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OPTICAL PROPERTIES AND CRYSTALLOGRAPHY OF ZONED PUMPELLYITE FROM THE WITWATERSRAND

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ABSTRACT

Pumpellyite from the Witwatersrand was found to be intensely zoned and to show exceptionally wide variations in optical properties. Refractive indices (β_D) varied from 1.693 to 1.726, birefringences from .0016 to .0103 and optic axial angles ($2V_X$) from 38° [optic axial plane perpendicular to (010)] to 71° [optic axial plane parallel to (010)]. These angles again were dispersed through a wide range, showing $r>v$ in the former case and $r<v$ in the latter. The following axial elements were determined for the mineral: $\beta=70°$, $c/a=.842$.

INTRODUCTION AND PHYSICAL PROPERTIES

Pumpellyite was first described by Palache and Vassar from the Lake Superior region in 1925. Since that time it has been found to be of fairly widespread occurrence, being reported from Haiti (Burbank, 1927), California (Irving, Vonsen and Gonyer, 1932), Eurasia (Waldmann, 1934; Quitzow, 1935; Quitzow, 1936), Japan (Tsuboi, 1936), and New Zealand (Hutton, 1937). Up to the present a fair amount of work has been done on the chemistry[1] and such properties as the refractive indices and optic axial angles of pumpellyite, but to the writer's knowledge precise data on the crystallography and very interesting dispersion is still scanty. The present study was undertaken mainly with a view of determining the relations between the various optical and crystallographic properties of the Witwatersrand material.

During January, 1940, a small sample, stated to have come from the City Deep mine, Johannesburg, was submitted to the Geological Survey for identification. It consisted of aggregates of lustrous, greenish-black needles of pumpellyite in white vein quartz. Only about 1 gram of the mineral was available for study and all efforts to obtain a further supply have thus far proved unsuccessful. The specimens received were up to 1.5 cm. in diameter and were penetrated by coherent aggregates having a cross section of a few millimetres. The aggregates consisted of individual crystals, averaging about 1 mm. in diameter, in parallel or semi-parallel grouping. No terminating crystal faces were observed on the

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[1] For a table of analyses see Irving, Vonsen and Gonyer (1932); also Tsuboi (1936).

needles but a series of prism faces, usually striated and somewhat rough, were present.

The pumpellyite was tentatively identified from its optical properties and a qualitative chemical analysis. In the optical measurements however certain serious divergencies from the known properties[2] of pumpellyite were found so that quantitative chemical data were required to place the identity of the mineral beyond doubt. Mr. C. F. J. van der Walt kindly undertook a partial analysis of about 350 milligrams of the cleaned mineral and obtained the following result: SiO_2 36.9%, Al_2O_3 27.7%, Fe_2O_3 (total) 9.0%, CaO 22.4%, MgO 1.0%. Total 97.0%. In addition 0.6% B_2O_3 was determined by the writer. The result shows a fairly good agreement with analyses of pumpellyite from other localities and although the percentage of alumina is somewhat high, this is probably due to the presence of an alteration product in the material analyzed.

The following physical properties were determined on the mineral: $H. = \pm 6$. $G. = 3.226 - 3.318$. Magnetism, weakly magnetic to the electromagnet. Crystals of pumpellyite removed from their quartz matrix were found to be unsuitable for accurate measurement on the goniometer and only one angle of about 63°, measured in the direction perpendicular to the elongation, appeared to be fairly constant.

About 15% of the mineral examined showed evidence of alteration, the final product being a semi-opaque, cryptocrystalline aggregate with a mean index of refraction of about 1.65. This material usually formed layers showing a rude parallelism with crystal outlines and cleavages and, in the small amount of material available, was impossible to separate satisfactorily by means of heavy liquids. Purification of the mineral for chemical analysis (apart from the removal of adhering quartz) was not attempted as it was further found that the heavier fractions were richer in fragments of comparatively high birefringence. Of the two values for the specific gravity given above, the lower figure represents the gravity of a small cleaned fragment and the higher that of the heaviest grains in a crushed sample. These figures were obtained by immersing the mineral in methylene iodide and diluting with acetone until neither floating nor sinking occurred. The specific gravity of the liquid was measured immediately and the result corrected for water at maximum density.

