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## U—Pb AND K—Ar ISOTOPIC DATING OF SINEC (RIMAVICA) GRANITES (KOHÚT ZONE OF VEPORIDES)

(Figs. 3, Tabs. 2)



**Abstract:** New results of radiometric dating of Sinec (Rimavica) granites by U—Pb and K—Ar method are presented in the paper. The K—Ar method (muscovite and biotite) gives the age of 94 m. y., which can be interpreted by tectonics. The U—Pb dating of zircons gave a concordant age of  $350 \pm 5$  m. y., which is a more reliable age than the one obtained from the same samples by Rb—Sr isochrone (Cambel et al., 1988). The increased age of Sinec (Rimavica) granites determined by Sb—Sr method could be caused by a supply of radiogenic Sr from the surrounding older metamorphic rocks to the granites during retrograde metamorphism synchronous with granite autometasomatism.

**Резюме:** В статье приводятся результаты радиометрического датирования Синецких (РимаVICких) гранитов U—Pb и K—Ar методом. K—Ar метод (мусковит и биотит) дает возраст 94 млн. лет, который можно интерпретировать тектонически. U—Pb датирование цирконов дает конкордантный возраст  $350 \pm 5$  млн. лет, который является более надежным чем возраст полученный из тех же проб методом Rb—Sr изохроны (Cambel et al., 1988). Повышенный возраст Синецких (РимаVICких) гранитов определенным методом Rb—Sr может быть результатом приноса радиогенного Sr из вмещающих более древних метаморфических пород в граниты в течение ретроградного метаморфизма синхронного с аутометасоматозом гранитов.

### *Object of the investigations and geological problems*

The aim of the paper is a complex geochronological dating of Rimavica Granites penetrating the crystalline strata in the southeastern part of Veporides (Kohút Zone). Existing opinions on the age of granites and the surrounding metamorphic rocks are in the present time contradictory. Thus, the age estimation of the sedimentary-metamorphic basement penetrated by the granites varies according to palynological data from Lower Paleozoic to Upper Carboniferous (Rimava Formation). The time of granite intrusion is determined by various authors as Variscan to Alpine cycle. Assumptions about young, Alpine

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age of Sinec (Rimavica) granitoids are founded on the existing K-Ar dating on micas, giving an interval of 118—88 m. y. (Kantor, 1960, 1979 ex Vozárová — Vozár et al., 1979; Camel et al., 1979; Camel et al., 1980).

In regions with polymetamorphic history, to which belong also West Carpathians, there is always a dilemma: do K-Ar datings reflect the true age of crystallization of the mineral and the rock containing it, or is this an apparent age, reflecting a partial or complete loss of radiogenic argon. In this case the problem lies in the following: are the granitoids and the synchronous metamorphism Alpine and the surrounding strata, even though only partially. Upper Paleozoic, or are all the rocks products of Variscan orogeny but they underwent intensive radiologic rejuvenation (loss of Ar) during the following Alpine tectonogenesis? (Král, 1977).

For the purpose of solving this problem, 4 samples have been collected from the Sinec (Rimavica) Granites for a simultaneous study by three methods: U-Pb isotopic on zircons, Rb-Sr isochronic on whole rock and K-Ar method on micas. All samples have been collected on the area between Mútnik and Krokava, from peripheral parts of the Rimavica Granite Massif, in the zone of injection or intrusive contact of the granites with garnet-mica schists and paragneisses of Hladomorná Valley Formation. All typical granite varieties have been sampled — from biotite granodiorite to muscovite-rich leucogranites. Characteristic features of the collected samples as well as of the Rimavica Granitoids generally are:

1. Predominance of plagioclase over potassium feldspar;
2. Permanent presence of small garnet grains;
3. Varying, but often high muscovite contents;
4. Strong metasomatic effects, with the substitution of biotite and plagioclase by large-flaked muscovite, of plagioclase by albite with clinozoisite and sericite inclusions, of biotite by chlorite and sericite; this is usually accompanied by a number of hydrothermal veinlets of quartz, quartz-muscovite, quartz-albite composition.

The results of Rb-Sr isochronic age determination of the granites are presented in the paper Camel et al. (1988). In this paper only data from the U-Pb and K-Ar method and a general interpretation of the results obtained by all the three methods from same samples are presented.

