ON β-URANOTILE

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(Optical investigations by V. Steinocher, chemical analyses and general description by R. Nováček.)

In 1935 the name β-uranotile was given by R. Nováček (7) to a new mineral from Jáchymov (Joachimstal) which was chemically identical with uranotile (=uranophane) and possessed the formula $CaO \cdot 2UO_3 \cdot 2SiO_2 \cdot 6H_2O$, but was distinctly different in physical, and especially in optical properties. It has been shown that the optical data of the new mineral agree with those given by Larsen and Berman (5, 6) for schroeckingerite. Evidently Larsen determined the optical constants on a sample of β-uranotile mixed with carbonates, and therefore regarded it as a carbonate (schroeckingerite). The true schroeckingerite is described in detail and its identity with dakeite shown in the preceding article by R. Nováček (10).

The data in Larsen's tables have caused the occurrences of two β-uranotiles from localities in the United States to be described as schroeckingerite. In 1935, Miss E. J. Armstrong reported this mineral as occurring in minute needles in the fissures of a pegmatite at Bedford, N. Y., and likewise the specimen from Spruce Pine, Mitchell Co., N. Carolina, in the British Museum of Natural History, London (No. 191, B. M. 1929) was labeled schroeckingerite. Both minerals are β-uranotile.

A third new locality of β-uranotile (besides the original locality of Jáchymov) is the well known fluorite occurrence at Wölsendorf, Bavaria. From this place the first crystallized uranotile was described by E. Bořický in 1870 (3), and not long ago A. Schoep and A. Scholz discovered (12) an unusual association of uranium minerals: uraninite, ianthinite, kasolite, fourmariérite, parsonsite, dewindtite, becquerelite and schoepite. At the end of their paper the writers state, that beside the above-named minerals, which they were able to identify definitely, they found on one specimen a small quantity of an unknown mineral, and record approximately its optical properties. As will be mentioned later, this mineral likewise has been shown to be β-uranotile.

In the present paper are given the optical and, where it has been possible, also the chemical data of the β-uranotile from the named localities.

The optical investigations have been carried out on an universal stage, modified according to Emmons, with five axes. The large dispersion of the extinction directions and high refraction made the use of monochromatic light necessary. This was produced by means of filters, so combined as to give the maximum amount of transmitted light with the greatest possible approximation to the required wave length. The filters were checked in advance spectroscopically.

The refractive indices were determined by means of the special Leitz arrangement in combination with the universal stage. This is a glass hemisphere with a small cavity of hemispherical shape into which the investigated mineral is placed and covered with an immersion liquid of a refractive index a little higher than that of the mineral. Upon heating by means of an electric current the refractive indices of the mineral and of the surrounding liquid become equal and the value determined from the angle of total reflection, which is measured directly on the stage. A more detailed description of this method is given by Rinne and Berek (11). According to the statement of Leitz, the accuracy of the determination of the refractive indices is ±0.002. Naturally it is necessary, as we have done in our investigations, to take a mean value of several measurements.

Quantitative chemical analyses could be made in only two instances, by microchemical methods on minute quantities. SiO_2 was determined by treatment with nitric acid, evaporating several times to dryness, filtering and igniting in an electric furnace. The ignited and weighed impure silica was evaporated with hydrofluoric acid as SiF_4 and the residue subtracted from the original weight of the impure SiO_2. The uranium was precipitated from the acetate solution and weighed as oxychinolate (oxine); calcium was determined as the oxalate. The more detailed data on the analytical procedure are given in the paper of Nováček (7, p. 3–5), where also more complete references to the literature are included (Hecht, Schwarz-Bergkampf, etc.).

β-Uranotile from Jáchymov (Joachimstal)

The β-uranotile from this original locality is described in detail in the papers of Nováček (7, 8, 9), where also a chemical analysis is given. The data are here briefly summarized, together with a more complete report on the optical properties.

