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DISTRIBUTION AND CONTENT OF URANIUM IN THE GRANITOID GLASSES

(Fig 1—5)

Abstract: Method of fission tracks has been used for measuring the uranium concentration in glasses of melted granitoid rocks of the West Carpathians crystalline. Concentration varies from 0,7 ± 0,1 ppm to 3,5 ± 0,2 ppm, with two values of 8,5 ppm and 17,2 ppm. Uranium distribution approaches to the normal distribution in majority of granitoid glasses. The portion of zircon on total activity of rocks has been examined.

Резюме: Методом следов по продуктам распада измерялась в стеклах плавленных гранитоидных пород западокарпатских кристаллических массивов концентрация урана. Концентрация этого элемента от 0,7 ± 0,1 ppm до 3,5 ± 0,2 ppm с двумя данными 8,5 ppm и 17,2 ppm. Дистрибуция урана в большинстве случаев приближается нормальному распределению. Непрямо наблюдалась доля циркония на общей активности пород.

Introduction

Method of fission tracks enables to determine the age of fission tracks (R. L. Fleischer — P. B. Price 1964), which can correspond to the "true age" of the examined geological subjects by investigation of the fission tracks (after spontaneous and induced fission of ^{238}U and ^{235}U) in dielectric mineral subjects (minerals and glasses) optically. One of the main contribution of this method is the possibility to determine low uranium concentrations in small amounts of mineral samples (P. B. Price — R. M. Walker 1963), or pulverised rocks (A. N. Komarov et al. 1969, F. D. Fisher 1970), and the possibility to examine optically the uranium distribution (in this case distribution of tracks after induced decay of ^{235}U) in the examined subject.

The distribution of uranium is described in this paper, as well as the uranium content in glasses, which arose by melting of the original granitoid rocks. The authors have attempted to correlate the content of some accessoric minerals (zircon, apatite) with uranium concentration in granitoid glasses.

Mineral and chemical composition of the investigated samples

Modal composition of the investigated samples (tab. 1) of the original rocks were computed by means of linear integration table. Minimal measured area was 8 cm^2, maximal area 12 cm^2. Total length of measured line depended on the grain size of the sample, and measured length varied from 800 to 1200 mm. Feldspars were determined by staining. Mineral composition of samples NT-1, NT-2 was assumed from the papers of J. Koutek (1930, 1931), samples MK-1 from the paper of B. Cambel and J. Valach (1956). Rock glasses were made from original material kindly given us by the above mentioned authors. Petrographic division and classification of the original rocks (fig. 1) was done in sense A. Streckeisen (1973). The index of

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Table 1

Modal composition of the original granitoid rocks

S. Nr.	Quartz	Plagioclase	K-feldspar	Biotite	Others
ZK-1	32,5	45	traces	21	1,5
ZK-2	35	33,5	25	6	0,5
ZK-3	30	34	22	11	3
ZK-4	—	—	—	—	—
ZK-20	25	55	6		14
ZK-24	27,5	48,5	11,5	9	3,5
ZK-25	36	33	18	11	2
ZK-26	30	34	25	4,0	7,0
ZK-28	22,5	52,5	7,0	13	5
ZK-29	31,5	39,5	13	4	12
ZK-30	30—31	44	14—15	11	0
ZK-33	45	15	25	2	13
ZK-42	36—37	30	25—26	—	8
NT-1	27	46	8	16	3
NT-2	5—6	40—42	1	45—48	6
MK-1	13	47	1	39	