#### *Sample localisation, their geological position and petrographical description*

Sample KV-1 — on the right side of a creek, on a footpath, 1.3 km SE of Mútnik. The sample was collected 10 m from the contact with garnet-mica schists of Hladomorná Valley Formation.

The rock is medium-grained, biotite  $\pm$  garnet-muscovite-plagioclase  $\pm$  quartz granodiorite. Secondary processes: substitution Bi  $\rightarrow$  Mu, Pl  $\rightarrow$  Ab + Mu + Clz, Bi  $\rightarrow$  Chl\*. The intensity of muscovitization is medium.

Samples KV-2, 3, 4 — all are collected from a small outcrop at the road Krokava-Hnúšťa, 300 m downwards by the road from the Pioneer camp "Krokava". In the outcrop there are biotite-muscovite granites and granodiorites (samples KV-3 and 4), with a small vein of muscovite leucogranite (sample

\* Symbols of minerals: Ab — albite, Bi — biotite, Cal — calcite, Chl — chlorite, Clz — clinozoisite, Mu — muscovite, Pl — plagioclase.

KV-2). Next to them are garnet-biotite gneisses of Hladomorná Valley Formation with injections of the same granites.

KV-2 — is medium-grained muscovite-garnet-plagioclase-K-feldspar-quartz leucogranite. It practically does not contain biotite (substituted by muscovite?). Extremely intensive autometasomatism with complete loss of Ca from plagioclase:  $Pl \rightarrow Mu + Ab$ ; interveined by muscovite veinlets.

KV-3 — medium-grained biotite  $\pm$  garnet-muscovite-plagioclase  $\pm$  K-feldspar-quartz granite. The rock is inhomogeneous, with mafic sections of diorite-like composition. Autometasomatism of medium intensity:  $Pl \rightarrow Ab + Mu + Clz$ .

KV-4 — coarse-grained biotite  $\pm$  garnet-muscovite-plagioclase-quartz granodiorite. Very weak autometasomatism:  $Pl \rightarrow Ab + Mu + Clz$ ;  $Pl \rightarrow Cal + Mu + Ab$ .

### *Results of K-Ar method*

Biotites and muscovites were separated from the samples; the results of age determination are presented in Tab. 1.

The coincidence of the determinations is good, so that it is possible to assume the existence of a real geological event in the studied region, with a minimal age of 93–94 m.y.. This event corresponds to the Alpine cycle of tectonogenesis, and according to the age scale to Lower Cretaceous. Our data agree very well with the data of Kantor, 1979 ex Vozárová—Vozár et al. (1979), Cambel et al. (1980) and Bagdasaryan et al. (1977) obtained for the same region by K-Ar dating (118–88 m.y.), as well as with the data of J. Burchart, B. Cambel and J. Král (Burchart et al., 1987), who, on the basis of an evaluation of all available K-Ar datings of West Carpathians, presented a K-Ar isochrone reflecting a tectonic or thermal event in Veporides with an age of  $94 \pm 18$  m.y..

### *Results of U-Pb dating of zircons*

#### Characterization of zircons

Zircons from the studied samples of Rimavica Granites mostly form idiomorphic pencil-shaped crystals and only rarely isometric grains. Up to 20 % of the zircons are transparent, pink, with diamond lustre. By a similar quantity are represented the relatively youngest metamict zircon forms, which are dark-brown and have lower lustre. A majority of zircons is slightly metamict, slightly clouded and of light-brown colour (coffee-colour).

Relatively more zircons contain biotite — inclusions in the Rimavica Granites from Kokava region. No inclusions of U-minerals, not even in metamict zircons, have been discovered on sections of selected zircon representatives by reflected electrons of SEM. Zircon sections on SEM made possible — besides

the study of inclusions — also the investigation of the grade of zircon zoning caused by variations in Hf contents in the structure of zircon. Older zircon forms determined in the sense of Pupin (1980) as mostly  $S_{12}$  zircon types have a more intensive oscillation structure suggesting a longer-termed zircon growth at relatively higher temperatures. Lower-temperature zircon forms — according to Pupin (l.c.)  $L_5$  and  $G_1$  types — are slightly zonal, frequently long-prismatic, suggesting their more rapid growth in a narrower interval of low temperatures.

