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URANIUM IN ACCESSORY MINERALS OF GRANITE ROCKS IN VEPORIDES

(Figs. 1—4)

A b s t r a c t: Content and distribution of uranium in apatite, zircon, monazite and allanite have been studied by the fission tracks method. The results showed the highest uranium content (1600—2600) in monazite among the minerals studied. The contents are lower in other minerals (apatite: 15—67 ppm, allanite: 100—410 ppm). In zircon the distribution of uranium is frequently inhomogenous. Peripheral zones of inhomogenous grains (500 ppm in average) have approximately thrice a higher uranium content than their centre (150 ppm in average). The average content 300 ppm is in zircon grains with homogenous uranium distribution.

Р е з ю м е: Содержание и распределение урана в апатите, цирконе монаците и алланите были изучены методом треков осколков деления ядер урана. Из изучаемых минералов, результаты показали самое высокое содержание урана в монаците (1600—2660). В других минералах содержания ниже (апатит: 15—67 ppм, алланит: 100—410 ppм). В цирконе распределение урана часто не гомогенное. В периферийных зонах не гомогенных зерн (500 ppм в среднем) бывает содержание урана трижды выше чем в их центрах (150 ppм в среднем). В цирконовых зернах с гомогенным распределением урана среднее содержание 300 ppм.

Introduction

In the last years, on the origination of the Čs. uranový priemysel (Czechoslovak uranium industry) a detailed investigation of granite rocks in the Veporides was carried out in respect of possible occurrences of uranium ore-mineralization. On the ground of the study of accessory minerals and measurements of total β-activity of heavy fractions, monazite, orthite and xenotime are referred to as possible bearers of uranium by M. C h o v a n (1973). On the other hand, zircon, apatite, titanite, epidote and other accessory minerals are not regarded interesting as for radioactive elements. In this paper the results of the study of content and form of uranium occurrences in apatite, zircon, monazite, allanite are presented from the following types of Veporide granite rocks:

1. biotite granodiorite of Sihla type. Locality: road-cut Čierny Balog-Sihla, at elev. point 1020,2 — Tlstý javor.

2. biotite granodiorite. Locality: northern slope of the Nízke Tatry (mountains), Holičná, 400 m west of elev. point 1299,0.

3. leucocrate granite of Rimavice type. Locality: České Brezovo, a quarry 200 m northwest of the periphery of the village.

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4. pegmatite-biotite granite. Locality: Mlynová valley, 200 m north of elev. point 910,0.

5. biotite granite of Hrončok type. Locality: Kamenistá dolina valley, at the valley mouth of Spády.

7. metasomatic porphyric biotite granite of Vepor type. Locality: Ipeľ r. valley, 320 m southeast of elev. point 769,1.

8. biotite granodiorite. Locality: eastern ridge of Trestník mountains, 100 m west of elev. point 1300,3.

Data obtained from the study of accessory minerals of the above rock types are presented in a paper by M. Chovan – J. Határ (1978) Total uranium contents in the rocks were determined by gammaspectrometry in the labs of CSUP in Příbram, the values are taken from J. Kamenický (1973).

Accessory minerals were analysed by the fission tracks method. In cases of higher uranium contents the method was combined with examination by X-ray microanalyser of JXA – 5A type by the analyst RNDr. J. Krištín, CSc. Uranium contents were determined by means of external detector. For this purpose "Estrofol" (A. Skowronski, 1974) was applied. Radiation of the samples by thermal neutrons (in IBJ in Swierk, Poland) was followed by etching of the "Estrofol" in standard manner. Uranium contents were determined in single grains. The total uranium content in minerals does not include uranium associated with inclusions.

Results and discussion

Results of measurements are in Tables 1, 2, 3. The value of concentration coefficient illustrates a comparison between uranium contents in apatite and in the respective rock. The value is omitted with other minerals because of the lack of grains measured and because of inhmogenous distribution of uranium in the minerals.

Apatite – is in all rocks studied. It is the most frequent accessory mineral. Apatite occurs in the form of typical column crystals, mostly limpid, occasionally yellowish. In apatite uranium appears in three optically distinguishable forms. Most frequently it is homogenously dispersed over the entire grain analysed. The second form is represented by uranium associated with inclusions. Apatite, zircon and a dark uncertain phase were found by optical examination. Uranium concentrations are lower in inclusions in apatites than in the grain alone. Different uranium concentrations in zircon inclusions in apatite depend upon the morphological type of zircons. Uranium contents above the so-called pleochroic cores did not change in concentrations in relation to the entire grain. The third form of uranium occurrences in apatite is that of uranium infiltrations. They are regarded as a secondary phenomenon. The infiltrations form narrow belts with max. width of 15 μm, formed by etched pits after a high concentration of tracks. Table 1 shows that the mean uranium content in apatite is low – 36 ppm, the contents ranging from 15 to 67 ppm. The mean value of the concentration coefficient is 2,6. U. V. Ljachovič (1973) quoted 96,4 ppm as the mean uranium content in apatites of granite rocks, and 28,0 as the concentration coefficient. These data show that uranium contents in apatites of the Veporide granide rock are very low as well as concentration coefficient in respect of increased uranium content in the rocks. As for uranium

