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GEOCHEMICAL CONSTRAINTS OF HYDROTHERMAL ALTERATIONS OF TWO-MICA GRANITES OF THE MOLDANUBIAN BATHOLITH AT THE OKROUHLÁ RADOUŇ URANIUM DEPOSIT

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ABSTRACT

Hydrothermal alteration of two-mica granites at the Okrouhlá Radouň uranium deposit is accompanied by the formation of episyenites. Episyenites originated from these granites typically show very intensive leaching of quartz, albitization of magmatic plagioclase and K-feldspar and chloritization of biotite. Later stages of alteration are characterized by significant carbonatization of episyenites. Mass balance constrains of episyenitization involve a loss of silica and a gain of sodium, calcium and CO_2. A significant part of the uranium mineralization (coffinite and pitchblende) was formed during the Permian extensional phase of late Variscan evolution of the Bohemian Massif.

KEYWORDS: uranium, alterations, two-mica granite, geochemistry, Moldanubian batholith, Bohemian Massif

INTRODUCTION

Uranium mineralization is frequently accompanied by hydrothermal alteration connected with quartz dissolution. Hydrothermally altered rocks depleted in quartz are referred to as episyenites as defined by Lacroix (1920). Episyenitization is a well-known type of subsolidus alteration in Hercynian granites of the French Massif Central (Limousin, Marche, Forez) and/or the Armorican Massif (Vendee) (Poty et al., 1986; Cathelineau, 1986, 1987). Other occurrences of episyenites, also evolved in granites connected with uranium mineralization, were described by Halenius and Smellie (1983), Smellie and Laurikko (1984) and Öhlander (1986) from Sweden. In the Bohemian Massif, episyenitization of uranium-bearing granites has been reported from the Smrčiny pluton (Hecht et al., 1994), the Bor pluton (Fiala 1980a, b; Romanidis, 1980) and the Central Bohemian pluton (Pivec and Langrová, 1982). In the Bohemian Massif, episyenitization and the associated uranium mineralization occur not only in granites, but also in high-grade metamorphic rocks of the Moldanubian Zone (Mrázek and Fiala, 1979; Fiala and Čadek, 1981; Dill, 1983; Křibek et al., 2002; René, 2002). Episyenitization of metamorphic rocks of the Moldanubian Zone from the Okrouhlá Radouň uranium deposit was described by Fiala and Čadek (1981). The evolution of episyenites in two-mica granites from the same deposit was mentioned by Romadinis (1980) and Fiala and Králík (1989).

The present study focuses on geochemical constraints of evolution of episyenites in two-mica granites in the southern part of the Okrouhlá Radouň uranium deposit. The two-mica granites belong to the Klenov granite body and represent a distinct type (the so-called Deštná granite) of two-mica granites in the Moldanubian (South Bohemian) batholith (Fig. 1) (Klečka et al., 1991; René et al., 1999, 2003). This paper also presents some results of a project on the Klenov granite body, supported by the Grant Agency of the Czech Republic (Project No. 205/97/0514).

GEOLOGICAL SETTING

The Okrouhlá Radouň uranium deposit, lying some 15 kilometres north of Jindřichův Hradec, was mined in the 1970s and was ranked among smaller uranium deposits in the Bohemian Massif (Arapov et al., 1984). The total amount of uranium obtained from this deposit was 1340 metric tons (Šuráň and Veselý, 1997). The deposit was opened by two shafts to a depth of about 600 metres and developed along the strike at a distance of about three kilometres. The wider area of this deposit was subjected to extensive exploration by boreholes and other exploratory works (Mrázek, 1972), with boreholes drilled to the depths of 300–1200 metres.

The Okrouhlá Radouň uranium deposit lies on the northeastern margin of the Klenov granite body (Fig. 2). This isolated magmatic body, called the Klenov massif by Zelenka (1923), represents the largest granitic body west of the Central Moldanubian pluton. Both magmatic bodies are a part of the Moldanubian (South Bohemian) batholith (Klečka et al., 1991; René et al., 1999, 2003). The two-mica

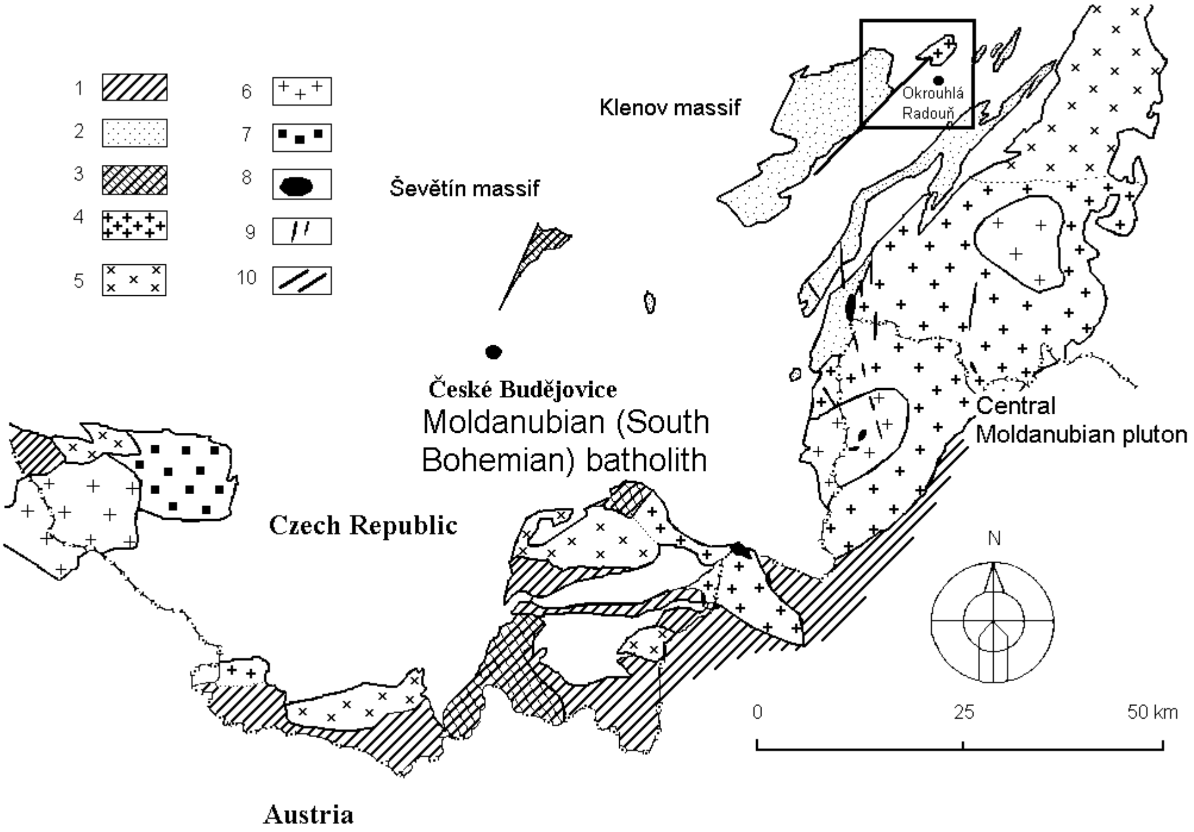

Fig. 1 Sketch map of the Moldanubian (South Bohemian) batholith. After Breiter and Sokol (1997), modified by author.
1 – granite of the Weinsberg type, 2 – granite of the Deštná - Lásenice type, 3 – granodiorite of the Freistadt and the Mauthausen types, 4, 5 – granite of the Eisgarn type, 4 – the Číměř granite, 5 – the Mrákotín granite, 6 – granite of the Zvůle (Landštejn) type, 7 – durbachites, 8 – granite of the Homolka type, 9 – dykes of granite porphyries, 10 – faults.

granites of this magmatic body have been recognized to represent an independent geochemical type of two-mica granite, because of their particularly low contents of Zr and Th (Klečka et al., 1991; René et al., 1999, 2003). They are, therefore, different from the two-mica granites of the Eisgarn type, which build most of the Moldanubian batholith in the Czech Republic.