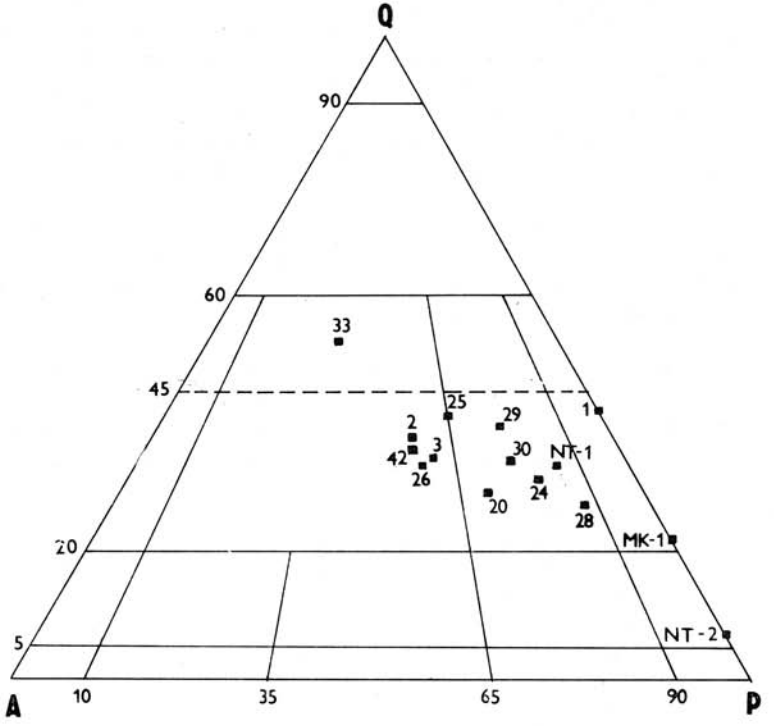


Fig. 1. Position of the studied rocks in A. Streckeisen's (1973) diagram.

refraction of the rock glass was measured by immersion method at the room temperature, reproduced accuracy of measuring was 0,005. Density of glass was calculated from chemical analysis on the basis of specific refractive energy of the individual oxydes. The results are summarized in table 2. Chemical analyses were assumed from the paper of L. V. Tauson et al. (1974).

Method of work

Granitoid glasses were obtained according to the procedure described in paper by M. Dyda and J. Macek (1973). A tablet was made from 100—200 g of homogenized rock under the pressure, and then it was melted by temperature of 1600 °C in dependence on the acidity of a rock during 4—12 hours. Presence of crystalline phase has not been found either optically or by X-ray diffraction.

Glass fragments (mean weight 0,2 g) were packed into the aluminium foil and put into reactor.[1] After irradiation by thermic neutrons, glasses were embedded in the synthetic bitumen and then ground and polished. After being perfectly polished, the preparate was etched in 38 % HF at proper temperature 20 °C during the time of 15—17 sec.

Density of fission tracks was counted in optical microscope by magnification of 550 times in reflected light. Tracks were counted on the 200 unit areas (a part of viewing field), randomly chosen. Minimally 626 and maximally 7 965 tracks were counted in dependence on density of fission tracks.

Table 2

Index of refraction (n) and density (h) of the granitoid glasses. Q is value of quartz amout from modal analysis calculated on 100, P — feldspar index

S. Nr	n	h	Q	P	Nomenclature according A. Streckeisen (1973)
ZK-1	1,527	2,53	41,5	100	tonalite
ZK-2	1,498	2,41	37,4	57,2	granite
ZK-3	1,506	2,40	34,7	60,8	granite
ZK-4	1,501	2,45	—	—	granite
ZK-20	1,521	2,44	29,0	90,0	granodiorite
ZK-24	1,503	2,42	31,4	80,9	granodiorite
ZK-25	1,504	2,42	41,3	65,0	granite
ZK-26	1,499	2,37	33,6	57,7	granite
ZK-28	1,518	2,55	27,2	87,5	granodiorite
ZK-29	1,498	2,38	39,6	75,5	leukogranodiorite
ZK-30	1,503	2,41	34,0	76,0	granodiorite
ZK-33	1,486	2,35	53,0	37,5	leukogranite
ZK-42	1,487	—	37,4	57,2	granite
NT-1	1,513	2,42	33,3	85,0	granodiorite
NT-2	1,573		12,0	100,0	quartz diorite
MK-1	1,555	2,53	21,4	100,0	tonalite

[1] Samples were irradiated in INR, Řež near Prag.

A formula of G. A. Wagner (1973) has been used for calculation of uranium concentration:

$$U_{g/g} = \frac{A.\ p_i}{g_r\ .\ \rho\ .\ I.\ L.\ \varphi\ .\ \sigma\ .\ f} \qquad (1)$$

where:

A — atom weight of investigated element
p_i — density of tracks per 1 cm^2
g_r — factor of registration area of detector. The value $g_r = 2$ has been used in calculation
ρ — density of investigated sample
I — isotopic ratio $^{235}U : {}^{238}U$ (used was $7{,}26 . 10^{-3}$)
L — Loschmidt's number
φ — integral dose of thermic neutrons ($1{,}97 . 10^{16}$ of neutrons per cm^2) which has been determined by the internal glass uranium standard and has been chosen in order to a resulting density of fission tracks provided comfortable and precise calculation, i. g. mean value x . 10^5 tracks per cm^2.
σ — fission cross section of ^{235}U for thermal neutrons (value used is $582 . 10^{-24}$ cm^2).
f — factor of output. It is most critical variable in equation (1). Changes of this variable are dependend on the kind of track detector, conditions of observation (sort of lightning, magnification used, and so on). Value of f can be obtained by two ways, either by calculation it from sample with known uranium content, or by measuring of R (mean length of fission tracks able for etching) and δ_c (critical angle, under which fission track is not registrated any more). In calculation, R — 8 μm, measured on the subject $\delta_c = 30°$ (R. L. Fleischer, P. B. Price 1964). Value of f is then substituted by expression $R/2 . \cos \delta_c$.

Shape and size of etched fission tracks. Shape of etched tracks in minerals is strongly influenced by structure of mineral and a kind of crystallographic area. As it can be seen from fig. 2, two observed sections of etched tracks on the surface of preparate are characteristic for granitoid glasses, and they are the result of a different position of canal-shaped fission track to observed surface — circular and elliptical shape.

Size of etched tracks is not equal in various granitoid glasses. The authors, measuring the diameter of tracks perpendicular to observed surface, found out that this parameter is dependent most expressively on SiO_2 content, the diameter of etched track is decreasing with increasing content of SiO_2.

Results of measurements and discussion

Obtained uranium concentration in granitoid glases (tab. 3) is lower than mean values reported by A. P. Vinogradov (1962) —$4{,}10^{-4}$ %, and by B. Mason (1971) — $3{,}7.10^{-4}$ % for mineralogically and chemically similar original rocks. The increased uranium content has been observed in glasses ZK-26 and ZK-3, where uranium content in the last sample has an anomalous value, because values of gamma-spectrometric measurements of J. Bartošek et al. (1972) are more than 10 times lower for similar rocks types and from similar localities. Investigation of melted quartz-feldspar fraction from the mentioned sample showed very low uranium content (approximate calculation above 1 ppm).

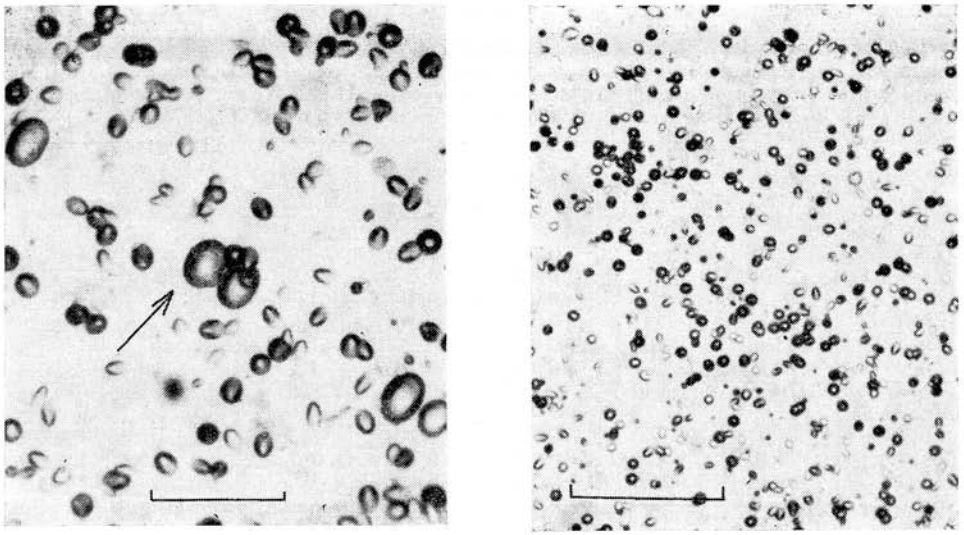


Fig. 2. Fission tracks in granitoid glasses. Tracks (pits) marked by arrow are of non-nuclear origin. Scale on fig. 2 a is 30 μm, on fig. 2 b 100 μm.

Relation of U to SiO_2 and K_2O content (data taken out from the paper by L. V. Tauson et al. (1974) is demonstrated on on fig. 3 a, b. A partial dependence of the uranium content on the above mentioned oxydes content in the investigated samples results from the calculated correlation coefficient r ($U/SiO_2 = 0{,}30$; $U/K_2O = 0{,}31$).