OPTICAL AND CRYSTALLOGRAPHIC PROPERTIES

Universal stage measurements on thin sections were in agreement with monoclinic symmetry, the direction of elongation of the prisms being the symmetry axis. Pleochroism was observed as follows: X = pale greenish-yellow, Y = blue-green, Z = brownish-yellow. Absorption $Y > Z > X$. The pleochroism was found to vary somewhat. For fragments showing the

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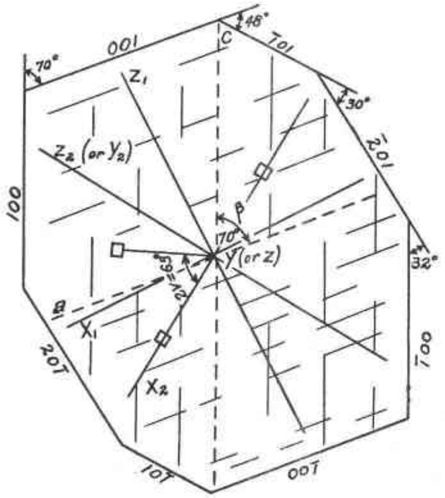
[2] For a summary of the optical properties of pumpellyite see Hutton (1937).

highest interference colours, Y is a lighter green and the absorption of the Z and Y directions about equal. It is of interest to note here that Quitzow (1936) found the intensity of the colour of the mineral to vary with the strength of the dispersion.

Zoning parallel to the *b*-axis is marked and highly irregular although there is a tendency for the zones to show some parallelism with crystal outlines and for the mantles to have a higher birefringence than the cores. The zones may have sharply defined boundaries or grade into one another. At best, areas that are suitable for optical measurements may be .3 mm. in diameter, measured across the length of the crystals, but frequently fragments are so intricately zoned that they are completely unsuitable for such measurements. The ether axis corresponding to *b* is common for all zones in a single individual but any axis perpendicular to this may differ by as much as 32° from the corresponding axis in an adjacent zone.



Optic orientation of pumpellyite

In thin sections cut parallel to (010) and accurately oriented by means of the universal stage, crystal outlines were more easily measurable than on the goniometer. Of the two imperfect cleavages observed, the better [considered as (001) after Palache and Vassar, 1925] was not found to correspond to a crystal face, while the other [considered as (100)] was paralleled by a well-developed face. The mean of a number of measurements for the angle between the cleavages gave a value of 70°. In addition hemiorthodomes ($\bar{1}01$) and ($\bar{2}01$), at angles of 48° and 78° respectively to (001), were noted. The positions of these faces and their relations to the optical directions are shown in Fig. 1.

On account of the imperfect development of the cleavages the accuracy of the crystallographic measurements is not considered to be greater than ±2°.

The axial elements of pumpellyite as far as these could be determined are thus as follows:

$$\beta = 70^\circ$$

$$\frac{c}{a} = \frac{\sin(001 \wedge \bar{1}01)}{\sin(\bar{1}00 \wedge \bar{1}01)} = .842.$$

Twinning is fairly common in the material examined but difficult to distinguish from the ordinary zoning. The composition plane although somewhat irregular corresponds in general with (001) while a stereographic projection of the ether axes shows that the twinning axis may be either the pole of this face or the *a*-axis. It should be observed here that the possibility of finding a twinning axis for any two lamellae is not sufficient criterion to establish a twinned relationship as opposed to a zonal one. In the material examined both twinning and zoning occur in the [010] zone and it was possible to find twinning axes in both cases. Even where the "twinning axis" coincides with a crystallographic axis or the normal to a crystal face, a twinned relationship could not be said to be proved since the correspondence may be fortuitous.

The property employed to distinguish between zoning and twinning was the dispersion of the ether axes. When these axes were plotted on the stereographic net for different colours of light it was observed that the red and green of all the lamellae usually succeeded each other in the same direction on the great circle corresponding to [010]. In rare cases, however, the order of the colours was reversed for certain components as compared to others. In these cases it was evident that a different type of relationship from the usual obtained, and when the "twinning axis" was transferred to the centre it could be seen (by referring to the positions of the ether axes for red and green light) that only where the dispersion of one component was reversed relative to the other, the relationship was one of twinning.

