

TYOLOGY, ZONING AND CHEMISTRY OF ZIRCON FROM MAIN TYPES OF GRANITIC AND RHYOLITIC PEBBLES IN CONGLOMERATES OF THE PIENINY KLIPPEN BELT CRETACEOUS FLYSCH (WESTERN SLOVAK SEGMENT, WESTERN CARPATHIANS)

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Abstract: The pebbles of granitic and rhyolitic rocks from conglomerates of the Pieniny Klippen Belt Cretaceous flysch can be divided on the basis of zircon study (typology, zoning of crystals, chemical composition) into three main groups: 1 - granites, granite porphyries and rhyolites of the exotic Upohlav type with the tendency to A-type granite suite probably analogical to some post-orogenic (Permian to Triassic) intrusions of Alpine-Mediterranean Belt; 2 - granites, granodiorites to leucotonalites and rarely pegmatites of the Krivá type similar to some orogenic Hercynian granitic rocks from the Central Western Carpathians (Tátric and Veporic Units), and 3 - porphyric granite of the Lubina type which can be compared with orogenic (Hercynian ?) calc-alkalic intrusion.

Key words: Pieniny Klippen Belt, Cretaceous flysch conglomerates, granitic rocks, rhyolites, pebbles, zircon.

Introduction

Polymict conglomerates of various, often exotic rocks occurring in flysch turbidites of Middle to Upper Cretaceous (to Paleogene in eastern Slovakia and western Ukraine) age in the Pieniny Klippen Belt are a subject of longlasting discussion and belong to intensively studied problems of the Carpathian geology. In addition to the dominating assemblage of various types of sedimentary rocks, mainly carbonates (Mišík & Sýkora 1981; Mišík et al. 1991b) and less common metamorphic rocks (metaquartzites, orthogneisses), also various types of basic rocks can be found along with their HP-LT metamorphosed equivalents (blueschists) and acid rocks: granitoids, granite porphyries and rhyolites. The most peculiar among the above-mentioned magmatic rocks are exotic granites and porphyries which exhibit petrographical and geochemical properties not corresponding to any known occurrence in the Western Carpathians (Zoubek 1931; Kahan 1965; Krivý 1969; Kamenický et al. 1974; Šímová 1985). On the other hand, a part of the pebbles resemble the orogenic Hercynian granitic rocks from the Tátric and Veporic Unit of the Central Western Carpathians (Tátra granite sensu Zoubek l.c., granite I - Šímová l.c.).

The question of types, provenance and age of the granitic and rhyolitic pebbles in flysch sequences of the Pieniny Klippen Belt (PKB) is still unresolved, even though it seems to be of utmost importance for the understanding of paleogeographical and geotectonic development in the area between Outer and Central Western Carpathians. Therefore, we have tried to contribute to the solution by means of a some geochemical and mineralogical investigation of these acid magmatic rocks. We present the results of study of a most informative accessory mineral - zircon.

Geological settings

The Pieniny Klippen Belt belongs to the fundamental tectonical elements both the Western and Eastern Carpathians. This extremely complicated and variegated geological structure between Carpathians centralides and externides runs along the arc of orogenic belt with approximately 800 km length and its width reaches only several hundred meters to a few km (maximum approximately 15 km). The PKB consists of various Mesozoic to Paleogene sedimentary sequences with rare manifestations of synsedimentary volcanism (Mišík et al. 1991a). The most typical property of the belt is its intensive shortening and tectonical reworking into the typical klippen style. Middle to Upper Cretaceous flysch with occurrences of exotic conglomerates belongs to dominant facies of the western Slovak segment of the PKB. Conglomerate bodies of the maximum extent are found in two flysch cycles of the Klape Unit (Albian to Cenomanian and Upper Cenomanian to Santonian) and one cycle of the Pieniny Unit s.s. (Upper Cenomanian to Santonian). The conglomerate occurrences are of various thickness (0.X - 65 m) forming younger phases of flysch cycles which belong to the upper parts of sub-marine alluvial cones, i.e. proximal wild flysch facies at the basin flank (Marschalko 1986).

The conglomerates consist of relatively well-rounded pebbles of a few cm size up to 3.5 m large blocks, resedimented probably from submarine delta fans and transported by long rivers from a large source area (Mišík & Sýkora 1981).

Pebbles of granitic and rhyolitic rocks

Granitic rocks and their porphyric varieties amount about 4 % of pebbles, in several localities even more (10 - 18 %; Nosice, Vrtižer, Zemianska Dedina - Marschalko l.c.). Two basic