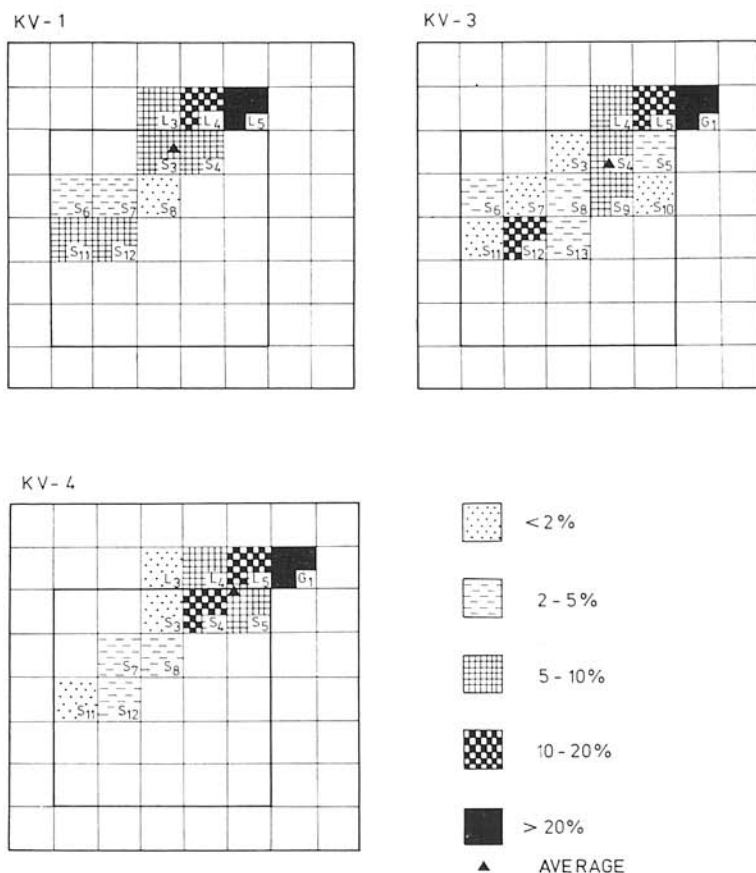


Fig. 1. Distribution of zircon types in Sinec (Rimavica) Granitoids (according to the classification of Pupin — Turco, 1972; Pupin, 1980).

A maximum amount of zircons in the samples of Rimavica Granites is according to the typological classification of Pupin (l.c.) represented by the types  $L_5$  and  $G_1$ . Only in the sample KV-3 it is possible to establish another maximum in zircon numbers in the area of the type  $S_{12}$  (Fig. 1). The Trend of Evolution Typology (T.E.T.) in all samples begins with relatively higher-

