

## CONTRASTING ORIGINS OF THE MIXED (NYF + LCT) SIGNATURE IN GRANITIC PEGMATITES, WITH EXAMPLES FROM THE MOLDANUBIAN ZONE, CZECH REPUBLIC

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### ABSTRACT

Two granitic pegmatites with mixed (NYF + LCT) signatures from the Moldanubian Zone, Czech Republic, were examined to determine their origin. On the basis of geological setting, paragenesis, geochemical modeling and chemical composition of rocks and minerals, two contrasting modes of origin were discerned. The Li-bearing (lithium micas + elbaite) Kracovice pegmatite, with “amazonite”, samarskite and fergusonite, is the product of the strong fractionation of pegmatite-forming melt derived from the orogenic I-type Třebíč pluton. Geological, petrological, compositional and isotopic data from this ultrapotassic melasyenite–melagranite point to mixing of a mantle-derived magma with (leuco)granitic melt from crustal rocks. The granite–pegmatite system of the Třebíč pluton and the mainly Li-bearing Kracovice pegmatite are typical examples of the mixed (NYF + LCT) petrogenetic family. The contaminated elbaite-subtype pegmatite Bližná I, Český Krumlov Unit, with lithium tourmalines (elbaite, liddicoatite, uvite), primary REE minerals [bastnäsite-(Ce), parisite-(Ce), allanite-(Ce)] and Ca(Mg)-rich minerals (diopside, uvite, titanite), gained its mixed signature through a unique pre-emplacment process involving the external contamination with Ca, Mg and REE of an evolved (LCT) pegmatite-forming melt by distal carbonatite-like marbles with an NYF signature. Despite the clearly mixed-signature mineralogy and geochemistry, the Bližná I pegmatite represents a special type of contaminated LCT pegmatite rather than a member of the mixed family.

*Keywords:* Li-bearing pegmatites, mixed (NYF + LCT) signature, origin, contamination, mixing test, Moldanubian Zone, Czech Republic.

### INTRODUCTION

The current classification of granitic pegmatites includes two main petrogenetic families, LCT (enriched in lithium + cesium + tantalum) and NYF (enriched in niobium + yttrium + fluorine) (Černý 1991a, Černý & Ercit 2005, Ercit 2005, London 2008); however, some pegmatites share mixed (NYF + LCT) geochemical and mineralogical characteristics, *e.g.*, Høydalen near Tørdal, Telemark, Norway, and the O’Grady Batholith, Selwyn plutonic suite, N.W.T., Canada (Černý & Ercit 2005). These pegmatites contain minor to accessory Li-bearing minerals (lithium micas, elbaite) along with

various accessory Y,REE minerals [*e.g.*, (Nb,Ta,Ti) oxides, silicates] and they are assigned to a mixed family. The following possibilities have been considered for the genesis of the mixed granite–pegmatite systems (Černý 1991b, Černý & Ercit 2005): “(i) a pristine NYF magma from depleted crust may become contaminated by digestion of undepleted supracrustal lithologies, (ii) the crustal protolith may have been only partially depleted, and (iii) the anatexis may have affected a mixed range of depleted and undepleted protoliths”. The spectrum of genetic possibilities may considerably expand once various models of NYF-granite derivation and diverse potential modes of LCT enrichment

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or external contaminations of the pegmatite-forming melt are considered (Černý & Ercit 2005, Martin & De Vito 2005).

The two pegmatites are characterized by the presence of typical LCT-signature minerals (lithium tourmalines, lithium micas) and typical NYF-signature minerals [allanite-(Ce), Y, oxides of REE, Nb, Ta and Ti, primary Y,REE fluorcarbonates]. They occur in the Moldanubian Zone, Czech Republic. The lithium-bearing pegmatite (lithium micas + elbaite) from Kracovice, with Y,REE minerals, is related to the Třebíč pluton (Němec 1990, Novák *et al.* 1999a, Škoda *et al.* 2006); the contaminated elbaite-subtype pegmatite Bližná I (Novák *et al.* 1997, 1999b) contains common primary REE minerals. To reveal the factors controlling the origin of their mixed signatures, we describe (i) the geological setting of the pegmatites, including geochemistry of parental granites and host-rock complexes, (ii) results of a paragenetic study, (iii) the chemical composition of rocks and minerals, and (iv) mixing tests based on the whole-rock chemical data.

#### GRANITIC PEGMATITES OF THE MOLDAUBIAN ZONE AND A SUMMARY OF THE GEOLOGICAL SETTING OF THE REGIONS EXAMINED

The Moldanubian pegmatite field is characterized by numerous granitic pegmatites of different origin and mineralogy (Černý & Ercit 2005, Novák 2005, Breiter *et al.* 2010; Fig. 1). Pegmatites of the rare-element class are the most abundant and exhibit a high variability in size, textural differentiation, degree of fractionation and mineralogy, from barren to highly fractionated pegmatites with both LCT and NYF signatures. Most pegmatites of LCT signature are typically enriched in B with tourmaline (dravite – schorl – foitite – elbaite – rossmanite – liddicoatite; Novák & Povondra 1995, Selway *et al.* 1998, 1999, Novák *et al.* 1999b, 2004) as an omnipresent accessory to minor mineral. Both lepidolite- and elbaite-subtype pegmatites are typical (Fig. 1). Pegmatites of the NYF family, varying from primitive allanite-type to more evolved pegmatites of euxenite type, occur exclusively in the Třebíč and

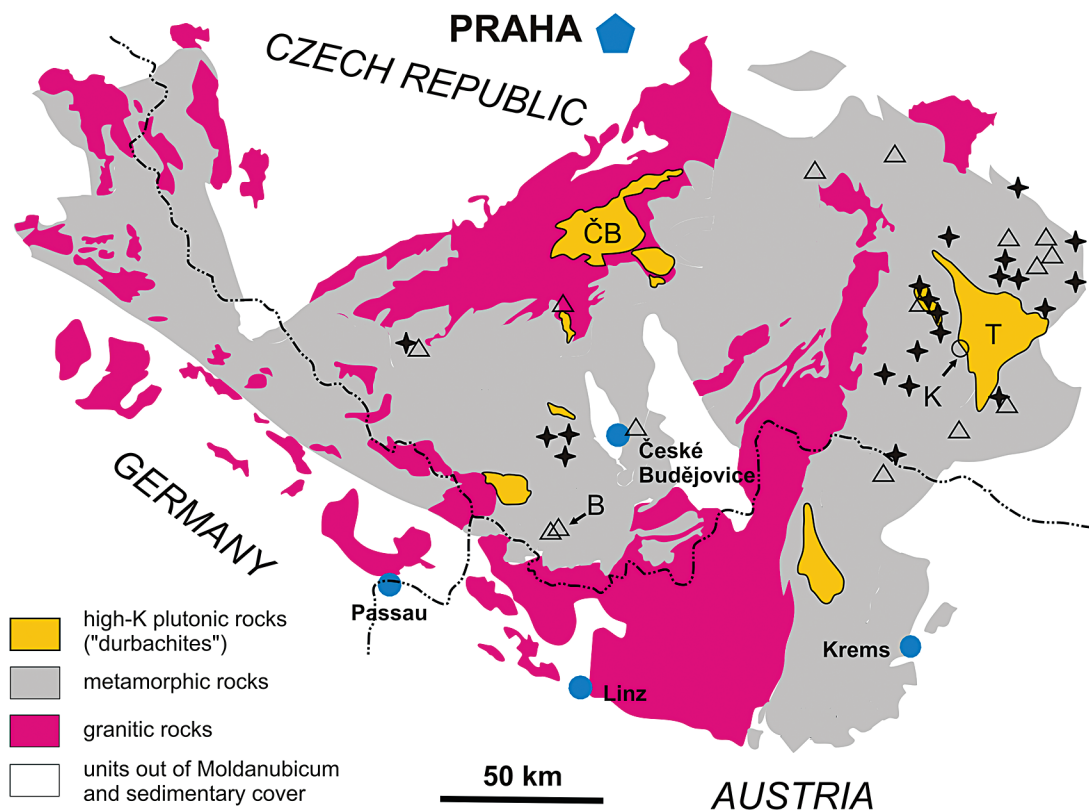


FIG. 1. Schematic geological map of the Moldanubian Zone showing the major occurrences of complex Li-bearing pegmatites (Breiter *et al.* 2010). Stars: lepidolite-subtype pegmatites, triangles: elbaite-subtype pegmatites, Bližná I (B), circle: Kracovice (K) pegmatite. ČB: the Čertovo břemeno pluton, T: the Třebíč pluton.

