pumpellyite twinned on {001}, with intergrown lawsonite, shown in diagrammatic section parallel to {010}. Sonoma County, California.

angle between  $\alpha$  and the twin plane (001) in adjacent parts of twins is 12° as reported by Irving and collaborators. An interpretation of the cusped 'oak leaf' twins is illustrated in fig. 7. The cusps appear to be due to a combination of (100) and  $(\bar{1}02)$  faces which theoretically make an angle of 46° with each other. According to an alternative explanation they might be due to (100) and  $(\bar{1}0\bar{2})$  at an angle of 39°. Actual measurements of the cusp angle on the universal stage were usually 40–45°. The extinction angle between  $\gamma$  and the face making a large angle with (001) was consistently found to be close to 7°. If the face is (100), the extinction angle  $\gamma : (100)$  should be 4½° according to the

figured interpretation, but  $19\frac{1}{2}^\circ$  according to the alternative explanation. This supports fig. 7, although the agreement is not perfect. The anomalies are considered to be due partly to difficulties of measurement and partly to disorientation of various parts of the aggregates, and perhaps to vicinal faces making small angles with (100) and ( $\bar{1}02$ ).

*Asahine, Titibu-gōri, Saitama prefecture, Japan.*—A large hand-specimen was provided by Professor Tsuboi, who described the material in 1936 (analysis D, table III). Bladed crystals of pumpellyite, 1 cm. or more long, make up veins cutting metagabbro, itself rich in pumpellyite. Thin sections show that the large pumpellyite 'crystals' are closely twinned and slightly disoriented aggregates. 'Oak leaf' twinning is sometimes developed. Very slight variation was observed in optical properties,  $\beta$  ranging from 1.687 to 1.690. Typical refractive index figures agree closely with those of Tsuboi, but significant discrepancies were found in the optic axial angle, and in the extinction angle which was measured on twinned crystals. The revised values only have been utilized in the graphs showing the relation between optical properties and chemical composition (figs. 8, 9, p. 131).

		Revised values.	Data due to Tsuboi (1936)
$\alpha$	...	1.686	1.686
$\beta$	...	1.688	1.690
$\gamma$	...	1.699	1.698
$\gamma - \alpha$	...	0.013	0.012
$2V_\gamma$	...	$49\frac{1}{2}^\circ$	$36-40^\circ$
$\alpha : (001)$	...	$15\frac{1}{2}^\circ$	$11-12^\circ$

*Howell's Point, Riverton, Southland, New Zealand.*—This material was collected by Professor C. E. Tilley. Small prismatic deep bluish-green crystals occur embedded in chlorite in albite-filled amygdales in a spilitic pillow lava. The crystals are dominated by the faces (100), ( $\bar{1}01$ ), and ( $\bar{1}02$ ). The optical properties were found to be remarkably uniform:

$\alpha$	...	1.694	colourless
$\beta$	...	1.698	deep bluish-green
$\gamma$	...	1.713	colourless
$\gamma - \alpha$	...	0.019	
$2V_\gamma$	...	$65^\circ$	
Extinction angle $\gamma : c = 17^\circ$ in acute angle $\beta$ .			

*North Range, Taringatura survey district, Southland, New Zealand.*—Pumpellyite is occasionally present as a reconstituted mineral in the greywackes of the district, and also as a constituent of metasomatic bodies of quartz, albite, adularia, and pumpellyite which locally replace



laumontitized vitric tuffs. It occurs as aggregates of tiny prismatic crystals less than 0.05 mm. long and of rhombic cross-section due to the dominance of (100) and (001) faces. In a typical specimen the optic axial plane is parallel to (010) as in all the pumpellyites described above,  $\gamma:c$  is approximately  $27^\circ$ ,  $\beta$  1.70–1.72,  $2V_\gamma$  is large, dispersion strong with  $r < v$ , and there is normal pleochroism  $\alpha = \gamma =$  almost colourless,  $\beta =$  bluish-green to yellowish-green. Such properties are those of ordinary, moderately iron-rich pumpellyite. In some cases a transition was observed from greenish to brownish absorption tints. A golden-brown, pleochroic mineral in a metasomatic rock (O.U. 8811) of negative sign and unusually high refractive indices and birefringence was also proved to be pumpellyite, a powder photograph showing a pattern indistinguishable from that of Calumet pumpellyite. Unfortunately this interesting material was much too fine-grained and poorly crystalline to allow detailed measurement of optical properties. Nevertheless two distinct optical orientations were observed, as follows:

(i) Optic axial plane parallel to (010);  $\beta$  1.71–1.73;  $\gamma - \alpha = 0.02$ ;  $2V_\alpha$  variable,  $80-20^\circ$ ; dispersion  $r > v$  strong; pleochroic with  $\alpha = \gamma =$  very pale bluish-green to brownish-green,  $\beta =$  pale to deep golden-yellow.