Considering that we have had thin-sections, chemical analyses and glasses from original material, we have attempted to express quantitatively to what degree uranium

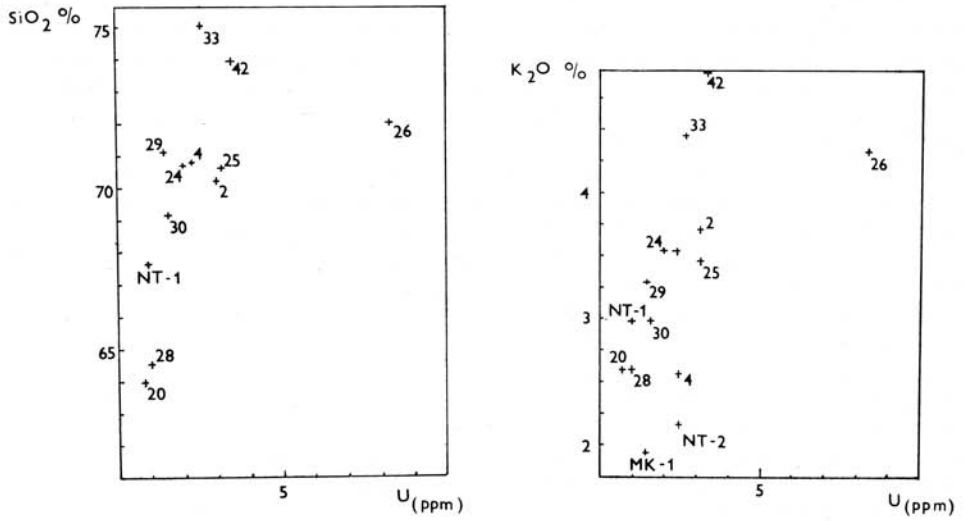


Fig. 3. Relation of U — concentration in granitoid glasses to weight percent: a — SiO_2, b — K_2O. Value of correlation coefficient (r) for $U/SiO_2 = 0{,}30$; for $U/K_2O = 0{,}31$.

Table 3

Weight percent of SiO_2 and K_2O, areal percent of accessoric minerals and uranium concentration. The error in calculation of uranium content is calculated from counting the fission tracks only. In all the glasses it varies between 6—11 % (Weight percent of SiO_2 and K_2O is taken from the paper by L. V. Tauson et al. (1974).)

No.	SiO_2	K_2O	Apatite	Zircon	ZR_p/ZR	U (ppm)
ZK-1	62,44	2,12	0,100	0,005	1,0	2,6 ± 0,2
ZK-2	71,08	3,70	0,027	0,006	2,0	3,1 ± 0,3
ZK-3	69,25	3,28	0,029	0,017	9,0	17,2
ZK-4	70,77	3,52	0,003	0,011	1,7	2,3 + 0,2
ZK-20	63,96	2,60	0,076	0,003	only ZR_p	0,7 ± 0,1
ZK-24	70,72	3,52	0,026	0,012	0,5	2,0 ± 0,2
ZK-25	70,58	3,46	0,045	0,004	6,6	3,1 ± 0,3
ZK-26	71,98	4,32	0,027	0,012	6,0	8,5 ± 0,9
ZK-28	64,53	2,60	0,119	0,016	0,3	1,1 ± 0,1
ZK-29	71,16	3,28	0,020	0,008	0,1	1,4 ± 0,1
ZK-30	69,14	2,56	0,021	0,007	0,95	1,5 ± 0,1
ZK-33	75,04	4,46	0,013	0,005	—	2,5 ± 0,1
ZK-42[1]	73,95	4,40	—	—	—	3,5 ± 0,2
NT-1[1]	67,71	2,97				2,3 ± 0,2
NT-2[1]	51,70	2,14				1,1 ± 0,1
MK-1[1]	54,00	1,87				1,3 ± 0,1

[1] In relation to lack of thin sectioned material, we have not calculated the areal % of accessoric minerals.

concentration is dependent on the content of some of accessory minerals in the investigated rocks. Zircon and apatite were correlated only, because they occured in the all samples. The areal % content of apatite and zircon can be found in tab. 3, values are obtained optically from thin-sections. In the same table, ratio of two groups of zircon — ZR_p and ZR is described. ZR_p includes such zircons, which form intensive pleochroic halos in biotite, ZR includes zircon forming inexpressive or any halos at all. Two groups of rocks were distinguished on the basis of mutual relation ZR_p/ZR:

1. This group consists of rocks with predominance of active zircons, and mutual ratio ZR_p/ZR is greater than 1.

2. Zircons with low activity prevail in this group, ZR_p/ZR is lower than 1.

Supposing that qualitative differences between pleochroic halos of zircons in biotite reflect the quantitative differences of uranium content in zircons, we can recognize two groups of rocks in the studied set of samples (fig. 4), in what the primary uranium content has not been the same.