Čertovo břemeno syenite plutons (Fig. 1) and also contain accessory tourmaline (dravite – schorl – uvite; Novák *et al.* 2011). The pegmatites of the rare-element class (both LCT and NYF) crystallized in a short period of ~338–333 Ma (Novák *et al.* 1998, Ertl *et al.* 2004) and closely followed an episode of high-grade metamorphism. The NYF pegmatites are accompanied by contemporaneous mafic calc-alkaline (orogenic lamprophyres) to peralkaline (orogenic lamproites) dike intrusions derived from the different parts of the metasomatized mantle (Krmíček 2010, Krmíček *et al.* 2011).

Porphyrific, amphibole–biotite melasyenite to quartz melasyenite and melagranite (durbachite) of the Třebíč pluton are emplaced in medium- to high-grade metamorphic rocks of the Moldanubian Zone at an upper-crust level at  $P \approx 2\text{--}4$  kbar (Novák & Houzar 1996, Houzar & Novák 2006) and at  $334.8 \pm 3.2$  Ma (Kotková *et al.* 2010). Its bulk composition is metaluminous (ASI in the range 0.85–0.93), with high concentrations of  $K_2O$  (5.2–6.5 wt.%),  $MgO$  (3.3–10.4%),  $P_2O_5$  (0.47–0.98%), Rb (330–410 ppm), Ba (1100–2470 ppm), U (6.7–26.2 ppm), Th (28.2–47.7 ppm), Cr (270–650 ppm), Cs (20–40 ppm), and high K/Rb = 133–171, but with unusually low CaO and Sr contents, and a high Rb/Sr value, 0.8–1.3 (for more details, see Holub *et al.* 1997, Janoušek *et al.* 2000). The geochemical signature as well as isotopic Sr ( $^{87}Sr/^{86}Sr_{337}$  in the range 0.709–0.7125) and Nd ( $\epsilon Nd_{337} = -6.3$ ) data require an origin from anomalously enriched mantle domains (Holub 1997, Wenzel *et al.* 1997, Parat *et al.* 2010) perhaps contaminated by the deeply subducted mature crust (Janoušek & Holub 2007, Lexa *et al.* 2011). Subsequently, these magmas were subjected to hybridization by crustally derived, leucogranitic melts (Holub 1997).

The pegmatite district Černá v Pošumaví, including the more or less contaminated elbaite-subtype pegmatites Bližná I and Bližná II, is located in the southern part of the Český Krumlov Unit, Moldanubian Zone, Czech Republic (Novák *et al.* 1997, Černý 2004, Novák 2005, Fig. 1; for details of the regional geology, see Dallmeyer *et al.* 1995). In analogy with rare-element pegmatites elsewhere in the Moldanubian Zone (Ackermann *et al.* 2007, Breiter *et al.* 2010, Gadas *et al.* 2012), the pegmatite-forming melts intruded relatively cold rocks at T in the range 300–450°C and  $P \approx 2\text{--}4$  kbar, as indicated by common primary petalite at several South Bohemian pegmatites (Fig. 1). The area features medium- to high-grade plagioclase–biotite paragneisses with common intercalations of quartzitic paragneisses, quartzites, calc-silicate rocks, amphibolites, abundant metacarbonates and closely associated graphite beds. Small bodies of leucocratic granites, locally with nodular tourmaline and with thin tourmaline-rich veinlets of pegmatite, cut this rock complex ~1.5 km northwest of the Bližná pegmatite bodies; however, the parental granite of the Bližná pegmatites is not known. Two distinct types of metacarbonates were distinguished

in this region (Drábek *et al.* 1999, Houzar & Novák 2002). (i) Abundant calcite to calcite–dolomite marbles locally with common silicates form large nodular to tabular bodies, up to 30 m thick and 1 km long. (ii) Less common carbonatite-like calcite–dolomite marbles have been found as relatively thin layers, up to 3–4 m thick (Drábek *et al.* 1999, Houzar & Novák 2002). They are interpreted as metacarbonates from a shallow marine environment containing a considerable proportion of volcanoclastic material derived from alkaline volcanism (Drábek *et al.* 1999). The amphibolite-facies metamorphism of carbonatite-like marbles was dated on molybdenite using the Re–Os method at ~495 Ma (Drábek & Stein 2003). Both types of metacarbonate rocks form bodies concordant to the volcanosedimentary sequence and characterized by a highly heterogeneous chemical and mineralogical composition.

#### METHODS AND SAMPLES

The chemical compositions of minerals were obtained on the Cameca SX 100 at the Joint Laboratory of Electron Microscopy and Microanalysis, Department of Geological Sciences, Masaryk University, Brno and the Czech Geological Survey. Analytical conditions used were: accelerating voltage 15 kV, and beam current 30 nA. As standards, we used synthetic minerals (for more details, see Škoda & Novák 2007, Novák *et al.* 2011). A few electron-microprobe analyses of tourmaline (Rečice, Ctidružice, part of Bližná and Kracovice) and micas (Kracovice) were done using the Cameca Camebax SX–50 instrument, University of Manitoba, Winnipeg, with a beam diameter of 4–5  $\mu m$  and an accelerating potential of 15 kV (for more details see Selway *et al.* 1999).

Bulk chemical analyses of rocks were carried out at the Acme Chemical Laboratories Ltd, Vancouver, Canada. Concentrations of the major elements were established by ICP–ES after fusion with lithium borate flux; concentrations of the trace elements, including the REE, were established by ICP–MS with lithium tetraborate fusion.

We used a graphical expression of the general mixing equation of Langmuir *et al.* (1978) for the Bližná I pegmatite:  $C_M = X_A \times C_A + (1 - X_A) \times C_B$ , where  $C_M$  is the concentration of an element in the contaminated melt,  $C_A$  is the concentration of the same element in the pegmatitic component,  $C_B$  is the concentration of the same element in the contaminant component, and  $X_A$  corresponds to the fraction of the pegmatitic component participating in mixing and contamination (0–1). The mixing equation can be rewritten as:  $X_A = (C_M - C_B) / (C_A - C_B)$ . If mixing occurs, each element considered should define a straight line in the  $(C_M - C_B)$  versus  $(C_A - C_B)$  diagram. Subsequently, the slope of the line gives the mass proportions of the mixture (Fourcade & Allègre 1981).

Samples of rocks and minerals from the Třebíč pluton and related pegmatites were collected on outcrops and sporadically from fragments on fields (some pegmatites). Pegmatites and both types of metacarbonate rocks from Bližná were accessible in the abandoned graphite mine Václav in 1991–1997 (Bližná I pegmatite and host marbles) and in 1994–2007 (Bližná II pegmatite and carbonatite-like marbles), respectively; currently, both underground outcrops of the pegmatites are not accessible. The samples ~0.5–1.0 kg of the individual units of the granitic pegmatite Bližná I (BLAD, BLA, BLB) and ~3 kg of metacarbonate rocks, respectively, were crushed for whole-rock chemical analysis. The samples of ordinary marble (BLM–1, BLM–2, BLM–3) were sampled invariably at a distance of 0.2, 0.5 and 1 m from the contact with Bližná I pegmatite; the samples of carbonatite-like marble (BLK–1, BLK–2, BLK–3) come from an outcrop located ~200 m northwest from the Bližná I pegmatite.

#### GRANITIC PEGMATITES RELATED TO THE TŘEBÍČ PLUTON

The Třebíč pluton (Fig. 2) is characterized by abundant intragranitic NYF pegmatites with common biotite and tourmaline of two distinct varieties (Škoda *et al.*

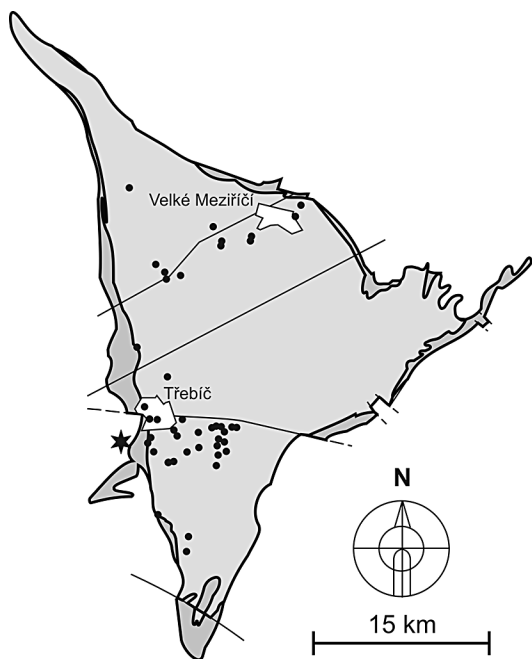


FIG. 2. Schematic geological map of the Třebíč pluton and pegmatite districts (slightly modified from Škoda *et al.* 2006). Light grey: melagranite to melasyenite; dark grey: leucocratic granites locally with tourmaline; dots: pegmatite occurrences; Kracovice: star.