Table 1

Concentration of uranium in apatite and in entire rock, in ppm. Ko – concentration coefficient calculated as a ratio of uranium concentration in apatite and in entire rock Uranium concentration in apatite was calculated after G. A. Wagner (1969). Integral dose of thermal neutrons 1.63×10^{15} nvt. cm^{-2}.

No.	Apatite (ppm)	Whole rock (ppm)	K_o	Number of grain counted
1	23 ± 2	9,9	2,3	70
2	29 ± 3	10,2	2,8	103
3	47 ± 3	34,6	1,4	70
4	41 ± 3	5,2	7,7	181
5	67 ± 12	19,9	3,4	52
7	15 ± 2	4,0	3,7	70
8	25 ± 2	6.7	3,7	101

associations, apatite in the samples studied, is not significant mineral.

Zircon – is a common accessory in the samples studied, although in lower amounts than apatite. Zircon occurs most frequently in the form of perfect crystals of small size (to 0,1 mm). In zircon grains with homogenous distribution of uranium the mean content of it is 300 ppm. The distribution of uranium in zircon is frequently inhomogenous, the mean uranium content in marginal zones of in homogenous grains is 504 ppm, i. e. approximately thrice as much as the mean uranium content in the centre of grains – 152 ppm. The uranium content in the centre of zonal grains is always lower than in external parts of the grains (Fig. 1). Zonal distribution of uranium in zircon may be controlled by the presence of old cores with a lower uranium content or the increased uranium concentration in marginal zones of crystals was caused by altered uranium concentration around the growing crystal. Examination by X-ray microanalyser did not show any chemical inhomogenity of grains studied. In the grains analysed, the Hf and Zr distributions are homogenous, La, Ce, U contents are low below the limit of determinability (0,1 % U). Contents in single grains are presented in Table 2.

Allanite – is a common accessory mineral of more basic types of Veporide granodiorites. Its content is extremely high in the sample 1. Most frequently it forms irregular grains, occasionally shortcolumm crystals of brownblack colour. The distribution of uranium in alanite is homogenous, there are, however, marked differences in uranium concentrations in the two samples (Table 3). Low uranium content in sample 1 – granodiorite of Sihla type – and high uranium content in allanite of biotite granite – 4 – are in good accordance

Table 2

Concentrations of uranium in zircons in ppm. Presented are concentrations of uranium in zonal grains (two data), and in grains with homogenous distribution of uranium (one datum). Concentration of uranium in zircon was calculated according to J. P. Carbonnel — G. Poupeau's (1969) formula. Integral dose of thermal neutrons $2{,}24 \times 10^{14}$ nvt. cm^{-2}.

No. 4			No. 5			No. 7			No. 8		
Grain	Centre	margin	Grain	Centre	margin	Grain	Centre	margin	Grain	Centre	margin
1	66	513	1	588		1	61	499	1	48	386
2	103		2	174		2	97		2	109	574
3	199		3	157		3	263		3	302	
4	984		4	345		4	174		4	397	561
5	42	100	5	126		5	50	287	5	109	759
6	271	475	6	184	667	6	43	359	6	98	
7	89	615	7	54	443	7	59	499			
8	353	541	8	144	739	8	320				
9	533	820									
10	513										
11	242	402									
12	336										
13	41	328									
14	271										
15	70										

with U. V. Ljachovič's (1973) finds. Ljachovič quoted the mean uranium content in allanite of granodiorites — 112,3 ppm and of biotite granites — 368,8 ppm. There is a tendency of increas ing uranium content in later crystallization products. Contents of uranium in single grains in granodiorite (1) range from 40 to 177 ppm, in granite (4) from 175 to 882 ppm.

Monazite —is in very small amounts in most samples. Its higher content was in samples 3 and 7. Crystals are mostly idiomorph, yell ow or yellow-brown in colour. The surface of monazite is rough, opa que. Grain size ranges up to 0,2 mm. In monazite studied, uranium is comparatively homogenously dispersed. Differences in concentrations of uranium in grains of the same sample are

Table 3

Concentration of uranium in allanite and monazite, in ppm. Concentration of uranium was calculated with the value of the mean etchable length of track $R = 8.10^{-4}$ cm. Integral dose of thermal neutrons 5.56×10^{14} nvt. cm^{-2}.