Any dependence has not been found between the areal % content of apatite in thin-sections and uranium content in glasses.

Distribution of uranium in granitoid glasses. Graphically obtained parameter of the standard deviation s and mean w from the probability diagram (fig. 5 a, b, c) are in a good accordance with the calculated values (tab. 4) in most cases, which means that distribution of uranium approximates to the normal distribution in most cases. However, in many cases there are mixed populations, often bimodal or polymodal, which is demonstrated on the probability diagram by changing the dip of line, or by shifting of a part of line in certain distance according to what degree population has similar parameters.

Table 4

Calculated parameters: s^2 — dispersion, (s) — standard deviation, w — mean; and parameters obtained graphically from the probability diagram — s_g and w_g. C — variation coefficient

No.	distribution	s^2	Scale	s_g	Weale	w_g	C
ZK-1	polymodal	0,835	0,914	0,9	2,4	2,5	0,381
ZK-2	bimodal	0,242	0,492	0,6	3,0	2,6	0,164
ZK-3	heterogeneous				17,2		
ZK-4	biomodal	0,767	0,876	0,8	2,3	2,0	0,381
ZK-20	unimodal	0,097	0,311	0,3	0,7	0,7	0,444
ZK-24	polymodal	1,416	1,190	0,9	2,0	2,1	0,598
ZK-25	heterogeneous	1,538	1,240	1,0	3,20	2,5	0,388
ZK-26	heterogeneous				8,5		
ZK-28	biomodal	0,280	0,529	0,5	1,0	0,8	0,529
ZK-29	unimodal	0,439	0,663	0,6	1,5	1,4	0,442
ZK-30	polymodal	0,578	0,760	0,7	1,5	1,2	0,507
ZK-33	biomodal	0,384	0,620	0,9	2,5	2,4	0,248
ZK-42	unimodal	1,020	1,01	1,1	3,4	3,4	0,297
NT-1	unimodal	0,411	0,641	0,7	2,4	2,3	0,267
NT-2	unimodal	0,116	0,340	0,2	0,9	0,9	0,378
MK-1	biomodal	0,073	0,270	0,3	1,3	1,3	0,208