2006, Škoda & Novák 2007, Novák *et al.* 2011): (i) subhomogeneous allanite-type pegmatites form small irregular nests and segregations, commonly with a transitional contact to the host syenogranite; (ii) lenses, dikes and irregular bodies of simply zoned euxenite-type pegmatites (with aeschynite- or euxenite-group minerals; Škoda & Novák 2007) are up to 2 m thick and several meters long. The euxenite-type Klučov I pegmatite is characterized by a higher degree of fractionation, manifested by more abundant B-bearing minerals and the chemical composition of the tourmaline (Novák *et al.* 2011). The symmetrically zoned northwest-trending pegmatite dike in Kracovice, ~1 m thick and 30 m long, cuts graphitic gneiss ~300 m west of the edge of the Třebíč pluton (Figs. 1, 2; Němec 1990, Škoda *et al.* 2006). In its most differentiated part, the pegmatite consists from outer to inner units of: a coarse-grained granitic unit (Kfs + Pl + Qtz + Bt + Ms), an abundant graphic unit (Kfs + Qtz ± Bt), evolving to minor blocky K-feldspar, and an albite complex situated close to a small quartz core. Major, minor and accessory minerals typical of the individual types of pegmatites are given in Table 1.

The most primitive allanite-type pegmatites contain, along with major quartz, K-feldspar, plagioclase and biotite, several accessory minerals including tourmaline (Table 1). Euxenite-type pegmatites are characterized by presence of Y,REE oxide minerals and several accessory minerals (Table 1); some pegmatites contain beryl. The most evolved euxenite-type pegmatite Klučov I is slightly B-, Mn- and Sn-enriched, and the Kracovice pegmatite contains Li-bearing minerals, muscovite and several accessory minerals. An increasing degree of fractionation from allanite-type pegmatites to the Kracovice pegmatite is evident in the mineral assemblages (Table 1).

Tourmaline-super group minerals are common accessories in all pegmatites; hence, their compositional evolution is a useful tool to reveal the fractionation trend from primitive allanite-type pegmatites through euxenite-type pegmatites, including the more evolved Klučov I pegmatite to the Li-bearing Kracovice pegmatite. Black tourmaline (dravite – schorl – uvite) generally shows two trends distinct for the allanite- with euxenite-type pegmatites and for the euxenite-type pegmatite Klučov I (Fig. 3). Tourmaline from Kracovice (Al-rich schorl to Mn-rich elbaite) shows very limited variation in <sup>IV</sup>Al, and moderate to high contents of Mn and F, all increasing from schorl to elbaite. All diagrams in Figure 3 illustrate the compositional evolution from intragranitic, less evolved allanite- and euxenite-type pegmatites through the Klučov I pegmatite to the distal Li-bearing Kracovice pegmatite.

The compositional trend in micas [only biotite with moderate variation in Fe/(Fe+Mg) is known in all allanite- and euxenite-type pegmatites] to more complex assemblage in Kracovice with biotite, zinnwaldite and masutomilite + Mn-rich polyolithionite (Novák *et al.*

1999a) yielded high Mn/(Fe+Mn) values,  $\leq 0.87$ . The micas also exhibit high F contents, increasing with the progress of fractionation just as in tourmaline. The concentrations of Rb and Cs and the Cs/Rb values in the lithium micas from Kracovice are similar to those from complex LCT pegmatites in the Moldanubicum (Černý *et al.* 1995, Novák & Povondra 1995, Novák & Černý 1998a, Novák *et al.* 1999c). The primary Y,REE minerals involve common allanite-(Ce) in all intragranitic pegmatites, common  $AB_2O_6$  minerals (aeschnite-group and euxenite-group minerals), and very rare monazite-(Ce) in euxenite-type pegmatites (for details see Škoda & Novák 2007). The rare  $ABO_4$  oxide minerals (fergusonite, samarskite) and monazite-(Ce) + xenotime-(Y) were identified in the Kracovice pegmatite. Compositional trends in tourmalines and (Y, REE, Nb, Ta, Ti)-rich oxide minerals (Figs. 3, 4) as well as overall mineral assemblages (Table 1) corroborate the genetic relationship of all pegmatites including the

Kracovice pegmatite to the single granite–pegmatite system of the Třebíč pluton.

CONTAMINATED ELBAITE-SUBTYPE  
PEGMATITES FROM BLIŽNÁ AND ASSOCIATED  
METACARBONATE ROCKS

*The metacarbonate rocks*

Two distinct types of metacarbonates include (i) abundant calcite to calcite–dolomite marbles and (ii) less common carbonatite-like calcite–dolomite marbles. (i) Ordinary metacarbonate rocks are texturally and compositionally heterogeneous, banded, fine- to medium-grained and locally rich in silicates (diopside, tremolite, phlogopite > quartz, feldspars, scapolite, muscovite, forsterite, spinel, tourmaline). (ii) Less common carbonatite-like calcite–dolomite marbles also are characterized by a highly heterogeneous chemical and mineralogical composition (Table 2). They are locally layered and silicate-rich (diopside, tremolite, phlogopite > quartz, forsterite), with sulfide-rich portions (pyrrhotite, pyrite, molybdenite, chalcopyrite, galena, sphalerite). Locally common accessory REE-bearing minerals (Th,REE-rich betafite, euxenite, uranothorite), barite, ilmenite and zircon (Drábek *et al.* 1999) are rather randomly distributed. Ordinary marbles from outcrops near the Bližná I pegmatite are enriched in silicates; however, almost silicate-free metacarbonates also are common in this region (Houzar & Novák 2002). Comparing ordinary marbles and carbonatite-like rocks, the latter exhibits higher Si/Al and Na/K values, and higher contents of Fe and Mn (Table 2). The carbonatite-like marbles are characterized by variable but locally high concentrations of Ba, Sr, Mo, Pb, Cu, Y, Zr, Nb and REE (see Table 2). The two types of marble also show distinct REE patterns (Fig. 5).

*Geology, internal structure and mineralogy of the Bližná I and Bližná II pegmatites*

The Bližná I pegmatite dike, ~4 m thick and several tens of meters long, generally northeast-trending and dipping ~35° to the southeast, cuts silicate-rich ordinary calcite–dolomite marble (Fig. 6). The dike is enclosed in the rock sequence overlying a graphite bed, and the closest known layer of the carbonatite-like marble is at a distance of ~200 m to the northwest. The contacts with the host marble are typically sharp except for a tourmaline-rich exocontact zone described in detail by Novák *et al.* (1999b), limited exclusively on the foot-wall margin of the pegmatite (Fig. 6).

The textural-paragenetic units include a volumetrically predominant, commonly coarse-grained schorl unit (BLA) (Fig. 6) built of grey to bluish grey perthitic microcline, subordinate quartz, albite<sub>An4-0</sub> and minor black to brown-black Mg-bearing schorl to Al-rich schorl. This unit contains randomly distributed

TABLE 1. PRIMARY MINERALS FROM THE INDIVIDUAL TYPES OF PEGMATITES IN THE TŘEBÍČ PLUTON

	allanite-type pegmatites	euxenite-type pegmatites	Klučov I	Kracovice
quartz	+++	+++	+++	+++
K-feldspar	+++	+++	+++	+++
amazonitic Ksp		++	++	++
plagioclase $\leq An_{18}$	+++	+++	+++	+++
albite		+++	+++	+++
dark micas	+++	+++	+++	+++
muscovite				++
lithium micas				+++
garnet				++
topaz				++
actinolite	++	+		
dravite	++	++	+	
schorl	+	++	+++	+++
elbaite				++
ilmenite	++	++	++	
pseudotitile		+	+	
titanite	++	++	+	
rutile		+	+	+
niobian rutile		+	+	++
columbite-(Fe)		+		+
"wolframioxiolite"				+
aeschnite group		++	++	
euxenite group		++	++	
samarskite group				++
fergusonite-(Y)				+
scheelite				+
allanite-(Ce)	++	++	++	
monazite-(Ce)	+	+		++
xenotime-(Y)				+
zircon	+	++	++	++
beryl		+		+
bazzite		+		+
phenakite		+		+
hambergite				++
herzenbergite			+	
cassiterite			+	+
tinzenite			+	
fluorite		+		+
pyrite	+	+	+	+

Abundance of minerals: +++: major and minor minerals, ++: common accessory minerals, +: rare accessory minerals.

