VII.

Study on some secondary uranium minerals.

(Etudes sur quelques minéraux secondaires de l'uranium.)

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The decomposition-products of the primary uranium ores, usually of the uraninite, are very numerous; as they have beautiful colours, they arose the interest of mineralogists in the beginning of the past century. The reports, however, are very incomplete due to many factors which make an exact study of these products very difficult. Among these the most important ones are: The mentioned secondary minerals are relatively scarce; they are mixed with other secondary minerals, e. g. sulphates with gypsum, carbonates and silicates with calcite, wherefore mechanical separation of the pure material was necessarily hindered. Another one, which to-day has to be considered in particular in evaluating the old analyses is the imperfect analytical method used more than fifty years ago, usually an insufficient description of the analytical procedure, finally the apparent fact that the analyst could not distinguish between a homogeneous substance and a mixture of various minerals. These facts are of the greatest importance for the study and revision of the secondary uranium-minerals as even to-day, where some milligrams of the substance are sufficient for a complete quantitative analysis, due to them often a definite examination becomes impossible. For these reasons as well as many other less important ones, still at the present time the orientation in the extended group of the secondary uranium minerals is insufficient, which is demonstrated by the fact that the same species of mineral has several synonymes and that some different species are found in the collections under the same name.

Many very keen observations of mineralogists, published in easily accessible papers, were neglected, new names were formed abundantly or two or more different species were united under one name. Only new analyses and extensive comparative studies could therefore lead out of this chaos of various names.

Věstník Král. Čes. Spol. Nauk. Tř. II. Roč. 1935.

The research of all the minerals belonging to this work requires, however, much time, especially if it is desired to prove them by as many new analyses as possible. Therefore I am dividing the whole study into several parts, the first of which is published in the present paper and deals with the secondary sulphates and silicates, which I was able to study more completely. Even so, there remain of course many problems for further studies.

Before this work was started, my teacher Dr F. SLAVÍK, professor at the Charles IV.-University, Prague, was asked by Professor CH. PALACHE of Harvard University, Cambridge, Mass., if he could have an analysis made in his laboratory of johannite, a rare hydrous uranium-copper sulphate from Jáchymov, or at least send a suitable material for analysis so that the accurate study of M. A. PEACOCK (46) could be completed. I was appointed by Professor SLAVÍK to perform the analysis. I beg to express my heartiest thanks for his numerous advices, his great interest and for his help in procuring the necessary material for research and comparison and in obtaining the literature-references.

In studying the papers concerning johannite and the adjoining minerals the incompleteness of our knowledge about all the groups of the secondary uranium sulphates became apparent. This was why I began to study this group as well as the silicates of the uranotile group in particular and I reached results given in the present paper. I again emphazize that there remains very much for the definite solution of these problems even in this limited section.

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Analytical procedure.

As was mentioned above, the greatest difficulty in examining the secondary uranium minerals is caused by the small quantity of material which is available, being furthermore usually impure. Single pure components can be separated only mechanically by means of election under the magnifying glass or under the microscope, as the considerable solubility of all these minerals in dilute acids, in many cases even in water, makes a removal of the impurities by dissolving impossible. One cannot separate these minerals by heavy liquids because the water in the secondary uranium minerals is bound similarly as in the zeolites, the content of it being easily variable; it could even be exchanged eventually — as in the zeolites — by another liquid or by a gas.

For a quantitative analysis of such small quantities of the pure material as are available, a special microchemical method has been worked out by Hecht (17, 18, 19, 20), which was mostly used in this work. Sometimes instructions as given by Schoep (64), Hueber (27), Schwarz-Bergkampf (67), and Goldschmidt (14) as well as by general textbooks of microchemistry (Emich 10) or by the papers of Benedetti-Pichler were followed.

The water in the sulphates as well as in the silicates was determined by heating the sample slowly in a small bottle (1 cc) with ground-in stopper up to 250—280° C (the silicates still higher).

That no considerable quantity of SO_3 does volatilize was proved by the analysis of a dried sample which contained nearly as much SO_3 as the undried substance (the content of water being subtracted). I did not attempt to construct a dehydratation-curve, having only very small quantities at my disposal, but I intend to obtain one with some of the sulphates. — Usually about 4—12 mgs of the sample were taken; according to the supply of the material 1—3 analyses were carried out. Where it was possible, SO_3 was determined separately. In the sulphates, the copper was determined at first by microelectrolysis (lead was also determined in this way, but the results were always checked by a determination as $PbSO_4$). Iron was separated from uranium by precipitating with ammonia after adding hydroxylamine-hydrochloride (HECHT 17, 18, 19); in a few cases, where the quantity of the material was very small, I was satisfied by the qualitative test, showing that only traces of iron were present. This precipitation of iron was made twice in a porcelain crucible, filtered by a porcelain filterstick (Berlin), ignited and weighed as Fe_2O_3. After the decomposition of the hydroxylamine-hydrochloride, the uranium was determined by precipitation with 8-hydroxyquinoline according to HECHT (17, 18, 19, 20). In order to compare the results, the uranium was precipitated in a porcelain crucible sometimes as ammoniumuranate, ignited, and weighed as U_3O_8 (SCHOEP 64), but the results were not so good, because the precipitation was incomplete though a great excess of ammonium nitrate was used and the ammonia was free from CO_2. The uranium oxine was precipitated in a Jena micro-filterbeaker (SCHWARZ-BERGKAMPF 68), as well as the CaO, which was precipitated by the ammonium oxalate, dried at 110° C, and weighed as $CaC_2O_4 . H_2O$. The new method of the determination of Ca as Ca-pikrolate according to DWORZAK and REICH-ROHRWIG, recommended by HECHT, was not applied for some technical difficulties and will be tested later. Magnesia was found only in traces. SO_3 was determined by the usual method, precipitating with $BaCl_2$; the presence of As was investigated by the method of SANGER-BLACK (TREADWELL 73). For the determination of silica the method of SCHWARZ-BERGKAMPF (67) was used, which proved to be very good¹) being connected with the separation of SiO_2 by HNO_3 and H_2O_2 according to GOLDSCHMIDT (14). It gives better results than

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¹) The normal macrochemical determination of SiO_2 in the natrolithe from Vrahožily (Bohemia) made with 1 g of substance showed 46·69% SiO_2, compared with a microchemical determination of the same sample, where 12 mgs were taken, which shows 46·54% SiO_2.

double evaporation with HCl and filtration with a platinum filterstick. The residue in the platinum crucible, after SiF_4 was volatilized, was usually very small and, as the iron was found in the analysed silicates only in traces, was added as U_3O_8 to uranium. Platinum crucibles of a capacity from 1 to 8 ccs were most advantageous; the filtering was done by a normal glass filterstick according to EMICH, into which was packed a tightly rolled piece of quantitative filterpaper as described by SCHWARZ-BERGKAMPF (67).

In most cases it was necessary to limit the physical research to the optical characteristics. I was able to study crystallographically merely the uranotile of Jáchymov, which is different from the material studied by SCHRAUF (65) and PJATNICKIJ (PJATNITZKY 48, 49), and which I want to call β-uranotile for the present. The above mentioned sulphates, excepting the johannite, described in detail recently by JEŽEK (28) and by PEACOCK (46), are not suitable for exact crystallographical observation. The optical research is limited to the determination of the refractive indices (by immersion), of the optical orientation and the pleochroism. Density determinations of the powdery and fine hairy minerals are very inaccurate, especially when very small quantities of substances are available. Some very interesting phenomena of luminescence could be observed in the ultraviolet light of the Hg quartz lamp (HERAEUS), which allowed to distinguish, at least in some cases, minerals which were very similar macroscopically. So far only some sulphates (zippeite, uranopilite) were examined with X rays by ULRICH and TROUSIL, who employed the method of DEBYE-SCHERRER-HULL, but until now the results were not very satisfactory. The structural examination of uranotile and β-uranotile has to be postponed until sufficient and suitable material is available, though this examination is very desirable in order to decide whether there are two modifications of one compound.

The radioactivity was tested many times, even of the secondary uranium minerals (BARDET 1, KOLBECK-UHLICH 36 etc.), and frequently a very strong activity could be found. I did not repeat these experiments as they are of no special significance for our purpose.

According to quotations from the literature gathered up to now in the mineralogical textbooks (HINTZE 23, DOELTER 9, DANA 6, DANA-FORD 7, NIGGLI-FAESY 44), next to the well defined aqueous copper-uranium sulphate, i. e. the johannite, another, very closely related mineral, called gilpinite is described and further some basic uranium sulphates, containing usually copper or calcium (voglianite, uranochalcite = Urangrün, medjidite, zippeite, uranopilite, uraconite); the last three are united under one name Uranoker. Frequently we find (especially in the German textbooks) the names Uranvitriol, Basisches Uransulphat, Uranblüthe, etc. I propose to treat these minerals, as far as it is possible to identify them, later on; here I wish to mention only that many analyses, especially those of LINDACKER in VOGL's monography on Jáchymov (75), were certainly carried out with impure materials. LINDACKER divides almost all species into a variety with Ca and variety with Cu, though these accessories are not in a stoechiometric relation to the other components. Neither did I find such a high amount of water (over 20%) nor such a low one (5%) as LINDACKER, with any of the secondary sulphates, using either old specimens from the collections or the fresh material from the mines. These facts make the identification very difficult, especially when much of the material, the so-called original, has proved to be quite another than it was labeled.

Johannite.