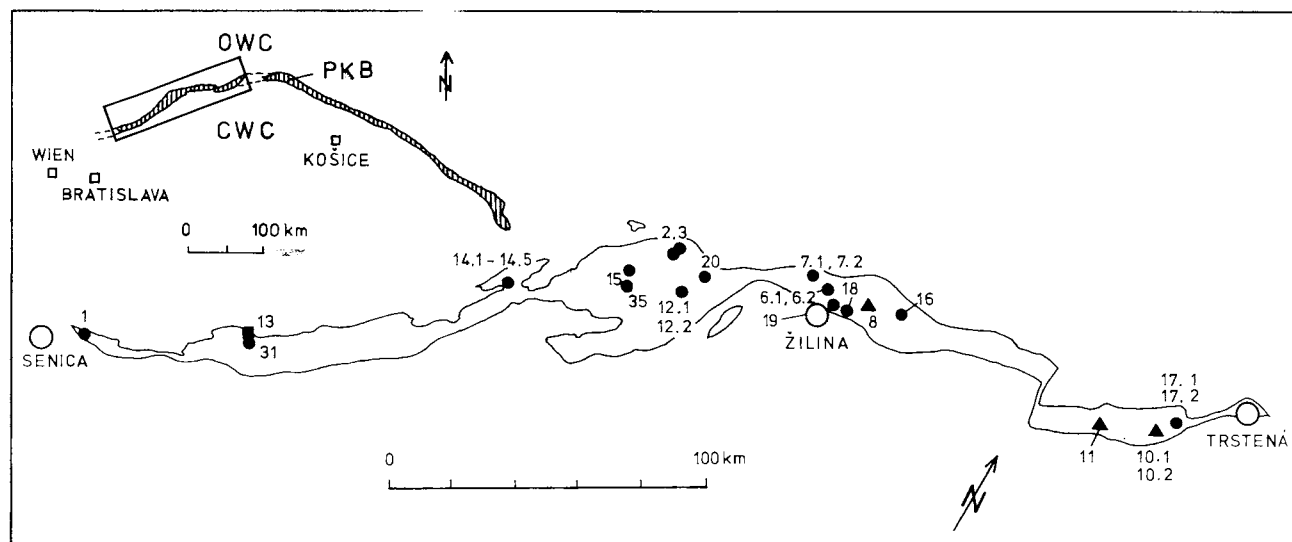


Fig. 1. Geological sketch of the position of western Slovak segment of the Pieniny Klippen Belt. Studies samples are marked out.

Explanation: PKB - Pieniny Klippen Belt; CWC - Central Western Carpathians; OWC - Outer Western Carpathians; circles - granites, porphyries, rhyolites of the Upohlav type; triangles - granitoids of the Krivá type; square - granite of the Lubina type. Numbers of localities refer to Tab. 1.

types can be distinguished already in the field: the exotic and Tatra granite (Zoubek 1931). The division is fully confirmed by their petrographical, mineralogical and geochemical properties, which were studied by various authors (Zoubek l.c.; Šířmová 1985, etc.). They are also the subject of our further study (Uher et al., in prep.). One, or both granitoid types can be found at particular occurrence in variable ratios.

Acid volcanic rocks - rhyolites are also relatively frequent. In addition to above types we have also found another differing granite type near village of Lubina (settlement Lukáč). Therefore, in the following text, we distinguish three basic types of acid magmatites occurring in the western Slovak segment of the Pieniny Klippen Belt (Fig. 1, Tab. 1.):

1 - The Upohlav type include exotic granites, granitic porphyries and rhyolites. The granites are leucocratic rocks with pink, often porphyric K-feldspar dominating usually over strongly sericitised oligoclase, and probably younger, fresh albite. Green to green-brown biotite occurs as fine-grained xenomorphic aggregates, rarely as subhedral crystals. The average modal composition of the Upohlav granitic rocks contains ($n = 13$): quartz 34.4 ± 6.7 , K-feldspar 36.9 ± 13.2 , plagioclase 23.3 ± 11.9 , biotite 4.8 ± 2.7 (in vol. %). The granitic porphyries show micrographic or allotriomorphic texture with K-feldspar phenocrysts.

To the Upohlav type we range also rhyolites with aphanitic matrix and magmatically corroded quartz and albite.

2 - The Krivá type: biotite to two-mica leucocratic granites, granodiorites, leucotonalites and granitic pegmatites. These rocks resemble some Hercynian granitoids in the Central Western Carpathians (Tatric and Veporic Unit). The granitic rocks show the even-grained texture with predominance of plagioclase over K-feldspar and subhedral brown biotite, muscovite and garnet is sometimes frequent. The average modal composition of the Krivá granitic rocks contains ($n = 4$): quartz 35.8 ± 1.8 , K-feldspar 11.5 ± 5.7 , plagioclase 45.5 ± 9.2 , biotite 4.4 ± 2.7 , muscovite 1.0 ± 0.5 (vol. %).

The granitic pegmatites consist of coarse-grained pink K-feldspar and quartz, sometimes with muscovite.

3 - The Lubina type is a coarse-grained porphyric granite with pink perthitic K-feldspar, strongly sericitised plagioclase and rare

anhedral to subhedral yellow-brownish biotite, often chloritised. The definition of this type is inaccurate as only a single pebble has been found so far.

Methods of zircon study

Approximately 5 to 7 kg solid pebbles of the size between 20 - 30 cm were used for zircon separation. Samples were crushed, powdered and sieved up to 0.5 mm. Zircon concentrates were obtained using shaking table, heavy liquid (bromofom) and electromagnetic separation techniques.