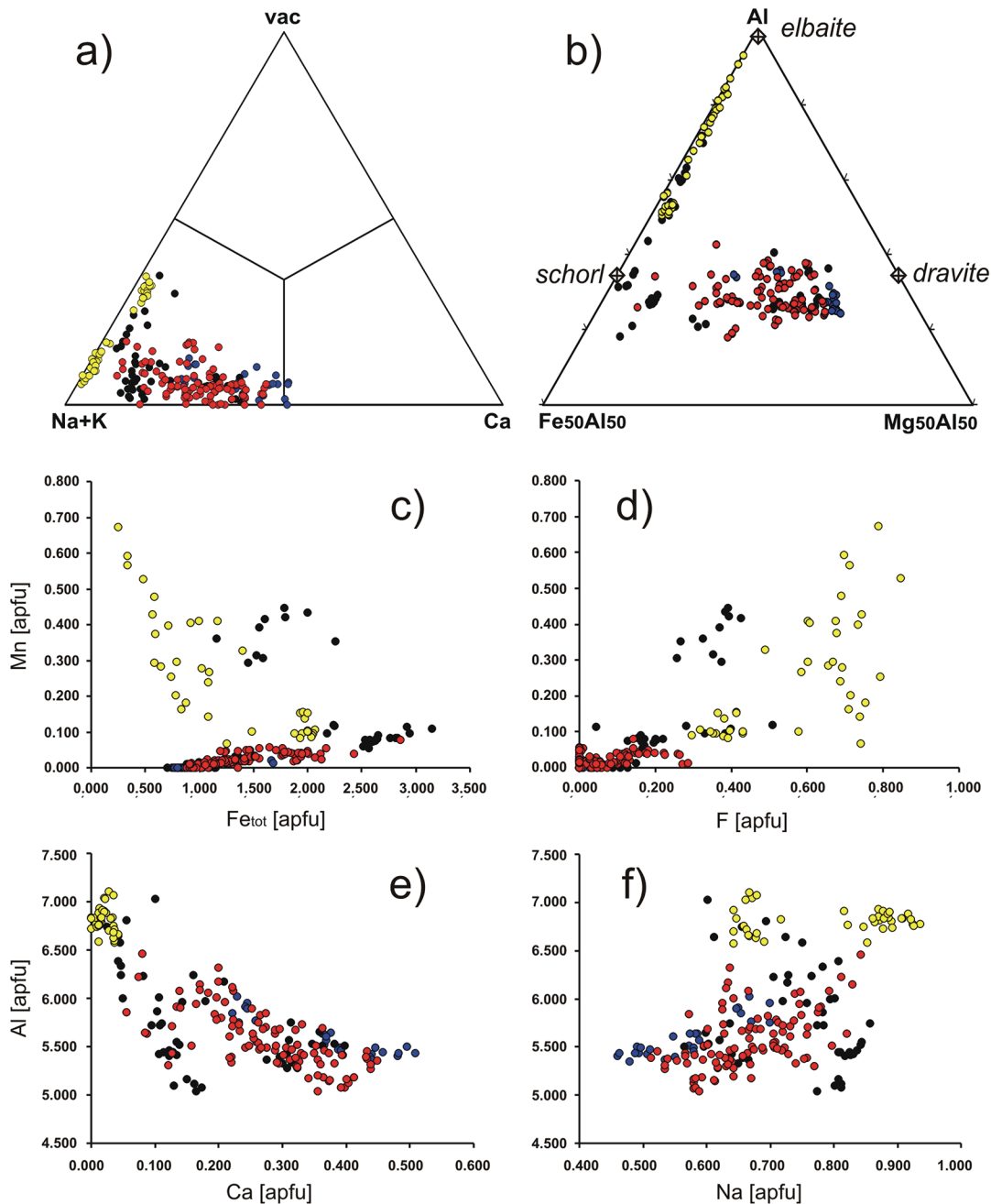


FIG. 3. Composition of tourmaline from allanite-type pegmatites (blue symbol), euxenite-type pegmatites (red symbol), the Klučov I pegmatite (black symbol), the Kracovice pegmatite (yellow symbol): a) (Na+K) – Ca – X-site vacancy plot; b) Fe<sub>tot</sub>–Mg–Al triangle; c) Fe<sub>tot</sub> versus Mn; d) F versus Mn; e) Ca versus Al; f) Na versus Al. (in part modified from Novák *et al.* 2011).

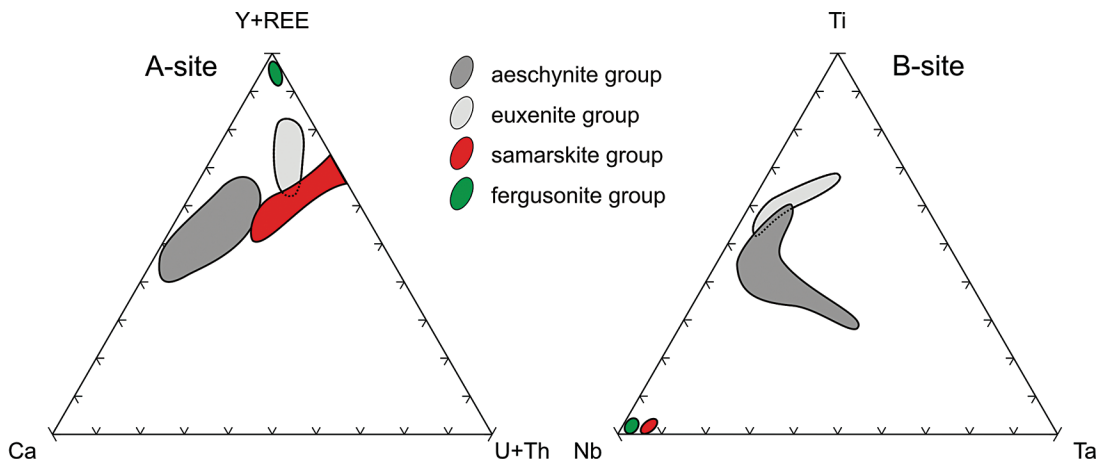


FIG. 4. Chemical composition of aeschnite-, euxenite-, samarskite-group minerals and fergusonite. (A-site and B-site occupancy). (in part modified from Škoda & Novák 2007).

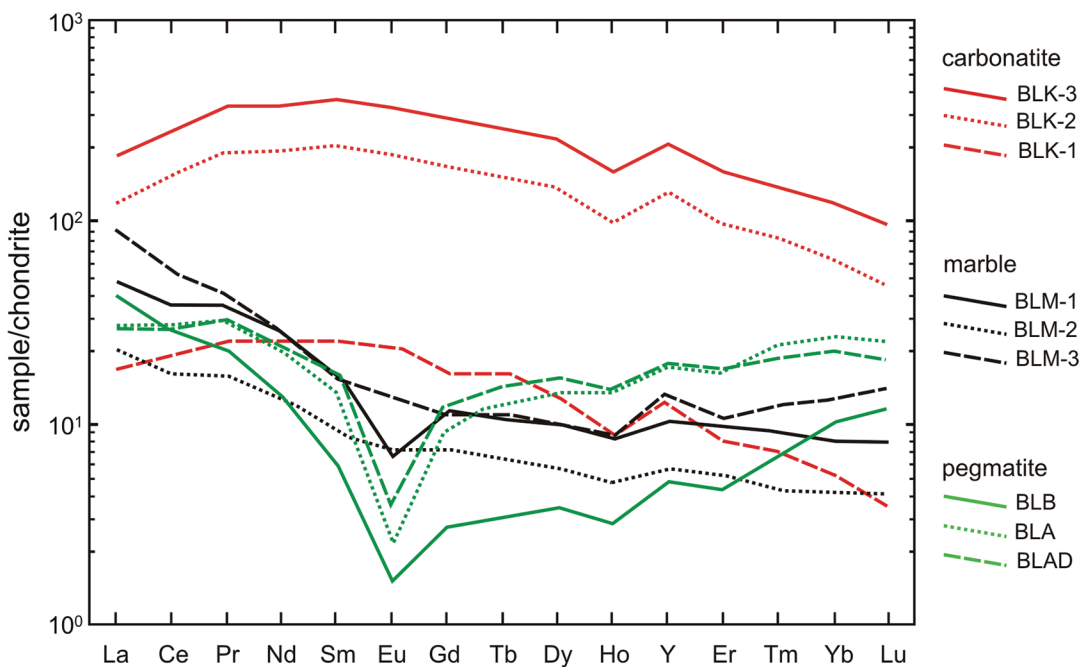


FIG. 5. Chondrite-normalized REE patterns of the rocks from the Bližná area. Chondrite data from Taylor & McLennan (1985).

diopside-bearing nests (BLAD), ~20 cm in diameter (Fig. 6), composed of feldspars, quartz, subhedral, elongate grains of white diopside, up to 3 cm in long. The schorl unit evolves to the elbaite–liddicoatite unit (ELU) as nests, up to ~30 cm in diameter (Fig. 6). The grain

size increases from ~1–3 cm in the schorl unit to ~10 cm in the elbaite–liddicoatite unit. Blocky K-feldspar, quartz and graphic textures (K-feldspar + quartz) and brown-yellow-pink tourmaline are locally present, as well as exceptional small fragments of massive graphite.

