# **COMPOSITIONAL EVOLUTION OF ZONED TOURMALINE CRYSTALS FROM POCKETS IN COMMON PEGMATITES OF THE MOLDANUBIAN ZONE, CZECH REPUBLIC**

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## **ABSTRACT**

Common pegmatites with pockets lined with crystals of smoky quartz, feldspars, muscovite, and black tourmaline commonly occur in migmatized biotite–sillimanite gneisses and felsic granulites of the Strážek Unit, Moldanubian Zone, in the Czech Republic. Mostly concordant dikes or irregular bodies (0.1–2 m thick) consisting of an outer granitic unit, a graphic unit and a pocket unit with large crystals (up to 1 m long) of minerals show transitional to locally sharp contacts to the host migmatized rocks. Thin muscovite-rich veins, 1–10 cm thick, with the assemblage muscovite ≥ albite, quartz and tourmaline are closely associated spatially. Variable prismatic to lens-shaped tourmaline crystals and their aggregates, up to 30 cm across, from pockets and muscovite-rich veins exhibit striking zoning in the optical microscope (distinct colors and pleochroism) and in BSE images: core (foitite – schorl – magnesio-foitite), intermediate zone (schorl – dravite) and narrow outer rim (schorl – dravite); tourmaline from muscovite veins is relatively homogeneous (dravite – magnesio-foitite). A slight decrease in *X*-site vacancy, in  $Fe_{tot}/(Fe_{tot}$ + Mg) and Al contents but sharp increase of Ti from the core to intermediate zone and outer rim, respectively, are typical. Also, the LA–ICP–MS study of a single crystal of tourmaline from a pocket demonstrates a very similar chemical composition of core and intermediate zone, but a distinct composition of outer rim; Mn, Ni, Zn, Sc and Ga decrease, and V, Cr, Co and Sr increase in the rim. Nearly all Fe was determined as  $Fe^{2+}$  (<3% of  $Fe^{3+}$  in both core and intermediate zones of the tourmaline) using Mössbauer spectroscopy. The mineral assemblages (andalusite + cordierite) of common pegmatites and their geological and petrographic features indicate P–T conditions of ~500–650°C and less than ~3–4 kbar for massive units, but lower for tourmalines and other minerals from pockets and in particular for associated muscovite-rich veins. The distribution of pegmatites in migmatitic rocks, their concordant and transitional (diffusive) contacts to the country rocks, simple assemblages of minerals, primitive chemical composition of minerals (all typical for abyssal pegmatites), and common large pockets lined with well-developed crystals of smoky quartz, feldspars, muscovite, and tourmaline (which is typical of miarolitic pegmatites), do not fit the current classifications of granitic pegmatites.

*Keywords*: foitite, schorl, dravite, magnesio-foitite, compositional evolution, zoning, pocket, common pegmatite, Moldanubian Zone, Czech Republic.

#### **INTRODUCTION**

Compositional trends in tourmalines have been studied in a variety of granitic pegmatites from barren (*e.g.,* Novák *et al*. 2004a, Roda-Robles *et al.* 2011, Gadas *et al*. 2011) to highly fractionated complex (Li) ones (*e.g.,* Jolliff *et al*. 1986, Federico *et al*. 1998,

Selway *et al*. 1999, Tindle *et al*. 2002, Ertl *et al*. 2010). During the primary crystallization of a pegmatite, tourmaline crystals generally exhibit an increase in Al, Mn and Li, and a decrease in Mg, Ti and Fe in the simplified sequence dravite–schorl–foitite  $\rightarrow$  schorl–olenite  $\rightarrow$  $schor$ l–elbaite  $\rightarrow$  elbaite  $\rightarrow$  rossmanite. Compositional variations in tourmaline crystals from pockets were

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examined almost exclusively in Li-rich pegmatites (*e.g.,* Ertl *et al*. 2007, 2010, Lussier & Hawthorne 2011, Lussier *et al*. 2011, and references therein), where a complex zoning (oscillatory and sectoral) was generally observed. Tourmaline crystals from pockets in primitive, Li-poor pegmatites have only rarely been examined (Novák *et al*. 2004a).