The Klenov granite body is about 25 km long, elongated NE-SW. It stretches from Lomnice nad Lužnicí to the SW surroundings of Kamenice nad Lipou. Its average width is 6–8 km. It is hosted by regionally metamorphosed rocks of the Moldanubian Zone, particularly sillimanite-biotite paragneisses. It is dissected into separate tectonic blocks by NW–SE-striking faults. At its eastern margin, the Klenov granite body is limited by an expressive NE–SW-striking shear zone termed the Northeastern shear zone (Pletánek, 1979). The marked tectonic deformations of the northeastern margin of the Klenov granite body are also concentrated to NNW–SSE-striking shear zones, usually containing uranium mineralization and/or quartz-carbonate mineralization (Figs. 2, 3). The most significant shear zone in area of the Okrouhlá Radouň uranium deposit is the so-called Main Radouň zone (OR-5), which was explored in along-strike direction for a distance of about two kilometres. The richest uranium mineralization developed along this shear zone at a depth of 250–400 m beneath the present surface. The OR-5 shear zone strikes 350–0° and dips 65–80° W. Its thickness is highly variable, from 30 centimetres to about seven metres. The largest thickness was observed in the central part of the Okrouhlá Radouň uranium deposit, where this shear zone is present in high-grade metamorphic rocks of the Moldanubian Zone. In this part, the shear zone is filled with cataclastites formed by host rocks, altered to clay minerals-rich and chlorite-rich assemblages. Albitization of the original plagioclase, chloritization of biotite and enrichment in hematite are common. The cataclastites in the central part of the deposit are sometimes enriched in graphite and sulphides, and host uranium mineralization enriched in coffinite, partly also pitchblende. In the southern

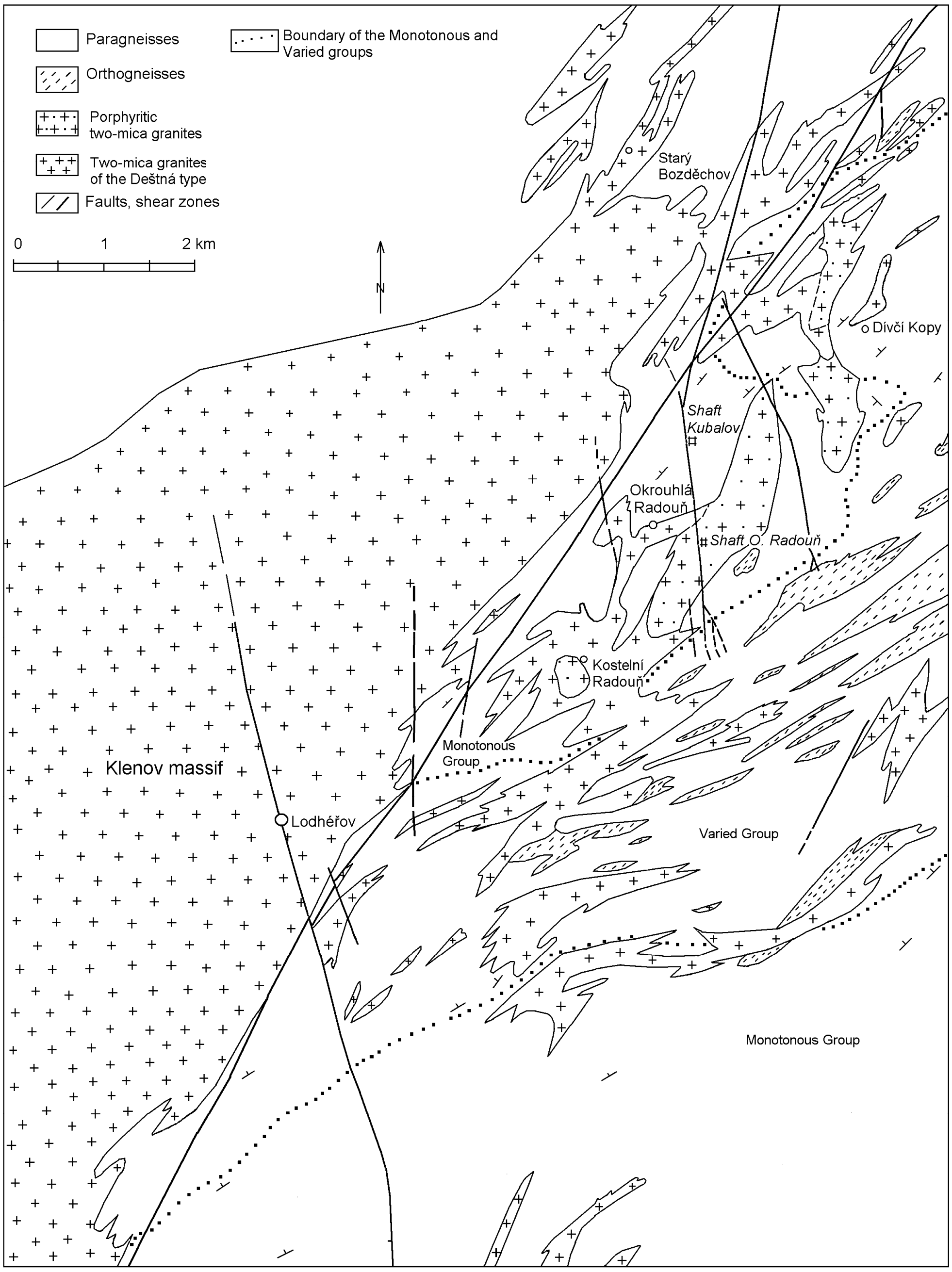

Fig. 2 Geological map of the northeastern margin of the Klenov granite body (after Pletánek 1979, modified by author).

part of the deposit, the OR-5 shear zone splits to a higher number of thinner shear zones (OR-5, OR-5a, OR-3, OR-3b) (Figs. 2, 3), partly developed in two-mica granites of the Deštná type. In the proximity of OR-5 and OR-3b shear zones, the two-mica granites are altered to produce wide metasomatic zones composed mainly of albite, carbonates, hematite, hydromuscovite and chlorite. The highest thickness of metasomatite bodies and lenses is reached at places of changing strike of the shear zone. At such places, episyenite bodies form lenses or very irregular shapes. Very thick episyenite bodies rich in uranium were found in the OR-3b ore zone near the sampling sites of Re-509 and Re-510. Other portions of two-mica granites rich in uranium are present in gallery V-2 in the central part of the deposit (Fig. 3). This gallery yielded samples for mass-balance analysis of episyenitization (Fig. 4). Larger bodies of episyenites, sometimes following faults, are accompanied by lenses or very irregular veins of carbonates with quartz and/or sulphides (pyrite, galena, chalcopyrite, sphalerite). The thickness of these carbonate-rich lenses is usually 1–50 centimetres.