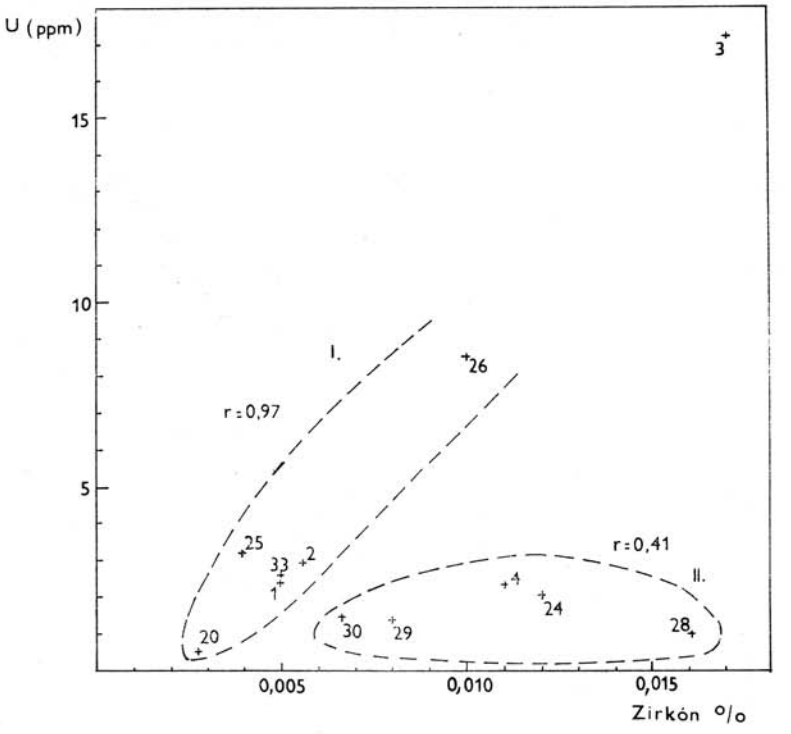
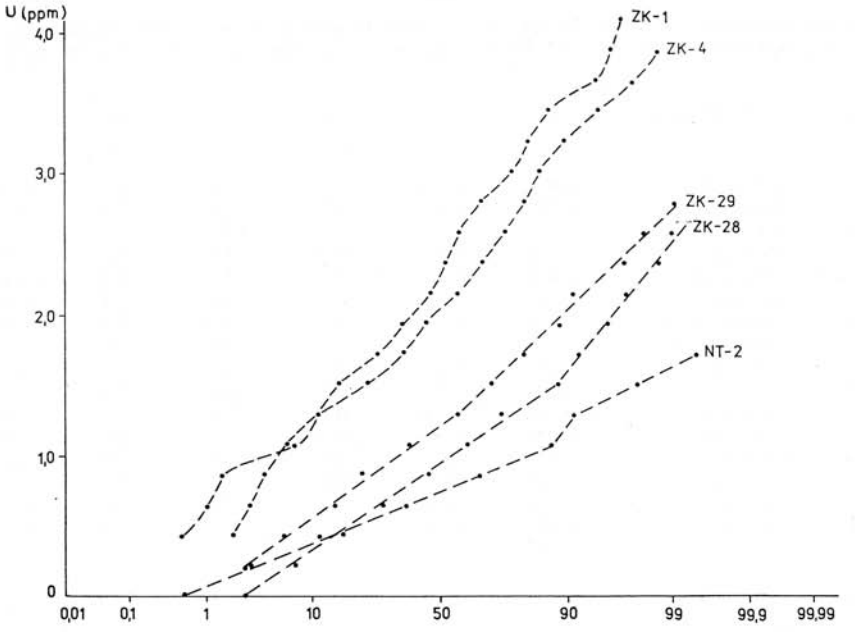
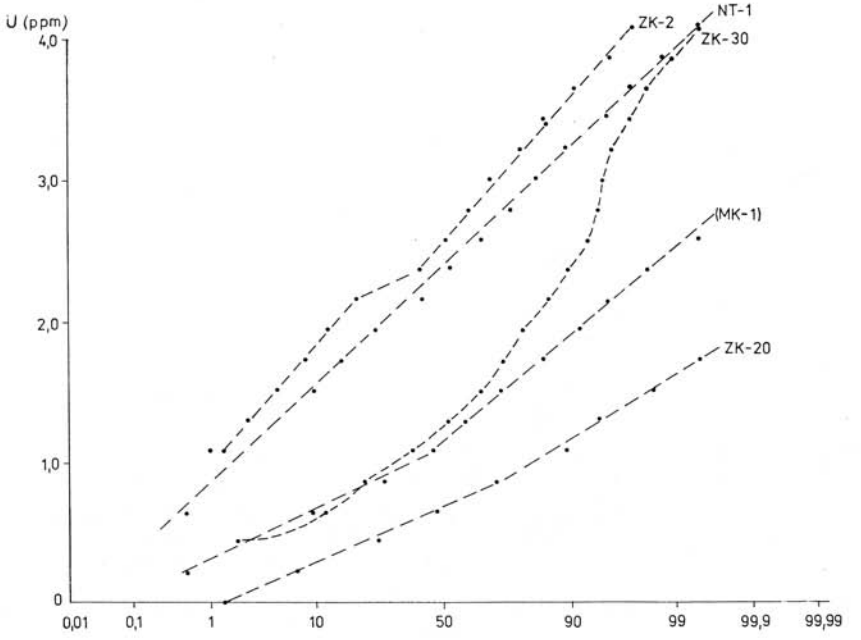


Fig. 4. Dependence of uranium concentration on areal percent of zircon in the investigated samples.