Common pegmatites, with the mineral assemblage  $(Pl + Kfs + Qtz + Ab + Bt + Ms + Tur \pm Ap, And, Crd)$ are enclosed in migmatized gneisses and granulites, and occur irregularly but commonly in the Moldanubian Zone, Czech Republic. Their constituent minerals, mainly tourmaline, were studied from the 19<sup>th</sup> century (*e.g.,* Kolenati 1854, Slavík 1904, Sekanina 1928), but few analytical data were published (Povondra 1981, MacBride 2002, Novák *et al*. 2004a, Míková *et al*. 2010). Many common pegmatites contain pockets with zoned crystals of black tourmaline, which typically show the compositional trend foitite  $\rightarrow$  schorl  $\rightarrow$  dravite  $\rightarrow$  magnesio-foitite from core to rim, both parallel and perpendicular to the **c** axis, showing a reverse evolution to that in ordinary pegmatites (Gadas *et al*. 2011). We present here compositional data (EMPA, LA–ICP–MS, Mössbauer spectroscopy) to document tourmaline crystals from common pegmatites with pockets and associated muscovite-rich veins. We also discuss the compositional evolution found within individual crystals of tourmaline, as well as the classification of these pegmatites.

## Geological Setting and Outline of Granitic PEGMATITES FROM THE STRÁŽEK UNIT

Common pegmatites with pockets lined with crystals of smoky quartz, feldspars, muscovite and tourmaline occur in several regions comprised of migmatized gneisses and migmatized felsic granulitic rocks within the Moldanubian Zone (Fig. 1). The pegmatites are concentrated mainly in the Strážek Unit, but rather isolated occurrences were found in the Moldanubian Zone west of the Třebíč pluton, in the area adjacent to the Kutná Hora – Svratka Unit (Fig. 1), and in the Austrian part of the Moldanubian Zone. The metamorphic rocks of the Strážek Unit underwent a polyphase metamorphic evolution; a HT–HP event in the upper amphibolite to granulite facies at  $T_{\text{max}} \sim 850-900^{\circ} \text{C}$ and  $P_{\text{max}} = 12 - 18$  kbar was overprinted during a rapid decompression by an MP–HT event at  $T < 700^{\circ}$ C and P ~4–6 kbar (*e.g.,* Pertoldová *et al*. 2009, and references therein), and this stage very likely produced a partial melting of rocks. Mineral assemblages of migmatized portions of biotite–sillimanite gneisses or granulitic gneisses are simple (Pl + Qtz + Kfs + Bt  $\pm$  Sil  $\pm$  Crd  $\pm$ Grt  $\pm$  Hc  $\pm$  Tur) and very similar to those in common pegmatites.