As was mentioned in the introduction of this paper, the first impulse to the studies of the secondary uranium minerals was given by the question of Prof. PALACHE requiring the analysis of johannite for the work of PEACOCK (46), which appeared during the printing of the present paper. Therefore I shall give only a short review of the researches on johannite and the discussion about its chemical composition. The first two quotations of JOHN (31) from the year 1821 tell us about the chemical composition of the johannite (called by John »Uranvitriol«, »Natürliches schwefelsaures Uranoxydul«). HAIDINGER (15) in 1830 carried out the morphological examination, determined further the presence of copper in this mineral and called it johannite. JOHN (30) returned in 1845 to the same problem with a notice concerning the occurrence, LINDACKER in VOGL's monography (75) published the first two quantitative analyses of this rare mineral from Jáchymov, and FRENZEL (12) gave Johanngeorgenstadt in Saxony as its locality. After a longer interval — omitting the researches on its radioactivity — the johannite was thoroughly studied in 1915 by JEŽEK (28), of course only from the crystallographical and physical point. As for the chemical composition, he assumed the old analyses of LINDACKER and gave a formula, not corresponding to the proportions of valency. Soon after, LARSEN and BROWN (41) described the gilpinite from Colorado with a chemical quantitative analysis, carried out with very impure material and disagreeing with LINDACKER's analyses. On the basis of crystallographical measurements, LARSEN and BERGMAN (39) declared in 1926 gilpinite as identical with johannite and state only that the analyses did not agree with those of LINDACKER. Finally the study of PEACOCK (46) explained many of the morphological problems as well as the chemical composition.

I have carried out the analysis of johannite from Jáchymov, which is quoted in PEACOCK's paper. This was done with the material from the collections of Národní Museum, Prague, Nrs. 4888 and 4889; 7, 9 and 11 mgs of the substance were taken with the following results:

	%		
CuO	8·07	1015 = 1 × 1015	
PbO	0·00		
UO_3	61·34	2143 = 2 × 1071	
$FeO + Fe_2O_3$ . .	—		
SO_3	16·59	2072 = 2 × 1036	
H_2O	13·84	7682 = 7 × 1097	(8 × 960)
CaO, MgO . . .	—		
	99·84		

The analysis gives the formula **$CuO \,.\, 2\,UO_3 \,.\, 2\,SO_3 \,.\, 7\,H_2O$**, which corresponds to 8,48% CuO, 61,01% UO_3, 17,07% SO_3 and 13,44% H_2O. The analysis differs much from both analyses of LINDACKER (see I., II. of Table 1.) as well as from the analysis of gilpinite (III.) and its evaluation (IV.), quoted by LARSEN and BROWN (40) and interpreted for the formula of gilpinite $RO \,.\, UO_3 \,.\, SO_3 \,.\, 4\,H_2O$ ($R = Cu, Pb, Fe, Na_2, K_2$).

It seems impossible to suppose, that in the so-called gilpinite the alkalies, copper, iron, and lead can substitute each other isomorphically, but it seems necessary to eliminate a considerable part of these compounds with the gangue and a part of the water. Perhaps copper can be substituted only by iron, of course not by the total quantity (3,84%) mentioned in the analysis. As according to LARSEN and BROWN (41), the gilpinite was dissolved in dilute HCl and the gangue formed nearly 20%, it is probable that the iron was chiefly extracted from it and that similarly the alkalies and eventually lead do not belong to the gilpinite, but to the accessory mechanical impurities.

I realize that it is a difficult task to evaluate the analysis with a good conscience, but it is unavoidable to try it if not another better analysis is available. Therefore in column V. of Table 1. the evaluated analysis of the gilpinite is given in which not only the gangue and the same quantity of water is subtracted as has been done by LARSEN and BROWN (in order not to complicate matters), but also all the alkalies, lead, and $^2/_3$ of the iron. Although there is no proper basis for this calculation, especially concerning the iron, one must admit, that it has certain merits, as the percentage proportions found are very near to my analysis and thus to the theory.

Table 1.

	I.	II.	III.	IV.	V.
	%	%	%	%	%
UO_3	67·63[2]	67·81[2]	45·67	56·72	60·75
CuO	6·10	5·88	4·67	5·80	6·21
FeO	0·23	0·17	3·84	4·77	1·70
PbO	—	—	0·67	0·82	—
K_2O	—	—	0·56	0·70	—
Na_2O	—	—	1·55	1·93	—
SO_3	20·24	19·79	12·44	15·45	16·55
H_2O + ...	5·62	5·56	10·08	12·15	14·79
H_2O —			1·50	1·66	
gangue	—	—	19·64	—	—
	99·82	99·21	100·62	100·00	100·00

Uranochalcite (Urangrün) and Voglianite.

The first thorough descriptions of these minerals with the quantitative analyses are given in the book of VOGL (75). The uranochalcite, also called Urangrün is mentioned by BREITHAUPT (4),[3] ZIPPE (87)[4] and more precisely in the above book of VOGL. HERMANN (21) describes under the name uranochalcit (resp. uranocalcit) something quite different which

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[2] given as »Uranoxidoxidul«, i. e. U_3O_8.

[3] p. 173: »Uranochalzit. Schimmernd. Gras- bis apfelgrün. Strich apfelgrün. Uiberzug aus zarten bis haarförmigen Fasern zusammengesetzt, die jedoch mit einander verwachsen sind, in nierenförmige Gestalten übergehend. Härte 3—3¾. Besteht aus basisch schwefelsaurem Uran- und Kupferoxyd mit etwas Wasser... Ist jedoch mit dem Uranvitriol nicht zu verwechseln...«

[4] »... findet sich als kleinnierenförmiger mitunter zart sammtartig drusiger Überzug von licht grasgrüner ins Apfelgrüne übergehender Farbe...«

will be treated more in detail in the chapter on silicates. Next to Jáchymov is mentioned Annaberg (Saxony) as the locality of the uranochalcite (Frenzel 12). There are only two analyses by Lindacker (Vogl 75) — see Table 2. (I., II.) — together with two analyses of a mineral (III., IV.) called voglianite by Dana (6, edit. 1868) and described by Vogl (75) as »Basisch-schwefelsaures Uranoxidoxidul«. Vogl distinguishes two varieties of this mineral, one containing Ca, the other containing Cu.

Table 2.

	I.	II.	III.	IV.
	%	%	%	%
U_3O_8	36·20	36·08	79·50	79·691
CuO	6·61	6·50	—	2·243
CaO	10·13	10·08	1·66	0·048
FeO	0·12	0·16	0·12	0·362
SO_3	19·88	20·18	12·34	12·125
H_2O	27·09	27·24	5·49	5·253
	100·03	100·24	99·11	99·722

Vogl (75, p. 119) describes the uranochalcite in similar words as Zippe; the voglianite according to the author forms botryoidal and spheroidal aggregates of various green tints and occurs rarely together with the johannite which it resembles in shape. Both varieties (the one with Ca and the one with Cu) differ from each other by a gradation in tint.[5])

In all of the collections of Prague I found only one sample of the uranochalcite from Jáchymov, which was unsuitable for an accurate analysis (collections of the Charles IV. University, No. 2624), while no voglianite at all could be found. Therefore I had to borrow the material from the collections of the Naturhistorisches Museum and the University in Vienna, from the collections of the Academy of Mines in Freiberg (Saxony) or those of the Convent Teplá.

All specimens designated as uranochalcite or Urangrün and the only specimen of »voglianite« from the Museum in Vienna (No. G 7257) proved upon further examination by no means to be sulphates but in the most part silicates of the uranotile-group, i. e. the cuprosklodowskite, described recently

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[5]) »... sich die kalkhältige Verbindung durch eine pistaziengrüne, die kupferoxidhältige aber durch eine gras- bis spangrüne Farbe auszeichnet.«

by VAES (74). This mineral shall be treated in the second part of this paper (p. 28). The uranochalcite from Johanngeorgenstadt in Saxony, examined by LARSEN (37, p. 149 and 220), see also LARSEN-BERMAN (38), p. 119, kept in U. S. Nat. Museum in Washington (No. 85178) corresponds by its optical and chemical properties also the cuprosklodowskite (see p. 29). One of the specimens from Jáchymov (Museum Vienna No. H 401, XXI, 1900), signed »Urangrün«, was a phosphate from the uranium-mica group. The »Uranochalcite« from the collections of the Directory of Mines in Příbram, found on the heap of the mine »Jánský« (No. 862) contains nearly 50% CuO, no uranium; it belongs probably to the langite-herrengrundite group.

The problem of the existence of the uranochalcite and voglianite therefore had to be left unsolved. Perhaps it will be possible to locate in a collection the material corresponding to VOGL's description of these minerals so that the revision of the old chemical analyses can be carried out.

The medjidite (described originally by SMITH 70) could not be obtained for the investigation; however, I shall publish, if possible, the results of the revision later on. According to TETZNER-EDELMANN (71) the same was found in Seiffen (Schwarzwassertal); one of these specimens is kept in Dresden (Zwinger). TETZNER-EDELMANN ascribe their locality erroneously to Saxony, but it is really on the territory of the Czechoslovak Republic.

Uranopilite.

The most frequent decomposition-products of the uranium ores are the yellow aqueous and basic uranium sulphates, generally called uranium ocker, sometimes also uraconite (uraconise) or zippeite. Though long ago there were two species known to be very different, i. e. the uranopilite and zippeite, usually we find the inaccurate name »ocker« etc. or both names mixed up. The chief cause of errors is the lack of exact optical data, made on the analysed material. I have carried out ten analyses of the yellow secondary uranium sulphates and determined the optical properties of all. I found that six of them correspond fully to the uranium ocker from Johanngeorgenstadt, described by WEISBACH (84), who called it uranopilite, using two analyses of N. SCHULZE. This is identical with the mineral from Jáchymov, analysed in 1854 and called »Basisches Uransulphat« by

DAUBER (8). TETZNER-EDELMANN (71) mention the uranopilite from Seifen from the mine »Glück mit Freude« near Jáchymov, from where the material of SLAVÍK (69), which was a little different optically, was taken. It is possible that there belongs the fibrous uranium-ocker from Příbram, mentioned by REUSS (53). In Table 3. the former analyses of uranopilite are repeated: I. and II. are the analyses of the specimens from Johanngeorgenstadt, analysed by SCHULZE (WEISBACH 84), III. »Basisches Uransulphat« from Jáchymov, analysed and described by DAUBER (8):

Table 3.