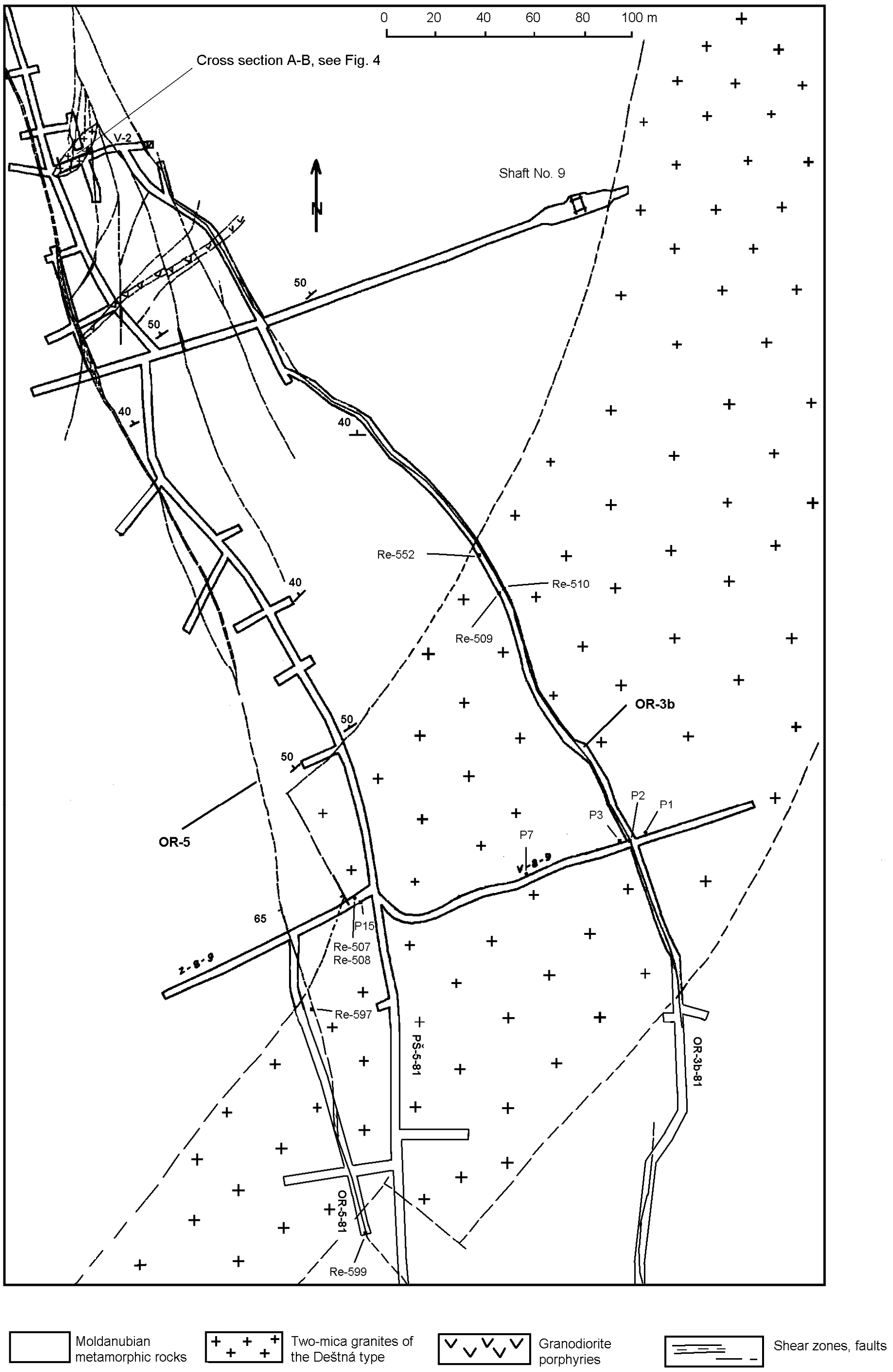

Fig. 3 Mining map of the southern part of the 8. level at the Okrouhlá Radouň uranium deposit (after Patočka et al., 1977, modified by author).

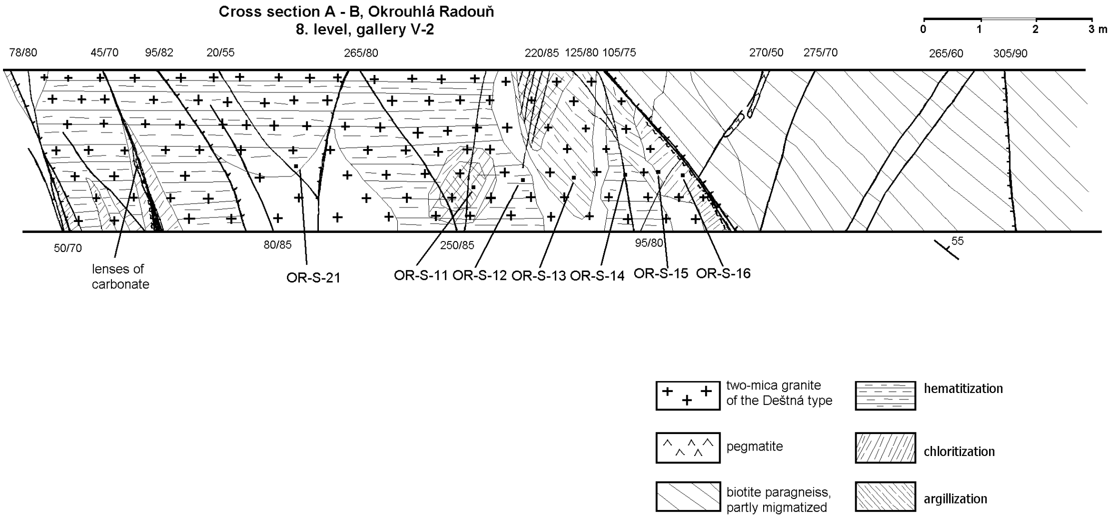

Fig. 4 Cross section of the episyenite body at the V-2 gallery (northern wall of gallery).

ANALYTICAL METHODS

Sampling focused mainly on the episyenites and partly altered two-mica granites of the southern part of the Okrouhlá Radouň uranium deposit. Unaltered two-mica granites from the southern part of this deposit were also sampled for comparison. Samples of rock material 2–5 kg in weight were taken for chemical analyses. The rocks were crushed in a jaw-breaker and an agate ball mill. Major elements and some trace elements (Ba, Rb, Sr, Zr) were determined by conventional X-ray fluorescence spectrometry on the Siemens SRS-1 spectrometer at MEGA Ltd. laboratory. Major elements were analysed on fused glass disks, analyses of trace elements were obtained on pressed rock powder pellets. FeO content was determined by titrimetric method. U and Th were determined by gamma-ray spectrometry using the multi-channel gamma-ray spectrometer Canberra also in MEGA Ltd. laboratory. REE and Y contents were determined by ICP MS at Activation Laboratories Ltd., Ancaster, Canada on Perkin Elmer Sciex ELAN 6100 ICP mass spectrometer. The decompositions of rock samples for ICP MS analyses involved lithium mataborate-tetraborate fusion. Precision of these analytical methods was tested by duplicate analyses.

Analyses of minerals (plagioclase, K-feldspar, biotite) were performed on a CAMECA SX-100 microprobe in WDX mode at the Institute of Mineralogy, University of Hannover, and at the Institute of Geology, Academy of Sciences of the Czech Republic. Accelerating voltage was 15 kV and beam current was 10 nA. The correction of raw analytical data employed the X-PHI procedure. Mineral formulas were recalculated using the Minpet 2.0 software.

PETROGRAPHY

COMPOSITION OF UNALTERED TWO-MICA GRANITES

The Deštná two-mica granite in the southern part of the Okrouhlá Radouň uranium deposit is a fine- to medium-grained, equigranular monzogranite (Fig. 5). Very often it also contains schlieren or small nodular accumulations of older biotite (sometimes together with sillimanite), which probably represent restite. The modal composition is quite variable: quartz 28–42 vol.%, K-feldspar 21–33 vol.%, plagioclase 23–34 vol.%, primary magmatic muscovite 1.2–7.0 vol.% and biotite 1.4–4.1 vol.%. Accessory minerals are andalusite, zircon, apatite, ilmenite, monazite, xenotime and very rare cordierite. K-feldspar (microcline) with composition $Or_{81\text{-}97}$ $Ab_{3\text{-}19}$ $An_{0.1\text{-}0.4}$ forms irregular, anhedral to subhedral grains, usually 0.6–1.2 mm in size. Plagioclase (oligoclase, mostly $An_{11\text{-}24}$) forms subhedral to euhedral tables, 0.4–0.8 mm in size, sometimes slightly zoned with an albite rim ($An_{0.3—7.0}$) and oligoclase core. Biotite usually forms subhedral tables, 0.2–1 mm in size, with significant pleochroism; yellowish to yellow-brown along X and red-brown to dark brown along Y and Z. Its composition corresponds to siderophyllite with Fe/(Fe+Mg) ratio 0.64–0.68. Andalusite occurs as subhedral to euhedral, pink-coloured grains 0.1 mm in size, and is probably of magmatic origin.

COMPOSITION OF EPISYENITES

Episyenitized granite and carbonate-rich episyenite is light rose to red-brown, medium grained, equigranular rock. Episyenite is, as a result of hydrothermal leaching of original magmatic quartz, characterized by medium to high porosity of the rock.