β-uranotile from Jáchymov usually forms coatings on uraninite. The minute needles, generally shorter than 1 mm., are of yellowish-green or more rarely of a pure yellow color and include minute black grains, perhaps of uraninite. The needles are either intergrown with the matrix or are grouped in fan-like and radial tufts in cavities, from which single crystals may be relatively easily isolated. The needles of β-uranotile are less elongated than those of normal uranotile from Jáchymov and other localities (length to breadth = 5:1, 3:1, etc.); sometimes their shape is more tabular.

The goniometrical investigations (Nováček (7), p. 26) could not be carried out accurately because of the imperfect character of the crystal faces. The microscopical data, given below, are more reliable. Nevertheless, even these imperfect measurements prove the monoclinic symmetry of the β-uranotile, with a perfect clinopinacoidal cleavage and with the angle β nearly equal to 90°. In the original description by Nováček (7) the value of ρ (001) is given as 2°, or $\beta=92°$. The microscopical measurements by Steinocher show a greater inclination, $\beta=$about 94°. Of the domatic forms, only one orthodome face, with the angle $\rho=48°36'$, has been established on the goniometer.

The β-uranotile crystals from Jáchymov are almost without exception twinned parallel to the orthopinacoid (Figs. 1, 2, 4). A more detailed description of the twinning is given later.

The density was determined by suspension in Clerici's solution and found to be 3.953.

V. Billiet (2) investigated the structure of β-uranotile crystals roentgenometrically, but only Debye-Scherrer diagrams could be made, so that a conclusion concerning the crystal structure of this mineral is not possible.

The quantitative chemical analysis is given in the papers quoted by Nováček (7, 8). The result agrees closely with the theoretical formula for normal uranotile, $CaO \cdot 2SiO_2 \cdot 2UO_3 \cdot 6H_2O$. That all minerals of the uranotile group, viz. uranotile, sklodowskite, and cuprosklodowskite have six molecules of water, has been confirmed by Billiet through roentgenometric studies (2). Schoep originally attributed to these minerals seven molecules of water. With reference to β-uranotile, it does not seem probable that a different degree of hydration could be the cause of its properties differing so considerably from normal uranotile.

ANALYSIS OF THE β-URANOTILE FROM JÁCHYMOV

	I	II Theoret. values
	%	%
SiO_2	13.11	14.02
UO_3	66.29	66.81
CaO	7.32	6.55
H_2O	12.87	12.62
CO_2	not determined, but present in small quantity	—
	99.59	100.00

Under the microscope β-uranotile shows a prismatic or needle-shaped habit; at times its crystals are rather tabular with (010) predominating. The dimensions vary, usually being about 0.1×0.03 mm. In the vertical zone the two pinacoids (010) and (100) are developed; the prisms are terminated by faces of the orthodiagonal zone. One of these, taken as (001), is almost perpendicular to (100), having the angle ρ about 4°, making $\beta=94°$. Besides this face, we find on all crystals positive and

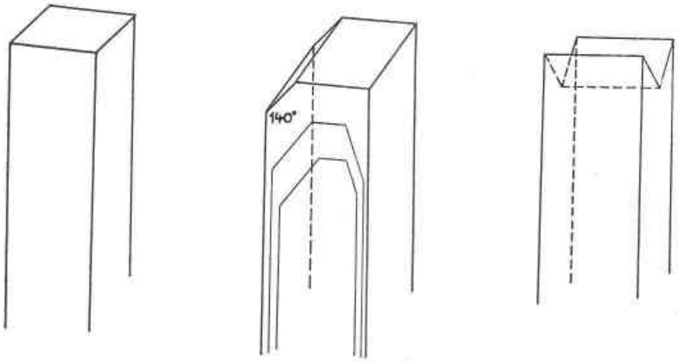

FIG. 1. Common types of β-uranotile from Jáchymov. The first type (left) is the rarest at Jáchymov, whereas it is the most common type on the β-uranotile from Wölsendorf. On the second type (center) a face of a positive orthodome and zonal character are seen. The third type (right) is a contact twin after (100).