Microprobe analyses of zircon were made by JEOL JCSA 733 Superprobe microanalyser (Geological Institute of D. Štúr, Bratislava) at following conditions: accelerating voltage 25 kV, probe current 40 nA, electron beam diameter 5 μ m, standards used zircon (for Si, Zr, Hf) and YAG (for Y), ZAF correction used.

Internal zoning of zircon was studied by JEOL JSM 840 scanning electron microscope (Geological Institute of D. Štúr, Bratislava) using 26 kV accelerating voltage.

Zircon typology

Typological analysis of zircon (Pupin 1980, 1988) has recently become a wide-spread informative method of petrological study of magmatic rocks, mainly granitoids. The I.A and I.T typological parameters correspond to alkalinity or agpaicity, $(Na + K)/Al$ and crystallization temperature, respectively. This, in turn, may indicate geochemical and geotectonic magma type, its relation to the orogen and indirectly characterizes the geotectonic situation in the time of intrusion (Pupin 1988).

The Upohlav granites and granite porphyries (21 samples) show practically identical typograms with unambiguous distribution of zircon types with high I.A and I.T values (Tab. 1, Figs. 2a, 3a). The P₅ zircon subtype dominates, the D type is frequent, and P₃₋₄, S₂₃₋₂₅, J₃₋₅ subtypes are present, but rare. In two cases (loc. Podbranč, Vrtižer) also a metamict, low-temperature bipyramidal A type of zircon is present (Fig. 2b).

Table 1: Localization, rock types, typological means points and rocks Zr concentrations.

Localization	Rock Type	I \bar{A}	I \bar{T}	Zr (ppm)
BP-1 Podbranč	G(U)	700	706	263
BP-2 Stupné	G(U)	699	704	234
BP-3 Brvnište	G(U)	700	701	182
BP-6.1 Považský Chlmec	P(U)	697	712	278
BP-6.2 Považský Chlmec	G(U)	687	707	330
BP-7.1 Divinka	G(U)	698	712	252
BP-7.2 Divinka	G(U)	685	699	370
BP-8 Záštranie	GD(K)	275	300	102
BP-10.1 Krivá	GD(K)	254	293	102
BP-10.2 Krivá	GD(K)	245	301	64
BP-11 Široká	T(K)	278	312	79
BP-12.1 Vrtižer	G(U)	699	731	410
BP-12.2 Vrtižer	P(U)	688	710	229
BP-13 Lubina	GP(L)	408	366	180
BP-14.1 Vrš. Podhradie	G(U)	681	714	450
BP-14.2 Vrš. Podhradie	G(U)	696	710	282
BP-14.3 Vrš. Podhradie	P(U)	699	676	191
BP-14.4 Vrš. Podhradie	P(U)	688	717	302
BP-14.5 Vrš. Podhradie	P(U)	700	707	295
BP-15 Upohlav	G(U)	690	712	316
BP-16 Vadičov	R(U)	678	700	252
BP-17.1 Podbiel	G(U)	699	709	191
BP-17.2 Podbiel	G(U)	700	718	190
BP-18 Tepličkan.V.	R(U)	697	705	209
BP-19 Zádubnie	G(U)	700	713	209
BP-20 Beňov	G(U)	693	715	340
BP-31 Cetuna	G(U)	700	721	224
BP-35 Nimnica	G(U)	699	711	410

The overall number of types and subtypes varies in one sample between 3 and 9 (5 in average). There are no differences between typograms of granites and granite porphyries, only in the one case of Vršatské Podhradie porphyry (BP-14.3) a moderately lower I.T is observed along with the more significant presence of P₄ subtype (Tab. 1).

The rhyolites from Vadičov and Teplička nad Váhom have their typograms almost identical with Upohlav type granites and granite porphyries. The Vadičov sample shows the presence of a small amount (5 %) of zircon with low I.A and I.T indices (Fig. 3b) which might indicate e.g. a contamination of rhyolite magma by another magmatic material.