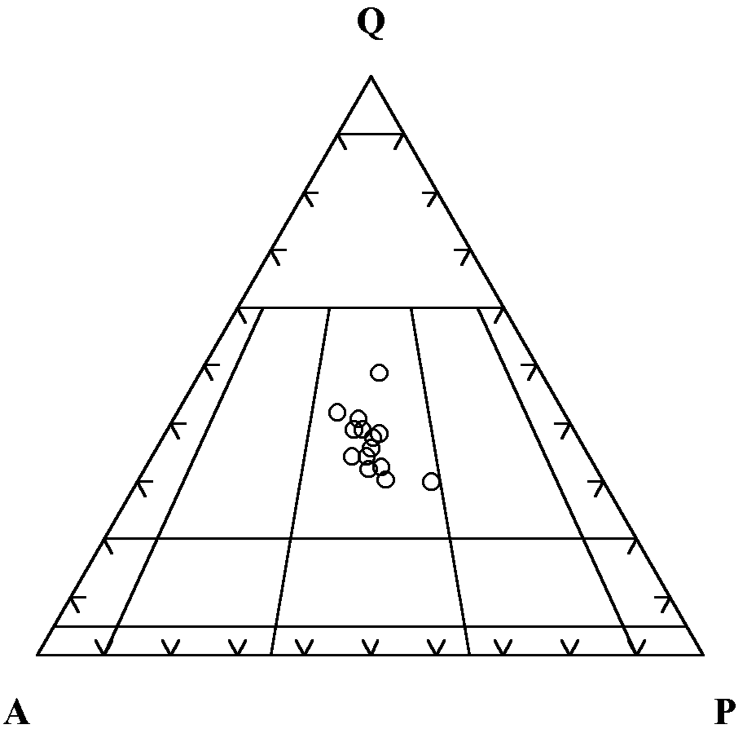

Fig. 5 Modal composition of two-mica granite of the Deštná type from the southern part of the Okrouhlá Radouň uranium deposit in the IUGS diagram.

Except for rose to red-brown colour of feldspars and higher porosity, the macroscopic features of episyenite are similar to those of the original unaltered granite. Thus the original magmatic textures are relatively well preserved, even in the case of total leaching of quartz. The first stage of episyenite formation (pre-ore stage of mineralization, see Fig. 6) is marked by the occurrence of vugs resulting from the dissolution of original magmatic quartz. In later stages, mainly in post-ore carbonate stage, these vugs were filled with carbonates. In the pre-ore mineralization stage, some of the vugs were filled with newly formed albite (albite II).

The transitional zone between altered and unaltered granite is obviously gradual over a few tens of centimetres to one metre. Commonly, the transitional zone displays a weak red colouring due to the presence of fine-grained hematite tables irregularly distributed in albitized original magmatic plagioclase (albite I) and K-feldspar. Albite and carbonate are the main constituents of the episyenite, together occupying 65–85 vol.% of the bulk rock. Albite, as complete pseudomorphs after magmatic plagioclase (albite I), is lacking in the deuteric assemblages commonly observed in the unaffected granite. The albitized plagioclase exhibits relict polysynthetic twins inherited from the magmatic precursors, but not the classical chessboard twinning typical of secondary albite. Albitized original plagioclases are also characterized by high contents of very fine-grained (0.001–0.1 mm) tables of hematite. Authigenic generations of albite (albite II and albite III) also occur as epitaxial overgrowths on the albite pseudomorphs of magmatic plagioclase (albite I). These overgrowths, chiefly albite III, may display polysynthetic twinning. In some cases, the most strongly albitized episyenites contain also microscopic druses of epitaxy-crystallized albite IV. All three to four generations of hydrothermally originated albite are part of the pre-ore stage of hydrothermal mineralization (Fig. 6). Newly formed albites have near end-member composition ($An_{2.0\text{-}6.0}$).

In albitized granites, the original magmatic K-feldspar is generally only sericitized or kaolinized. During later episyenitization, also K-feldspar is leached, and the rest of potassium is bonded in hydromuscovite and/or illite. Total leaching of K-feldspar is also significant for the pre-ore stage of hydrothermal mineralization. In the early hydrothermal stage of alteration, biotite is chloritized and altered to chlorite I, which forms pseudomorphs after biotite. In later stages, new, hydrothermal generations of chlorite (chlorite II and III) are formed. These later generations of chlorite are very rare in episyenites originated by alteration of two-mica granites of the Deštná type due to the small amount of magmatic biotite. Hydrothermal alteration of biotite is associated with the formation of hydrated titanium oxides (leucoxene?), resulting from the liberation of titanium bounded in the original magmatic biotite.

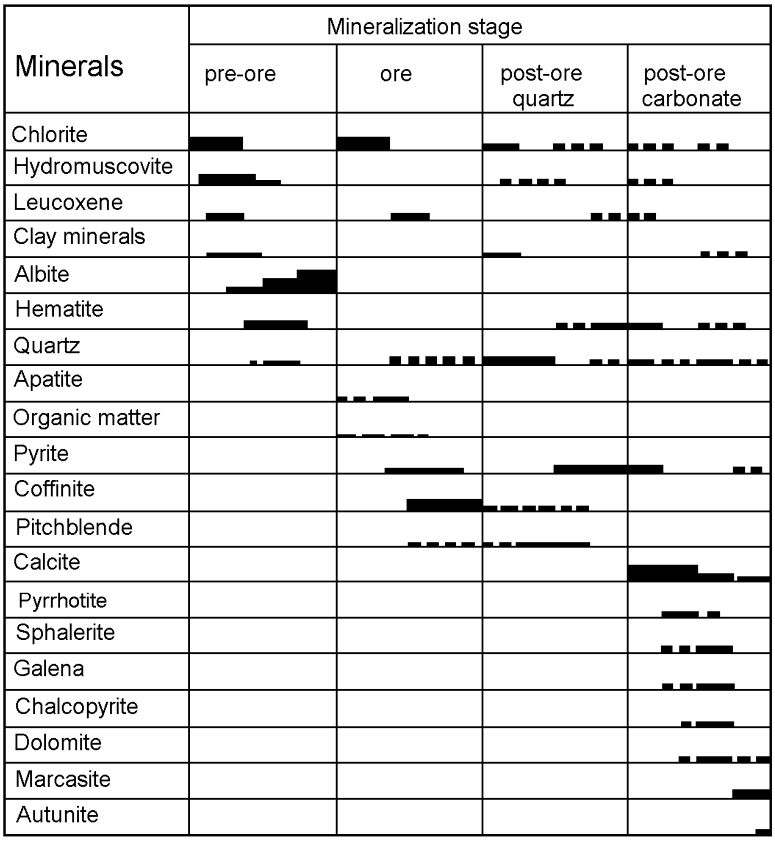

Fig. 6 Paragenetic sequence of the mineralization stages in the Okrouhlá Radouň uranium deposit (after Arapov et al., 1984).

Almost all carbonates were formed during a later, post-ore stage of mineralization. Carbonates fill vugs formed during leaching of magmatic quartz and/or form lenses and veins along smaller faults in episyenites (Fig. 4). The thickness of these lenses and veins is 1–25 centimetres. The origin of carbonates was subdivided into four substages by Brodin (1971). Three groups (PK1–PK3) are formed by calcite, carbonates of PDK group are formed by dolomitized calcite. The oldest substage is formed by PK1 group of carbonates. These carbonates form stalked or columnar, grey- or rose-coloured grain aggregates. Some of the grains are tectonically deformed. PK1 carbonates are characterized by relatively higher contents of MgO, but very low content of MnO (Fig. 7). Carbonates of group PK2 are very abundant. This group is formed by white or rose calcite, sometimes with small inclusions of sulphides (pyrite, chalcopyrite, galena, sphalerite). Dolomitized calcites of the PDK group were formed by later dolomitization of calcites of groups PK1 and PK2. The amount of the dolomite component sometimes reaches 80 vol.% (Solnický, 1972; René, 1998). The youngest carbonate substage is formed by calcites of group PK3. These calcites are white, sometimes rose-like carbonates, usually forming the above mentioned lenses and veins along smaller faults in episyenites. These carbonates typically show relatively high contents of manganese (Fig. 7).

CHEMICAL COMPOSITION OF ROCKS

COMPOSITION OF UNALTERED TWO-MICA GRANITES

The two-mica granites of the Deštná type are characterized by relatively low contents of MgO, TiO_2 and CaO, particularly when compared with the Eisgarn group granites of the Moldanubian batholith (René et al., 2003). This corresponds well with the relatively low amounts of biotite in these granites and with their felsic character. All analysed samples are peraluminous with A/CNK (molecular $Al_2O_3/CaO+Na_2O+K_2O$) ratios of 1.13 to 1.29. The higher peralumosity of these granites is also expressed by the higher contents of normative corundum (CIPW norm): 2.0–4.0 wt.%. The peraluminous character results in a high amount of muscovite and a considerable presence of andalusite or cordierite. Due to their higher A/CNK ratios, the two-mica granites from the southern part of the Okrouhlá Radouň uranium deposit are S-type granites in the sense of the classification of Chappell and White (1974), like the other two-mica granites of the Moldanubian batholith.