	I.	II.	III.[6]
	%	%	%
CaO	2·08	1·96	—
UO_3	77·17	77·46	79·9
SO_3	3·18	4·56	4·0
H_2O	16·59	14·69	14·3
residue	0·39	1·33	—
	99·41	100·00	98·2

My analyses of the uranopilites, given in table 4. (the description of them will be given later on) were carried out on material from the following localities:

I. St. Just, Cornwall, collections of Národní Museum, Prague, No. 17.363.

II. Johanngeorgenstadt, Saxony, Naturhist. Mus. Vienna, No. Aa 3247 (labeled »Uranblüthe«).

III. Příbram, Bohemia, mine Anna, vein »Jánská«, Directory of Mines Příbram, No. 859.

IV. Jáchymov, Mineralog. Institute of Charles IV. University, Prague, No. 10.067.

V. Jáchymov, Mineralog. Institute of Charles IV. University, Prague, No. 10.068.

VI. Jáchymov, Mineralog. Institute of Charles IV. University, Prague, No. 3884.

In the same table are given the refractive indices α, β and γ for the sodium light and the extinction on (010) $(= \beta : c)$.

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[6] In the book of Doelter (9, IV-2, p. 652) this analysis is given with the title »Uranvitriol« and by a misprint the values for UO_3 and SO_3 are changed.

Table 4.

	I.	II.	III.	IV.	V.	VI.
	%	%	%	%	%	%
UO_3	79·89	81·14	80·78	81·20	80·19	81·43
Fe_2O_3	traces	—	traces	0·00	1·17	traces
CaO	1·63	0·18	traces	0·93	0·60	0·00
SO_3	4·24	3·78	4·18	4·04	4·15	3·62
H_2O	13·98	14·68	14·32	14·03	13·81	13·88
CuO	0·00	—	—	0·00	0·00	—
PbO	0·08	—	—	0·00	—	—
As	—	—	—	—	traces	—
	99·82	99·78	99·28	100·20	99·92	98·93
α_{Na}	—	1·6220	—	1·6228	1·6237	—
β_{Na}	1·6248	1·6240	1·6257	1·6248	1·6254	1·6254
γ_{Na}	1·6333	1·6325	1·6339	1·6336	1·6339	1·6339
$\gamma - \alpha$	—	0·0105	—	0·0108	0·0102	—
$\beta : c$	18°	17°—18°	18°	18°	18°	18°

All my analyses are in a good agreement among themselves and with the analysis of DAUBER (8); there is a greater discrepancy in comparison with the analyses of SCHULZE (WEISBACH 84), but even this is negligible for the evaluation of the chemical character of the uranopilite. I am sure that the presence of calcium is caused chiefly by the admixed gypsum, which is only separated with great difficulty from the uranopilite; the isomorphical substitution of the uranyle by calcium is of a secondary importance. The iron (chiefly in the analysis V.) comes from the brown limonitic crusts, which — especially in the case of sample No. V. — cover the pure needle-shaped aggregates of the uranopilite. After all it is probable also that a small part of the iron enters directly into the uranopilite compound, the proof of which being the colour of the specimens. Those containing traces of iron are of a golden-yellow colour (I., III., V., VI.), whilst the others are rather yellowish-green; furthermore, specimens III., V. and VI. have refractive indices which are a little higher.

The amount of water varies, as can be expected with compounds of this type and origin, but even the greatest difference (II—IV) does not mean more than a difference of one molecule of H_2O in the formula of the uranopilite. On the whole, the chemical composition of the uranopilite corresponds to the formula

$$6UO_3 \, . \, SO_3 \, . \, xH_2O,$$

where x is either 16 or 17. The agreement between the analyses and the theory is perfect as is evident from the following table 5., where the theoretical values in % are given for the compound with 20, as well as for those with 17 and 16 molecules of H_2O. In the fourth line are given the data of the compound $6UO_3 . SO_3 . . 10H_2O$, called by me the β-uranopilite which will be treated later on (p. 15). The percentage values of the compound with 20 molecules of water are given so that the first analysis of SCHULZE, indicating 16,59% H_2O, could be interpreted. Thus could possibly be explained the deviating results of WEISBACH (84) on the extinction of the uranopilite, who found it to be 9°, whilst my observations showed always 18°.

Table 5.

$6UO_3 . SO_3 . xH_2O$

x:	$\%UO_3$	$\%SO_3$	$\%H_2O$
20 H_2O	79·59	3·71	16·70
17 H_2O	81·63	3·81	14·56
16 H_2O	82·34	3·84	13·82
10 H_2O	86·84	4·05	9·11

It is not possible to identify any of LINDACKER's analyses quoted by VOGL (75) with one of the uranopilites.

One can characterise the paragenetical, morphological and physical properties of the uranopilite as follows: The pure uranopilite has a clear yellow colour with a greenish tint (e. g. the material of analyses I., II., IV. of table 4.), if traces of iron are present, the colour is golden-yellow. It forms very fine needly and hair-like crystals of a silky lustre, which aggregate woolly into kidney-shaped and grape-like masses,[7]) or it coats over and fills out the fissures in the decomposed ore-material. It is always accompanied by needles of gypsum, often by zippeite (see p. 16) and usually is covered by brown limonitical crusts. Under the microscope we see a very perfect cleavage (010), parallel to which the crystals are also tabular. The extinction on this face, which is terminated by 1 or 2 oblique faces with varying angles (see fig. 1. in the text and fig. 1. on pl. I.), is 18° to the elongation for Na-light in the obtuse angle β; the elongation is negative (normal to the flat-face (010) emerges the obtuse bisectrix α, the direction β makes an

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[7]) In accordance with the description of Dauber (8): »... bildet schön citrongelbe mikroskopische Krystalle... oft zu Kugeln zusammenhäuft auf Uranpecherz...« and Weisbach (84): »Die Aggregate bestehen aus kurzen zarten Krystallhaaren, die, nur sehr locker verwachsen, durch den schwächsten Fingerdruck auseinanderfallen«.

angle of 18° with the elongation — see fig. 1.). The pleochroism is not perceptible on the flat-faces; in the sections normal to the cleavage it can be noticed that the absorption in the direction α becomes a little smaller. The dispersion ($\varrho > v$) is strong and causes abnormal interference colours especially in the sections normal to γ. The needles of this orientation are not extinguished in white light. The determined optical data are in perfect agreement with the observations of Larsen (37), carried out with material from the U. S. Nat. Museum: he found for uranopilite from Jáchymov (No. 84.651) $\alpha = 1{,}621 \pm 0{,}003$, $\beta = 1{,}623$ and $\gamma = 1{,}631$, the extinction $\beta : c = 15^0 \pm 2^0$; for the other sample (l. c. 37, p. 160, C) he gives $\beta = 1{,}627$ and the same optical orientation. In the new edition of the book (Larsen-Berman 38), he has not mentioned uranopilite at all, but has given the data for uranopilite under the name of zippeite (p. 112 and 113) with the following small changes: the extinction $\beta : c = 22^0$ (Cornwall) or 23^0 (Jáchymov). The data given by Slavík (69) for the »zippeite« i. e. uranopilite from Seifen near Jáchymov are differing from ours, inasmuch as the refractive indices are a little higher (1,635 and 1,645) and the extinction is parallel. It is probable, however, that only the needles oriented normal to γ were observed and that by a partial dehydratation something close to β-uranopilite was produced and thus the refraction increased. I have examined next to the analysed uranopilites a series of other »ockers« from diverse collections and have found that e. g. the »Uranocker«. Nos. 1531 and 7136 from the collections of the University in Vienna, No. 18.737 from the Mineralog. Inst. of the Academy of Mines in Freiberg are typical uranopilites, whilst the »uranopilite« No. 18.741 is really the silicate uranotile. The uranopilites Nos. 18.724 and 18.725 were correctly determined

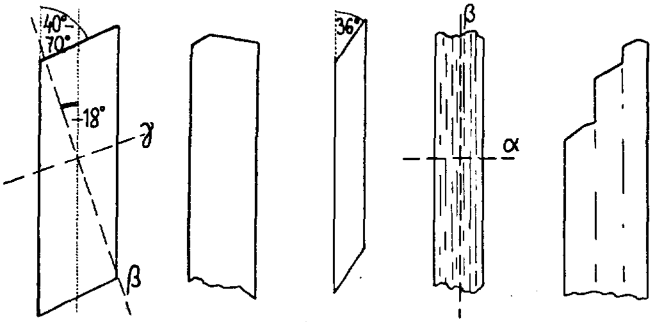

Fig. 1.

as such. The uranopilite exists together with the zippeite on the sample No. 595 from the collections from the German University in Prague. The »Uranblüthe« described by ZIPPE (86) was, seemingly, also a uranopilite, which had carbonates as impurities and therefore was taken for a carbonate by ZIPPE. The sample No. 15.697 from Jáchymov, kept in the collections of the Národní Museum in Prague and labeled »Uranblüthe« (collected by ZIPPE) is the silicate cuprosklodowskite (see p. 29).

On microscopical preparations of uranopilite, which were dipped into the Canada-balsam in the cold and were left there for a longer time an interesting phenomenon could be observed. The needles lose their pretty yellow colour probably by dehydration and become dirty greyish brown; the birefringence decreases till it totally disappears; the extinction, as long as it is perceptible, is almost parallel.

I will carry out an accurate study of this phenomenon, which was also observed with zippeite (see below), if I shall gain suitable material. — It seems also, that only by dehydration from 16 or 17 molecules H_2O to $10H_2O$ the **β-uranopilite $6UO_3 . SO_3 . 10H_2O$** could be produced, which I have succeeded to determine only on the specimen from Jáchymov, kept in Národní Museum, Prague (No. 17.365, labeled »Uranocker«). I have carried out the chemical analysis, however, only once, and with a very small quantity of the substance (this was why I was unable to determine Ca). I found that the chemical composition corresponds to the above formula as is evident from table 5. of various hydrations of the uranopilite (p. 13).