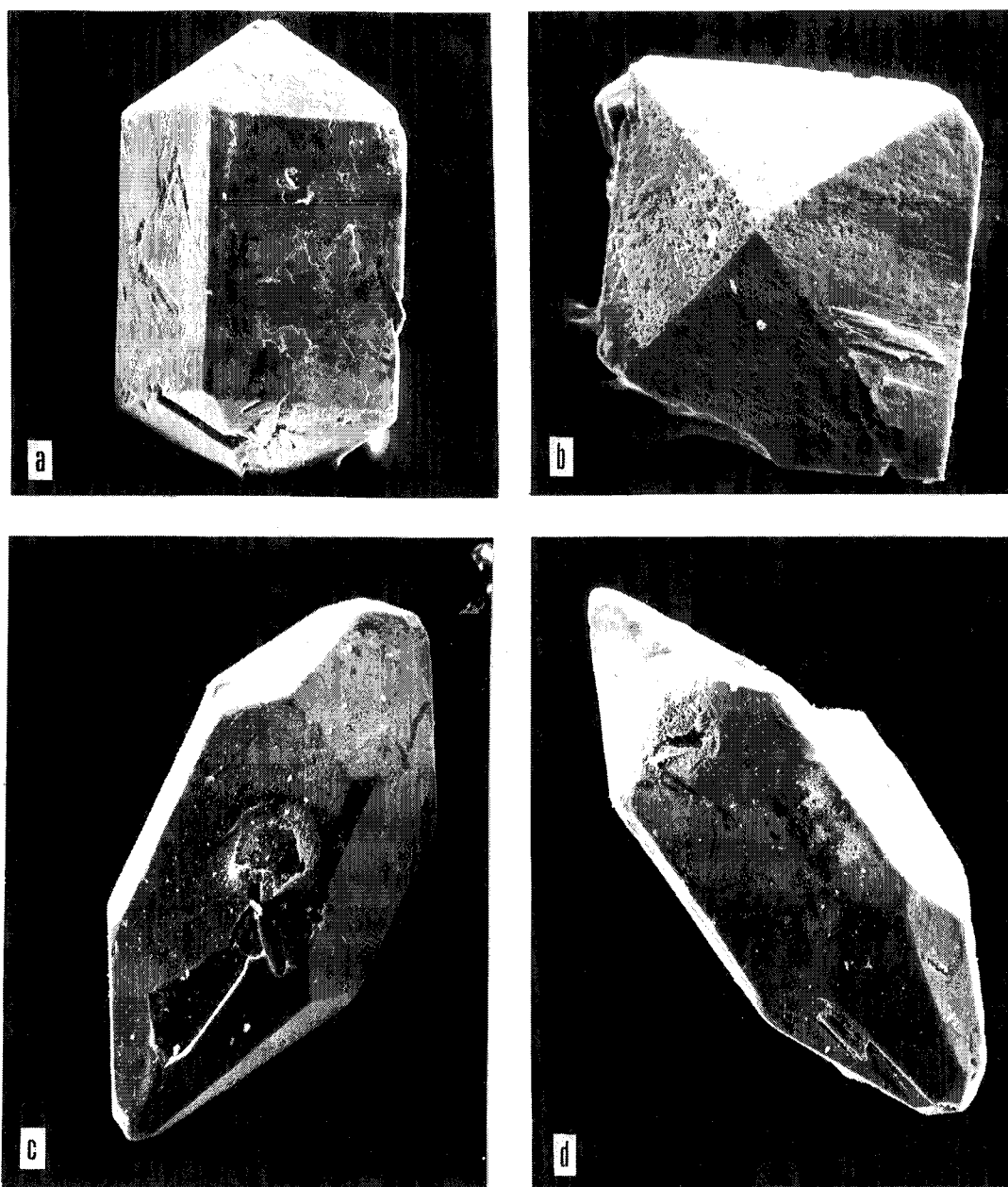


Fig. 2. Zircon morphology (SEM). **a** - P₅ subtype Upohlav locality (BP-15), Upohlav type, the length of crystal 0.24 mm; **b** - A, Vrtižer locality (BP-12.1), Upohlav type, 0.12 mm; **c** - S₁₋₂, Široká locality (BP-11), Krivá type, 0.33 mm; **d** - S₆, Krivá locality (BP-10.1) Krivá type, 0.28 mm.

The granitic rocks of the Krivá type (4 samples) differ in typology significantly from the previous group. Zircons with low I.A and I.T indices (S₁₋₂ subtypes) strongly prevail, the L₁₋₂, S₆₋₇ subtypes are presented to a lesser extent and other subtypes are rare (Tab. 1, Figs. 2c, d, 3c). The total number of types and subtypes is 7 to 11 (9 in average). The typograms almost do not differ each other, slight differences are in the I.A index (Tab. 1).

The Lubina type porphyric granite (1 sample) shows a specific zircon typogram with high frequency of the S₂ subtype and lower frequency of S₁₂₋₁₃, S₃, S₇₋₈ and P₁ subtypes. A characteristic feature of the Lubina granite is high total number of identified zircon subtypes ($n = 23$, Fig. 3d).

Chemical composition and zoning of zircon

The internal structure - zoning of individual zircon crystals belongs also to the important source of information regarding the P-T-X conditions of crystal growth, presence of relict generations of zircons from preceding orogenic cycles (Broska & Caňo 1987; Vavra 1990), etc.

Zircons from the Upohlav type granites and porphyries show only moderate regular oscillatory (Fig. 4a), less diffusional zoning, or they are practically unzoned. In a few cases zircon crystals exhibit irregular internal structure with replacement phenomena by a younger zircon generation with high HfO₂ (4.15 wt. % - Podbranč) and Y₂O₃ (0.89 wt. % - Stupně) contents. The yttrium contents, however, vary within