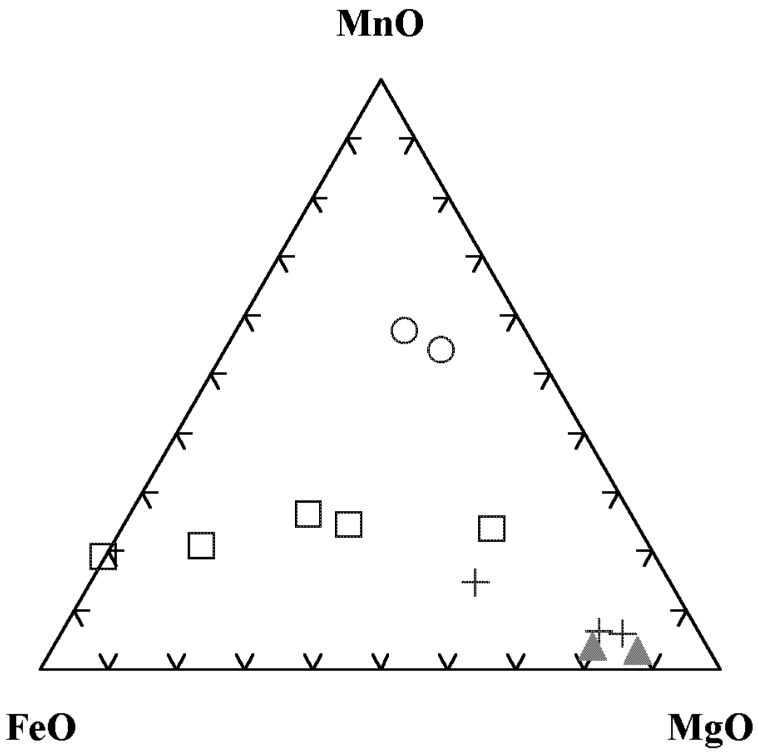

Fig. 7 Distribution MnO, FeO and MgO in carbonates of the Okrouhlá Radouň uranium deposit. Crosses – carbonates of the PK1 sub-stage, quadrangles – carbonates of the PK2 sub-stage, circles – carbonates of the PK3 sub-stage, full triangles – carbonates of the PDK sub-stages.

These granites are characterized by low total contents of REE and a depletion in high-field-strength elements (HFSE – Zr, Nb, Ta, Hf, U, Th) (Table 1, Figs. 8, 9). Particularly significant is the low content of Th and Zr (Fig. 10). The examined two-mica granites are characterized by low LREE/HREE ratios (La_N/Yb_N = 4.5–11.4) and by the absence of the europium anomaly (Eu/Eu* = 0.90–1.14) (Table 2, Fig. 8).

COMPOSITION OF HYDROTHERMALLY ALTERED GRANITES AND EPISYENITES

Hydrothermal alteration of two-mica granites from the southern part of the Okrouhlá Radouň uranium deposit is characterized by a higher Fe_2O_3/FeO ratio and by a significant depletion in SiO_2 contents (Fig. 11). Episyenites typically show high contents of Na_2O and usually low contents of K_2O. The content of K_2O is higher only at the presence of higher amounts of hydromuscovite and/or illite. The later carbonatization of quartz-depleted episyenites is characterized by high contents of CaO and CO_2 (Table 3).

The origin of episyenites is, due to the dissolution of K-feldspar, connected with a depletion in rubidium and sometimes also with the evolution of a prominent negative europium anomaly (Eu/Eu* = 0.11) (Fig. 8). The higher content of HREE in episyenites (Fig. 8) is connected with the origin of uranium mineralization and a higher concentration of HREE in coffinite. Albitization of granites is in some cases accompanied by the origin of the lanthanide tetrad effect. The origin of this effect is connected with higher mobility of REE in hydrothermal fluids rich in strong complexing components (fluorine and phosphorus) (Bau, 1996; Irber, 1999). This effect in episyenites from the Okrouhlá Radouň uranium deposit was quantified by the calculation of the first and the third tetrad ($TE_{1,3}$), as proposed by Irber (1999). The calculated values of the tetrad effect in episyenites from the southern part of the Okrouhlá Radouň uranium deposit are generally low ($TE_{1,3}$ = 0.98–1.02).

MASS BALANCE CONSTRAINTS OF THE ORIGIN OF EPISYENITES

The origin of hydrothermally altered rocks is essentially connected with the chemical losses and gains produced during alteration of the parent rocks. At the presence of a significant mass or volume change accompanying the alteration, it is not possible

Table 1 Chemical analyses of two-mica granites and episyenites from the Okrouhlá Radouň uranium deposit (wt.%).

	Re-507	Re-508	Re-509	Re-510	Re-552	Re-597	Re-599	P1	P2	P3	P7	P15
SiO_2	72.50	71.04	73.04	53.81	74.61	54.92	76.89	72.29	53.56	72.45	73.14	74.05
TiO_2	0.11	0.09	0.08	0.08	0.04	0.06	0.06	0.13	0.12	0.10	0.08	0.05
Al_2O_3	15.21	13.45	15.24	18.35	13.88	15.60	10.51	15.33	16.41	14.60	15.17	14.09
Fe_2O_3	0.28	0.37	0.24	1.17	0.42	0.42	0.39	0.21	0.89	0.56	0.10	0.18
FeO	0.75	0.47	0.69	0.18	0.40	0.32	0.04	1.02	0.49	1.13	1.10	0.87
MnO	0.04	0.06	0.04	0.14	0.04	0.09	0.02	0.03	0.20	0.04	0.02	0.03
MgO	0.42	0.45	0.34	0.55	0.12	2.81	2.61	0.30	0.44	0.28	0.26	0.09
CaO	0.86	2.77	0.86	8.39	0.92	8.09	2.03	0.68	9.92	0.77	0.66	0.56
Na_2O	3.70	2.66	3.04	6.24	4.74	8.25	4.06	3.69	7.66	3.65	3.66	3.99
K_2O	5.10	5.97	5.12	1.10	4.00	1.87	2.17	4.20	0.90	4,72	4.53	3.88
P_2O_5	0.16	0.28	0.20	0.20	0.44	0.20	0.16	0.20	0.13	0.15	0.12	0.49
H_2O^+	0.66	0.39	0.71	1.67	0.37	0.75	0.79	0.90	0.50	0.40	0.50	0.48
H_2O^-	0.08	0.15	0.15	1.16	0.17	0.12	0.14	0.53	0.26	0.20	0.14	0.12
CO_2	0.17	1.76	0.14	6.49	0.13	6.39	1.30	0.10	8.00	0.14	0.08	0.05
Total	100.04	99.91	99.89	99.53	100.28	99.89	101.17	99.61	99.48	99.18	99.56	98.93
Ba (ppm)	335	354	356	n.d.	79	49	48	375	190	340	363	50
Rb (ppm)	209	153	200	55	223	9	21	168	22	197	202	231
Sr (ppm)	118	130	141	296	35	303	72	100	153	119	110	191
Zr (ppm)	58	31	37	51	n.d.	60	16	94	88	65	44	99
U (ppm)	4.1	9.1	4.4	353	14.4	44	1.3	4.1	174	5.2	7.4	18.7
Th (ppm)	4.2	2.2	2.4	6.7	2.0	4.6	1.7	7.3	1.5	4.8	5.0	1.5

Re-507 - muscovite-biotite granite, Okrouhlá Radouň, 8. level, gallery Z-8-9; Re-508 - hydrothermally altered muscovite-biotite granite, Okrouhlá Radouň, 8. level, gallery Z-8-9; Re-509 - muscovite-biotite granite, Okrouhlá Radouň, 8. level, gallery OR-3b-81; Re-510 - episyenite, Okrouhlá Radouň, 8. level, gallery OR-3b-81; Re-552 - biotite-muscovite granite, Okrouhlá Radouň, 8. level, gallery OR-3b-81, Re-597 - episyenite, Okrouhlá Radouň, 8. level, gallery OR-5-81; Re-599 - hydrothermally altered muscovite-biotite granite, Okrouhlá Radouň, 8. level, gallery OR-5-81; P1 – biotite-muscovite granite, Okrouhlá Radouň, 8. level, gallery V-8-9; P2 – episyenite, Okrouhlá Radouň, 8. level, gallery V-8-9; P3 – biotite-muscovite granite, Okrouhlá Radouň, 8. level, gallery V-8-9; P7 – biotite-muscovite granite, Okrouhlá Radouň, 8. level, gallery V-8-9; P15 – muscovite-biotite granite, Okrouhlá Radouň, 8. level, gallery Z-8-9. n.d. – not determined