It was further noted that in the cases where the dispersion indicated a twinned relationship the twinning axis was always the normal to (001) or the *a*-axis. In cases of zonal structure the "twinning axis" had no fixed direction.

Extinction angles and optic axial angles were measured in sections nearly perpendicular to X and the results are shown in graphical form in Fig. 2. In connection with these results it must be stated that especially in the measurement of extinction angles a high degree of accuracy was unattainable. Cleavages even in what appeared to be the same zone sometimes diverged by as much as 8°. Optic axial angles larger than 40° were measured on the universal stage, while the smaller angles were measured by conoscopic observation according to Mallard's method. The accuracy is ±2° for measurements on the universal stage and becomes progressively less as the optic axial angle and birefringence decrease. It is indeed extremely difficult even with a strong monochromatic light to distinguish the difference between 20° ($Y=b$) and 20° ($Z=b$). In rare cases it has been observed that the optic axial plane is apparently parallel to (010) for green light and perpendicular to it for red.

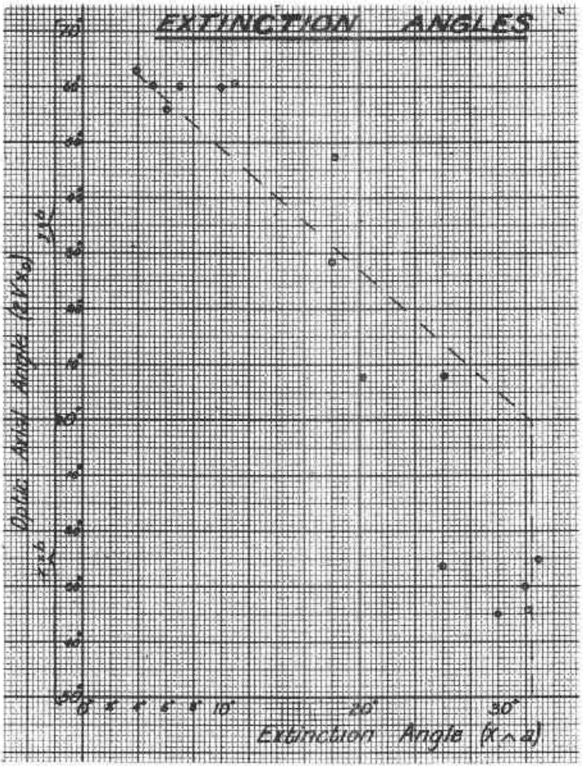


Fig. 2

The extinction angle ($X \wedge a$) was found to vary from 4° to about 32° in the acute angle β. A relationship between the variables in Fig. 2 has been suggested by means of a line ruled on the graph. This may be stated as follows: "As the optic axial angle becomes smaller, one axis continuously approaches the other which within small limits remains fixed. When zero is reached, the optic axial plane from being parallel to (010) becomes perpendicular to it and the position of the acute bisectrix remains more or less constant." Inspection of the optic axes in thin sections on the universal stage confirmed this statement. It has further been noted that the "fixed" optic axis is always the one that is least dispersed.