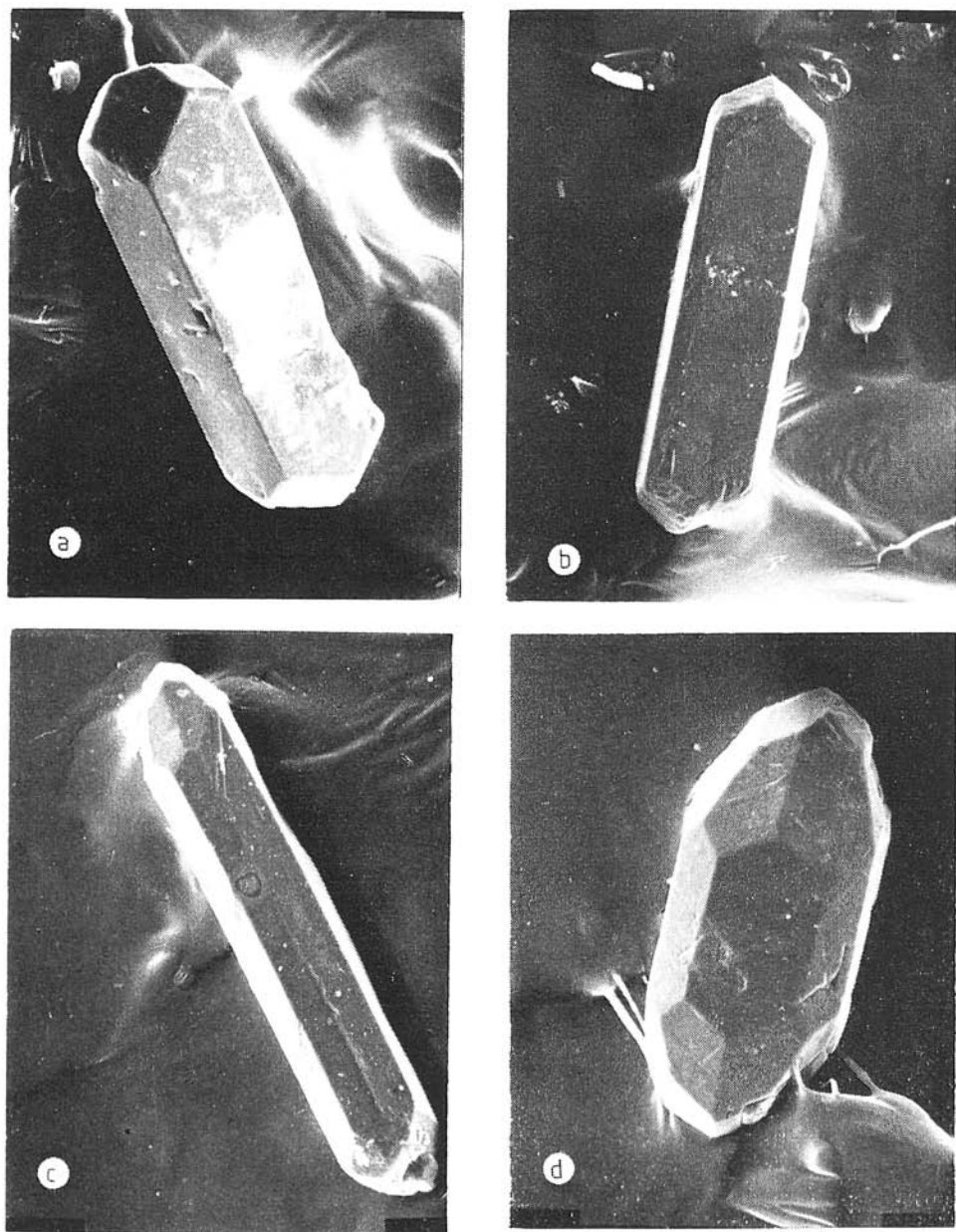


Fig. 2. In the sample KV-3, maximum of zircons is developed as types  $G_1$  or  $G_1$  and  $L_5$ . A second less marked maximums is represented by the type  $S_{12}$ . Increased content of  $S_{12}$  types in the sample KV-3 is probably caused by the presence of more basic inhomogeneities in the granitoid.

*Explanations:* a — type  $G_1$ , magnif. 400x; b — type  $L_5$ , magnif. 280x; c — type  $S_{12}$ , magnif. 270x; d — type  $S_{12}$ , magnif. 240x (Photo I. Holický).

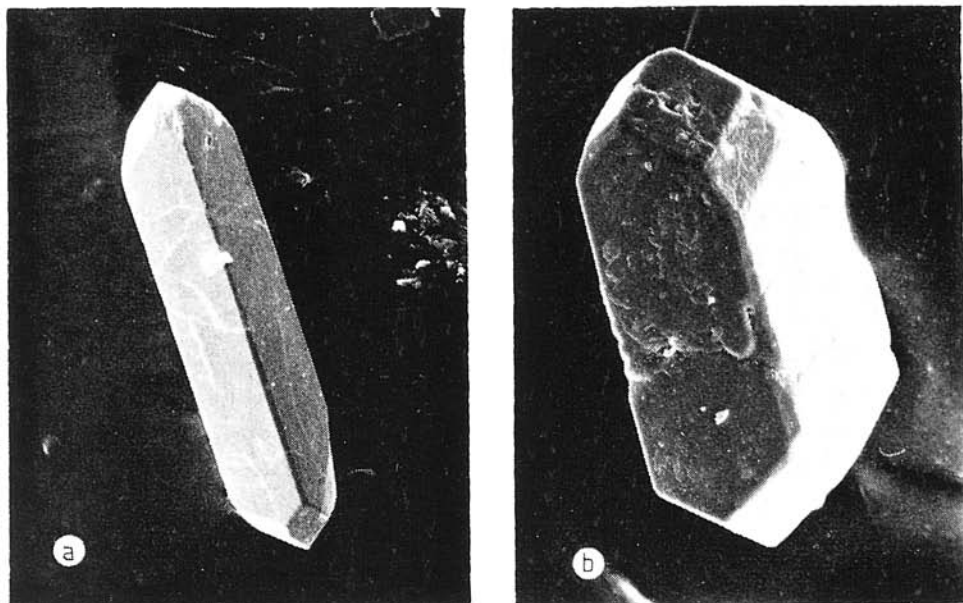


Fig. 3. Principal zircon types in the sample KV-4 represented mostly by the types  $G_1$ , or  $G_1$  and  $L_3$ .