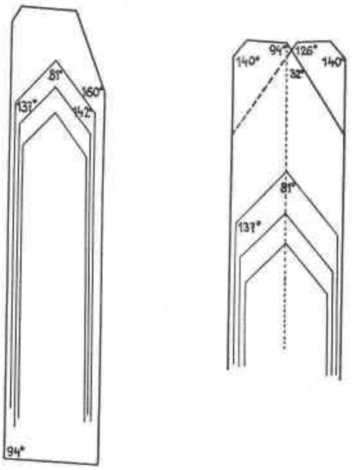

FIG. 2. Zonal character and twinning of β-uranotile from Jáchymov. The angular values are only approximate.

negative orthodomes with ρ values of 50° for $(\bar{h}0k)$ and 58° for $(h0k)$; the former corresponds to the orthodome found by goniometric measurements with the angle $\rho = 48°36'$. Another orthodome has an angle ρ of about 70° (Figs. 1, 2).

The β-uranotile from Jáchymov differs from that of other localities by the zonal development of its crystals, which is best observed on the clinopinacoid. The zones are sharply outlined by the orthopinacoid (100) and orthodomes, which are generally identical with those observed as faces with $\rho = 50°$ and 58°. The basal pinacoid (001) as a rule is absent in the outline of the zones (Fig. 2). In each zone the optical constants change continually from the center to the outside boundary. At the opposite side the sequence is repeated in the same or in a slightly different manner.

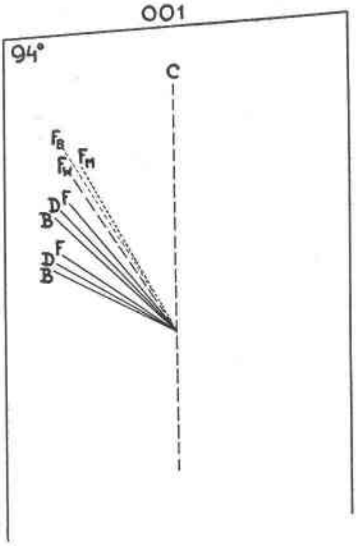

FIG. 3. Extinction angles $c{:}\gamma$ on the clinopinacoid of β-uranotile. The maximum and minimum extinctions of β-uranotile from Jáchymov (spectrum lines B, D, and F) are indicated by full lines, the extinction for green light (line F) of β-uranotile from Bedford (F_B), Mitchell Co. (F_M) and Wölsendorf (F_W) by dotted lines. To avoid confusion, the extinctions for red (B) and yellow (D) light are not given for β-uranotiles from the latter localities.

As stated above, almost all crystals of β-uranotile from Jáchymov are twinned on (100) and frequently they grow together in the same plane (Fig. 4). In more complicated cases they resemble the Carlsbad twins of feldspars, one individual being partially covered by the other (Fig. 2).

The optical symmetry of β-uranotile is monoclinic, agreeing perfectly with the results of the goniometric investigations. The pleochroism is very distinct in sections other than those parallel to (010). In the orthodiagonal direction the crystals are entirely colorless or of a faintly yellowish color. In the directions of β and γ, which lie in the plane of (010), the color is lemon-yellow without a perceptible difference of absorption between the two directions.

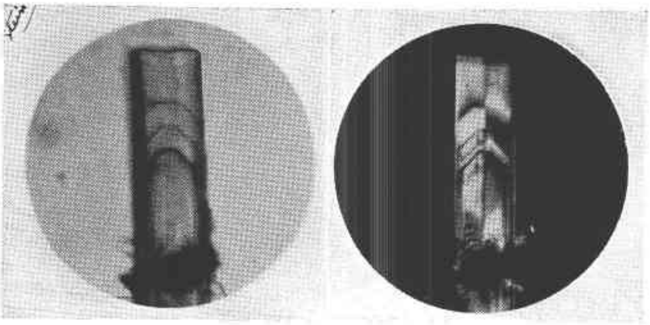
(a) (b)

FIG. 4. (*a*) Photomicrograph of a twinned and zoned crystal of β-uranotile from Jáchymov in ordinary light. The re-entrant angle at the contact of the basal pinacoids is visible. In the outlines of the zones, the faces of orthopinacoid and orthodomes are very prominent, but the (001)-faces are in this case also distinct. ×100.