TABLE 2. CHEMICAL COMPOSITION OF ROCKS IN BLIŽNÁ

	BLAD	BLA	BLB	BLM-1	BLM-2	BLM-3	BLK-1	BLK-2	BLK-3	extreme values
SiO <sub>2</sub> wt%	69.16	75.93	63.39	42.74	33.29	35.80	21.14	13.74	12.96	
TiO <sub>2</sub>	0.28	0.27	0.08	0.39	0.14	0.15	0.04	0.05	0.05	
Al <sub>2</sub> O <sub>3</sub>	13.67	13.29	18.38	8.14	2.75	1.82	0.42	0.44	0.29	
Fe <sub>2</sub> O <sub>3</sub> (tot)	0.25	0.10	0.15	4.12	1.99	1.45	7.59	3.05	3.36	
Cr <sub>2</sub> O <sub>3</sub>	0	0	0	0	0	0	0	0	0	
CaO	5.87	3.56	4.71	24.22	34.01	35.16	32.37	39.10	44.42	
MgO	2.34	0.31	0.50	10.90	4.83	9.58	17.53	11.54	5.38	
MnO	0.27	0.04	0.07	0.70	0.07	0.13	0.24	0.35	0.43	
Na <sub>2</sub> O	2.29	3.07	7.53	0.28	0.22	0.35	0.65	0.82	0.58	
K <sub>2</sub> O	4.12	2.34	1.12	2.10	1.09	0.19	0.07	0.20	0.07	
P <sub>2</sub> O <sub>5</sub>	0.04	0.03	0.20	0.80	0.15	0.06	0.05	0.03	0.03	
LOI	1.60	1.00	4.00	6.80	21.30	15.10	19.50	29.90	31.30	
Total	99.89	99.94	100.13	101.19	99.84	99.79	99.60	99.22	98.87	
W ppm	7.2	118.8	0.7	1.5	1.2	3.2	0.8	0	2.8	
Nb	17.2	26.9	15.4	6.4	2.8	4.5	6.1	10.9	10.6	393 (a)
Ta	4.9	7	9.7	0.5	0.3	0.3	0	0	0	58 (a)
V	30	0	0	54	28	36	30	49	68	
U	10.1	42.4	11	2.4	1.3	7	0.2	1	2.8	461 (a)
Th	1.7	14.4	6.3	5.6	2.3	2.7	3.6	24.7	127.8	
Zr	67.2	23	21.4	7.6	26.4	37.6	8.7	20.4	19.8	4214 (a)
Hf	2.2	1.1	2.3	2.1	0.8	1.3	0.2	0.2	0.2	
Sn	10	5	15	2	1	5	0	1	0	
Sc	81	10	5	9	3	4	0	0	0	
Ga	18.8	2.9	35.5	1.8	3.4	9.6	1.2	2	1.9	
Sr	54.7	44.5	28.8	163.3	237.9	11.4	273.4	1264	2001	
Ba	105	17	13	201	81	35	316	1648	4195	
Be	16	19	6	1	0	8	5	11	12	
Rb	26.8	14.8	77.1	86	47.5	12.9	1.1	3.7	1.4	
Cs	7.6	4.3	7.9	5.1	2.6	0.5	0.1	0.1	0	
Y	33	31.7	8.7	17.4	10	23.8	21.3	239.9	416	
La	9.9	10.3	14.7	16.9	7.8	31.4	6.2	41.4	71.3	
Ce	26.3	28.1	26.4	34.9	15.9	52.4	19.3	151.1	260	
Pr	3.84	3.98	2.77	4.44	2.05	5.1	3.13	26.13	45.31	
Nd	15.4	14.3	8.7	17.7	8.6	18.3	15.7	140.8	239.7	
Sm	3.44	2.81	1.23	3.34	1.76	3.23	5.04	45.52	78.55	
Eu	0.28	0.18	0.12	0.51	0.54	1	1.78	15.66	27.04	
Gd	3.23	2.53	0.79	2.98	1.9	2.85	4.63	48.19	83.75	
Tb	0.71	0.6	0.16	0.49	0.31	0.53	0.8	7.94	13.49	
Dy	4.98	4.21	1.14	3	1.75	2.85	3.96	43.44	76.68	
Ho	1.14	1.1	0.25	0.65	0.39	0.67	0.68	7.77	13.94	
Er	3.71	3.54	0.93	1.94	1.09	2.14	1.61	19.33	35.42	
Tm	0.66	0.77	0.22	0.29	0.15	0.39	0.23	2.58	4.7	
Yb	4.93	5.89	2.19	1.8	0.99	2.86	1.2	13.91	27.15	
Lu	0.69	0.86	0.4	0.27	0.15	0.5	0.13	1.6	3.29	
As	5	0	0	6.7	2.1	0.9	0	0	0	
Se	0	0	0	0.7	0	0	0.7	1.4	1.9	
Bi	0.1	0	0	0.3	0.1	0.8	3	5.8	1.9	
Co	0.5	0.3	0.5	10	9.4	3.8	13.8	3.9	4.5	
Mo	0.2	0	0	0.9	1.9	8.4	25	315.1	377.2	1930 (b)
Cu	0.4	0.3	0.9	35.1	13.8	6.5	257.6	12	7.2	302 (b)
Pb	4	4.2	1.2	5.7	4.3	25.6	258	323.1	272.7	1909 (b)
Zn	5	3	0	12	4	18	66	69	21	255 (b)
Ni	0.6	0.8	0.8	24.8	14.1	16.8	4.3	4.8	2.1	
Cd	0	0	0	0	0	0	0.5	0.9	1.9	
Ag	0	0	0	0	0	0.2	0.5	0.9	0.7	

Samples: pegmatite: BLA: schorl unit, BLAD: diopside-bearing nests, BLB: uvite-liddicoatite unit, BLM-1, BLM-2, BLM-3: samples of ordinary marble collected from the body cut by pegmatite, BLK-1, BLK-2, BLK-3: carbonatite-like marbles. The extreme values for selected trace-elements from carbonatite-like marbles taken from: a) Houzar & Novák (2002) and b) Drábek *et al.* (1999).



The uvite–liddicoatite unit (ULU) adjacent exclusively to the tourmaline-rich reaction zone (Fig. 6) differs from the other units by lower amount of quartz and K-feldspar, olive green to pink tourmaline, but mainly by minor primary calcite; the Y,REE-bearing minerals are absent. Chemical compositions of rocks (Table 2) imply apparent contamination by Ca in all individual pegmatite units. The normalized REE patterns indicate a pronounced negative Eu anomaly and a slight enrichment in the middle REE mainly for the samples BLAD and BLA (Fig. 5).

The simply zoned elbaite-subtype pegmatite Bližná II, a dike up to 6 m thick and ~100 m long, generally northwest-trending and dipping ~70° to the northeast, is hosted mainly in graphite-bearing biotite gneiss. It crops out in two places: the chimney K15 and on the surface outcrop (Černý 2004) ~200–250 m south of the Bližná I pegmatite. Three distinct textural-paragenetic units were found rather randomly distributed: a dominant coarse-grained unit (Kfs + Ab + Qtz) with abundant brown to black dravite–schorl and rare pink elbaite, a minor graphic unit (Pl + Qtz) with dravite–schorl and olenite, a blocky unit (Kfs + Qtz) and a rare albite-rich unit with primary muscovite, most accessory minerals and black schorl to pink elbaite. The Bližná I and Bližná II pegmatites contain abundant tourmaline along with major K-feldspar, plagioclase and quartz; Bližná I is mica-free and contains more common Ca(Mg)-enriched minerals (Table 3).

Two rather distinct assemblages with REE minerals were distinguished in the Bližná I pegmatite. Yellow-brown tabular crystals of bastnäsite-(Ce), up to ~1–2 mm in diameter, are enclosed in (Ca,Mg,Mn)-rich schorl to uvite and associated with allanite-(Ce), titanite and parisite-(Ce) in a diopside-bearing nest (Fig. 7a). Well-developed, yellow to wax yellow, tabular crystals of bastnäsite-(Ce), up to 5 mm in size (Fig. 7b), from elbaite–liddicoatite unit are completely enclosed or closely associated with zoned grains of brown (Ca,Mg,Mn)-rich schorl–elbaite to pink Mn-rich elbaite–liddicoatite. Most minerals contain very thin veinlets of late minerals (REE fluorocarbonates, REE-bearing epidote, stillwellite-(La) (?) and dravite; Fig. 7b). In the Bližná II pegmatite, very rare, small inclusions of thortveitite associated with tourmaline were found at the surface outcrop (Černý 2004) and represent along with rare accessory monazite-(Ce) and zircon the only (Y,REE)-bearing mineral.