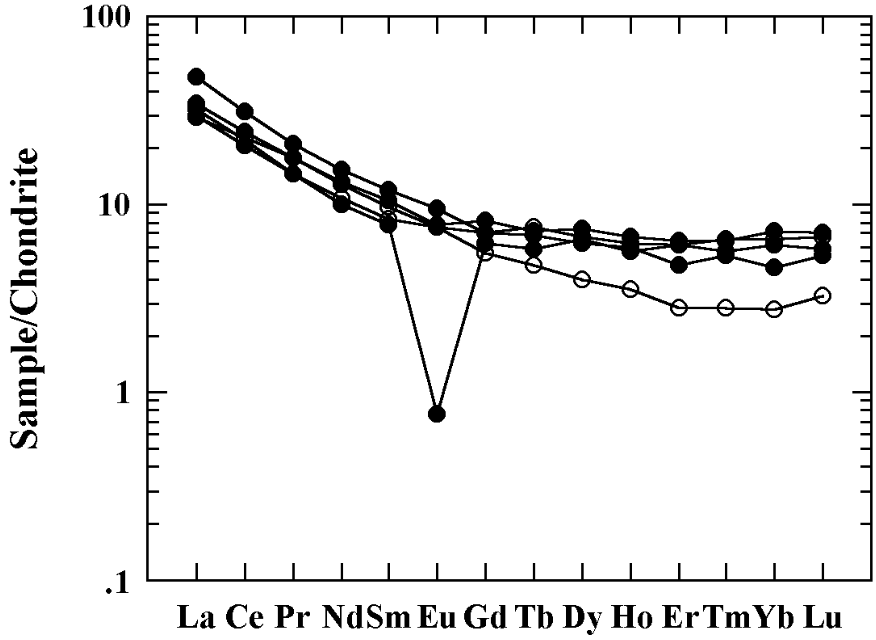

Fig. 8 Chondrite-normalized REE patterns for unaltered granites of the Deštná type and episyenites. Open circles – unaltered two-mica granites of the Deštná type, full circles – episyenites from the southern part of the Okrouhlá Radouň uranium deposit. Normalising values of the chondrite from Taylor and McLennan (1985).

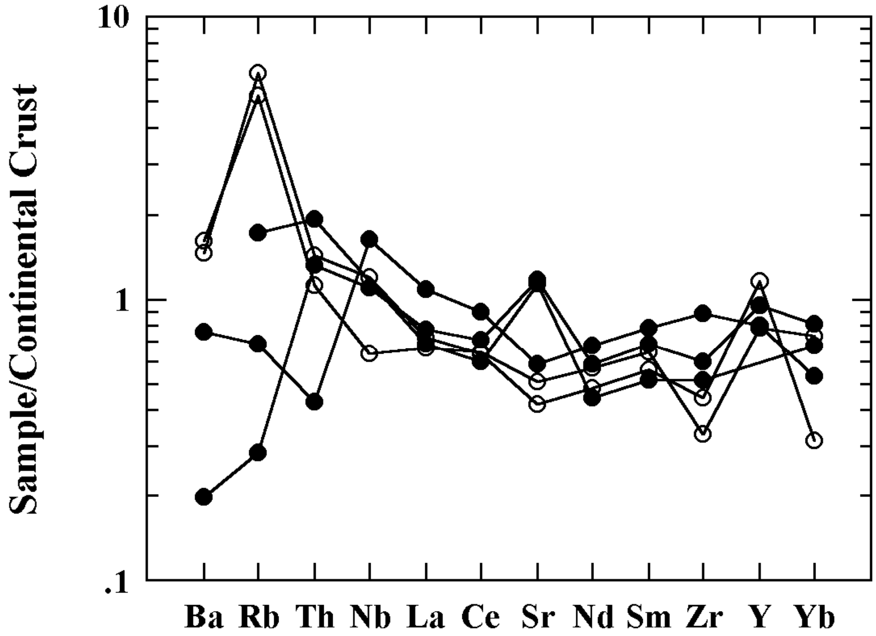

Fig. 9 Normalised trace elements patterns for unaltered granites of the Deštná type and episyenites from the southern part of the Okrouhlá Radouň uranium deposit. Normalising values of the continental crust from Taylor and McLennan (1985). Symbols see Fig. 8.

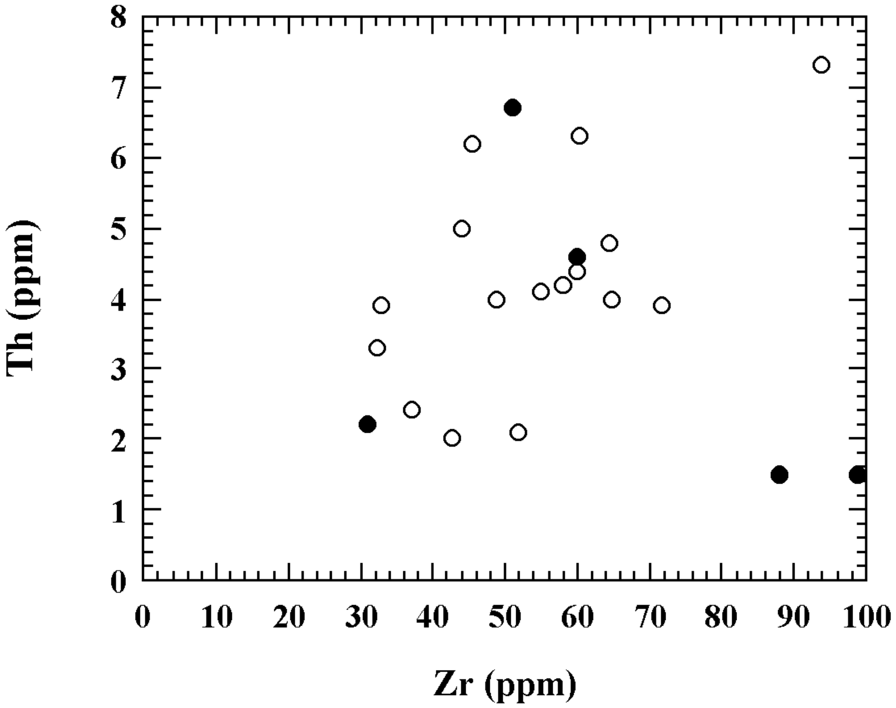

Fig. 10 Th vs. Zr diagram with plots of unaltered granites of the Deštná type and episyenites from the southern part of the Okrouhlá Radouň uranium deposit. Symbols see Fig. 8.

Table 2 Content of rare earth elements in episyenites and two-mica granite from the Okrouhlá Radouň uranium deposit (ppm).

	Re-510	Re-597	P2	P7
La	11.0	12.5	17.3	11.6
Ce	19.5	23.6	29.9	21.1
Pr	2.0	2.4	2.9	2.0
Nd	7.1	9.3	10.9	7.7
Sm	1.8	2.4	2.8	1.9
Eu	0.07	0.68	0.82	0.67
Gd	1.9	2.5	2.1	1.7
Tb	0.34	0.42	0.40	0.28
Dy	2.5	2.8	2.4	1.5
Ho	0.48	0.57	0.51	0.30
Er	1.5	1.6	1.18	0.71
Tm	0.2	0.23	0.19	0.10
Yb	1.5	1.8	1.16	0.69
Lu	0.22	0.27	0.21	0.12
La_N/Yb_N	4.96	4.69	10.05	11.38
Eu/Eu*	0.11	0.84	1.03	1.14

Re-510 – episyenite, Okrouhlá Radouň, 8. level, gallery OR-3b-81; Re-597 – episyenite, Okrouhlá Radouň, 8. level, gallery OR-5-81; P2 – episyenite, Okrouhlá Radouň, 8. level, gallery V-8-9; P7 – biotite-muscovite granite, Okrouhlá Radouň, 8. level, gallery V-8-9.