(ii) Optic axial plane perpendicular to (010) with  $\gamma$  parallel to the length of the fibre;  $\gamma$  variable, up to  $1.75$ ;  $\gamma - \alpha$  estimated on thin sections with Berek compensator as about  $0.028$ ;  $2V_\alpha$  variable from almost uniaxial to  $60^\circ$ ; dispersion  $r < v$  strong; pleochroic with  $\alpha = \beta =$  pale bluish-green to pale brownish-green,  $\gamma =$  golden-yellow to yellow-brown.

It is likely that there is a continuous series between orientations (i) and (ii) with virtually uniaxial intermediate varieties.

Irving, Vonsen, and Gonyer (1932) have shown that the brownish-yellow absorption tint for iron-poor pumpellyite from Skaggs, California, can be correlated with the presence of 1.03 % MnO, but in other respects the optical properties of the Skaggs mineral are normal. Quitzow (1936) has described pumpellyite from Braunau, Kellerwald, having all transitions from the normal green variety of positive sign,  $r < v$ , to material having negative sign and very small optic axial angle,  $r > v$ , and the pleochroic scheme  $\alpha = \gamma =$  faintly brownish,  $\beta =$  yellowish-brown (parallel to the length of the fibres). The double refraction of the brown variety is greater than that of the green. These properties are comparable with those of the North Range pumpellyite no. 8811, orientation (i). Lacroix (1942) has also described transitions from green to

brown varieties. Quitzow suggested that the brown colour of his material, like that of Irving and collaborators, might be due to the presence of manganese. This can, however, hardly be the explanation of the anomalous properties of the North Range pumpellyite. The rock 8811 was estimated to contain 8% pumpellyite, and a partial analysis revealed the presence of 0.025% MnO. Hence even assuming that all the MnO is contained in the pumpellyite, the mineral can hardly contain more than the order of 0.3% MnO. On the other hand, the high indices and birefringence suggest high iron.

#### RELATION OF OPTICAL PROPERTIES TO CHEMICAL COMPOSITION.

The systematic nature of the variations of the optical properties of pumpellyite has been pointed out by de Roever (1947b). In eastern Borneo, he found a continuous series from colourless varieties with low refractive index (1.67), low birefringence (0.010), small  $2V_y$ , and feeble dispersion, to dark-green varieties with high refractive index (1.72), higher birefringence (0.018), large  $2V_y$ , and strong dispersion. All these conclusions are confirmed in the present study and they are correlated with varying iron content. Furthermore, it appears that the increase in  $2V_y$  with refractive index can be continued to values greater than  $90^\circ$  when the sign becomes negative ( $\beta$  1.72–1.73). It is probable that the optic axial plane changes from parallel to (010) to perpendicular to (010) as the refractive index for vibrations parallel to the  $b$ -axis, which is normally  $\beta$ , continues to increase at a greater rate than those in (010) and eventually exceeds them for values of about 1.74 to 1.75. An explanation of the properties of the yellow-brown Taringatura pumpellyite would thus be afforded, but until analysable material is found with these properties, the question cannot be decided satisfactorily. The suggested explanation is shown diagrammatically in fig. 8 as an extrapolation of the refractive index curves for more normal pumpellyites, based on four published analyses and four new partial analyses. Many measurements on single crystals show a close correlation between optic axial angle and refractive index, and this is shown in fig. 9.

*Extinction angles.*—The measurement of extinction angles in (010) sections of pumpellyite is usually a matter of some difficulty, and measurements are probably not very reliable except where made on twinned crystals. The limited data available indicate an increase in extinction angle with increasing iron content (fig. 9).

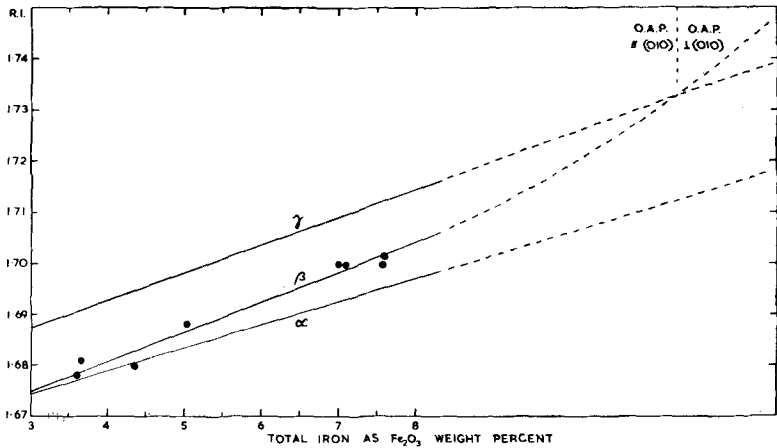


FIG. 8. Refractive indices of pumpellyite related to iron content. Points represent data from published analyses and new partial analyses,  $\beta$  alone being plotted. Dashes—probable form of refractive index curves beyond the analysed range.