*Explanations:* a — type  $L_3$ , magnif. 200x; b — type  $G_1$ , magnif. 550x (Photo I. Holický).

-temperature zircons ( $750^\circ \pm 50^\circ\text{C}$ ) and culminates with forming of zircons of the types  $L_3$  and  $G_1$  at temperatures lower by approx.  $150^\circ\text{C}$ . The higher content of  $S_{12}$ -type zircons in the sample KV-3 can most probably be connected with macroscopical inhomogeneities of a more basic character in this sample.

In the separation process of zircons for U—Pb dating, their morphological features and visually determined metamictization grade were taken into account. The samples KV-3 and KV-4 were separated into three fractions, in which zircons of the same metamictization grade or of a selected morphological feature were merged.

In the sample KV-1, insufficient quantity of zircons rendered U—Pb dating impossible; in the sample KV-2, zircons have been found only as separate grains.

Zircons of the sample KV-3 have been divided into:

"S types" — pink, transparent zircons, with equal number of (100) and (110) surfaces; they are relatively the oldest, forming at higher temperatures ( $750^\circ \pm 50^\circ\text{C}$ ).

"L—G types" — pink zircons, transparent, from the end of the evolution typological trend; with predominant surfaces (100) and (101).

"M types" — metamict zircons, completely opaque, relatively the youngest ones.

In the sample KV-4, zircons have been divided according to their metamictization grade:

Table 1  
Values of K-Ar age of micas

Sample No.	Mineral	Contents			Age m.y.
		Potassium $^{40}\text{K}$	Radiogen. argon ng g	Ar $^{40}$ rad. $^{40}\text{K}$ total	
KV-1	muscovite	$8.61 \pm 0.08$	$57.9 \pm 1.2$	42; 55	$94 \pm 4$
KV-2	muscovite	$8.65 \pm 0.03$	$70.5 \pm 1.5$	43; 48	$114 \pm 4$
KV-3	biotite	$7.28 \pm 0.07$	$48.7 \pm 1.4$	33; 39	$94 \pm 5$
KV-4	biotite	$7.02 \pm 0.07$	$46.7 \pm 1.0$	32; 46	$93 \pm 4$

The calculation of age was made with the use of unified constants accepted on the Geological Congress in Australia:  $\lambda_K = 0.581 \cdot 10^{-10} \text{ year}^{-1}$ ,  $\lambda_\beta = 4.962 \cdot 10^{-10} \text{ year}^{-1}$ ,  $^{40}\text{K} = 0.01167 \text{ at. } ^{40}\text{K}$ .

"L—G types" — non-metamict zircons, transparent, pink, with diamond lustre.  
 "1/2 M types" — semi-metamict zircons, light-brown and slightly clouded.  
 "M types" — metamict zircons of brown colour, completely opaque.

The zircons separated according to their morphological characteristics and metamictization grade have been treated by U—Pb isotopic analysis.

#### Method of analysis

The method of U—Pb isotopic analysis used in the laboratories of isotopic geochemistry of GEOCHI, Academy of Sciences of the U.S.S.R., is completely after Krogh (1973). The work was carried out in a clean geochemical laboratory, with filtered air and overpressure, securing a level of pollution of 0.5 ng of lead for zircon analyses.

A charge of 5–10 mg of unpulverised zircon (to exclude a possible pollution by pulverization), is decomposed hydrothermally in a teflon capsule in concentrated HF, at a temperature of 200 °C. After decomposition the charge is transferred into 1N HBr and divided into two aliquot parts. From one part, after addition of a combined ( $^{208}\text{Pb} + ^{235}\text{U}$ ) tracer, the determination of uranium and lead contents is carried out by the method of isotopic dilution, with a precision of  $\pm 1\%$ . The second part is used for the determination of the isotopic composition of lead. Uranium and lead are separated for the isotopic analysis by chromatography on resin — BioRad 1 X 8. The isotopic composition of lead was measured on the mass spectrometer TSN—20GA, CAMECA, with the use of silicagel as ion-emitter. The precision of the measurement is  $\pm 0.15\%$ . The results of U—Pb dating of zircons are presented in Tab. 2.