—	UO_3	82,40%	—
—	Fe_2O_3	2,03%	—
—	CaO	??	—
—	SO_3	4,17%	—
—	H_2O	9,40%	—
		98,00%	

There are very remarkable differences between the β-uranopilite and the normal uranopilite with regards to their optical properties. The needles, apparently of the same shape as those of the uranopilite are not as beautifully clear-yellow or lemon-yellow, but somewhat greyish, dirty green or of a brown tint. The pleochroism of the β-uranopilite is also not very perceptible. The extinction is parallel in all positions of the needles and the birefringence is considerably higher. Especially striking is the high refraction, the indices of which are about $\alpha = 1,72$, $\beta = 1,76$ and $\gamma = 1,76$. The elongation is parallel with the optic normal β, the obtuse bisectrix γ is lying in the flat-face (010). It is probable that the β-uranopilite is the same mineral as was mentioned by LARSEN (37, p. 150) with the uranopilite; according to LARSEN this mineral occurs together with the normal uranopilite and gypsum, its β being parallel to the elongation and the refractive indices being $\alpha = 1,68$, $\beta = 1,71$. The angle of the optical axes (2V) is small. The only discrepancy (except the differences of the refractive indices) is the optical sign, described by LARSEN as positive, whilst that of our β-uranopilite is negative. Perhaps the »uraconite« of LARSEN (37, p. 149) from Gilpin Co., Colo. (U. S. Nat. Mus. 85.007), belongs also to these minerals, the indices being $\alpha = 1,75$, $\beta = 1,79$ and $\gamma = 1,85$ and the elongation identical with γ. The »uraconite« from Telegraph mine, Colo., kept in Národní Museum, Prague (No. 2007), is, according to LARSEN (37, p. 160), closely related to the zippeite.

According to VOGL's description (75, p. 119—124) of the »Uranblüthe« called also »zippeite« by the author, a part of the material seems to have been uranopilite[8]) though the analyses (see below table 6., column I.—II.) are very different from those of the normal uranopilite.

The uranopilite and to a lesser extent the β-uranopilite show a very strong yellow-green luminescence in the ultraviolet light. In this way they can be distinguished from the zippeite, which is macroscopically very similar, but does not show the described luminescence.

Zippeite.

The other most frequent secondary sulphate is the yellow zippeite. I use this name of HAIDINGER (16) in spite of the fact that the first analyses of zippeite which were carried out by LINDACKER (VOGL 75) and are given in the following (6.) table of analyses (Nos. I.—II.), concern another compound, closer resembling the uranopilite. I did not yet succeed to locate this VOGL-LINDACKER

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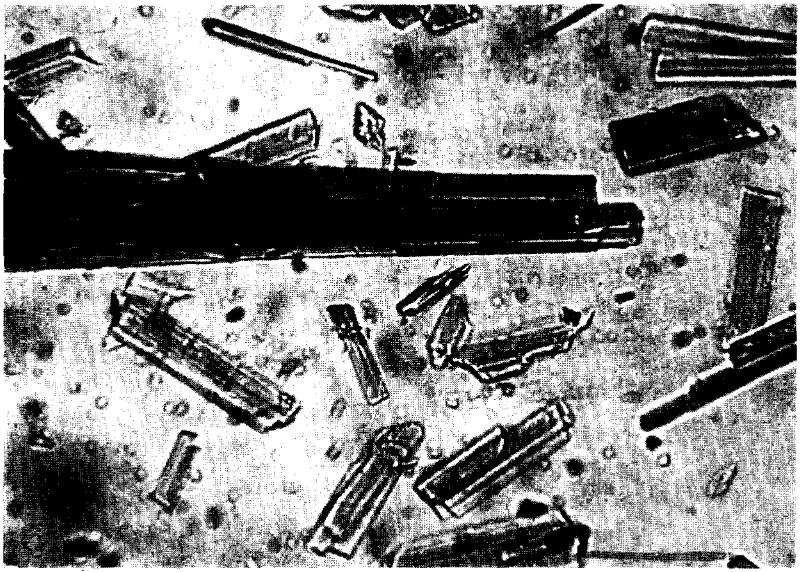
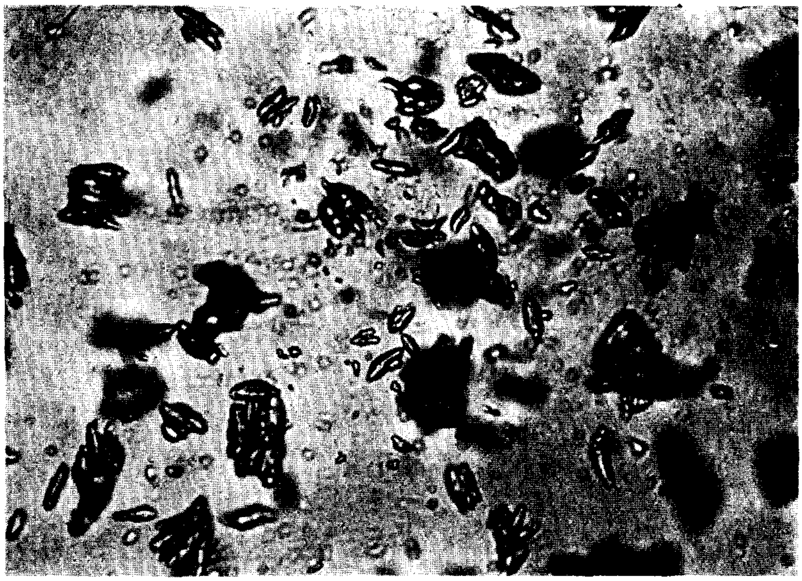
[8]) p. 122: »...Die Uranblüthe in beiden Variationen kommt immer in Gesellschaft des Uranokers und Gypses vor. Vom Uranoker ist sie durch die reine hochschwefel- oder schön orangegelbe Farbe und durch eine in kleinen Schuppen und Nadeln auftretende Kristallisation verschieden. — Diese Kristalle treten entweder einzeln auf oder sind zu kugligen oder länglich runden Partien angehäuft...«

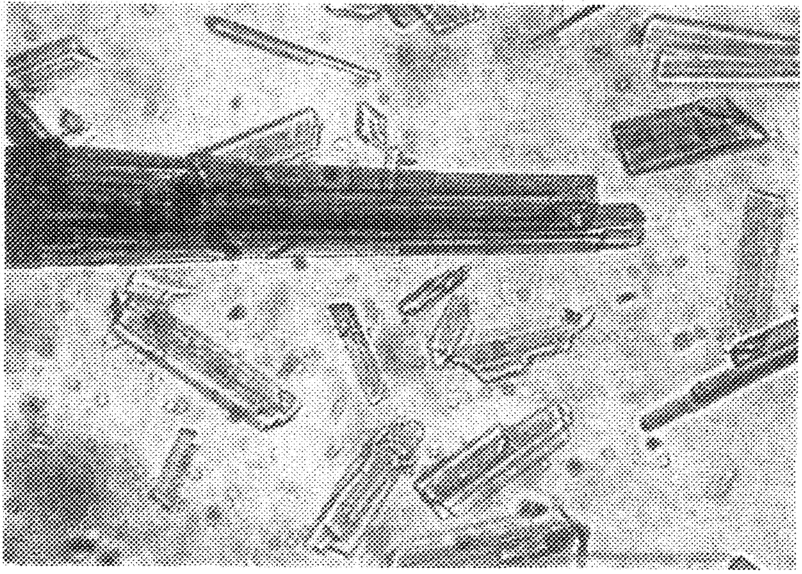


1. *Uranopilite* from Jáchymov; needles and laths terminated by faces with varying angles. Enlarged 200 diameters.

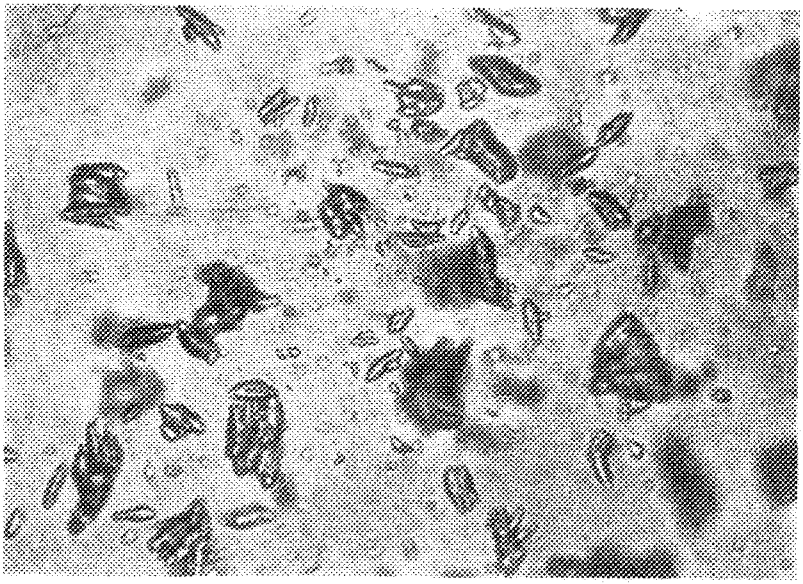


2. *Zippeite* from Jáchymov; tiny spindle- and lense-shaped crystals, partly contorted, forming rosettes and parallel rows. Enlarged 200 diameters.

Phot. by *F. Ulrich.*

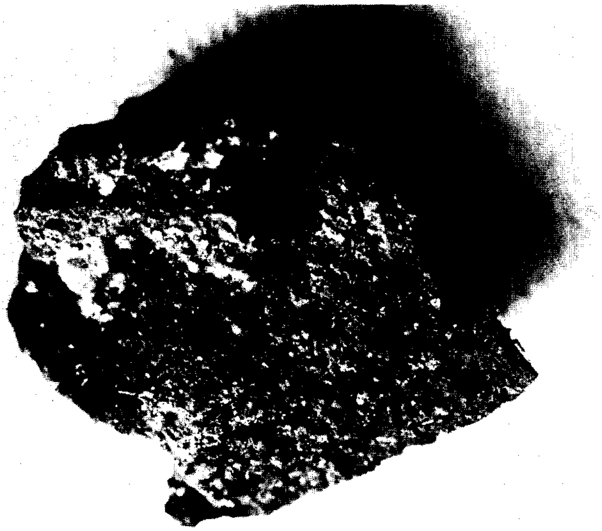


1. *Uranopilite* from Jáchymov: needles and laths terminated by faces with varying angles. Enlarged 200 diameters.