The pegmatites differ in chemical composition of tourmaline (Fig. 8) as well. Tourmaline from Bližná I is typically Ca-enriched and has elevated Mn in yellow to brown elbaite (Fig. 8d). Tourmaline from the surface outcrop at Bližná II is similar to that at Bližná I, showing limited to strong enrichment in Ca. In the underground outcrop at Bližná II, tourmaline is less contaminated by Ca (Figs. 8b, c) but strongly Mn-enriched ( $\leq 1.28 \text{ apfu}$ ; Fig. 8d). In general, the tourmaline crystals exhibit chemical compositions similar to those from ordinary

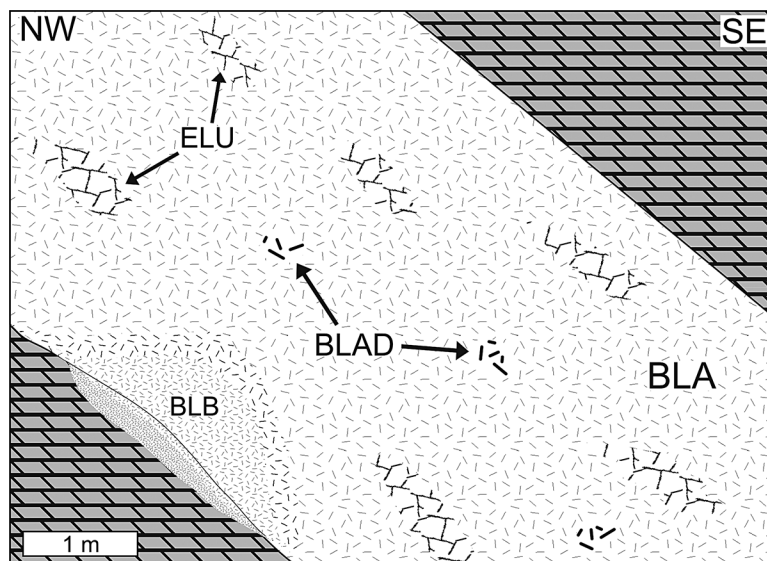


FIG. 6. Cross section through the Bližná I pegmatite enclosed in ordinary marbles; BLA: schorl unit, ELU: elbaite–liddicoatite unit, BLAD: diopside-bearing nests, BLB: uvite–liddicoatite unit with narrow tourmaline-rich contact zone (modified from Novák *et al.* 1999b).



FIG. 7. BSE images of REE minerals from the Bližná I pegmatite. a) Homogeneous equidimensional grain of bastnäsite (Bsn) enclosed in uvite (Uv) and associated with titanite (Ttn), allanite (Aln) and altered parisite (Pst). b) Fragment of euhedral grain of heterogeneous bastnäsite (bright) in elbaite-liddicoatite. Late veining: dravite (dark) and stillwellite-(La)? (bright).

TABLE 3. PRIMARY MINERALS FROM THE INDIVIDUAL PEGMATITES AND THEIR TEXTURAL-PARAGENETIC UNITS (OUTCROPS) IN BLIŽNÁ I AND BLIŽNÁ II

unit-outcrop	Bližná I				Bližná II	
	BLA	BLAD	ELU	BLB	under-ground	surface
quartz	+++	+++	+++	+++	+++	+++
K-feldspar	+++	+++	+++	+++	+++	+++
albite	+++	+	+++	+++	+++	+++
plagioclase $\leq$ An <sub>33</sub>	+	+++	+		+	+++
muscovite					++	
spessartine					++	+
schorl	++		+	+	+++	+
dravite		+		+	++	+
uvite		+		++		
elbaite			++	+	+	
liddicoatite			+	+		
olenite						+
axinite		+	+		+	
datolite			+			
dumortierite						+
diopside		+++	+			
titanite		++			+	+
epidote		+				
calcite				+		
allanite-(Ce)		+				
bastnäsite-(Ce)		+	+			
parisite-(Ce)		+				
thortveitite						+
zircon	+	+	+		+	
monazite-(Ce)	+				+	+
apatite	+				+	
columbite-(Mn)					+	
pyrochlore			+			
rutile					+	+
niobian rutile					+	
scheelite		+				
cassiterite					+	
pyrite					+	+

Abundance of minerals: +++: major and minor minerals, ++: common accessory minerals, +: rare accessory minerals.

elbaite-subtype pegmatites of the Moldanubian Zone (Fig. 8; Novák & Povondra 1995, Novák *et al.* 1999b, Novák 2000). The Bližná I pegmatite and the Bližná II surface outcrop are close to the Ca-contaminated Řečice pegmatite enclosed in pyroxene gneiss (Novák 1999, Novák *et al.* 1999c), with a variable degree of external contamination by Ca and Mg.

## DISCUSSION

*Comparison of the Kracovice pegmatite with similar Li-bearing (NYF + LCT) pegmatites and origin of the mixed signature in the Třebíč granite-pegmatite system*

The Li-bearing Kracovice pegmatite exhibits some mineralogical features (common lithium micas and elbaite) similar to spatially related complex (LCT) pegmatites of the Moldanubian Zone (Fig. 1). However, minerals from both groups have fairly distinct compositional trends and substitution mechanisms, signifying an absence or low participation of Al in tourmaline and a high participation of R<sup>2+</sup> (Fe, Mn) in the lithium micas in Kracovice (Černý *et al.* 1995, Novák *et al.* 1999a, 1999c, Novák 2000). In addition, the garnet has elevated contents of Y, the feldspars are P-poor, apatite is very rare, amblygonite or montebrazite is absent, and (Y, REE, Nb, Ta, Ti)-rich oxide minerals predominate over columbite-group minerals (Table 1). The Kracovice pegmatite also is entirely different from peraluminous, B-rich, (Li, F, REE)-poor anatectic pegmatites from this region (Cempřek *et al.* 2010, Gadas *et al.* 2012), for example in the isotopic composition of B in tourmaline (Míková *et al.* 2010).

The Kracovice pegmatite has mineralogical features similar with the mixed NYF + LCT pegmatite Høyaldalen near Tørdal, Telemark, Norway (Bergstøl & Juve 1988, Raade *et al.* 1993) and with the Leduc mine, Gatineau River area, Quebec, Canada (Hogarth *et al.* 1972, Ercit 2005). However, a lack of mineralogical and geochemical data mainly from the second locality does not enable a more detailed comparison. The Kracovice pegmatite, characterized by high activities of B and F, and very low activity of P, is a product of geochemical fractionation of mixed pegmatite-forming melt derived from the orogenic I-type Třebíč pluton. Contamination (LCT) from the host-rock sequence is rather unlikely

because of the absence of textural indications (sharp contacts with host gneiss, absence of any xenoliths) and estimated very short transport from parental granite to the site of emplacement. In addition, leucocratic tourmaline-bearing granites developed along the border of the Třebíč pluton (Fig. 2), which the pegmatite-forming melt very likely passed through, are rather primitive (Buriánek & Novák 2007) to cause external LCT-type contamination.

The pegmatites of the O'Grady Batholith, Selwyn plutonic suite, N.W.T., Canada, including the Li-enriched ones (Ercit *et al.* 2003), are very similar to the Třebíč pluton pegmatites and to the Kracovice pegmatite in