(*b*) The same crystal between crossed nicols. The composition plane and the imperfect extinction in different zones is quite distinct. ×100.

On (100) the extinction is parallel to the vertical and orthodiagonal axes and shows no dispersion. The longitudinal direction in this case is always positive. On the contrary, if the prismatic crystals lie on the clinopinacoid, which is the most frequent case with fragments, (010) being the plane of perfect cleavage and often the plane of tabular development, they show an oblique extinction and a very strong dispersion, so that in white light in no position is the extinction complete for all colors (Fig. 3).

In red light (approximately the B-line $= 687\mu\mu$) the extinction has been determined to be:

(a) $\gamma : c = 46°-50°$ in the zones of higher refraction
(b) $\gamma : c = 54°-60°$ in the zones of lower refraction.

For sodium light (line $D = 589\mu\mu$)

(a) 44°–48°
(b) 52°–57°

For green light, near the line F ($486\mu\mu$)

(a) 39°–43°
(b) 50°–55°

The extinction direction γ lies in the obtuse axial angle β (Fig. 3). With the higher refraction and smaller angle $c:\gamma$, the birefringence increases (see table of refractive indices and birefringence).

The conclusion to be drawn from the extinction angles in the different zones of growth is that the β-uranotile of Jáchymov is an isomorphous mixture of at least two components, the crystallization of which took place rhythmically and rather regularly. At the beginning of each crystallization phase the substance of lower refraction was formed, characterized also by a greater extinction angle $c:\gamma$, and subsequently the substance of higher refraction and a smaller extinction angle increased in amount. Therefore we always find the substance with lower refraction in the zone near the center of the crystal, the other substance with a higher refraction in the outer portions. Naturally these features are not absolutely equal in all crystals and in all zones, but nevertheless the crystallization sequence from the substance of a lower refractive index and greater angle, $c:\gamma$, to the substance of higher refraction and smaller angle, is constant.

The refractive indices have been determined by the method described above for red (B), yellow (D), and green (F) light and for the different parts of the growth zones. Unfortunately the zonal development and the forms of the zones, in most cases, have not allowed exact measurements of all refractive indices in each part of a zone.

In the accompanying table are given the mean values of a whole series of determinations of refractive indices.

The cause of the zonal development of the β-uranotile from Jáchymov and the anomalies in the optical properties in different parts of the zones remains uncertain. The variable water content alone can not be held responsible for so great differences in physical properties. It seems more probable that a relatively small admixture of another substance (a lead compound?) or a different degree of oxidation of the uranium may be the reason. The presence of lead has not been proved in the samples from Jáchymov, but a minute quanity of lead is not excluded and on the compact specimens of uranotile or of the so called "uranophane" from Mitchell Co., as will be pointed out below, the lead content has been established. A final solution of this question would be possible only if a sufficient quantity of pure material of both extreme portions could be obtained for an analysis.

α						β						γ					
B		*D*		*F*		*B*		*D*		*F*		*B*		*D*		*F*	
min.	max.	min.	max.	min.	max.	min.	max.	min.	max.	min.	max.	min.	max.	min.	max.	min.	max.
1.659	1.668	1.661	1.671	1.665	1.674	1.674	1.685	1.682	1.694	1.692	1.704	1.682	1.694	1.689	1.702	1.700	1.713

From these refractive indices we obtain the following values for the double refraction:

Δ_B min. $=0.023$ Δ_D min. $=0.028$ Δ_F min. $=0.035$
Δ_B max. $=0.026$ Δ_D max. $=0.031$ Δ_F max. $=0.039$

The optical axial angle has been determined on the universal stage for different wave lengths as follows:

$2V_B=73°$ $2V_D=71\frac{1}{2}°$ $2V_F=70°$

The angles calculated from the refractive indices for sodium light are:

$2V_{Na}$ for minim. refractive indices $=66°$
$2V_{Na}$ for maxim. refractive indices $=60°$

The great differences between the calculated and measured values seem reasonable, if we take into consideration the fact that a difference of 0.002 in the refractive index α (not exceeding the errors of observation) causes a difference of 3° in the angle $2V$. Moreover, as stated above, the refractive indices tabulated are mean values taken from many measurements.