Granitic pegmatites of various size, internal structure and geochemical signature and origin are common in the Strážek Unit and essentially in the Bory Granulite Massif and its close vicinity (Novák 2005, Cempírek *et al*. 2010). They are represented, following estimated P–T conditions of their formation, by: (i) a borosilicatebearing pegmatite veinlet (grandidierite–ominelite + boralsilite + werdingite + dumortierite + schorl) cutting leucogranulite, which postdates the development of a foliation in the host granulite at T  $\approx 750^{\circ}$ C and P  $\approx$ 6–8 kbar (Cempírek *et al*. 2010); (ii) coarse-grained, cordierite-rich leucosome in dark gneisses (cordierite  $[X<sub>Fe</sub>$  in the range  $0.44-0.50$  + biotite + tourmaline  $\pm$ dumortierite), locally showing pegmatitic textures but no pockets (Povondra *et al*. 1992, Kotková & Melichar 2003); (iii) the simply zoned common pegmatites, the focus of this study, with accessory andalusite, which mostly show transitional (diffusive) contacts to country rocks and locally contain large crystals of smoky quartz and feldspars in pockets (Gadas *et al.* 2011); (iv) crosscutting, symmetrically zoned pegmatites (muscovite + tourmaline + andalusite + sekaninaite  $[X_{Fe}]$  in the range  $0.74-0.97$ ] + biotite + apatite) with sharp contacts and containing primary diaspore, Fe–Mn phosphates, pyrite, löllingite, and numerous accessory minerals, commonly occurring in the most evolved, large bodies, up to 35 m thick and several hundred m long (Duda 1986, Staněk 1991, 1997, Novák & Taylor 2005); (v) discordant, symmetrically zoned, complex lepidolite-subtype pegmatites (lepidolite + elbaite + amblygonite + rossmanite) with sharp contacts to the host rocks, the most highly fractionated pegmatites of the region (Selway *et al*. 1999, Novák *et al*. 2004b). The U–Pb radiometric data on monazite from the discordant pegmatite (type iv) in Dolní Bory – Hatě yielded the ages of  $335.8 \pm$ 2 and 337.2 ± 2 Ma (Novák *et al*. 1998) comparable with the age of other pegmatites of the Moldanubian Zone (Ertl *et al*. 2004). Whatever the origin and degree of fractionation, all pegmatite types occurring in the Bory Granulite Massif and its envelope have similar geochemical signatures: peraluminous character, high activity of B, presence of accessory minerals with Ti, W, Sc, and As, and total absence of Be-bearing minerals. The pegmatites originated in a short period of  $\sim$ 5–7 Ma during rapid decompression and were sequentially generated under rather distinct P–T conditions (for more details see Cempírek *et al*. 2010).

## Internal Structure and Mineralogy of the Common Pegmatites

The common pegmatites with pockets have been rarely found in good outcrops, and are most common as fragments or isolated crystals found in agricultural fields. Classical localities include Bobrůvka, Cyrilov, Dolní Bobrová, Dolní Bory, Horní Bory, Meziboří, Netín, Rousměrov, Suky, Vídeň and Znětínek (Fig. 1). The pegmatites form concordant (Figs. 2a, 2b) and, less commonly, discordant dikes or irregular bodies with transitional to locally sharp contacts with the host migmatized rocks (biotite–sillimanite gneiss, migma-



Fig. 1. Selected occurrences of common pegmatites with pockets and muscovite-rich veins, plotted on the simplified geological map of the northeasternmost part of the Moldanubian Zone. (slightly modified from Gadas *et al.* 2011). Gneisses and granulites are commonly migmatized.

tized leucocratic granulite rocks). Transitional and sharp contacts between individual textural-paragenetic units may be developed within a single pegmatite body. The internal structure of pegmatites evolved from common, simply zoned thin dikes, several cm to several dm thick (Figs. 2a, 2b, 3), to rare, texturally more complicated dikes, up to 2 m thick. The pegmatites are commonly built of (i) an outermost, medium- to coarse-grained granitic unit (Pl + Qtz + Kfs  $\pm$  Bt  $\pm$  Ms), (ii) a graphic unit ( $Qtz + Kfs \pm Bt$  or Ms), and (iii) a pocket unit with large crystals of primary smoky quartz I (up to 1 m long), K-feldspar (up to 30 cm in size), albite, muscovite I (up to 5 cm in size) and commonly also tourmaline (up to 30 cm long; Figs. 2c, 2d). In some pegmatite dikes,

tourmaline is absent. Larger dikes contain (iv) blocky K-feldspar evolving gradually from graphic K-feldspar + quartz intergrowths and locally (v) a small quartz core (Fig. 3). The mineral assemblages of granitic and graphic units and in blocky K-feldspar include (along with major and minor minerals) andalusite replaced by muscovite, strongly altered cordierite, fluorapatite and rare accessory dumortierite, almandine–spessartine, pyrite, ilmenite, rutile, monazite-(Ce), xenotime-(Y) and Zn-rich hercynite.