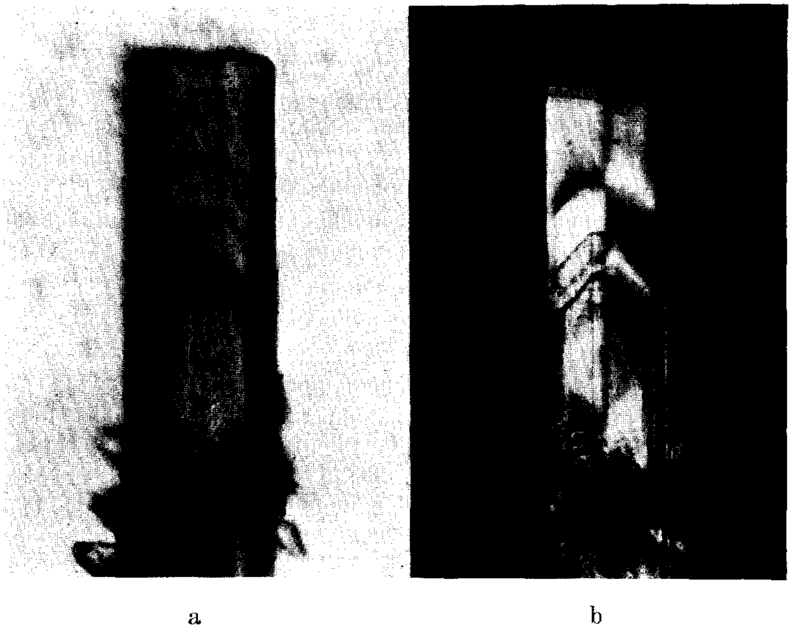


2. *Zippeite* from Jáchymov; tiny spindle- and lense-shaped crystals, partly contorted, forming rosettes and parallel rows. Enlarged 200 diameters.

Phot. by *F. Ulrich.*



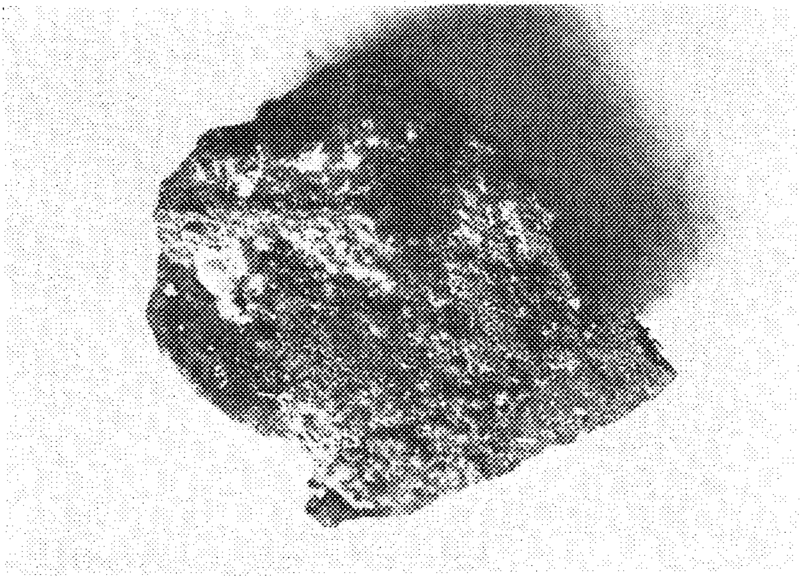
3. Kidney- and wart-shaped coatings of *uranopilite* and some *zippeite* (lower right) which quite recently must have been formed on limonite and a piece of wood (below) in an abandoned gallery. Sample from Jáchymov. Enlarged 2 diameters.



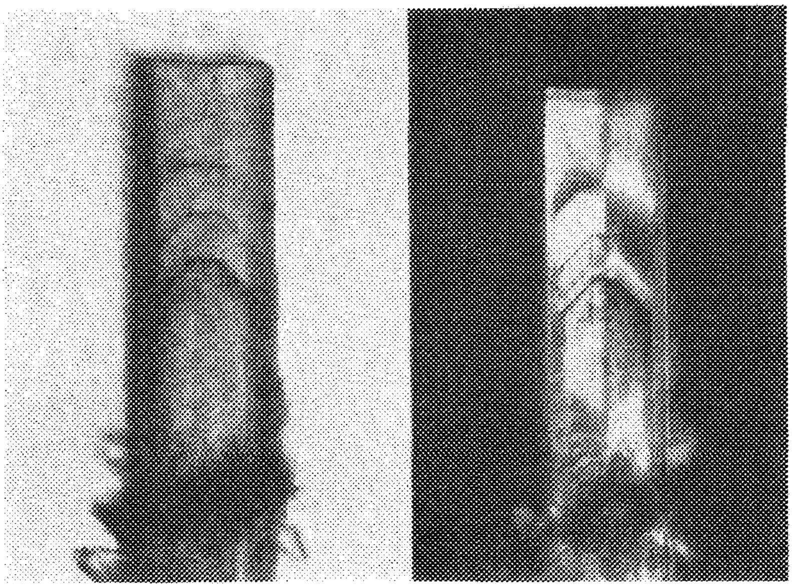
a b

4. Twinned crystal of *β-uranotile* from Jáchymov with a very distinct zonal structure and twinning line parallel with (100). a) in ordinary light, b) in polarized light between crossed Nicols. Enlarged 200 diameters.

Phot. by *F. Ulrich.*



3. Kidney- and wart-shaped coatings of *uranopilite* and some *zippeite* (lower right) which quite recently must have been formed on limonite and a piece of wood (below) in an abandoned gallery. Sample from Jáchymov. Enlarged 2 diameters.



a b

4. Twinned crystal of *β-uranotile* from Jáchymov with a very distinct zonal structure and twinning line parallel with (100), a) in ordinary light, b) in polarized light between crossed Nicols. Enlarged 200 diameters.

Phot. by F. Ulrich.

compound. The original specimen of Vogl's zippeite, kept in the collections of the Directory of Mines in Jáchymov No. 295, as well as the later quotations concerning zippeite (Hess 22, Larsen 37) are more or less concordant with the three sulphates, mentioned below, which were analysed by me (see table 7.). Therefore I suppose to be right in ascribing the name »zippeite« to these minerals and to annul the problematic compounds of Vogl's zippeite till the analyses of Lindacker can be checked. Much nearer to our zippeite is the »Uranoker« of Vogl (75, p. 124—127) given in table 6. in columns III.—V., differing essentially by the amount of water only, which is, however, in zippeite very variable.

Table 6.

	I.	II.	III.	IV.	V.
	%	%	%	%	%
UO_3 (U_3O_8?)	62·042	67·855	70·936	66·052	58·48
Fe_2O_3	—	0·172	0·413	0·863	2·46
CuO	5·208	—	0·235	—	—
PbO	—	—	—	—	2·21
CaO	—	0·607	—	2·622	3·03
MnO	—	—	—	—	0·35
SiO_2	—	—	—	—	1·46
SO_3	17·361	13·063	7·116	10·165	10·22
H_2O	15·232	17·693	20·880	20·057	20·58
	99·843	99·390	99·580	99·759	98·79

Doelter (9) ascribes the analyses I. and II. to zippeite and III., IV. and V., called by Vogl »Uranoker«, to uraconite.

My three new analyses (see table 7.) were carried out on specimens from Jáchymov, kept in Národní Museum, Prague (Nos.: I. 4891, II. 4890 and III. 2009).

Table 7.

	I.	II.	III.
	%	%	%
UO_3	71·98	73·47	74·76
Fe_2O_3	1·17	0·00	—
CaO	1·88	4·13	3·58
SO_3	10·02	10·19	10·15
H_2O	13·95	11·32	11·37
As	—	—	0·00
	99·00	99·11	99·86

α_{Na}	1·575	1·636	1·616
β_{Na}	1·615	1·694	1·677
γ_{Na}	1·646	1·732	1·700
$\gamma - \alpha$	0·071	0·096	0·084

There is to be noticed the amount of lime which varies much and is certainly not caused by impurities (gypsum etc.); further the considerably higher amount of water of specimen I. I have specially determined the water in a fresh specimen of zippeite from the mines. Much more water was found in it (15,11%), accordingly also lower refractive indices $\alpha = 1{,}570$ and $\gamma = 1{,}641$. The material, however, is not homogeneous; next to individuums with the mentioned low indices (and perhaps still lower) are others, morphologically of the same shape, but with much higher indices. One can observe on the analysed specimens (espec. II. and III.) similar, though not striking, phenomena. It seems, that a series of hydrates of the zippeite exists. Thus Lindacker's analyses Nos. III., IV. and V. (see table 6.) with more than 20% of water are entitled to be quoted there. An extreme on the other side — though the refractive indices do not differ much from ours — is the zippeite from Grand Wash, Fruita, Utah, analysed by Schaller and described by Larsen (37, p. 160, 3) and Hess (22), the analysis of which was carried out with a very impure material and gave the following data (I. column):

Table 8.

	1.	2.
	%	%
UO_3	72·50	80·3
SO_3	11·11	10·7
H_2O	8·77	9·0
CaO	0·97	—
$P_2O_5 + As_2O_5$	3·73[9]	—
CuO	0·96	—
SiO_2	1·96	—
	100·00	100·00

After subtracting the impurities (libethenite, gypsum etc.) Hess calculated the formula of zippeite from the percentages, given in column II. (table 8.) as $2UO_3 . SO_3 . 3H_2O$ though the amount of water is nearer to 4 molecules. Similarly, i. e. by over-calculating the old Lindacker analyses (see table 6., p. 17, col. I., II.), Hess writes the formulas $2UO_3 \ . \ SO_3 \ . \ 5H_2O$ and

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[9] by difference.

$2UO_3 . SO_3 . 8H_2O$. The agreement is approximate only, as is evident from the theoretical values, given in the following table:

Table 9.