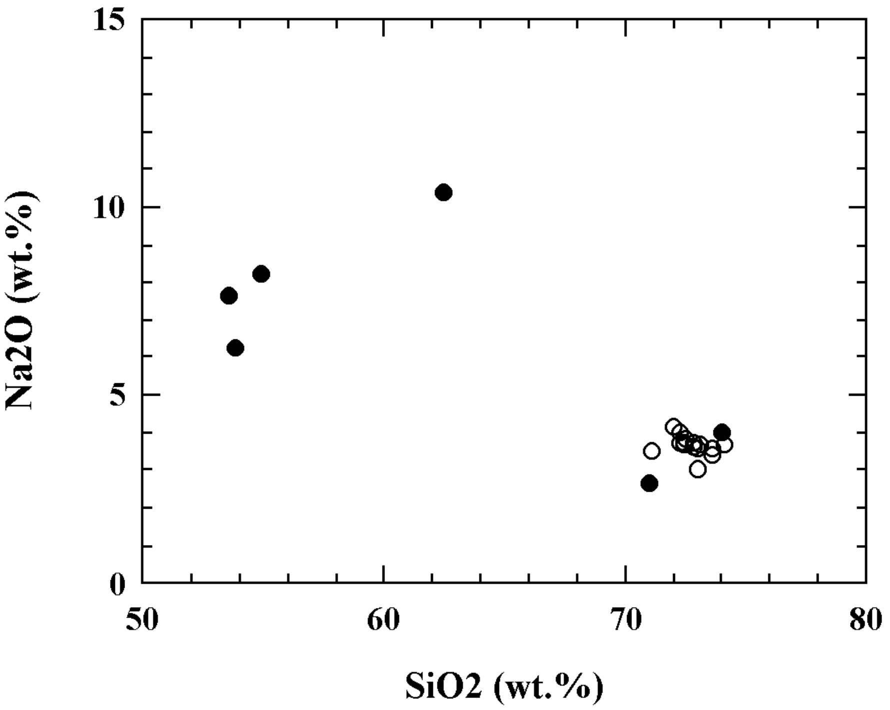

Fig. 11 SiO_2 vs. Na_2O with plots of unaltered granites of the Deštná type and episyenites from the southern part of the Okrouhlá Radouň uranium deposit. Symbols see Fig. 8.

Table 3 Chemical composition of episyenites, Okrouhlá Radouň, 8. level, gallery RV-2 (wt.%).

	OR-S-11	OR-S-12	OR-S-13	OR-S-14	OR-S-15	OR-S-16	OR-S-21
SiO_2	56.07	54.35	54.63	53.43	49.46	55.98	63.89
TiO_2	0.14	0.14	0.10	0.12	0.12	1.06	0.14
Al_2O_3	18.03	16.41	17.66	15.37	15.88	21.27	18.89
Fe_2O_3	0.59	1.13	0.43	0.71	1.31	1.43	0.57
FeO	1.20	0.28	0.73	0.82	2.48	3.45	0.73
MnO	0.15	0.10	0.09	0.16	0.19	0.05	0.06
MgO	1.00	0.20	0.40	0.38	0.94	1.35	0.57
CaO	4.88	9.75	8.63	10.50	10.88	2.51	1.54
Na_2O	3.77	8.09	7.48	7.48	6.57	3.47	9.84
K_2O	7.65	0.52	0.90	0.63	0.47	1.88	0.50
H_2O^+	2.15	1.09	1.60	1.34	2.40	5.55	0.83
P_2O_5	0.13	0.16	0.22	0.26	0.06	0.14	0.23
CO_2	4.43	6.88	6.62	8.13	8.53	1.43	1.57
Total	100.19	99.10	99.49	99.33	99.29	99.57	99.36

Table 4 Tabulated losses and gains (wt.%) for episyenites, Okrouhlá Radouň, 8. level, gallery RV-2. Volume factor (Fv) based on Al_2O_3.

	OR-S-11	OR-S-12	OR-S-13	OR-S-14	OR-S-15	OR-S-16	OR-S-21
SiO_2	-31.45	-28.64	-31.67	-26.36	-31.38	-38.08	-27.66
TiO_2	0.07	0.08	0.04	0.07	0.06	0.65	0.06
Al_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe_2O_3	0.03	0.54	-0.08	0.22	0.73	0.51	0.00
FeO	0.52	-0.16	0.17	0.34	1.77	1.85	0.14
MnO	0.08	0.04	0.03	0.10	0.13	-0.01	0.00
MgO	0.65	0.05	0.19	0.22	0.70	0.76	0.30
CaO	2.84	7.33	5.86	8.56	8.59	0.72	0.21
Na_2O	-1.84	2.10	1.14	2.01	1.00	-2.48	2.49
K_2O	1.89	-3.56	-3.29	-3.43	-3.59	-2.77	3.63
H_2O^+	1.29	0.55	0.89	0.84	1.73	3.25	0.24
P_2O_5	-0.34	-0.30	-0.27	-0.21	-0.39	-0.35	-0.27
CO_2	3.32	5.73	5.11	7.25	7.37	0.84	1.06
Sp.gr.(g/cm^3)	2.40	2.41	2.40	2.43	2.43	2.43	2.34
Fv	0.85	0.93	0.86	0.98	0.95	0.71	0.83

to directly compare chemical compositions of the altered rocks with those of the fresh rock. In his classic study of this problem, Gresens (1967) made the fundamental assumption that one or more components are immobile during alteration. Gresens' approach has been widely applied to hydrothermal deposits (Leitch and Lentz, 1994). The original method of calculation of losses and gains produced during hydrothermal alteration of two-mica granites from the southern part of the Okrouhlá Radouň uranium deposit was used for a set of chemical analyses from gallery V-2 in the central part of this deposit.

The volume factor (F_v) was determined from the ratio of relatively immobile Al_2O_3 and changes in specify gravity of unaltered and altered rocks (Table 4). The hydrothermal alteration of granites and the origin of episyenites in the southern part of the Okrouhlá Radouň uranium deposit were accompanied by significant loss of silica, usually also by a loss of potassium. The highest values of gain during episyenitization of granites are significant for the contents of calcium, CO_2 and sodium (Table 4).

DISCUSSION

The most characteristic textural feature of the altered granites and episyenites from the southern part of the Okrouhlá Radouň uranium deposit is the feldspar framework filled with younger generations of albite and several generations of carbonates. The initial formation of a vuggy reservoir rock, left from a leaching of magmatic quartz, is inferred as a part of the pre-ore stage in the hydrothermal mineralization sequence (Fig. 6). The initial quartz dissolution is combined with a complete albitization of magmatic plagioclase. The first stage of authigenic albite (albite II) is possibly simultaneous with the albitization of plagioclase. This albitization is accompanied by the origin of hematite forming fine inclusion in albitized plagioclase (albite I). The massive albitization accompanied by albitization of magmatic K-feldspar in episyenites is significant for episyenite classified as type-II by Cathelineau (1986). This type of episyenites was described by Cathelineau (1986) from Mortagne, Limousin, Escarpiére and Margeride in France and from Gérés in Portugal. The episyenitization with entire albitization of K-feldspar, significant for episyenites from the Okrouhlá Radouň uranium deposit, is relatively rare among West European uranium deposits. Most French uranium deposits in episyenitized two-mica granites are characterized by the stability of K-feldspar and/or their replacement by muscovite (Leroy, 1984; Cathelineau, 1986; Poty et al., 1986; Patrier et al., 1987). In Fichtelgebirge in the western part of the Bohemian Massif, K-feldspar is replaced by albite and/or sericite mainly at the uranium occurrences of Epprechtstein and Großschloppen. At the Hebanz uranium occurrence from the same area, K-feldspar is only partly replaced by sericite (Hecht et al., 1994). The replacement of the original magmatic K-feldspar by authigenic K-feldspar (adularia) was described from the Vendee uranium deposit in the Armorican Massif (France) by Poty et al. (1986), and from the Bernardan uranium deposit in the French Massif Central by Leroy (1984) and Patrier et al. (1987). The occurrence of authigenic K-feldspar was also reported from the Vítkov uranium deposit in the Bor pluton (western Bohemia) by Fiala and Králík (1989). The stability of K-feldspar during episyenitization is also suggested from some episyenite occurrences unrelated to uranium mineralization (Petersson and Eliasson, 1997).