The pockets are distributed close to the core of pegmatite bodies (Figs. 2a, 3), and the total volume of pockets relative to massive pegmatite units is highly variable within the individual bodies. The early



FIG. 2. Photographs of the outcrop of a pegmatite dike enclosed concordantly in migmatized gneiss, locality Věžná III (a) and its cut-out detail showing xenoliths of the country rock with a locally diffuse contact marked by the arrow (b). Tourmaline crystals from pockets in pegmatites: c) thick columnar crystal of tourmaline with smoky quartz covered in part by muscovite I, locality Krásněves, and d) thin columnar crystals of tourmaline intergrown on albite and muscovite II, locality Bobrůvka.

minerals from pockets (chiefly K-feldspar, tourmaline and quartz) extend their growth from the surrounding massive pegmatite (graphic unit and blocky K-feldspar), as is typical for miarolitic pegmatites (Černý 2000). In pockets, smoky quartz I, K-feldspar, albite, muscovite I and tourmaline are dominant minerals (Figs. 2c, 2d, 3). Large crystals of K-feldspar are commonly corroded and replaced by medium-grained secondary muscovite II and small aggregates of subparallel crystals of smoky quartz II. Rare, but perfectly developed crystals of fluorapatite, garnet, anatase, brookite and rutile and microscopic inclusions of pyrrhotite and "wolframoixiolite" in quartz and tourmaline crystals also were found. Fine- to coarse-grained aggregates and large flakes of muscovite and yellow to brown masses of clay minerals

typically fill the void space of pockets and represent the latest stage of mineral deposition (see Černý 2000).

Thin muscovite-rich veins,  $\sim$ 1–10 cm thick and differing from pegmatite dikes in shape, size and internal structure, have been sporadically found in close vicinity to common pegmatites with pocket or filling the cracks of pegmatite dikes. They are built of porous masses of fine- to coarse-flaked muscovite as a dominant mineral within veins cutting the host migmatized gneisses. Moreover, they exhibit a simple assemblage of minerals: muscovite  $\geq$  albite, quartz and tourmaline; K-feldspar and accessory minerals typical of common pegmatites, except for apatite, are commonly absent. According to similarities in mineral assemblages and apparent spatial relations, the muscovite-rich veins

seem to be late satellite veins genetically related to common pocket pegmatites (Gadas *et al*. 2011); however, poor outcrops do not enable us to attempt a detailed study of the relations between pegmatites and muscovite-rich veins.

#### Methods and Samples

The chemical composition of tourmaline from pockets and muscovite veins was determined using a Cameca SX100 electron microprobe at the Joint Laboratory of Electron Microscopy and Microanalysis, Department of Geological Sciences, Masaryk University, and Czech Geological Survey, in Brno. The measurements were carried out in wavelength-dispersive mode, with an acceleration voltage of 15 kV, a beam current of 10 nA, and a spot size of  $\sim$ 2–5  $\mu$ m. The following standards and  $\bar{X}$ -ray  $K\alpha$  lines were used: sanidine (Si, Al, K), albite (Na), olivine (Mg), andradite (Ca,