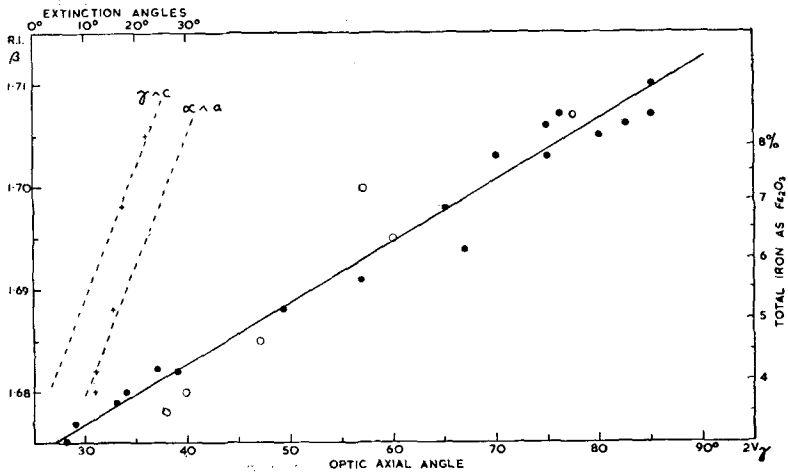


FIG. 9. Variation of  $2V$  and extinction angles of pumpellyite with refractive index and hence with iron content. Points—optic axial angle and  $\beta$  index determined on single crystals on the stage goniometer. Circles—optic axial angle data from the literature. Crosses—extinction angles.

## SYNONYMOUS VARIETIES.

In 1847 Whitney described under the name *chlorastrolite*, small rounded pebbles from the shore of Isle Royale, Lake Superior, consisting of a finely radiating, light bluish-green material. Hawes (1875) showed that this was not a homogeneous mineral, and on the basis of his analysis (table IV, E) he ascribed it to impure prehnite. Owing to the variable sign of elongation, Lacroix (1887) considered the material to be a supposed pleochroic variety of thomsonite. Professor C. Frondel has kindly pointed out that the green mineral is pumpellyite and he has provided several small pebbles. An X-ray powder photograph proves to be indistinguishable from photographs of type pumpellyite, and so confirms the identification. The refractive index  $\beta$  is 1.675–1.682. Hawes's analysis is seen to be in excellent agreement with pumpellyite analyses. In the pebbles examined by the writer, the pumpellyite is associated with some chalcedony and albite. In 1873 Foote described a massive mineral banded in different shades of green as a new species *zonochlorite*. It occurs both as shore pebbles and in situ in amygdaloidal trap at Neepigon Bay, Lake Superior. Hawes (1875) analysed it (table IV, G) and concluded that it was impure 'chlorastrolite'.

According to Kvasa (1950), Sobolev (1947) has shown that *lotrite* (Munteanu-Murgoci, 1900, 1901) is the same as pumpellyite. Sobolev's paper has not been seen by the present writer, but optical examination of material from the type locality (Lotru valley, southern Carpathians) kindly provided by the British Museum, shows that Sobolev's determination is undoubtedly correct. A single grain was mounted on the stage-goniometer and the following properties were determined:

New data.			Data due to Murgoci (1900)		
$\alpha$	...	1.674	$n$	...	1.67
$\beta$	...	1.675	$\gamma - \alpha$	...	0.014
$\gamma$	...	1.689	$2E_\gamma$	...	30°
$\gamma - \alpha$	...	0.015	$2V$	...	18°
$2V_\gamma$	...	28°	$\beta$ parallel to length		
$\beta$ parallel to length			Extinction angle to cleavage =		28°
Dispersion $v < r$	moderate				

Murgoci's analysis (table IV, H) agrees reasonably well with pumpellyite. His extinction angle seems rather high for a pumpellyite with about 3% total iron oxides as is indicated by the other optical properties, but as this is a difficult measurement to make on such poorly crystalline material, this is not a serious discrepancy. As Murgoci's description

and chemical analysis were both reasonably adequate, it is unfortunate that his work should have been overlooked. The name 'pumpellyite' is now firmly entrenched in the literature and 'lotrite' must lapse into synonymy, in spite of priority.

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