No.	Mineral	Number of grain counted	Average concentration	Individual concentration
1	allanite	14	107	73, 177, 122, 125, 57 104, 40, 93, 124, 121 110, 116, 124, 116
4	allanite	4	480	278, 586, 175, 882
3	monazite	5	1607	937, 2838, 1617
7	monazite	8	2657	1188, 1456 2970, 3322, 3234, 2244 2031, 2816, 2662, 1978

considerable (Table 3). Uranium contents in monazite, ranging within 1607—2657 ppm are regarded as average for monazites in granite rocks. P. M. Hurley — H. W. Fairbairn (1957) quoted 1866 ppm as mean uranium content in monazites of granites, U. V. Ljachovič (1973) found out 2198,5 ppm as the mean uranium content in monazites of granite rocks.

Examination by X-ray microanalyser showed that monazite of sample 3 is sometimes zonal, some grains are irregularly inhomogenous. It is proved by

Fig. 1. Zonal distribution of tracks after induced fission of ^{235}U in zircon. The figure shows evident increased concentration of uranium in external parts of the grain. Photographed by J. Kráľ.

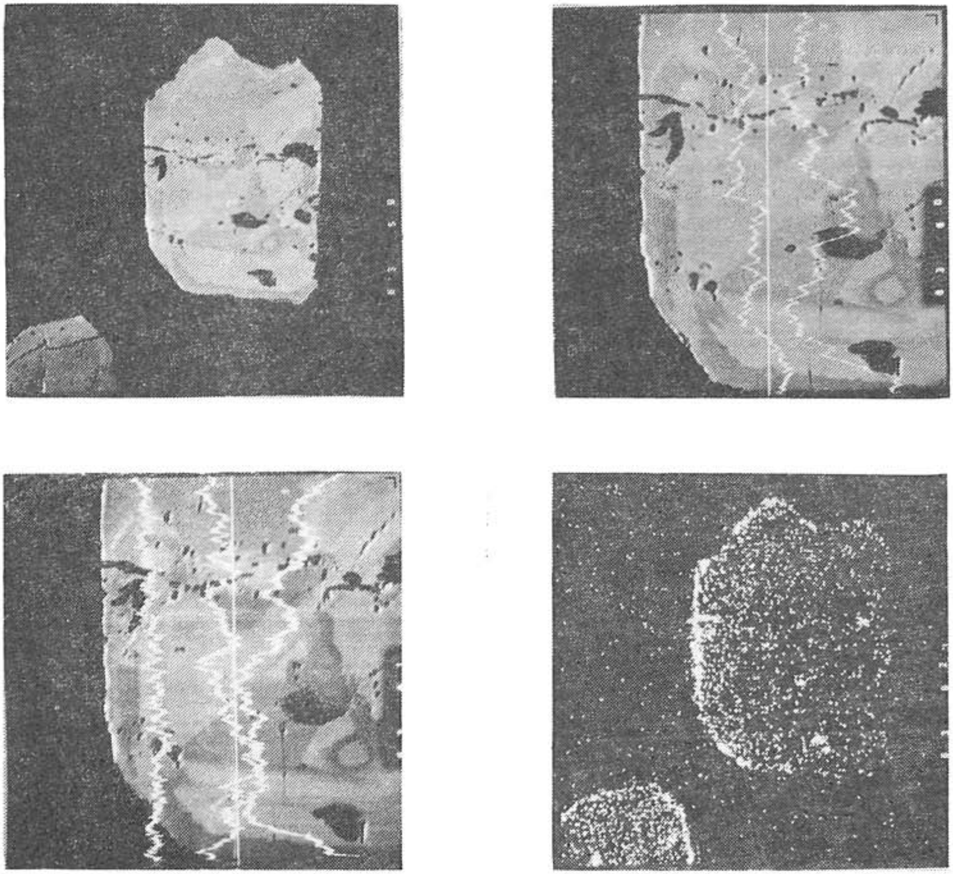
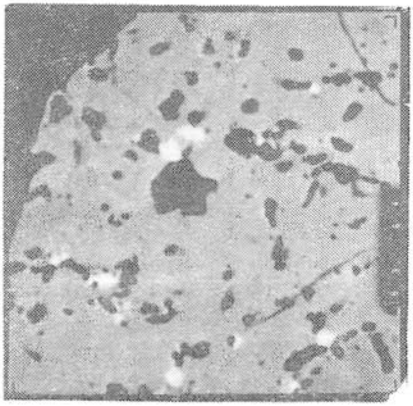