β-Uranotile from Bedford, New York

Miss E. J. Armstrong described in this Journal (1) a mineral, which she determined according to Larsen's tables (5, 6) as schroeckingerite. It is evident that this mineral is β-uranotile, as shown from the data given: Extinction 30° ±, strong dispersion $r > v$, negative, $2V = 40°$–$45° \pm$, $\alpha = 1.658$, $\beta = 1.682$, $\gamma = 1.685$; ± 0.003. Pleochroism X (α) = colorless, Y (β) = very pale greenish-yellow, Z (γ) = pale greenish-yellow.

In reply to a letter from one of the writers (R. Nováček) Miss Armstrong says: " . . . observed the CO_2 test through a 14× lens . . . and it seemed to effervesce slightly. This was probably due to admixed calcite." Another specimen, examined later, " . . . seemed to dissolve or disappear in the acid, but without noticeable effervescence." Consequently the mineral from Bedford also corresponds chemically to a silicate rather than to a carbonate though SiO_2 has not been determined because of the small quantity.

Miss Armstrong has kindly sent us a minute amount of the "schroeckingerite" from Bedford, and a sample of the pegmatitic rock with minute needles of the mineral in question. We have been able to verify its identity as β-uranotile.

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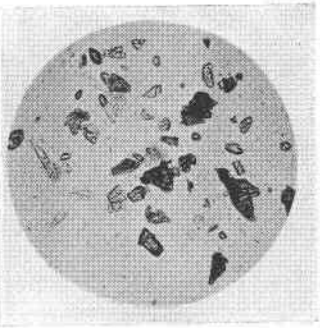

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FIG. 5. Photomicrograph of the β-uranotile from Bedford (crossed nicols): a radiated aggregate of β-uranotile with distinct twinning of some individuals after (100). ×30.

FIG. 6. Photomicrograph of the β-uranotile from Mitchell Co., N. Carolina; the admixture of quartz is visible. Ordinary light. ×15.

Under the microscope the needles and prisms of β-uranotile from Bedford are grouped to form radial aggregates (Fig. 5) and the crystals show in the vertical zone the two pinacoids (100) and (010). The terminal faces (001) are observed in some cases. The twinning after (100) is very frequent, the individual crystals of the twins, like the β-uranotile from

Jáchymov, either grow together in the plane of (100) or partially overlap each other.

The extinction on (010) is:

$$c:\gamma_B = 37\tfrac{1}{2}^\circ\text{–}40^\circ$$
$$c:\gamma_D = 35^\circ\ \text{–}37\tfrac{1}{2}^\circ$$
$$c:\gamma_F = 29^\circ\ \text{–}33^\circ$$

The extinction on (100) is parallel.

There is no distinct zonal structure similar to that shown on the crystals from Jáchymov. The apparent zoning of some prisms is caused by the mutual overlapping of the individuals of the twin-crystals.

β-Uranotile from Mitchell Co., N. Carolina

In the British Museum of Natural History in London there is a sample of pegmatite from Mitchell Co. (No. 191, B. M. 1929) with a yellow, compact, and crystalline mineral, designated as schroeckingerite (locality: Near Spruce Pine, Mitchell Co., N. Carolina). From a detailed study the crystalline parts of the yellow mineral also proved to be β-uranotile; the determination as schroeckingerite seems to have been made according to the data of Professor Larsen.