$2UO_3 . SO_3 . x\ H_2O$

x:	$\%UO_3$	$\%SO_3$	$\%H_2O$
$3H_2O$	81·02	11·33	7·65
$4H_2O$	79·00	11·05	9·95
$5H_2O$	77·09	10·78	12·13
$6H_2O$	75·26	10·53	14·21
$7H_2O$	73·52	10·28	16·20
$8H_2O$	71·86	10·05	18·09

From my analyses can be derived a similar formula with 5 or 6 molecules H_2O provided CaO forms an isomorphical mixture with UO_2O. It is very difficult to express the amount of lime stoechiometrically, as the quantity varies much so that the formulas become very complicated. We must consider that there is always a mixture of various hydrates of the zippeite, one of them being far prevalent; within limits of some tenths of percent other sulphates must be regarded to be present as impurities, and finally it cannot be overlooked that the microanalytical methods are no more precise using such small quantities. I do not exclude the possibility of the existence of one hydrate containing lime in a simple stoechiometrical proportion while other hydrates are nearly or absolutely free from this substance. This is only a suggestion which for its verification will require a larger quantity of material.

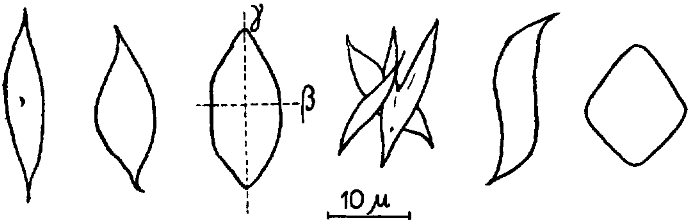

Fig. 2.

All zippeites show, as mentioned above, the same morphological character with constant optical properties except that the refractive indices decrease evidently with increasing hydration. Macroscopically the zippeite forms an earthy orange-yellow powder occuring usually with the uranopilite as crusts or kidney-shaped or grape-like coatings on the disintegrated ore-material. It is usually accompanied next to uranopilite with gypsum, limonite and sometimes by the uranotile and the uranium micas. By a slight pressure the aggregates of zippeite fall apart into small clods composed of a mass of tiny imperfect crystals not surpassing ever 0,02 mm. The crystals are usually spindle- or lense-shaped, variously stretched, crooked, or rounded off, forming frequently rosettes or parallel rows (see fig. 2. in the text and fig. 2. on the pl. I.). Plates with a rhombic outline are rare, and their angles vary much. The optical orientation is as follows: α is normal to the flat-face of the lenses, spindles and rhomboids, γ approximately bisects their acute angle. The dispersion is slight. Pleochroism appears strongly only perpendicular to the flat-face, where in the direction α the zippeite is colourless or very pale-yellow, and deep yellow in the direction γ. The tint of the yellow colour is a little paler in direction β. These observations agree with the data of Larsen for zippeite (37, p. 159—160), where for four samples the following data (for Na-light) are given:

	α	β	γ	$\gamma—\alpha$
1A (zippeite, Jáchymov)	1,630	1,70	1,720	0,090
1B (zippeite, Jáchymov)	1,620	1,680	1,720	0,100
2 (»uraconite«, Gilpin Co.)	1,660	1,710	1,760	0,100
3 (zippeite, Fruita, Utah)	1,630	1,689	1,739	0,109

The pleochroism as well as the optical orientation quoted agrees with my results, only the crystals of Larsen seem to be partially better developed. Next to the rhomboids Larsen gives laths with the outline of a parallelogram, the extinction being 32⁰—41⁰. The elongation according to Larsen is identical with the crystallographical axis c, the flat-face is then (010).

Next to Jáchymov and its vicinity as well as localities in Saxony (see Frenzel 12, who gives the name of the zippeite to the uranopilite, Schiffner 56 etc.) and in America (Gilpin Co., Fruita), the following localities may be mentioned for the occurence of zippeite (if the given descriptions may be trusted): Příbram, where Reuss (52, 53) describes a powdery ocker next to a fibrous one (i. e. probably uranopilite). I have identified microscopically the zippeite on specimen No. 860 from the collection of the Directory of Mines at Příbram from the vein Jánská (mine Anna). Slavkov (Schlaggenwald): Rücker (54) and J. Hoffmann (24, 25) mention the powdery uranium ocker from Slavkov without any description. »Uranblüthe« from Slavkov, mentioned by Hoffmann is of a green colour, contains SiO_2, CuO, P_2O_5, is amorphous, and therefore does not belong to our group. — Drmouly (Dürrmaul) and Schönficht: The occurences of uranium minerals in the neighbourhood of Mariánské Lázně (Marienbad), i. e. Drmouly and Schönficht, show another character, where phosphates and silicates prevale and neither the real zippeite nor uranopilite could be identified definitely. Perhaps a yellow coating from Jáchymov, described by JOHN (32), also belongs to the zippeite. Cornwall: PENROEP (47) mentions briefly the zippeite (probably uranopilite) and »uraconite« (»uranochre«) from Cornwall, SCHEERER (55) writes about an uraniumochre from Norway.

This closes, for the present, my research on secondary uranium sulphates. It is evident that much is missing to completely systematize the hitherto confused data on these minerals, but I hope that I shall be able to proceed in this work. It is certain, that the complicated calculations of the old analyses (RAMMELSBERG 51) cannot be trusted as the compounds are much simpler and vary only in the degree of hydration. I think that the great series of haphazard names will become useless and that there, where compounds only differ by the amounts of water, the different quantitative relations will be indicated sufficiently by suitable indices (α-, β-, γ- etc.) to the fundamental name of the mineral.

Silicates of the uranotile group.

The crystallized silicates of the uranotile group occur on the uranium-ore deposits less frequently than the sulphates. Although the amorphous silicates (gummite, eliasite) are comparatively frequent, the uranotile belongs to the mineralogical rarities, and further members of its group were still found quite recently. To these belongs firstly the sklodowskite from Belgian Congo described by SCHOEP (61), who showed immediately the analogy of this mineral with the uranotile (58, 59, 61, 62, 63). Furthermore recently the cuprosklodowskite from Kalongwe, Belg. Congo, was found, analysed only qualitatively and optically by VAES (74). This mineral was identified with one from Jáchymov and was analysed quantitatively for the first time by myself.

The uranotile, labeled also uranotyle, or uranophane, was investigated many times chemically as well as crystallographically since 1870, when a sample of it from Wölsendorf, Bavaria, was firstly described by BOŘICKÝ (2, cf. 3 and CECH 5). The most important investigations are: The works of WEISBACH (81, 82, 83) and WEBSKY (on so-called »uranophane« 78, 79, 80, see also SCHUCHARDT 66); the report of PREIS (50) on the analysis of the material from Wölsendorf; then the crystallographical observations by SCHRAUF (65) and PJATNICKIJ (PIATNITZKY) 48, 49. Finally a series of notices and studies from which especially that of LARSEN-HESS-SCHALLER (40) should be mentioned, where the identity of the so-called »lambertite« from Lusk, Wyoming, described by LIND and DAVIS (42) with the uranophane (i. e. uranotile) is proved by comparison of the optical properties with the material from Saxony. Of the elder authors, GENTH (13), KERR (34), FOULLON (11), WATSON (77), NORDENSKJÖLD (45) and HOFFMANN (26) were studying the uranotile and the associated minerals. I do not quote all their analyses partially because they were carried out on inhomogeneous material, and partially because they are compiled in the textbook of DOELTER (9, II-3, 421—424). Of recent date are only the works of SCHOEP (58, 59, 61 etc.) comparing this mineral with the sklodowskite and the investigation of the new minerals from Wölsendorf by SCHOEP and SCHOLZ (57); finally the work of THOREAU (72) on the uranotile from Katanga, which will be considered later in this paper. As for the radioactivity, it is treated in the work of BARDET (1) and others (see also MÜGGE 43, p. 440).

The occurrence of the uranotile in Jáchymov was supposed to be doubtful up to the present time according to some textbooks of mineralogy. Neither DANA (6) nor HINTZE (23) quote the uranotile from this famous locality of the uranium minerals. In the Czech textbook of JEŽEK (29) its occurrence in Jáchymov is doubted, although a part of the material of SCHRAUF (65) and PJATNICKIJ (48 ,49) was undoubtely from Jáchymov. I have found in the collections of the Mineralogical Institute of the University in Vienna both specimens from Jáchymov, mentioned by PJATNICKIJ (Nos. 8874, 8875), correctly labeled. In the Naturhist. Museum in Vienna is also kept a typical fine needly uranotile from Jáchymov (No. Aa 6741, 1873, L2); a similar uranotile exists in the collection in Freiberg (No. 26.582). In the collections in Prague I did not find such specimens and only from the Directory of Mines in Jáchymov I could obtain five little pieces on which a few uranotile needles can be seen. Tiny needles of the uranotile, having a little different shape, exist on the specimen No. 9761 in the collections of the Mineralog. Inst. of the Charles IV. University and No. 21.213 in the Národní Museum in Prague. I did not examine these specimens with the exception of making an optical identification. I paid much more attention to the specimens of uraninite from Jáchymov, coated with crusts of tiny greenish-yellow needly crystals, differing from the normal uranotile by a greater thickness of the needles as well as by the greenish colour-tint. One of these specimens was labeled »schröckingerite« in the collections of the Mineralog. Inst. of the Charles IV. University, Prague (No. 8100). The determination was made by means of LARSEN's tables (37) and further supported by the evolution of CO_2 in dilute acids. This phenomenon, however, is caused by the admixed calcite or perhaps also by the uranothallite. As could be confirmed since, the data of LARSEN (37, p. 131) were obtained with a similar material. By an extensive qualitative test it was determined that in the so-called »schröckingerite« next to a considerable quantity of uranium and water also lime and silica are present, while all the CO_2 came from the carbonate impurities. The optical properties, however, agree with those given by LARSEN and shall be discussed later; these were entirely different from those of the real uranotile. Therefore I decided to carry out a quantitative analysis as well. This was made possible by finding a similar mineral from Jáchymov in the collections of the High School of Mines in Příbram (labeled »uranotile«) and in the Naturhist. Museum in Vienna, where »uranotile« No. 3747 from Jáchymov is kept, which corresponds in every detail to my material. On this specimen, which was bought from Ing. MAUCHER (Munich), was a notice »Wahrscheinlich neues Uranmineral« with a pencil addition »Uranopilit?«. It is certain, that here uranopilite or another sulphate is out of the question. The specimen No. H 403 from Jáchymov (Schweizergang) has a similar character.