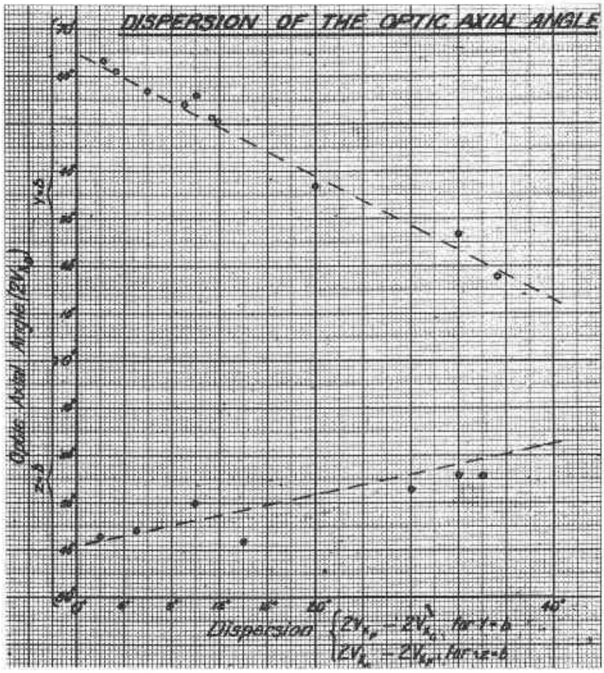


FIG. 3

DISPERSION

Figure 3 shows graphically the measurements of the optic axial angle for different wavelengths. For determinations of $2V_D$ a sodium lamp was employed as light-source while for $2V_F$ and $2V_C$ Lifa filters Nos. 391 and 215, respectively, were used in conjunction with a microscope lamp.

The dispersion of the optic axes varies from 2°, when the optic axial angle is large and Z or $Y=b$, to about 35° when the angle approaches zero. The bisectrices were found to be dispersed about $\frac{1}{2}°$ when the optic axial angle is large and $Y=b$ to about $4\frac{1}{2}°$ when $Z=b$.

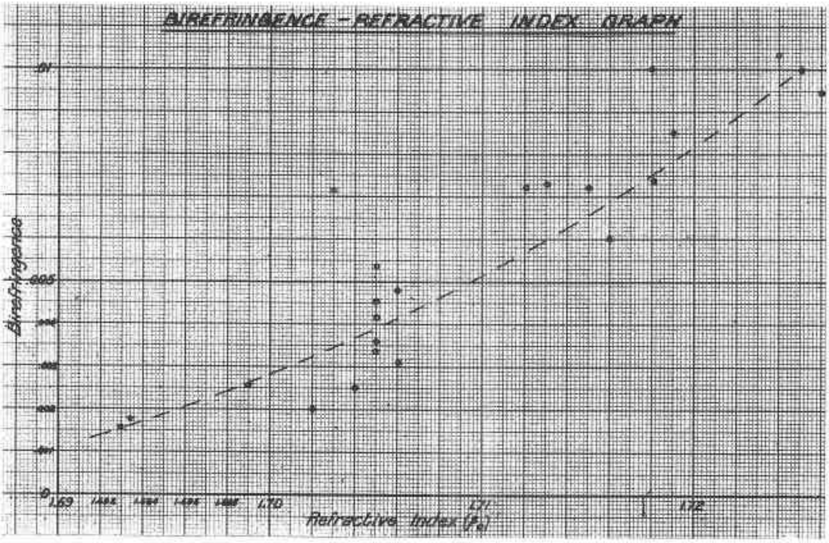

BIREFRINGENCE – REFRACTIVE INDEX GRAPH
Birefringence
.01
.005
.004
.003
.002
.001
0
1.69
1.692
1.694
1.696
1.698
1.70
1.71
1.72
Refractive Index