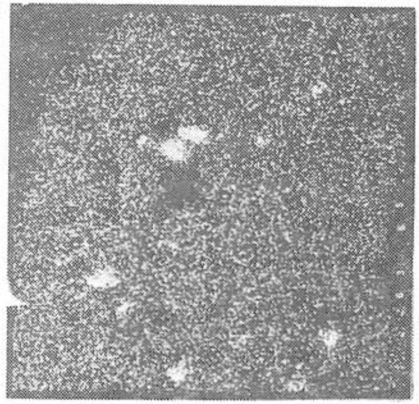
Fig. 2. Distribution of U, Th, Ca, Ce, La in monazite of sample 3 as found at examination by X-ray microanalyser. Photo J. Krištín; a) Composition Magn. 300×, b) Composition. Magn. 600×, Linear analysis of UI_α, ThI_α, c) Composition. Magn. 600×. Linear analysis of CeL_α, LaL_α, CaK_α. d) Distribution of CaK_α. Magn. 300×.

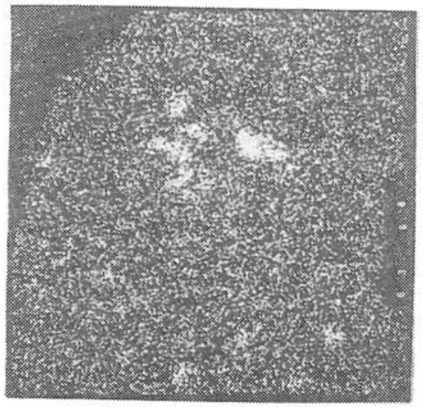
irregularly alternating lighter and darker phases. We have not found out for sure that some elements are associated with a certain phase. Distribution of thorium is markedly irregular and its content in monazite is higher than uranium content. Distributions of Ce and partially of La are inhomogenous (Fig. 2). Ca is accumulated on the margins of grains to form a calcium fringe there. Also thorium content is increased in the marginal zone. Minute inclusions of irregular shape with increased uranium content are also present. Such inclusions are particularly distinct in monazites of sample 7, fig. 3. They are light irregular enclosures with increased uranium and Si contents, and with decreased La, Ca, Fe contents. Perhaps they are coffinite inclusions. Presence of such inclusions is manifested by increased concentration of tracks (Fig. 4)



a

b





c

Fig. 3. Distribution of uranium in monazite of sample 7 as found by X-ray microanalyser. Photo J. Krištín; a) Composition. Magn. 1200×. b) Distribution of UL_{α}. Magn. 1200×. c) Distribution of SiK_{α}. Magn. 1200×.

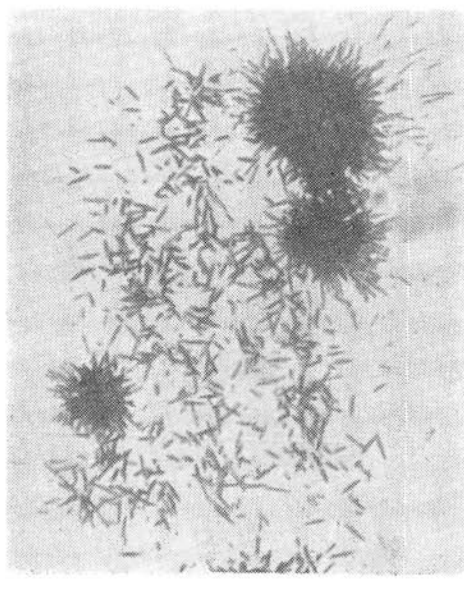


Fig. 4. Distribution of tracks after induced fission of ^{235}U in monazite. Markedly increased density of tracks is caused by inclusions richer in uranium than their surroundings. Photo J. Král.

Conclusion

Among accessory minerals studied for content and distribution of uranium, monazite shows the highest uranium content (2657 ppm). Distribution of uranium is homogenous. Present are coffinite inclusions, which may increase the total uranium content. Thorium content is higher. In allanite of granodiorite are lower uranium contents (107 ppm) than in allanite of biotite granite (480 ppm). Because of inhomogenous distribution of uranium in zircons the mean contents in single samples are not presented. Still they range within the mean values. The mean uranium content in apatites (34 ppm) is extremely low.

On the ground of the results quoted, in respect of uranium association, monazite is the most important accessory mineral. Allanite from granite shows a comparatively high uranium content. Higher thorium contents are presumed in both minerals. Uranium contents in zircons are mean and in respect of small amounts of zircon in rocks, the latter cannot be considered a significant mineral regarding the association of uranium. Also apatite is insignificant in this respect.

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