The pegmatite mother rock of β-uranotile consists of quartz, cyrtolite, muscovite and feldspar with massive "uranophane." The crystals are densely aggregated in small cavities and penetrate the quartz so intimately that they cannot be completely separated. Even minute fragments, which appear macroscopically quite homogeneous, show when crushed, a considerable admixture with quartz (see the photomicrograph 6). Therefore it has not been possible to determine the exact value of the density, nor to carry out a quantitative chemical analysis on perfectly pure material. A sample of 4.5 mg. of a mixture of β-uranotile and quartz was taken and the results are given below in column I. Even a separation using acetylene tetrabromide did not succeed in isolating a sufficient quantity of pure material. This would have necessitated crushing to a very fine powder in which the losses would have been too great. However, 1.7 mg. of β-uranotile were obtained for another microanalysis, the results of which are given in column II.

	I		II	
	%	Ratios	%	Ratios
SiO_2	31.9	2+3.7 (quartz)	29.3	2+3 (quartz)
UO_3	53.5	2	56.4	2
CaO	4.8	0.9	5.0	0.9
H_2O	9.9	5.9	10.2	5.8
	100.1		100.9	

Both analyses clearly show that the ratios of $UO_3:CaO:H_2O$ are the same as in other analyses of uranotile and β-uranotile, *i.e.*, 2:1:6. The higher silica ratio corresponds to the admixture of quartz, about 20%, according to the microscopical examination on the analyzed material.

The compact uranotile from the Flat Rock mine, Mitchell Co., was analyzed previously by F. A. Genth (4) and the results of both of his analyses (one incomplete) agree well with the theoretical values for uranotile.

ANALYSES OF MASSIVE URANOTILE (GENTH)

	I	II	Theoret. values
	%	%	%
SiO_2	13.55	13.88	14.02
UO_3	66.67	66.59	66.81
$Al_2O_3+Fe_2O_3$	traces	traces	—
CaO	6.23	7.11	6.55
H_2O	not determ.	12.02	12.62
PbO	0.74	0.45	—
SrO, BaO	0.42	0.48	—
P_2O_5	not determ.	0.29	—
		100.82	100.00

From several partial analyses of a compact "uranophane" from Mitchell Co. we obtained:

SiO_2	14.5%,	14.8%	and	15.0%
UO_3	61.5%,	62.0%		
H_2O	14.2%,	14.0%		

CaO was not determined; the sesquioxides, principally Al_2O_3, are present to the extent of 2.5% and PbO = 2.1%.

The "uranophane" from Mitchell Co. evidently contains a considerable quantity of impurities; the most interesting of them is lead (admixed gummite?), which might be able to cause essential differences in the optical properties of β-uranotile.

As a result of the comparison, we find that refractive indices of β-uranotile from Spruce Pine correspond to the zones of higher refraction in the crystals of the Jáchymov mineral, and the angle of extinction is smaller than in the zones of highest refraction (Fig. 3). No zonal change in optical properties, nor twinning, have been observed on the mineral from Spruce Pine.

In other features a complete identity with β-uranotile has been observed. The crystals show both vertical pinacoids and positive and negative orthodomes (the angles of the latter being in most cases $\rho=44°$ for the positive, and 48° for the negative orthodome). The base is often absent.

The density could not be determined accurately because of admixture with quartz; the result of a determination made by the suspension method is about 3.8.

The color of the crystals is a more intense yellow than in the β-uranotile from Jáchymov, with identical pleochroism (α = colorless, β and γ = yellow).

Extinction angles:

$$c:\gamma_B = 36°$$
$$c:\gamma_D = 32\tfrac{1}{2}°$$
$$c:\gamma_F = 29\tfrac{1}{2}°$$

The anomalies observed on different crystals are unimportant. The mean values of the refractive indices for different wave-lengths are:

α			β			γ		
B	D	F	B	D	F	B	D	F
1.661	1.663	1.668	1.684	1.688	1.693	1.691	1.697	1.707

The angle of the optic axes, determined graphically from the projection of the measurements on the universal stage, is

$$2V_B = 63\tfrac{1}{2}°$$
$$2V_D = 62°$$
$$2V_F = 60°$$

β-Uranotile from Wölsendorf

In 1931 Professor A. Schoep and Dr. A. Scholz described (12) several uranium minerals, new for this locality in Bavaria, known as the original locality for uranotile. Up to that time these minerals had been described almost without exception only from the Belgian Congo. On one specimen the writers observed several yellow needle-like translucent crystals whose identity with any known mineral had not been established. The brief description of the optical properties* points, here also, to β-uranotile.