U (ppm)
4,0
3,0
2,0
1,0
0
0,01
0,1
1
10
50
90
99
99,9
99,99
ZK-1
ZK-4
ZK-29
ZK-28
NT-2


U (ppm)
4,0
3,0
2,0
1,0
0
0,01
0,1
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50
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99
99,9
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ZK-2
NT-1
ZK-30
(MK-1)
ZK-20

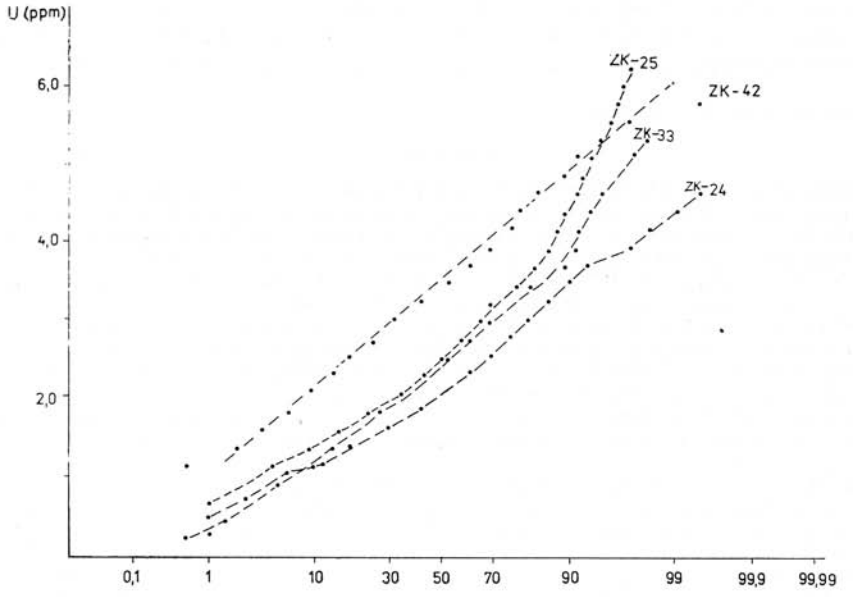

Fig. 5. Relative cumulative frequency curve of uranium content plotted on probability paper. (There ought to be the letter U instead of V on the y-axis).

Homogeneity of uranium distribution, expressed by value of dispersion (s^2), varies significantly. The highest dispersion value is in sample ZK-25, what appeares on diagram as a curve. The large heterogeneity of uranium distribution gives an evidence about complicated circumstances during the melting. It can be expected that resulting homogeneity of uranium content in glasses is dependent mostly on chemical composition of glass (SiO_2, alkalies), on temperature and time of melting and on grain-size of a sample. The influence of each of above mentioned parameters on the glass homogeneity would be possible to find out by precise investigation of melting conditions.

Conclusions

The authors presented in this paper the possibilities of measuring the uranium concentration in granitoid glasses by means of method of fission tracks. It can be stated, that such an application is possible on principle supposing that a great attentions is devoted to optimal conditions by the melting of rocks.

A partial dependence in the investigated set of samples is observed for uranium concentration on the total amount of SiO_2 and K_2O in the original rock. Zircon content in a rock can be correlated with uranium concentration as well.

The observed set of samples could be possible to divide into two groups on the basis of various qualitative and quantitative displays of zircon in biotites (pleochronic halos) and on the obtained uranium content in glasses. These two groups differ in supposed uranium content in zircon.

To confirm fully this conclusion would be possible by measuring of uranium content in the individual zircon grains from granitoid rocks.

At the end, we should like to express thanks to Academician B. Cambel for providing the investigated material and to Dr. J. Burchart for providing the uranium standard.

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Review by I. REPČOK.

Appendix
Localization of samples

ZK-1 Tribeč, N slope, valley near gamekeeper's cottage in the end of the road. ZK-2 Veporic, Chyžné, Hladomorná valley, Kohút zone. ZK-3 Chopok, Nízke Tatry Mts., 30 m WW from the rope railway station. ZK-4 Nízke Tatry Mts., Kralička. ZK-20 Veľká Fatra Mts., quarry, road on Smrekovica Mt. ZK-24 Nízke Tatry Mts., new road Sopotnica — Hronov. ZK-25 Nízke Tatry Mts., new road Sopotnica — Hronov. ZK-26 Veporic, Hrončok, nocar the end of the asphalt road. ZK-28 Veporic, Tlstý Javor Mt., road Čierny Balog — Hriňová, qiarry near the road. ZK-29 Veporic, Dobroč, Páleničnô valley. ZK-30 Vysoké Tatry Mts., Štrbské pleso near the Hotel FIS. ZK-33 Gemeric, Hnilec, bore hole HG-1, depth 300 m. ZK-42 Tribeč, Jánova Ves. NT-1 Nízke Tatry Mts., Ďumbier. NT-2 Nízke Tatry Mts., Lúžna (see J. Koutek 1930). MK-1 Malé Karpaty Mts., Bratislava, Hlboká cesta street.