FIG. 4

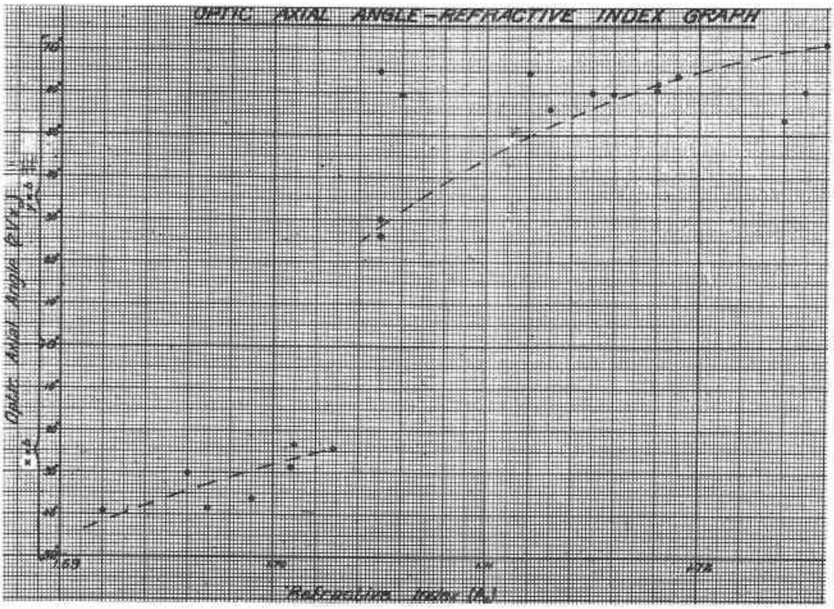

OPTIC AXIAL ANGLE – REFRACTIVE INDEX GRAPH
Optic Axial Angle (2V)
70
1.69
1.70
1.71
1.72
Refractive Index

FIG. 5

The relationship between the refractive index, birefringence and optic axial angle is shown in Figs. 4 and 5. To obtain these measurements the crushed mineral, mounted in hyrax, was investigated with the aid of the universal stage and monochromator. Grains were selected for which the *b*-axis was nearly horizontal and the optic directions X and Z (in certain cases X and Y) at angles of 40° to 50° to the vertical. On these grains the following properties were measured: (*a*) The retardation by means of Berek's compensator when Z or Y was vertical; (*b*) the diameter of the grain at right angles to this direction by means of a previously standardised micrometer ocular; (*c*) the optic axial angle; (*d*) the refractive index of the *b* crystallographic direction.

From (*a*) and (*b*) the birefringence $\beta-\alpha$ or $\gamma-\alpha$ could be calculated. Where $\beta-\alpha$ was obtained the complete birefringence was deduced graphically by taking into account the optic axial angle. After the measurements in hyrax had been completed a grain of known properties was removed from the mount and immersed in mono-iodonaphthalene. Again, the wavelength was read at which the index of the *b* direction equalled that of the immersion medium. With the aid of index dispersion curves of hyrax, mono-iodonaphthalene and pumpellyite it was then possible to deduce the refractive indices from the recorded readings in Ångstrom units.

The following were actual figures obtained. The refractive index (β) of the selected grain matched that of hyrax at 5850 Å. The portion of the cover-glass covering the grain was next removed, the exposed part of the mount dissolved in benzene and the grain recovered on a glass slide. This now matched with mono-iodonaphthalene at 4900 Å, indicating a refractive index of 1.727. From previously constructed dispersion graphs of pumpellyite it was found that at 5850 Å the grain would have an index of 1.7165. This was therefore the refractive index of hyrax at 5850 Å. One point on the hyrax dispersion curve having been established the whole curve could be constructed by referring to other hyrax curves near it.

The accuracy of these indices is not considered to be greater than .004 due to the difficulty in matching grains that were not ideally clear and unzoned. Further, the values of the optic axial angles given in Fig. 5 were obtained from only one optic axis and the acute bisectrix. Their accuracy is therefore less than the corresponding values in Figs. 2 and 3. For birefringence values a probable error of 15% is considered to apply.

Since indices lower than 1.702 could not be measured in the hyrax mount, ground sections of known orientation and thickness were immersed in mono-iodonaphthalene and the necessary optic angles, refractive indices and birefringences, to extend the graphs over the full range of refractive indices of the mineral, determined.

Dispersion graphs for the refractive index β of pumpellyite were constructed by immersing selected grains successively in liquids of different index and determining their indices for different wavelengths. The information in the table below was obtained from these graphs.

β_D	$\beta_F - \beta_C$
1.6935	.0140 (All ± .001)
1.7055	.0150
1.719	.0155
1.720	.0155
1.7265	.0160

Inspection of Figs. 2–5 shows that in general no sharply defined relationship between the respective variables can be made out. Whilst the lack of symmetry is to a large extent certainly due to experimental error, the divergencies in some cases (notably Fig. 5) appear to be too large to be explained by such error. A possible explanation is that the unit cell of the pumpellyite investigated contains more than two variable constituents, but detailed chemical work on fractions of known optical properties is needed before a definite conclusion can be reached. In the present case unfortunately not enough of the material was available for this work to be carried out.

Fig. 6. Section of pumpellyite parallel to (010) in quartz matrix. Note crystal outlines and semi-opaque alteration product. ×56.

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