Fe), rhodonite (Mn), hornblende (Ti), chromite (Cr), topaz (F) and ZnS (Zn). The counting time on peaks was 10 second for major elements and 20–40 seconds for minor to trace elements. The background counting time was one half of the counting time on peaks on the high- and low-energy background position. The raw data were reduced using appropriate PAP matrix corrections (Pouchou & Pichoir 1985). The crystal-chemical formulae of tourmaline were calculated on 31 anions to a general formula  $XY_3Z_6T_6O_{18}(BO_3)_3V_3W$ , where X  $=$  Na, Ca, K, vacancy,  $Y =$  Fe, Mg, Mn, Ti, Al, and V,  $Z = Al$ , and Mg;  $T = Si$ , and Al;  $B = B = 3$  *apfu*;  $V +$ *W* = OH + F = 4 (or 3.5, see Novák *et al*. 2004a), and using total Fe as FeO (see Mössbauer spectroscopy). Tourmaline crystals from the following localities were examined using the electron microprobe: tourmaline from massive pegmatite units (Znětínek), tourmaline from pockets (Bobrůvka, Cyrilov, Dlouhá Brtnice, Dolní Bory, Golčův Jeníkov, Hodíškov, Krásněves,



Fig. 3. Idealized cross-section through a common pegmatite with pockets from Znětínek. Note different mineral assemblages on the hanging-wall part (large crystals of smoky quartz I up to 25 cm long + albite I) and the footwall part of the pocket (large corroded crystals of K-feldspar up to 30 cm in size + muscovite II + subparallel crystals of smoky quartz  $II$  + tourmaline). a) Host migmatized biotite–sillimanite gneiss, b) tourmaline on the contact, c) medium- to coarse-grained granitic unit, d) graphic unit, e) blocky K-feldspar with massive quartz (grey), f) tourmaline (black), g) large crystals of smoky quartz I + albite I, h) quartz II + albite II + muscovite II.

Meziboří, Netín, Pikárec, Radenice, Sedletín, Sklenné, Strážek, Suky, Věžná III, Vídeň, Záseka, Znětínek), and from muscovite-rich veins (Hodíškov, Horní Bory, Radenice).

The transmission <sup>57</sup>Fe Mössbauer spectrum of powdered tourmaline samples (ground under acetone using an agate mortar) was recorded in constant acceleration mode at room temperature using a  ${}^{57}Co(Rh)$ source. The isomer shift value was calibrated against an  $\alpha$ -Fe foil. Spectra were folded and fitted by Lorentz functions using the computer program CONFIT2000 (Žák & Jirásková 2006). The effects of non-ideal thickness of the absorber and variable recoil-free fractions for iron atoms in non-equivalent structural sites were expected to be within experimental errors (hyperfine parameters  $\pm 0.01$  mm s<sup>-1</sup>, relative spectral area  $\pm 1\%$ ). Prior to analysis, all samples were washed in a cold dilute hydrochloric acid in order to remove any possible interstitial iron oxides and oxyhydroxides. The core and intermediate zone were examined on two zoned crystals from pockets (Hodíškov and Suky).

Instrumentation for the LA–ICP–MS at the Department of Chemistry, Masaryk University, Brno, consists of a laser-ablation system UP 213 (New Wave, USA) and an Agilent 7500 CE ICP–MS spectrometer (Agilent, Japan). A commercial Q-switched Nd:YAG laser-ablation device works at the fifth harmonic frequency, which corresponds to the wavelength of 213 nm. The ablation device is equipped with programmable XYZ stages to move the sample along a preset trajectory during ablation. The ablation cell was flushed with helium (carrier gas), which transported the laser-induced aerosol to the inductively coupled plasma – mass spectrometer (1 L/min). A gas flow of argon was admixed to the helium carrier gas flow after leaving the laser-ablation cell. Therefore, the total gas flow was 1.6 L/min. For LA–ICP–MS measurements, we used the hole-drilling mode (fixed position of sample during laser ablation) for the duration of 60 seconds per each spot. Laser ablation was performed with a laser spot-diameter of 80  $\mu$ m, laser fluence 4.5 J cm<sup>-2</sup> and repetition rate 10 Hz. We examined a zoned crystal of tourmaline with well-developed core, intermediate zone and outer rim from pocket in the pegmatite Suky.