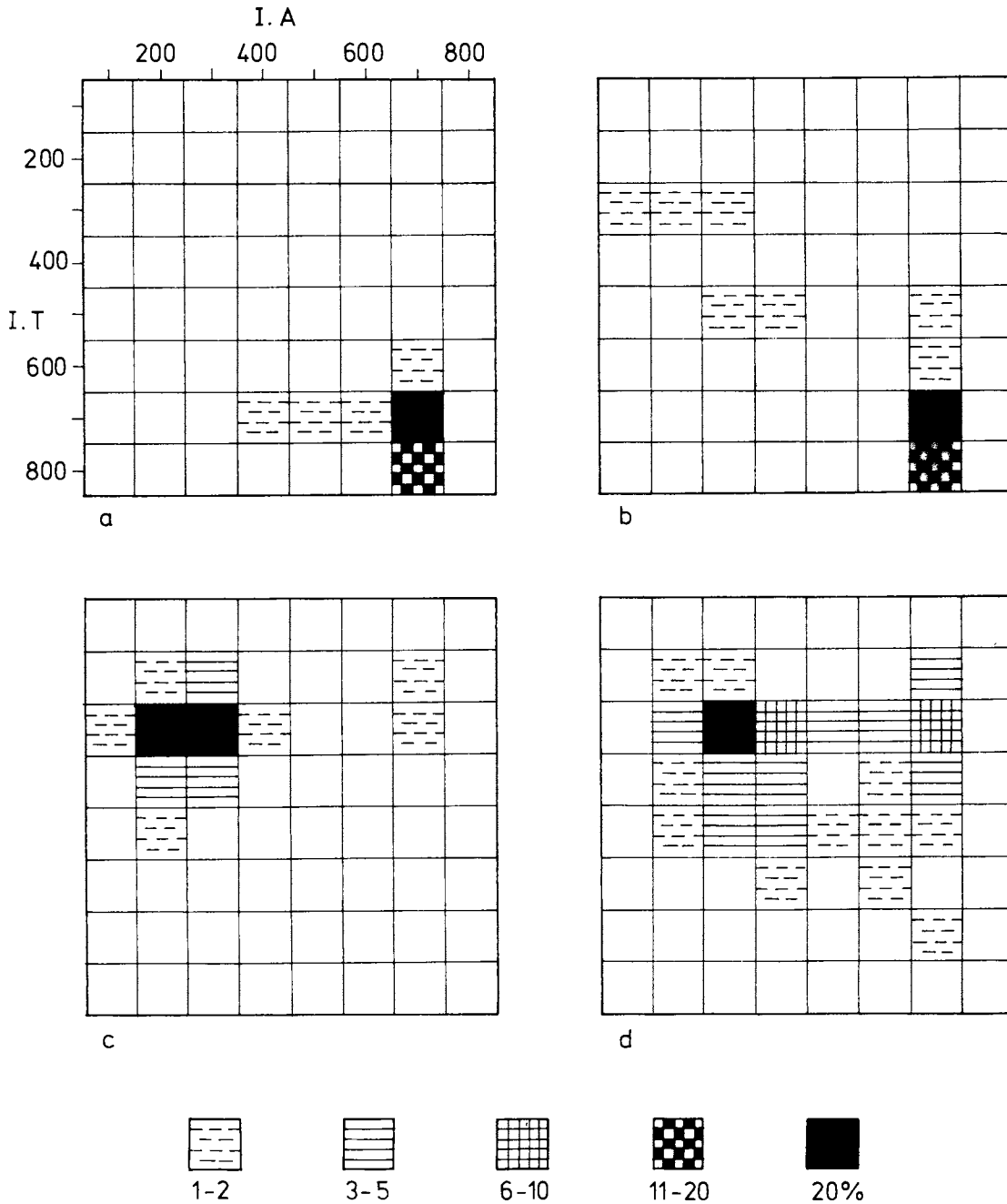


Fig. 3. Zircon typograms (Pupin 1980,1988). a - Beňov locality (BP-20), Upohlav type (granite); b - Vadičov locality (BP-16), Upohlav type (rhyolite); c - Zástranie locality (BP-8), Krivá type (granodiorite); d - Lubina locality (BP-13), Lubina type (porphyric granite).

the regular zoning (0.29 - 1.11 wt. % Y_2O_3 - BP-14.2, Vršatské Podhradie). The ZrO_2/HfO_2 ratios do not change significantly once rising or once declining (Tab. 2). Zircons are almost always perfectly transparent, light yellow or light pink, sometimes they contain small inclusions of albite, K-feldspar, biotite, apatite, ilmenite and rutile. Sporadically, a low temperature generation of metamict light brown zircon (A type) occurs which is unzoned with higher HfO_2 contents (1.9 - 3.4 wt. %) and often with numerous torianite (ThO_2) inclusions (Tab. 2, Fig. 4b).

The zircons from the Krivá type granitoids display a significantly different zoning. They have unzoned to weakly zoned cores often with irregular shapes (relict protolith cores, or due to a dissolution process) which are rimmed by weakly concentricly zoned outer parts with higher HfO_2 contents (up to 2 % - Tab. 2, Fig. 4c). The younger zones are slightly transparent, partly metamict with a black

pigment (organic matter?). ZrO_2/HfO_2 ratios always decline from core to rim. Torianite was locally observed among inclusions in the cores of partially metamict zircons (Fig. 4d).