I had at my disposition 7,5 mg of nearly pure material containing only traces of carbonates. The quantitative analysis gave following data (column I.):

Table 10.

	I. %	II. %
UO_3	66·29	66·81
SiO_2	13·11	14·02
CaO	7·32	6·55
MgO	—	—
H_2O	12·87	12·62
Fe_2O_3	—	—
CO_2	*)	—
	99·59	100·00

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*) not determined, but present in a small quantity.

The result of the analysis agrees with the theoretical values (II. column) of the compound

$$CaO . 2UO_3 . 2SiO_2 . 6H_2O.$$

A review of some better analyses of uranotile, given in table 11., shows that without doubt the chemical formula of the uranotile is identical with that of our mineral, but, as mentioned above and described further in detail, the optical properties of the analysed mineral are very different from those of the uranotile. There fore I propose to call this mineral β-uranotile, until an X-ray investigation will be possible.

I.—III. analyses by Bořický (2) of the uranotile from Wölsendorf.
IV. analysis by Preis (50) of a specimen from the same locality.
V.—VI. analyses by Winkler (Weisbach 81, 82), of the uranotile from the mine »Weisser Hirsch« near Neustädtel, Saxony.
VII.—VIII. analyses by Genth (13) of the uranotile from Flat rock mine, Mitchell Co., N. Carolina.

Table 11.

	I.	II.	III.	IV.	V.	VI.	VII.	VIII.
	%	%	%	%	%	%	%	%
UO_3	67·034	66·471	—	64·98	63·93	62·84	66·67	66·59
SiO_2	13·636	14·007	13·701	13·52	13·02	14·48	13·55	13·88
CaO	5·054	5·489	—	5·25	5·13	5·49	6·23	7·11
H_2O	12·48	12·84	12·679	14·18	14·55	13·79	—	12·02
Fe_2O_3	0·316	0·908	0·31	1·44	3·03	2·88	—	—
Al_2O_3					—	—	traces	traces
MgO	traces	—	—	0·20	—	—	—	—
P_2O_5	0·448	—	—	—	—	—	—	0·29
PbO	—	—	—	—	—	—	0·74	0·45
SrO	—	—	—	—	—	—	0·13	0·48
BaO	—	—	—	—	—	—	0·28	—
	98·968	99·715	—	99·57	99·66	99·48	—	100·82

Briefly the formula of uranotile and of its group in general shall be discussed. It is certain, that it corresponds to the simple proportions $CaO : 2UO_3 : 2SiO_2$ and one can have doubts only about the water of hydration as in the case of the sulphates whether it is normally 6 or 7 molecules. — Watson (77), whose analysis is not correctly evaluated (61,28% U_3O_8 corresponds to 62,44% UO_3 but not to 60,14%) gives the formula $CaO . 2UO_3 . 3SiO_2 . 7H_2O$, subtracting one molecule of SiO_2 and H_2O because hyalite was admixed to the material, so that the final formula agrees with ours. SCHOEP (58, 61, 62) on the basis of the sklodowskite analysis comes to a formula with $7H_2O$. He found in the sklodowskite 13,41% H_2O and increased this amount after evaluating his results up to 13,94%. Other estimations of H_2O (61, 63) gave 13,06%, 13,77%, 13,33% H_2O. The theoretical value for $7H_2O$ is 14,68% and 12,86% for $6H_2O$. — THOREAU (72) gives the analysis of the uranotile from Chinkolobwe (Katanga), carried out with 70 mgs of substance:

Table 12.

	I.	II.	III.
	%	%	%
Loss on ignition (250°)	—	15·4	13·6
»Insoluble«	9·0	6·8	7·5
UO_3	75·4	73·1	71·9
CaO (+ traces MgO)	—	7·4	7·0
Pb	—	traces	—
V, Se, Fe	—	0·0	—
		102·7	100·0

From I. and II. THOREAU derives the formula $CaO . SiO_2 . 2UO_3 . 6H_2O$ (theoretical values are given in column III.) and supposes the analysed mineral to be a special variety of the uranotile with only half of the usual amount of SiO_2. Considering the analysis and the procedure more in detail, we come to other conclusions, especially when all the optical and physical properties correspond totally with those of the uranotile. SiO_2 is given as »insoluble« and it is conceivable that a large amount of the silica remained in the solution and was precipitated with uranium or calcium, if the usual evaporation to dryness was not made. The sum of the components (102,7%) shows that large errors of observation have to be taken into account. As for the amount of water, corresponding in this case to nearly 7 molecules (theoretically 15,48% H_2O) no great weight may be given to it as the analysis shows insufficiences mentioned above.

The number of six molecules of water of hydration in the uranotile group is justifiable also by the analyses of the cuprosklodowskite (see below), giving 11,72% and 11,88% H_2O, i. e. still a little less than the theory requires for $6H_2O$ (12,28%).

The β-uranotile, analysed by myself, forms, as mentioned, comparatively thick needle-shaped crystals of yellow to yellow-green colour, troubled partially by tiny grains of a black substance, which could not be definitely determined. The length of the crystals is maximally 2—3 mms. Some measurements were carried out on the goniometre with two circles, but the quality of the material did not allow exact observations and I had to subsist on approximative data only. The crystals (fig. 3.) are in general somewhat tabular parallel to the face on which sometimes with the bare eye and always under the microscope (especially in polarized light) a sharp twinning-line, parallel with the elongation of the needles, and a zonar structure of the crystals can be seen (fig. 4. in the text and on the pl. II.).

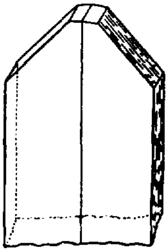

Fig. 3.

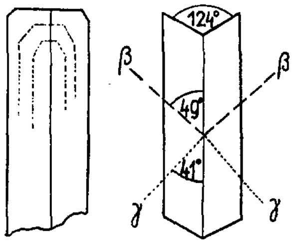

Fig. 4.

Untwinned crystals are very rare. The cleavage is parallel with the flat face. If the same is to be considered as a clinopinacoid *b* (010) as is evident from the optical orientation, also the orthopinacoid *a* (100) must be present on the crystals, being the twinning plane, further the orthodoma *d*, and rarely a small face *c* (001), nearly normal to both pinacoids. On the faces *a* vertical stripes may be seen as the effect of the perfect cleavage in the direction of *b* (010). There was only one measurable angle ϱ of the orthodomatical face *d*, the value of which is 48°36′ (average of 7 measurements, max. 49°0′, min. 48°19′). The signals of the pinacoidal faces *a* (100) and *b* (010) were generally indistinct, so that the measurements of the angle between them vary from 87° to 93°. ϱ of the face *c* (001) is no larger than 2°. It is interesting that the value ϱ of the face *d* 48°36′ is very near to ϱ of the face (103) on the sklodowskite 48°29′ measured by Schoep (58, 62). It is, however, very problematical to compare values of angles which are accidentally near to each other, but I mention this coincidence because the form of the crystals is after all very similar to the form of the sklodowskite and uranotile from Belgian Congo, though these are not twinned. Twinned crystals without any accurate description were also mentioned by Schoep (59, 60) in the case of chinkolobwite, which was proved to be identical with sklodowskite.

The most marked differences between β-uranotile and the typical uranotile are shown in the optical properties of the latter: On the faces (010) we see very abnormal interference colours; the sections of this orientation do not extinguish in the white light and only in the Na-light it is possible to determine the extinction to the twinning-line (i. e. to the elongation of the needles or to the axis c ($c : \gamma = 41^0$, $c : \beta = 49^0$) (fig. 4.). The direction α is normal to (010) so that the sections parallel with (100) show a parallel extinction. On (010) the optical figure in the convergent light is to be seen, $2E_{Na} =$ ca. 130^0, with a very strong crossed dispersion $\varrho > v$.

By means of the immersion method was determined for Na-light

$$\alpha = 1{,}665$$
$$\beta = 1{,}686 \qquad \gamma - \alpha = 0{,}031$$
$$\gamma = 1{,}696$$

On another, non analysed specimen, paler yellow, was found

$$\alpha = 1{,}662, \quad \beta = 1{,}686, \quad \gamma = 1{,}694; \quad \gamma - \alpha = 0{,}032.$$

The pleochroism is strong, α nearly colourless, β and γ deep yellow. The density, determined in the Clerici solution, is 3,953.

All these properties are in perfect agreement with the description of the so-called schröckingerite, given by LARSEN (37). After inquiring from the author, I was informed that the described mineral is a silicate and not a carbonate; therefore, the material of LARSEN is completely identical with mine, as to its chemical composition.

The original schröckingerite from the collections of the Národní Museum and the Charles University (Prague) forms thin sexangular scales of light yellowish-green colour, the refraction indices of which are quite little higher than that of Canada balsam 1·54). The schröckingerite is uniaxial or anormally weakly biaxial, with negative character of the birefringence. More detailed optical and chemical investigations will be published later.

For comparison I determined also the optical data of the typical uranotiles from the following localities:

Schneeberg, Saxony, $\gamma = c = 1{,}667$.

Wölsendorf, Bavaria, $\alpha = 1{,}647$, $\gamma = c = 1{,}669$.

Jáchymov (fine yellow needles) $\alpha = 1{,}648$, $\beta = 1{,}666$, $\gamma = c = 1{,}675$.

Jáchymov (tiny needles in the cavities of uraninite, collections of the Miner. Inst. of the Charles IV. Univ., Prague, No. 9761): $\alpha = 1{,}647$, $\beta = 1{,}661$, $\gamma = c = 1{,}668$.

Kasolo, Belgian Congo, originally labeled »sklodowskite«, Min. Inst. Univ. Prague, No. 9480: $\alpha = 1{,}649$, $\beta = 1{,}670$ ca., $\gamma = c = 1{,}674$.

One can assume that the β-uranotile is a mineral differing essentially physically from the normal uranotile, and representing perhaps a second modification of the compound $CaO \cdot 2SiO_2 \cdot 2UO_3 \cdot 6H_2O$. If I shall succeed to gain suitable material for the crystallographical and X-ray examination, it will be possible to decide about the mutual relation between both minerals.