## Description, Mineral Assemblages and Zoning of Tourmaline

Black tourmaline, a widespread accessory mineral in common pegmatites, is present in several morphological types in the texturally more evolved larger bodies of pegmatite (Fig. 3). Prismatic crystals and their aggregates are commonly enclosed in blocky K-feldspar, massive quartz and albite; tourmaline is commonly absent in the graphic unit. Radial aggregates of tourmaline were also sporadically found in country rocks near or directly on the contact with pegmatite (Fig. 3); nevertheless, the amount of tourmaline in

massive pegmatite units (least in the graphic unit, and in subequal proportions in the blocky K-feldspar and the massive quartz) is much smaller relative to pockets. In the muscovite-rich veins, well-developed isolated crystals of tourmaline and aggregates are present.

Tourmaline from pockets forms nearly euhedral long prismatic to short bipyramidal crystals (with a hexagonal or trigonal cross-section, Figs. 2c, 2d, 4) and their parallel to subparallel intergrowths, both up to 30 cm in size. Rarely, long doubly-terminated prismatic crystals are hemihedral with a pyramidal analogous pole, whereas a pedion usually terminates the antilogous pole. Crystals typically show prismatic striations and a strong vitreous luster (Figs. 2c, 2d, 4). Long prismatic crystals commonly grew from adjacent massive-pegmatite unit (mostly blocky K-feldspar) into the open space of pocket. In contrast, lens-shaped crystals, doubly-terminated prismatic to tabular crystals and some subparallel intergrowths grow on crystals of quartz or albite, and the whole volume of the tourmaline crystal was formed within pockets. Exceptionally, late hair-like fibrous aggregates of pale brown tourmaline occur in pockets (see Dutrow & Henry 2000). Tourmaline crystals and aggregates are generally overgrown by muscovite, quartz and albite, and all these minerals are present in two (or more) generations in pockets, where they crystallized with a more or less evident hiatus. The textural relations suggest that K-feldspar and tourmaline crystals extending from the massive pegmatite unit are the earliest minerals to crystallize into the pockets (see also Němec 1992).

Almost all crystals of tourmaline from pockets in common pegmatites exhibit striking zoning (transmitted light) and in the BSE images as well (Fig. 5), both parallel and perpendicular to the **c** axis. The core is volumetrically dominant and exhibits a sharp contact with the intermediate zone. The outer rim is usually narrow, from  $5-20 \mu m$  thick, only exceptionally up to 300  $\mu$ m (locality Suky, Fig. 5), typically with sharp contacts with the intermediate zone. The individual zones have different pleochroism (Fig. 5); in the core: O: greyish blue to khaki green, E: pale yellowish to colorless; intermediate zone: O: brown, E: pale brown to yellowish; outer rim: O: dark brown to brown, E: pale brown to yellowish. Within the core, a weak to moderate sector zoning and rare fine-oscillatory or patchy zoning were observed; oscillatory zoning is common in the intermediate zone (Fig. 5). Only locally, the core is partly replaced along the contact with the intermediate zone (locality Suky, Fig. 5). Also, late veins with a composition close to the outer rim replace core and intermediate zones (locality Suky, Fig. 5). The thin outer rim is commonly homogeneous, rarely with fine oscillatory zoning. On the contrary, tourmaline crystals from muscovite-rich veins are optically and compositionally rather homogeneous, and pleochroic (O: brown, E: pale brown).



Fig. 4. Crystals of tourmaline from common pegmatites; a) Dolní Bory, b) Meziboří, c) Cyrilov. Drawings of crystals from Sekanina (1928), photographs by P. Gadas.