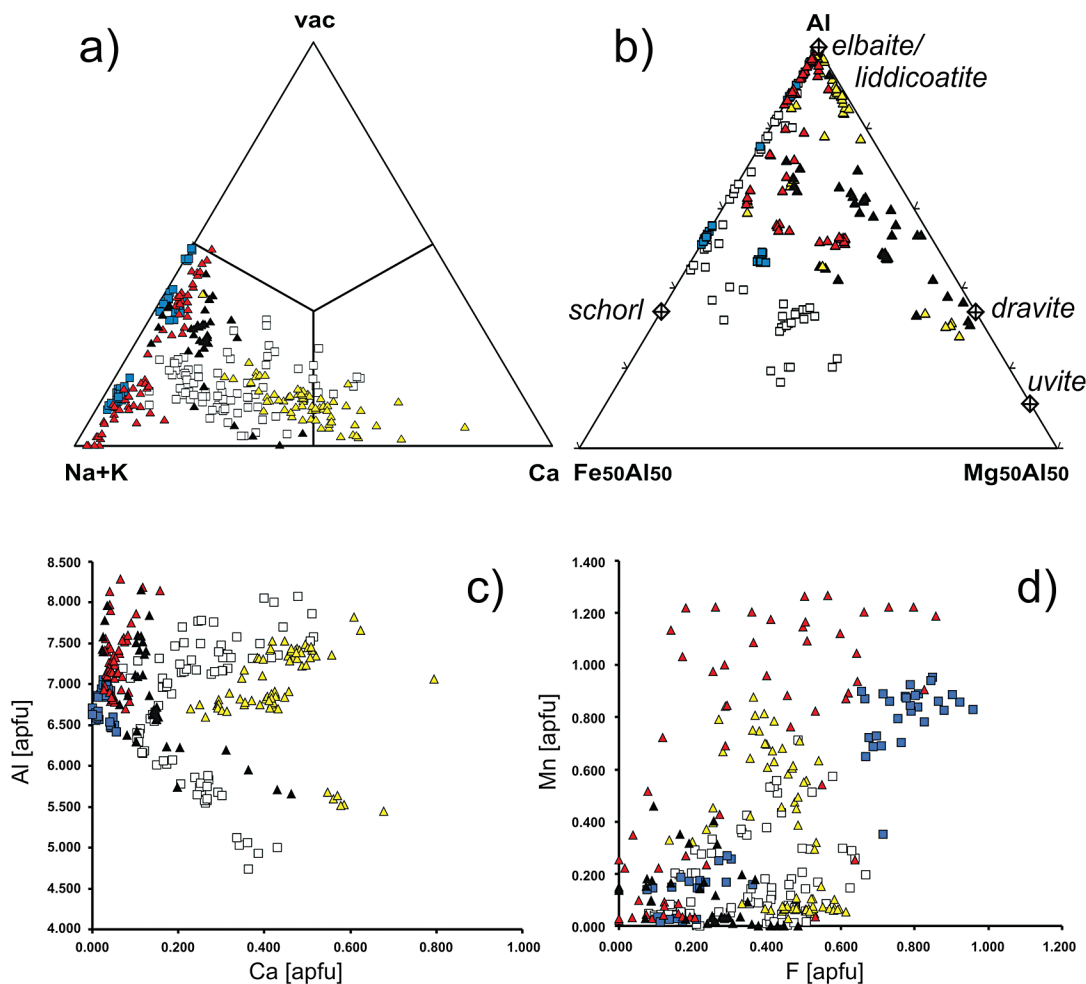


FIG. 8. Composition of tourmaline from selected elbaite-subtype pegmatites of the Moldanubian Zone: Bližná I (yellow triangles), Bližná II underground (red triangles), Bližná II surface (black triangles), Řečice (open squares), Ctíduzice (blue squares). Both latter localities are given to demonstrate typical examples of contaminated and non-contaminated elbaite-subtype pegmatites from the Moldanubian Zone: a) (Na+K) – Ca – X-site vacancy plot; b)  $Fe_{50}Al_{50}$ –Mg–Al triangle; c) Ca versus Al; d) F versus Mn.

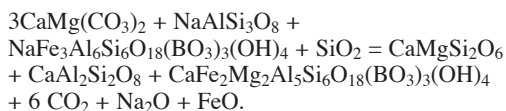
particular. This granite (hornblende quartz syenite) has geological, geochemical and petrographic features akin to the Třebíč pluton, such as major, minor and trace-element populations, porphyritic texture, depth of emplacement, and common scheelite-bearing skarns in the contact aureole. Its mixed geochemical characteristics are explained by a similar process of granite origin as in the Třebíč pluton: contamination of mantle-derived melt by undepleted crust (Coulson *et al.* 2000). Both pegmatite suites have a comparable evolution from less evolved pegmatites with mild to strong NYF signature to most evolved Li-enriched pegmatites with elbaite + lithium-dominant micas. Abundant Ti-rich minerals (ilmenite, rutile, pseudorutile, titanite) are characteristic of both suites. The enrichment in Cs (20–40 ppm: Třebíč pluton, 20 ppm: O'Grady Batholith) is manifested by a nanpingite-like mica, Cs-enriched chabazite and pollucite in pegmatites of the O'Grady Batholith (Ercit *et al.* 2003) and by Cs-enriched beryl (Novák & Filip 2010) and lithium-dominant micas with up to 1.34 wt.% Cs<sub>2</sub>O in the Třebíč pluton. Also, an enrichment in W is typical (7–8 ppm: Třebíč pluton, 8 ppm: O'Grady Batholith); scheelite and W-enriched stibiocolumbite are known in the O'Grady Batholith, whereas, scheelite, “wolframioxiolite” and W-enriched (Y,REE,Nb,Ta,Ti)-rich oxide minerals are found in the Třebíč pluton, respectively (Škoda *et al.* 2006, Škoda & Novák 2007). Common W-bearing minerals in both granite–pegmatite systems open a potential use of W as an indicator of the mixed signature of a pegmatite. However, W is a typical trace to minor element in (Nb,Ta,Ti)-oxide minerals in both NYF (Škoda *et al.* 2006, Škoda & Novák 2007) and in LCT pegmatites (*e.g.*, Novák & Černý 1998b, 2001, Černý *et al.* 2007, Novák *et al.* 2008, Cempírek *et al.* 2010) in the Moldanubian Zone.

The Třebíč pluton and O'Grady Batholith represent granite–pegmatite systems with a similar origin of parental granite and geochemistry. The evolution from less evolved but pristine NYF pegmatites to more evolved Li-bearing pegmatites with a mixed signature at both regions is likely enhanced by the fractionation process, in which the highly incompatible Li remains until the late stages of primary crystallization relative to the LREE, as is typical for more primitive NYF pegmatites. Based on the geochemical and mineralogical data, the granite–pegmatite system of the Třebíč pluton, Moldanubicum, may be established along with the O'Grady Batholith, Selwyn plutonic suite (Ercit *et al.* 2003), as typical examples of mixed granite–pegmatite systems (*cf.* Černý & Ercit 2005, Simmons & Webber 2008). Remarkable similarities in the concentrations of some trace elements (*e.g.*, Cs, W) in the parental granite and their presence in relevant accessory minerals from pegmatites imply that highly evolved pegmatites carry specific compositional features (fingerprints) of their parental granites throughout their overall crystallization up to the tail-end of primary crystallization

(London 2008). The granite–pegmatite system of the Třebíč pluton differs from most typical NYF and mixed granite–pegmatite systems (Černý & Ercit 2005) by evident orogenic character typical for all ultrapotassic intrusions of this region (*e.g.*, Žák *et al.* 2005, Verner *et al.* 2006, Janoušek & Holub 2007, Kotková *et al.* 2010, Krmíček *et al.* 2011). Such granite–pegmatite systems in ultrapotassic granites and related NYF-to-mixed granitic pegmatites have not yet been examined; they show that I-type synorogenic granites may generate pegmatites with a NYF-to-mixed signature.

*Origin of the mixed (NYF + LCT) signature in the Bližná I pegmatite and a role of external contamination in the origin of granitic pegmatite*

The evolved LCT pegmatite-forming melt from Bližná consumed and completely dissolved fragments of carbonatite-like marbles with a strong NYF-like geochemical signature during its ascent from the parental intrusion to the site of emplacement. Owing to a rather short transport of the melt from the site of contamination (likely hundreds of m; Fig. 9), the melt was not entirely homogenized, which can be seen from the heterogeneous distribution of diopside-bearing nests (BLAD; strongly digested fragments of carbonatite-like marbles) within the schorl unit (Fig. 6). Further evidence for pre-emplacment contamination by Ca, Mg, and the REE include the chemical composition of REE-bearing minerals rich in Ca(Mg) (parisite, allanite), and their close spatial association with Ca(Mg)-rich minerals (diopside, uvite, titanite, liddicoatite). For the origin of the diopside-bearing nests, a simplified reaction describing contamination of the pegmatite-forming melt by carbonate rock is given:



Dolomite is used as a contaminant, anorthite is present as a component in plagioclase (Table 3); for the tourmaline produced, a Fe-rich uvite composition is used for simplification, and minor Ti, Mn and F present in tourmaline (Novák *et al.* 1999b) are ignored.