#### Evaluation of the results of U—Pb isotopic study of zircons

The most interesting one for the study has been found to be the zircon of the sample KV-3, represented by three successively crystallizing varieties. In Tab. 2,



Table 2  
U-Pb isotopic study of accessory zircons from granites of Rimavica massif

Sample No. fraction	Contents, %		Isotopic composition of lead					Age acc. to $^{206}\text{Pb}/^{238}\text{U}$ m.y.	Isotope relations (radiogenic)		
	Pb	U	204	206	207	208	$^{207}\text{Pb}/^{235}\text{U}$		$^{207}\text{Pb}/^{206}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	
KV-3	0.0036	0.060	0.034	81.218	4.880	13.868	352	0.4180	0.05398	0.150	
KV-3	0.0050	0.087	0.023	83.453	4.930	11.593	352	0.4255	0.05501	0.129	
KV-3	0.0073	0.128	0.028	83.645	5.303	11.024	347	0.4472	0.05855	0.130	
KV-4	0.0050	0.085	0.054	82.895	5.414	11.637	349	0.4270	0.05570	0.116	
KV-4	0.0064	0.117	0.055	82.895	5.414	11.657	330	0.4028	0.05566	0.116	
KV-4	0.0071	0.143	0.033	84.003	5.494	10.464	304	0.3909	0.05876	0.103	

The following values of decay constant have been used:  $\lambda^{238}\text{U} = 1.55125 \cdot 10^{-10} \text{ year}^{-1}$ ;  $\lambda^{235}\text{U} = 9.8485 \cdot 10^{-10} \text{ year}^{-1}$ ;  $^{238}\text{U}/^{235}\text{U} = 137.88$  (Steiger-Jäger, 1977). Correction was made to common lead of 350 m. y.:  $^{206}\text{Pb}/^{206}\text{Pb} - 18.230$ ;  $^{207}\text{Pb}/^{206}\text{Pb} - 15.639$ .



a double increase of uranium content, from early to late zircon varieties, can be observed. Notwithstanding, the occurrence of rock-forming mineral inclusions, the content of common lead in zircons is low, so that the calculated isotopic relations are not very sensitive to the selected isotopic composition of admixing lead.

Since the isotope  $^{206}\text{Pb}$ , the product of  $^{238}\text{U}$  fission, strongly predominated in the lead of Phanerozoic zircons, especially the isotopic relation  $^{206}\text{Pb}/^{238}\text{U}$  is most reliable for dating of these zircons. The isotopic age according to the relation  $^{206}\text{Pb}/^{238}\text{U}$  in all three zircon types of the sample KV-3 corresponds to the limits of experimental error and is equal to  $350 \pm 5$  m.y.

A little different age of the metamict variety — 347 m.y. — could to a various extent be the proof of either a small loss of radiogenic lead from the metamict zircon structure, or of the time of crystallization and the following autometasomatism in not more than 5 m.y.. However, to make a definite conclusion with such a limited number of analyses is not possible.

In the sample KV-4, semi-metamict and metamict zircon with much higher uranium content was significantly predominant. Metamictization of zircon structure connected with increased uranium concentration caused a later loss of a part of accumulated radiogenic lead is reflected in lower "apparent" age values. At the same time, non-metamict zircon represented basically by the type L—G, is characteristic by the same age —  $349 \pm 4$  m.y. as the zircon in the sample KV-3, which is one more proof of their equal age.

When comparing the values of the isotopic ratio  $^{207}\text{Pb}/^{206}\text{Pb}$  in the studied zircons (Tab. 1) with the theoretical value for the age 350 m.y., i.e. 0.05350, it is evident that almost in all zircons this relation is noticeably higher than the theoretical value. The accepted explanation for this phenomenon mentioned by many authors in Phanerozoic zircons, is an insignificant admixture of an earlier radiogenic component. It is most marked in late metamict zircons. Only early-magmatic zircons in the sample KV-3 have concordant age values for all isotopic relations.

Thus, according to the results of our investigations, the time of crystallization of accessory zircons in Sinec (Rimavica) Granites is  $350 \pm 5$  m.y.. Taking into consideration the typical magmatic form of these zircons, the obtained age value should be also related to the age of crystallization of the granite melt itself.