Cuprosklodowskite.

In the begining of this paper (p. 8) I have mentioned that nearly all the samples which I found in the collections of various museums, labeled as »uranochalcite«, »Urangrün« or »voglianite« were by no means sulphates, but silicates of the uranotile group. Up to this time I did not know any hydrous silicate of copper and uranium, corresponding by its formula with the uranotile and sklodowskite; for this reason I have called the new mineral jáchymovite according to the locality Jáchymov (Joachimstal). During further studies, however, I was informed by the abstracts and, due to the kindness of Prof. BUTTGENBACH, by the original paper of VAES (74) about the detection of mineral of the uranotile group from Kalongwe, Katanga (Belg. Congo), containing copper. VAES was able to determine the optical properties and the qualitative composition only (CuO, SiO_2, UO_3, H_2O), whilst I have carried out two quantitative chemical analyses of the material from Jáchymov. The optical data determined by myself are not quite in agreement with those of VAES in the values of the refractive indices, but till now I was unable to procure the comparative material. I will call this mineral cuprosklodowskite, which name was given to it in the abstracts. But if it will be proved that the mineral of VAES is essentially different I would claim the name jáchymovite for the species from Jáchymov.

The cuprosklodowskite from Jáchymov forms very fine needly crystals, aggregated in radial rays, in silky coatings, kidney-shaped crusts and thin films of pale green colour on the disintegrated limonitical gangue. The green colour passes to greenish-yellow from the centre of the aggregates to their periphery. The needles are very tiny, 0,03 mm — 0,05 mm long, 0,005 — 0,01 mm thick, a little flattened. The laths end usually in a cross face with varying slants, some angles according to the observations of Vaes are approaching values of ca. 70°—75°. The pleochroism is perceptible, being of a greenish-yellow colour parallel with the elongation and yellowish or nearly colourless normal to the same. The needles extinguish parallel, only in a few cases I found an inclined extinction — probably another mineral was admixed. The elongation is generally positive, in which case the needles show abnormal interference colours. Crystals with negative elongation are nearly isotropic, showing no pleochroism and are vertically striped, which is caused by a cleavage parallel with (010). The refractive indices were the following (Na-light, all the material from Jáchymov):

»Urangrün« (analysis I., see table 13.), Vienna, University, No. 4549: $\alpha = 1{,}654$, $\beta \doteq \gamma = 1{,}664$.

»Voglianit« (analysis II., table 13.), Vienna, Naturhist. Mus. No. G 7257: $\alpha = 1{,}655$, $\beta \doteq \gamma = 1{,}667$.

Not labeled sample from the collections of the Convent in Teplá: $\alpha = 1{,}655$, $\beta \doteq \gamma = 1{,}667$.

Very near are also the refractive indices of the cuprosklodowskite from the collections of Freiberg Nos. 18.735, 18.732 and 18.731, labeled »uranochalcite«, from the University in Vienna »uranochalcite« No. 5201 and from the Naturhist. Museum in Vienna »uranochalcite« No. A1 733, VII, 54 (1836) -66.166. Typical cuprosklodowskite are also the samples: No. 1574 from the collections in Teplá, the »uranochalcite« No. 2624 from the Mineralog. Inst. of Charles IV. University (Prague) and from the collections in Zwinger (Dresden, Saxony) the »uranochalcite Nos. 16.161 and 21.724 and the »Uranocker« No. 10.183.

Among the »uranium carbonates« kept in the Národní Museum of Prague I found some other samples of cuprosklodowskite from Jáchymov, which were labeled: No. 15.693 as liebigite, 15.697 as »Uranblüthe« (Zippe's sample, see p. 15) and 15.698 as uranothallite.

The optical properties agree with those given by Larsen (37, p. 149) and assumed also by Larsen-Berman (38, p. 119) for the uranochalcite from Johanngeorgenstadt (Saxony), kept in U. S. Nat. Museum (No. 85.178): $\alpha = 1{,}655$, $\gamma = 1{,}662$. Only the optical sign is given by Larsen as positive and the elongation also as +. This is conceivable, as it is very difficult on examining such a small quantity of material, to find the needles oriented so, that the elongation is negative, i. e. turned on the edge, normal to the cleavage and to the flat face. According to the kind communication of. Mr. W. F. Foshag, Curator of Mineralogy and Petrology at the U. S. National Museum, Washington, this sample also contains SiO_2.[9]) The values of the refractive indices, given by Vaes (74), are essentially different. This author gives $n = 1{,}68$—$1{,}70$ as one of the most pronounced characteristics of the cuprosklodowskite, by which the same may be distinguished from the sklodowskite, whilst our data are only a little higher than those of the sklodowskite ($\alpha = 1{,}613$, $\beta = 1{,}635$, $\gamma = 1{,}657$) and nearly identical with those of the uranotile (see above). If the refractive indices of the material of Vaes are really so high as they are given and if the chemical composition corresponds quantitatively with our mineral, it is possible, that here also two modifications exist similar to the uranotile and β-uranotile.

The analyses of the two first samples mentioned above, i. e. I. of »uranochalcite« (University Vienna, No. 4549), 4,5 mg taken, and II. of »voglianite« (Naturhist. Museum Vienna, No. G 7257), 8,5 mg taken, make the formula of the cuprosklodowskite

$$\mathbf{CuO \,.\, 2SiO_2 \,.\, 2UO_3 \,.\, 6H_2O,}$$

as is evident from colum III. (table 13.), where the theoretical values are given.

Table 13.

	I.	II.	III.
	%	%	%
CuO	8·99	9·07	9·04
PbO	traces	0·18	—
UO_3	64·96	64·65	65·03
Fe_2O_3	traces	traces	—
CaO	traces	—	—
SiO_2	13·40	13·90	13·65
H_2O	11·72	11·88	12·28
SO_3	0·00	—	—
	99·07	99·68	100·00

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[9]) »...I have examined the sample of socalled uranochalcite from Johanngeorgenstadt in U. S. N. M. 85178 and find that it leaves a residue of silica upon treatment with acid. The mineral forms a pale grass green coating a fraction of a millimeter thick on the rock. It has a fibrous structure and a silky luster...«

It is interesting, that observations similar to mine were made by JOHN (30) in the year 1845, who, examining carefully a sample from Jáchymov, labeled »Urangrün«, found, that a silicate (»Kieselkupfer-Uranoxyd«) is present, which has similar properties as cuprosklodowskite.[10] This observation is quoted by VYSOKÝ (76), KENNGOTT (33, 1844—49, p. 69), ZEPHAROVICH (85, I., p. 225) and KLVAŇA (35); but vainly we search for this quotation in other textbooks. R. HERMANN (21) in 1859 published a note on the uranochalcite (he also calls it »uranocalcite«), which according to his description forms kidney-like amorphous masses of a metallic appearance containing SiO_2. The description as well as the analysis[11] show, that he had obviously not a homogeneous material, but i mixture, totally different from our cuprosklodowskite as well as from the uranochalcite of BREITHAUPT and VOGL. I quote these data only for completeness. The note of HERMANN was righteously criticised by KENNGOTT (33, 1859, p. 124—125). Whether the mineral, described by HOFFMANN[12] (24, 25) from Slavkov, is identical with the cuprosklodowskite, is not possible to be decided definitely.

In the collections of the Directory of Mines in Schneeberg-Neustädtl (Saxony) and in the Mineralienniederlage in Freiberg I found some samples labeled »uranochalcite« or »uranocalcite«. These are inhomogenous mixtures of the yellow and

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[10] »Das Mineral bildet einen sehr unvollkommen traubigen Überzug, oder es findet sich bloss in angeflogenen Theilen und in dünnen Rinden; die Farbe ist meistens apfelgrün, indessen auch zeisiggrün; es ist matt und etwas fettglänzend; schwach durchscheinend oder undurchsichtig. Das Gestein, auf dem es angeflogen vorkommt, ist sehr verwittert«... ...»Es ergibt sich aus diesen Versuchen, dass das Joachimsthaler Mineral kein basisches schwefelsaures Uran, sondern dass es zusammengesetzt sey aus:

Kiesel-Erde	ungefähr geichen Theilen	Phosphorsäure	einige Prozenten
Kupfer-Oxyd's		Arseniksäure?	
Uranoxyd's		Eisenoxydul's	
Wasser		Unbestimmten	
		Metall-Oxyd's	

[11] p. 321: Schwefel 5·79, Arsenik 7·23, Kupfer 10·21, Ni 0·97, Fe 2·31, Kieselerde 4·40, Wismuthoxyd 36·06, Uranoxyd 14·41, Eisenoxyd 11·95, Eisenoxydul 3·27, Wasser 2·40, Silber Spur. Sa. 100·00 (!! rightly 99·00).

[12] ...Eine dunkelgrüne Uranmasse, die im Anflug leicht kristallinisch wird, zeigt in kompakter Masse im polarisierten Lichte amorphe Eigenschaften; sie wurde vorläufig mit dem Namen Uranblüthe belegt; ...besteht...aus ..CuO, UrO_3, H_3PO_4, SiO_2.

green decomposition products of the uraninite, prevalently of the silicates, which are even observable in the thin sections under the microscope. They are mixed so intimately, that there can be no idea of a separation of the pure material. A little better is the yellow crystalline decomposition-product of the uraninite from Drmouly near Mariánské Lázně (ČSR), which seems to be homogeneous. A note concerning this mineral will be published later on.

Summary.

The chemical composition, physical properties and the occurrences of some secondary uranium minerals are treated. These minerals are:

Johannite $CuO . 2UO_3 . 2SO_3 . 7H_2O$.
Uranopilite $6UO_3 . SO_3 . 16$ (or 17) H_2O.
β-uranopilite $6UO_3 . SO_3 . 10H_2O$.
Zippeite $2UO_3 . SO_3 . 5—6H_2O$.
Uranotile and β-uranotile $CaO . 2SiO_2 . 2UO_3 . 6H_2O$.
Cuprosklodowskite $CuO . 2SiO_2 . 2UO_3 . 6H_2O$.

Further the names of some minerals, based on older analyses, were checked and the data concerning the optical properties of some of the mentioned minerals were corrected.

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