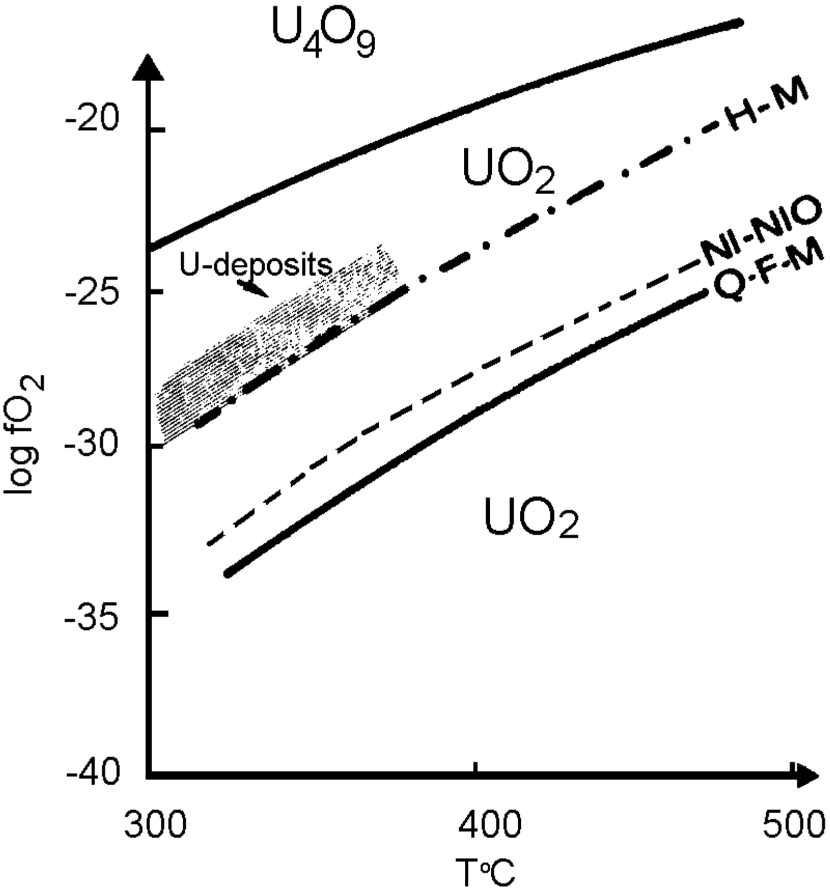

Fig. 12 Uranium oxides stability field (Nguyer Trung, 1985) and fluids associated with U deposits in the log fO_2 – temperature diagram at 1 kbar after Dubessy et al. (1987).

The well-ordered structural state of authigenic generations of albite (albite II to albite IV), as reflected by the absence of chessboard twinning, suggests that the albitization mechanism was mainly controlled by diffusion at conditions near to chemical equilibrium (see Morad et al., 1990; Slaby, 1992). Experimental synthesis of hydrothermal albite suggests that the presence of peralkaline fluids is essential for the ordering process to reach completion in authigenic albite (Martin, 1969).

Whole-rock mass balance changes expressed by the loss of Si and K and enrichment in Na, Ca and CO_2 are reconcilable with the above discussed mineralogical changes. Rb and K were lost during the albitization of K-feldspar and chloritization of biotite. Some potassium was later bonded in hydromuscovite or illite. Albitization of the original magmatic plagioclase was connected with the origin of hematite, which forms fine inclusions in albite I. The presence of hematite and whole alteration of magmatic ilmenite on hydrated Ti-oxides (leucoxene?) suggests a high fO_2 during the pre-ore stage of hydrothermal alteration. The stability of hematite within the pre-ore stage of hydrothermal alteration indicates that fO_2 was probably significantly higher than that fixed by the hematite-magnetite oxygen buffer (Fig. 12). A similar value of fO_2 is also characteristic for episyenites from the Western European uranium deposits (Cathelineau, 1987; Dubessy et al., 1987). The lack of muscovite in the presence of quartz, and the origin of authigenic albite indicate slightly basic pH (Montaya and Hemley, 1975). In neutral pH and oxidized conditions, the most stable phases are uranyl carbonate complexes and/or uranyl phosphate complexes (Sergeyeva et al., 1972; Nguyen-Trung, 1985). The significance of uranyl phosphate complexes for the origin of uranium mineralization at the Okrouhlá Radouň uranium deposit is supported by the presence of authigenic apatite in this mineralization stage (Fig. 6). During post-ore stages, remobilization of uranium mineralization was probably only very scarce (Anderson et al., 1989). The origin of uranium-bearing hydrothermal fluids is partly unknown; significantly meteoritic source of these fluids was recognized with help of oxygen isotopic ratios for the post-ore carbonate stage (Fiala et al., 1978).

The origin of episyenites at the Okrouhlá Radouň uranium deposit was accompanied by changes in the distribution of REE. Similar changes in REE distribution were described from either mineralized episyenites or episyenites not related to uranium mineralization (Cathelineau, 1987; Petersson and

Eliasson, 1997; Hecht et al., 1999). The REE may remain almost immobile during the origin of episyenites (Cathelineau, 1987; Leroy and Turpin, 1988; Dempsey et al., 1990; Hecht et al., 1994). If quartz leaching is associated with alkali metasomatism, especially albitization, moderate to strong mobility and fractionation of REE is observed (Taylor et al., 1981; Chatterjee and Strong, 1984; Cathelineau, 1987; Charoy and Pollard, 1989; Petersson and Eliasson, 1997). The depletion in LREE is most likely related to the dissolution of monazite and apatite. The HREE enrichment in episyenites can be explained by the formation of HREE-bearing uranium minerals (coffinite, uraninite; Cathelineau, 1987; Fayek and Kyser, 1997). The depletion of hydrothermal fluids in europium and the origin of negative europium anomaly may be connected with carbonate-fluid complexing (Lottermoser, 1992). Compared with the calculated values of the tetrad effect in REE chondrite-normalized pattern (Fig. 8) from episyenitized granites in the northern part of the Bohemian Massif ($TE_{1,3}$ = 0.97–1.11, Hecht et al., 1999), data for episyenitized granites from the Okrouhlá Radouň uranium deposit are very similar ($TE_{1,3}$ = 0.98–1.02). For comparison, calculated values of the lanthanide tetrad effect from highly fractionated topaz-albite granites associated with Sn-W mineralization in the Krušné Hory/Erzgebirge Mts. are significantly higher ($TE_{1,3}$ = 1.2–1.5; Irber, 1999). These differences can be probably explained by higher concentrations of complexing fluids (fluorine-bearing complexes) responsible for the albitization of the topaz-albite granites.

The origin of hydrothermal mineralization connected with the formation of episyenites in two-mica granites of the Okrouhlá Radouň uranium deposit was a relatively long-lasting process. The main uranium-ore stage characterized by the formation of coffinite and pitchblende was dated by the U/Pb method (Anderson et al., 1989). The age of pitchblende from this stage is 255±3 Ma. This age corresponds with U/Pb dating of the Variscan uranium mineralization from other hydrothermal uranium deposits of the Bohemian Massif (Anderson et al., 1988; Hein et al., 2002; Křibek and Hájek, 2005).

CONCLUSION

The results of the mineralogical and geochemical study of episyenized two-mica granites in the southern part of the Okrouhlá Radouň uranium deposit can be summarized as follows. The initial stage of hydrothermal alteration associated with significant quartz leaching left a vuggy, feldspathic framework. The enrichment in Na_2O with concomitant leaching of Rb is related to albitization of magmatic plagioclase and K-feldspar accompanied by the precipitation of authigenic albite in the presence of peralkaline fluids. The pre-ore hydrothermal alteration stage also involved the decomposition of the original igneous biotite and ilmenite. This alteration is connected with an oxidation of ferrous iron into ferric iron, mostly incorporated in hematite. Paragenetic relationships suggest that the fO_2 of the paleofluids was above the hematite-magnetite equilibrium. The ore stage was marked by the formation of coffinite and pitchblende, together with small amount of authigenic chlorite, hydrated titanium oxide (leucoxene?) and pyrite. A slight but marked HREE supply is noted during the episyenitization event. The post-ore mineralization stage is connected with a huge formation of carbonates represented by several generations of calcite. A significant part of the uranium mineralization was formed during the Permian extensional phase of the late Variscan evolution of the Bohemian Massif.

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