#### *General interpretation of the age values of Sinec (Rimavica) Granites according to three radiological methods*

The results of Rb—Sr isochrone dating of Rimavica Granites in whole rocks (samples KV-1, 2, 3 and 4) are presented in the paper Campbell et al. (1988). The value of isochrone age is  $394 \pm 6$  m.y. and it differs noticeably from the age determined on the same samples by U—Pb isotopic system of accessory zircons, i.e.  $350 \pm 5$  m.y.. Since both investigations have been carried out on a precise level of method, the correctness of the analytical data is above suspicion. Therefore, the difference of age values requires an explanation.

If we survey the Rb—Sr isochrone diagram (Campbell et al., 1988, Fig. 6), it is evident that the inclination of the isochrone as well as the low value of error ( $\pm 6$  m.y.) are completely determined by the position of the point of the sample KV-2 with maximum values of the relations  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{87}\text{Sr}$ .

From the petrographical description it follows that granites, particularly in this sample, underwent a maximal grade of autometasomatic acid leaching — muscovitization and albitization (Korzhinsky, 1955), with an especially strong migration of Ca and alkalis. The migration of Ca (geochemically related to strontium) is directly suggested by the presence of calcite in pseudomorphs after plagioclase in the granite sample KV-4 (see petrographical description of the samples).

The study of various types of granitoids and metamorphic suites of West Carpathians (Korikovskiy—Putiš, 1986; Cambel—Korikovskiy, 1986) as well as of Rimavica Granites has shown that autometasomatic retrograde muscovitization of the granites is frequently accompanied by intensive retrograde muscovitization of the surrounding garnet-mica schists, possibly pointing out to the synchronism of retrograde metamorphism and autometasomatism in granites and to a common source of fluids. In such a process, unilateral effect of granite fluids on the surrounding rocks is possible as well as an exchange of substance with the surrounding acid schists and gneisses — part an exchange of radiogenic Sr. Since the surrounding rocks are older than the crosscutting granites and this they should be at least 350 m.y. old, it is not possible to exclude that older strontium enriched in the isotope  $^{87}\text{Sr}$  goes to apical parts of granites. This is most probably the case in the sample KV-2 (and partly in the sample KV-1) which differ by a maximum grade of acid leaching.

Thus, if this assumption should be true, the obtained Rb—Sr age values appear to be somewhat higher in comparison with the age determined on magmatic zircons by the U—Pb method, i.e.  $350 \pm 5$  m.y..

At the same time, it is evident that the great difference between K—Ar datings of micas from Sinec (Rimavica) Granites and the determinations by U—Pb and Rb—Sr method clearly proves that the interval 93—114 m.y. does not correspond to the true age of micas and of the granites. The K—Ar determination fixed only an intensive argon isotope rejuvenation with a probable age 90—94 m.y.. The total sum of isotopical-geochronological and petrographical data proves that this was most probably only a purely thermal effect leading to a complete loss of radiogenic Ar, but without a new flows of diaphthoritic fluids and consequently without mineral alteration of the granite matter in the Alpine cycle.

This is above all supported by the fact that regardless of a substantial disturbance of the K—Ar system of micas, the U—Pb system of zircons as well as the Rb—Sr system of whole rocks (the most dependent ones on a repeated fluid effect) were completely unaffected in the Alpine age. In petrographical study of the rocks, no signs of coarse cataclasis or mylonitization, characteristic for Alpine dislocations in the crystalline suites (Cambel—Korikovskiy, 1986) have been found either. All secondary processes in granites — muscovitization of biotite and feldspars, deoxidation of plagioclases — have the character of typical autometasomatic leaching in the stage of granite cooling. Judging from the stability of the U—Pb age values, the time span of granite crystallization, including the stage of autometasomatism, hardly exceeded the interval of several million years, which coincides with the values of the error of measurement.

The effect of strong isotopic rejuvenation of Variscan rocks in the stage of Alpine tectonogenesis, partly or completely distorting only the K—Ar age va-

lues of minerals, is well known in West Carpathians, and proved on many objects in Veporicum, with numerous mylonitized zones, scales, overthrust faults etc. (Kantor, 1960; Burchart et al., 1987). The obtained results only corroborate this fact on the example of Kohút Zone of Veporides.

Thus, the results of a general geochronological study of Sinec (Rimavica) granites show that the most reliable age of their intrusion and crystallization is  $350 \pm 5$  m.y. — i.e. the age obtained according to U—Pb isotopic system of magmatic accessory zircons in samples unaffected by intensive autometamorphic processes.

Translated by K. Janáková

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