Zircons crystals from the Lubina type porphyric granite show mostly regular oscillatory zoning of various intensity and the decrease of ZrO_2/HfO_2 ratios and Y_2O_3 contents from cores to rims. Compared to the Krivá type they have significantly higher Y contents (Tab. 2). No inclusions of other minerals were observed.

Discussion

Results of the zircon study have shown significant differences in typology, zoning, and partly also in chemical composition of this mineral among various types of acid magmatic rocks pebbles

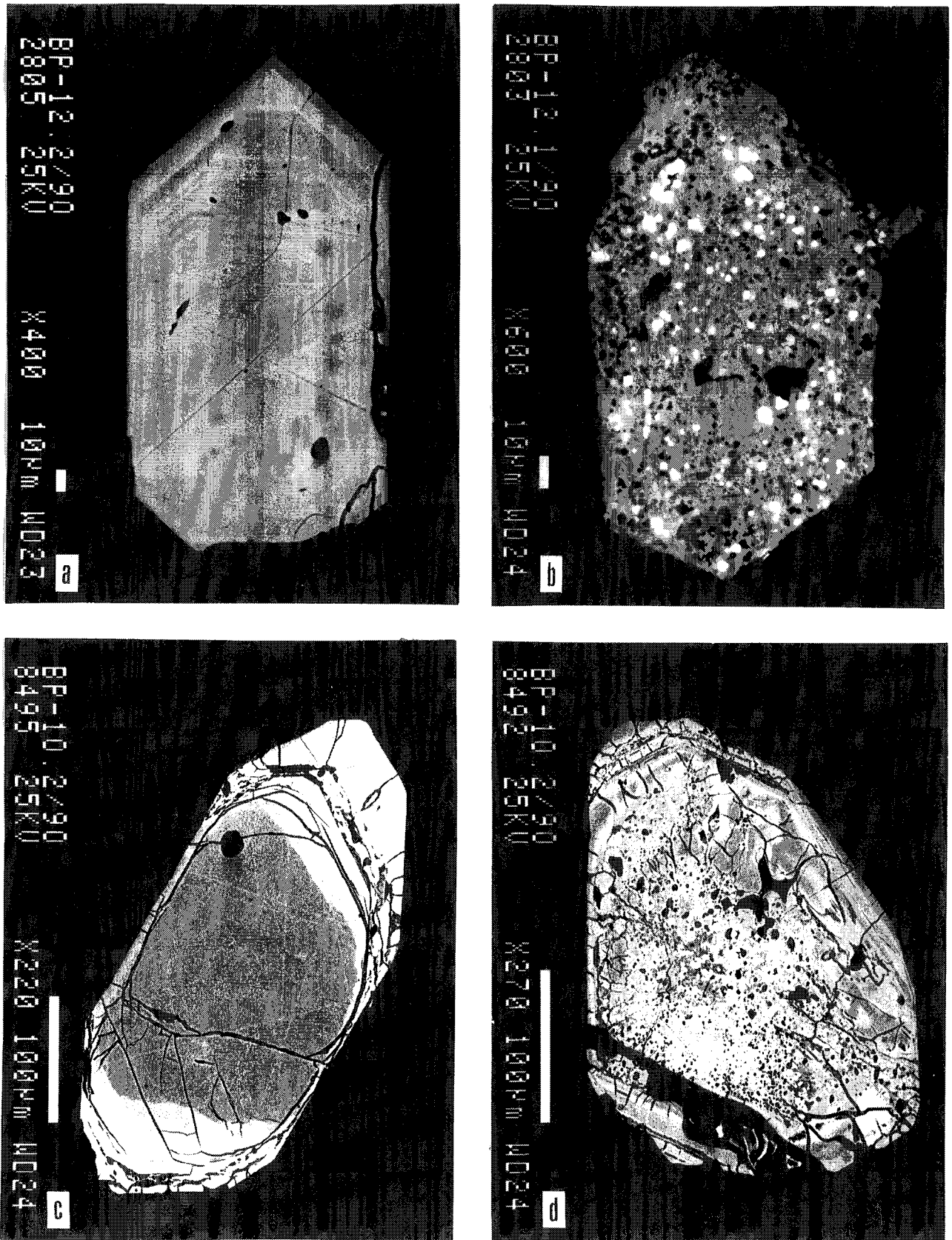


Fig. 4. Internal structure (zonig) of zircon (BEI). **a** - transparent crystal, Vrtižer locality, Upohlav type; **b** - metamict crystal with torianite inclusions (white), Vrtižer locality (BP-12.1), Upohlav type; **c** - transparent crystal with younger zone enriched in Hf (white), Krivá locality (BP-10.2), Krivá type; **d** - partly metamict crystal with torianite inclusions (white) in central parts, Krivá locality (BP-10.2), Krivá type. The length of bar is 10 μm (a, b), 100 μm (c, d).