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* "Yellow translucent crystals with a perfect cleavage parallel to a plane on which the acute bisectrix is observed. $2V$ relatively small, optical character negative. $N=1.69$, $n=1.68$. Between crossed nicols, in white light, the crystals show anomalous blue interference colors and the extinction is imperfect. The direction N forms an angle of 20°–30° to the longitudinal cleavage fissures and to the twin borders."

Our attention has been drawn to this mineral by Dr. Scholz, who kindly sent us a small quantity for investigation. In a letter from Dr. Scholz he has pointed out a great similarity in the properties of the Wölsendorf mineral and the "schroeckingerite" of Dr. Larsen, and has shown that it is a silicate.

The qualitative tests have shown the presence of SiO_2, UO_3, CaO and H_2O. In dissolving in acids sometimes a few gas bubbles escape, but it is not certain if they are CO_2. Gas inclusions are present also in the crystals, so that the crystals are cloudy. This is a distinct difference between the β-uranotile from Wölsendorf and the normal uranotile from this locality and the American and Jáchymov β-uranotiles.

The quantitative microanalysis (carried out on 2 mg. of material) has given the following data:

		Theoretical values:
	%	%
SiO_2	12.9	14.0
UO_3	66.9	66.8
CaO	7.1	6.6
H_2O	12.6	12.6
	99.5	100.0

The result of the analysis agrees closely with the theoretical values for β-uranotile. Only the per cent of SiO_2 is a little lower; for this difference probably an imperfect separation of silica is responsible as the evaporation with nitric acid was made only once.

The β-uranotile from Wölsendorf is not suitable for goniometric investigation. Besides both pinacoids, other faces also probably occur in the vertical zone. The orthodomes are mostly subordinate and often absent while the base is imperfectly developed. The density was determined in Clerici's solution to be 3.85.

Microscopically, besides the small cavities which cause the translucency of the crystals, the same features may be observed on the β-uranotile from Wölsendorf as on those from Jáchymov and Bedford. The individuals are relatively long (up to 2 mm.), often twinned (Fig. 7), show the same pleochroism (α=colorless, β and γ=yellow) and a strong dispersion of extinction. On the universal stage we have determined:

$$c:\gamma_B = 38\tfrac{1}{2}^\circ$$
$$c:\gamma_D = 36^\circ$$
$$c:\gamma_F = 33^\circ$$

The refractive indices for different wave-lengths are:

α			β			γ		
B	*D*	*F*	*B*	*D*	*F*	*B*	*D*	*F*
1.665	1.668	1.672	1.681	1.686	1.701	1.690	1.695	1.709(?)

$\Delta_B=0.025$ $\Delta_D=0.027$ $\Delta_F=0.037$(?)

According to a personal communication, Dr. Scholz has observed an interesting phenomenon which would support the view that β-uranotile is a rather labile modification of the substance $CaO\cdot 2UO_3\cdot 2SiO_2\cdot 6H_2O$. Several crystals, evidently β-uranotile, when crushed formed small pieces of acicular shape, the properties of which (extinction and refractive indices) correspond to normal uranotile. On a specimen sent us by Dr. Scholz we have noted the same change.

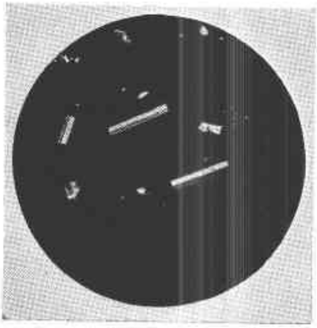

FIG. 7. Photomicrograph of twinned crystals of β-uranotile from Wölsendorf, Bavaria. Crossed nicols. ×15.

Concerning β-uranotile there still remains much to solve. Perhaps in the course of time we shall succeed in finding suitable material, which will enable us to finish the investigation of these unexplainable features.

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