Geochemical modeling offers further evidence for contamination of the pegmatite-forming melt by carbonatite-like marble. The best fit in the mixing test was obtained between the dominant schorl unit (sample BLA) as a pegmatitic component (SiO<sub>2</sub> = 76.74 wt.%; MgO = 0.31 wt.%; recalculated on a volatile-free basis) and carbonatite-like marble (sample BLK-1) as a contaminant, respectively (SiO<sub>2</sub> = 26.39 wt.%; MgO = 21.89 wt.%). The diopside-bearing nests are considered to be the product of contamination (sample BLAD; SiO<sub>2</sub> = 70.36 wt.%; MgO = 2.38 wt.%). In the

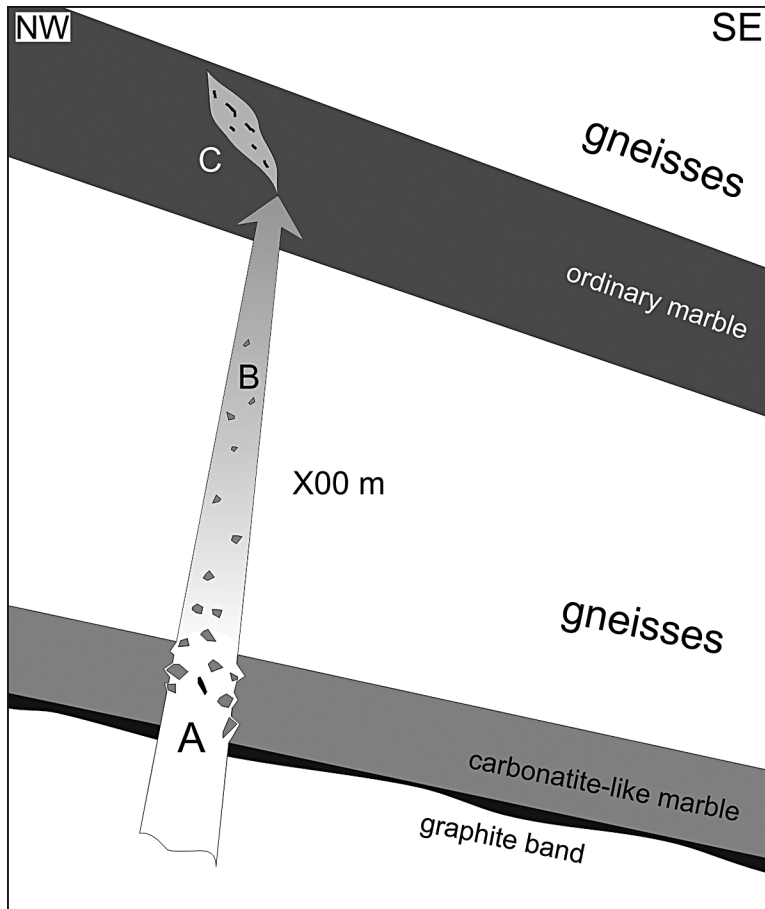


FIG. 9. Strongly schematic sketch showing pre-emplacment contamination of the pegmatite-forming melt of the Bližná I pegmatite by carbonatite-like marble. A: trapping of fragments of carbonatite-like marbles + very rare graphite fragments during forcible ascent of melt to the site of emplacement; B: dissolution of marble fragments and partial homogenization of the melt during the transport to the site of emplacement; C: crystallization of incompletely homogenized pegmatite-forming melt. The sketch is not in scale.

( $C_M - C_B$ ) versus ( $C_A - C_B$ ) diagram, the main oxides and REE define a straight mixing line with an excellent correlation ( $R = 0.998$ ; Fig. 10). Consequently, it is reasonable to consider that the diopside-bearing nests are products of melt contamination by marble with a carbonatite-like geochemical signature. The degree of contamination by this contaminant component is only ~10% in this particular case.

The Bližná I pegmatite is characterized by a high activity of B, but rather low activities of F and P. The pre-emplacment external contamination in Y and the REE of the LCT melt in the Bližná I pegmatite (Figs. 9, 10) is a unique process, which has no analogy in nature insofar as we know. Its scarcity may be explained by

very low probability to mobilize Y, REE, Nb, Ta, and other HFS elements in fluids at P-T conditions typical for generation and ascent of pegmatites with LCT signature in the upper crust (Martin & De Vito 2005), and the rarity of rocks with a strong NYF-signature such as carbonatite-like marble from Bližná in supracrustal complexes. The Ca(Mg)-contaminated Bližná II pegmatite enclosed in gneiss exhibits a lower but variable degree of pre-emplacment contamination within a single dike and obviously manifests high variability in external contamination of pegmatites within this region.

Martin & De Vito (2005) described several examples of hybrid NYF + LCT pegmatites from Madagascar, where dominant contamination of pegmatites in calcium

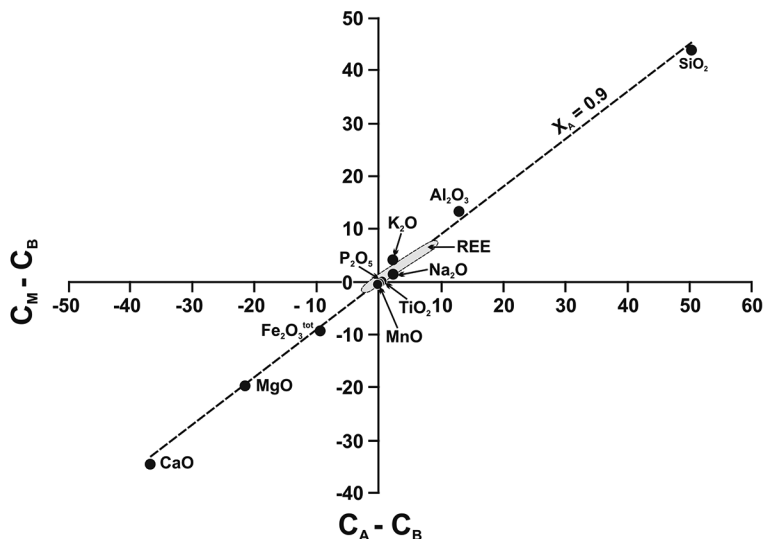


FIG. 10. Distribution of major and minor oxides and REE in the  $(C_M - C_B)$  versus  $(C_A - C_B)$  diagram of Fourcade & Allègre (1981). The pegmatitic component corresponds to the sample BLA, the contaminant component, to the sample BLK-1, and the mixed component, to the sample BLAD (see also Table 2 and Fig. 5).

from host rocks played a significant role. Although these pegmatites are geochemically quite strange, they likely represent products of ordinary external contamination from supracrustal host metasedimentary complexes rich in carbonates (Novák 2007). However, this contamination by Ca has geochemical character strikingly distinct from NYF or LCT characteristics. Similar contamination by Ca(Mg,Fe) was described from several pegmatites in Czech Republic: contamination in Ca and Mg from serpentinite and rodingite in Ruda nad Moravou (Novák & Gadas 2010); contamination in Mg from serpentinite in Věžná I pegmatite (Černý & Povondra 1966, Novák 1998, Novák *et al.* 2003, Dosbaba & Novák 2012), and contamination by Ca and Fe from a Fe-rich skarn in Vlastějovice (Kadlec 2007, Breiter *et al.* 2010). A role of contamination (commonly Ca, Mg) from host rocks or from channelways, through which the pegmatite-forming melts were passing to the site of emplacement (Novák 2007), should be routinely included in discussions of pegmatite origin in the cases where unusual bulk-compositions or, more commonly, strange compositions of individual minerals are observed. Up to now, significant contamination has been documented almost exclusively in relatively small bodies of pegmatite cutting rocks with contrasting chemical composition (*e.g.*, Černý & Povondra 1966, Martin-Izard *et al.* 1995, Novák *et al.* 1999b, Tindle *et al.* 2002, Breiter *et al.* 2010, Novák & Gadas 2010, Kuznetsova *et al.* 2010). Only minor contamination by Ca, Mg, and Ti in tourmaline and columbite was

described from the outer zones of the large Tanco pegmatite, southeastern Manitoba, enclosed in metagabbro (Selway *et al.* 2000, van Lichtervelde *et al.* 2006). Only contamination by major and important minor elements (*e.g.*, Ca, Mg, Fe, Ti) is feasible. If we compare the concentrations of Ca, Mg and Fe in host rocks (marbles, serpentinite, Fe-rich skarn) relative to their concentrations in minerals from the contaminated pegmatites (Novák *et al.* 1999b, Novák & Gadas 2010, Breiter *et al.* 2010), we can hardly expect detectable contamination by trace elements from host rocks without a significant contamination by major elements.

#### ACKNOWLEDGEMENTS

The authors thank the reviewers Saskia Erdmann and Mona-Lisa Sirbescu and the Guest Editor David London for constructive criticism that considerably improved the manuscript. The authors also thank J.B. Selway for carrying out chemical analyses of some minerals from Kracovice and V. Janousek for unpublished results of chemical analyses of the Třebíč granites. Milan Novák gratefully acknowledges his mentor, Professor Petr Černý, University of Manitoba, Winnipeg, for his outstanding contribution to research on granitic pegmatites, their minerals and origin, as well as to the way of thinking about them. His teachings also indirectly but significantly influenced the approach to research on pegmatites of his students, including those participated in this paper (RS, PG, PČ). This work was

supported by research project GACR P210/10/0743 to MN, RS and PG.

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Received July 2, 2011, revised manuscript accepted July 26, 2012.