from the Cretaceous flysch of the Pieniny Klippen Belt (western Slovak segment). Typological analysis of zircons from granites, granite porphyries and rhyolites of the Upohlav type with high I.A and I.T values suggests a relatively high crystallization temperature ($T \approx 800 - 900 \text{ }^\circ\text{C}$) as well as alkalinity (agpaicity) of the magma environment (cf. Pupin 1980, 1988). The zoning and chemical composition of the Upohlav type zircons points to a mainly single stage crystallization with moderate fluctuations within a single crystal, but with variations of Y_2O_3 contents and $\text{ZrO}_2/\text{HfO}_2$ ratios within rock. This may be connected with crystallization of a relatively mobile magma at different depth levels. Only locally a metamict Hf-rich zircon indicate the sub-solidus conditions. The Upohlav acid magmatites can be compared with small post-orogenic intrusions with higher alkalinity, temperature and Zr rock concentrations (cca 180 - 450 ppm, Tab. 1) in water-poor and F-enriched melt which solidified during the uplift along deep fault systems at various depth levels with granite - granite porphyry - rhyolite sequence (cf. e.g. Lameyre 1988; Bonin 1988, 1990; Whalen et al. 1987).

Although the signs mentioned ascribe these acid magmatites to A-type granites (Pitcher 1983; Whalen et al. l.c.), the absence of syenite members, alkalic pyroxenes and amphiboles as well as insufficient contents of some indicator elements in the Upohlav type (Uher, unpubl. data) do not allow to range these rocks to the products of evolved alkalic suites. An analogical granite type has not been described so far in the Tatric and Veporic Unit of the Western Carpathians. They do not resemble neither Sn-bearing granites of the Gemicic Unit with completely different zircon typology (Jakabská & Rozložník 1989). The zircons of the Rochovce and Hrončok type granites (Veporic Unit) appear more typologically similar with high I.A, but lower I.T indices (Fig. 5). More significant analogies are seen when comparing typology of the Upohlav acid magmatites with late-Hercynian leucocratic, higher alkalic granites and granite porphyries of the Velence Mts. (Transdanubian Range, Bakony Unit) in Hungary (Gbelský & Határ 1982; Uher & Broska in prep.) and particularly with a small intrusion of the Turčok granite at the northern boundary of the Gemicic Unit (Uher & Gregor 1992), Fig. 5. In addition, very similar zircon typology is known from Permian alkalic granites and rhyolites of the Western Mediterranean province, e.g. from Corsica (Pupin 1980, 1988).

On the other hand, our results are not in accord with present ideas on exotic magmatites of the Pieniny Klippen Belt flysch conglomerates as representing a Jurassic to Cretaceous volcanic-plutonic suite of the island arc or active continental margin type (Marschalko 1986; Birkenmajer 1988; Mišák & Marschalko 1988, etc.). Such subduction-related magmatic rocks with dominant intermediate, less acid composition (mainly andesites, dacites, diorites and granodiorites) show a characteristic zircon typology different from that of the Upohlav acid magmatites (cf. Pupin l.c.; Rozložník et al. 1985; Hodruša-Štiavnica intrusive complex in central Slovakia). To the contrary, the Upohlav type of granites and their (sub)volcanic products resemble mostly the post-orogenic (post-collisional) suite of A-type related rather to extensional than compressional tectonic setting (Pitcher 1983; Lameyre 1988, etc.). A possibility arises to compare the Upohlav acid magmatites with post-orogenic to early anorogenic suites in the Alpine-Mediterranean Belt associated with ceasing of the Hercynian orogen and with post-Hercynian consolidation between Middle Permian and Upper Triassic (Bonin 1988, 1990). Without doubts, it will be necessary to confirm our interpretation by further geochemical and, particularly, geochronological data. Besides, the problem of the provenance of exotic

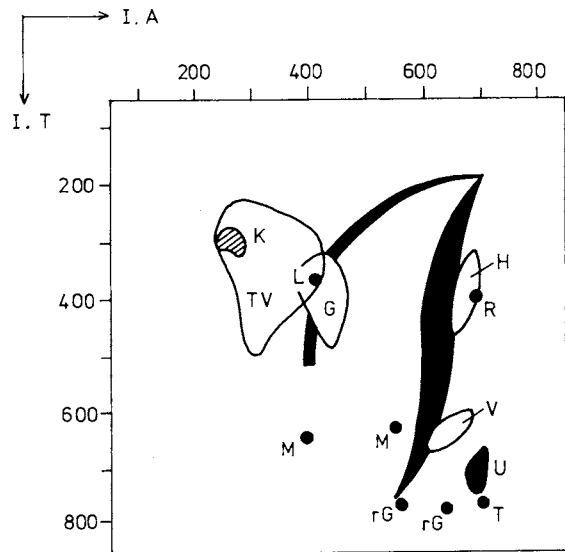


Fig. 5. Typological mean points of zircon (Pupin 1980, 1988) from various types of Hercynian granitoid rocks and rhyolites of the Western Carpathians, Velence and Mecsek Mts. compared with studied rocks.

TV - Tatric and Veporic granitoids (Broska & Uher 1991); G - Sn-bearing Gemicic granites (Jakabská & Rozložník 1989; Broska & Uher 1991); V,M - Velence and Mecsek granites (Gbelský & Határ 1982); T - Turčok granite (Uher & Gregor 1992) R - Rochovce granite; H - Hrončok and Vydrovo granite; rG - rhyolites of the northern part of Gemicic unit (Uher - unpublished); U - granites, porphyries and rhyolites of the Upohlav type; K - Krivá type granitoids; L - Lubina type granite.

rocks in the Cretaceous flysch of the Pieniny Klippen Belt remains still unresolved in spite of several suggestions of possible sources (Mišák & Marschalko 1988).

Contrary to the Upohlav type acid magmatic rocks, the Krivá type granitoid pebbles can well be compared with some Hercynian, especially Carboniferous, orogenic granites to leucotonalites of the Central Western Carpathians, the Tatric and Veporic Units, in particular, with monazite type granitoids (Broska & Uher 1991). They show very similar zircon typology (Fig. 5) and internal zoning (cf. Broska 1986) as well as trends and $\text{ZrO}_2/\text{HfO}_2$ ratios in zircons (Broska l.c.; Broska & Uher, unpubl. data).

The porphyric Lubina type can also be partially compared with the Krivá type granitic rocks, its typogram being more similar to lower temperature granites at the boundary of aluminous anatectic and calc-alkalic s.s. series (sensu Pupin l.c.).

The similarity of the Krivá and partly the Lubina type granitic rocks with Hercynian orogenic plutonites of the Central Western Carpathians, however, does not have to be a proof of their common origin. Analogical rocks may have originated also in another area of the Hercynian orogen.

Conclusions

Based on the study of typology, internal zoning and chemical composition of zircons from granitic and rhyolitic pebbles from conglomerates of the Cretaceous flysch of the Pieniny Klippen Belt (western Slovak segment) following conclusions can be formulated:

Table 2: Representative microprobe analyses of zircons from various PKB granitoids.

	BP-1C	BP-1R	BP-2C	BP-2R	BP-6.1C	BP-6.2R	BP-14.2C	BP-14.2R	BP-12.1M	BP-8C	BP-8R	BP-10.2C	BP-10.2R	BP-11C	BP-11R	BP-13C	BP-13R
SiO ₂	32.26	32.82	33.27	33.36	32.80	33.04	32.27	31.97	32.60	32.33	31.85	32.52	32.70	32.48	32.89	32.18	32.68
ZrO ₂	67.13	66.66	64.13	64.67	66.64	66.10	65.06	63.41	65.84	66.57	65.56	65.92	66.25	65.83	65.39	65.80	65.47
HfO ₂	1.27	1.38	2.03	1.76	1.40	1.31	1.49	1.31	3.42	1.48	2.08	1.43	1.45	1.32	1.64	1.41	1.44
Y ₂ O ₃	0.00	0.00	0.00	0.19	0.07	0.14	0.50	0.81	0.08	0.00	0.11	0.13	0.02	0.15	0.02	0.94	0.44
TOTAL	100.66	100.86	99.43	99.98	100.91	100.59	99.32	97.50	101.94	100.38	99.60	100.00	100.42	99.78	99.94	100.33	100.02
ZrO ₂ /HfO ₂	52.86	48.30	31.59	36.74	47.60	50.46	43.66	48.40	19.25	44.98	31.52	46.10	45.69	49.87	39.87	46.67	45.47
(Zr/Hf) _w	46.13	42.15	27.57	32.06	41.54	44.03	38.10	42.24	16.18	39.25	27.50	40.23	39.87	43.52	34.79	40.72	39.67
(Zr/Hf) _a	90.29	82.51	53.96	62.77	81.31	86.19	74.59	82.68	32.89	76.83	53.84	78.74	78.05	85.19	68.11	79.72	77.66

Explanations: C - core; R - rim; M - metamict zircon; w - weight ratio; a - atomic ratio. For locality see Tab. 1.

1 - Granites, granite porphyries and rhyolites of the Upohlav (exotic) type show zircons with high I.A and I.T typological indices, mainly regular oscillatory zoning and variable Hf and Y contents. They represent products of relatively high temperature dry magma with increased alkalinity which rised from a deep-seated source along fault systems and solidified at various deep levels of the crust. We suppose that a post-orogenic (post-collisional) A-type related magmatic suite can be concerned here with some analogies to post-Hercynian (Permo-Triassic) alkalic province in the Alpine-Mediterranean Belt (sensu Bonin 1988).

2 - Granites, granodiorites, leucotonalites and pegmatites of the Krivá type are characterized by zircons with low I.A and I.T indices, often with irregular cores and Hf-enriched, Y-depleted rims. They can well be compared with some Hercynian (Carboniferous) crustal-anatectic orogenic intrusions in the Central Western Carpathian crystalline complex (monazite type, Tatric and Veporic Unit).

3 - Porphyric granite of the Lubina type gives zircons with mainly lower I.A and I.T indices, regular oscillatory zoning and the decline of Zr/Hf and Y from cores to rims of crystals. This type shows the specific features making it similar to orogenic granitoids with affinity to calc-alkalic series. It is only partly comparable with Hercynian orogenic acid plutonites of the Central Western Carpathians.

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