The Chemical Composition of Tourmaline

Wet-chemical analyses of tourmaline crystals from the common pegmatites at the localities Bobrůvka, Dolní Bobrová, Cyrilov, Dolní Bory, Horní Bory, Meziboří, Pikárec and Rousměrov were published by Povondra (1981). The following minerals of the tourmaline supergroup (*cf*. Henry *et al*. 2011) were identified: schorl, "oxy-schorl", dravite, "oxy-dravite" and foitite; however, these data represent bulk compositions of



Fig. 5. Photomicrographs combining BSE images of zoned crystals of tourmaline from pockets in common pegmatites. The cuts are perpendicular to the **c** axis of the crystals; C: core, I: intermediate zone, R: outer rim or late veining (Suky). Note the sharp contacts between the individual zones and the alteration of core in the crystal from Suky.

whole crystals, and evident optical and compositional zoning (Fig. 5) was mostly overlooked. Only different density fractions from the individual zoned crystals were analyzed separately at some localities. Generally, low Fe<sup>3+</sup> (<0.2 *apfu*), Si (6.01–5.84 *apfu*) and a content of B in the range 2.91–3.07 *apfu*, close to the stoichiometric value (3) were found in these tourmalines, along with highly variable  $Fe/(Fe + Mg)$ , in the range 0.80–0.40 (Povondra 1981, Novák *et al*. 2004a).

#### *Electron-microprobe data*

In the Znětínek pegmatite (Fig. 3), the following paragenetic types of tourmaline were examined: grains from host rock and the needle-like crystals from contact with country rock (both Mg-rich schorl), prismatic crystals of foitite in blocky K-feldspar, small prismatic crystals occurring in pockets (foitite  $\approx$  schorl  $>$  dravite) associated with quartz II and muscovite II, respectively. They exhibit a great variation in  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$ (0.52–0.56, 0.80–0.81, 0.44–0.75); foitite from blocky K-feldspar is Ca- and Ti-poor with  $≤0.02$  and  $≤0.03$ *apfu*, whereas tourmalines from both contact and host rock are slightly enriched in these elements, with ≤0.10 *apfu* Ca and ≤0.10 *apfu* Ti relative to tourmalines from pegmatite.

The optical zoning and compositional zoning in individual crystals from pockets are only partly consistent (Fig. 5, Table 1). The core (foitite  $\approx$  schorl > magnesiofoitite) with  $Fe<sub>tot</sub> / (Fe<sub>tot</sub> + Mg)$  in the range 0.99–0.41) and intermediate zone (schorl > dravite) with  $Fe<sub>tot</sub>$ /(Fe<sub>tot</sub>)  $+$  Mg) in the range 0.99–0.39) are compositionally similar, showing mostly a slight to moderate decrease (rarely also a slight increase) in  $Fe<sub>tot</sub>$  (Fe<sub>tot</sub> + Mg) and in Al, and a slight decrease in the *X*-site vacancy (Fig. 6). Rims of crystals from pockets (schorl  $\approx$  dravite) are generally more homogeneous, with  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$ in the range 0.71–0.36. The only exception in Fe/Mg behavior is a zoned crystal from Bobrůvka (Fig. 5), where the intermediate zone has evidently higher  $Fe<sub>tot</sub>/$  $(Fe<sub>tot</sub> + Mg)$ , in the range 0.88–0.81, relative to the core, with 0.61–0.62, and the rim, with 0.56–0.60. On the other hand, tourmaline from Suky shows almost identical  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$  in core and intermediate zone  $(0.98-0.97)$ , but very low  $Fe_{tot}/(Fe_{tot} + Mg)$   $(0.36-0.37)$ in outer rim (Fig. 6). A sudden increase of Ti from the core to the intermediate zone and locally from the intermediate zone to the outer rim (Fig. 6c) is a typical feature of all crystals. Variation in *X*-site vacancy shows a moderate decrease from core (0.63–0.28 *pfu*) to intermediate zone (0.51–0.23 *pfu*) and rim (0.47–0.12 *pfu*). Low Ca contents (≤0.05 *apfu*, exceptionally up to 0.1), are typical of the core and intermediate zone; rims are slightly Ca-enriched, up to ≤0.15 *apfu*.

Chemical variability of tourmaline from muscoviterich veins is generally low, with high variability in *X*-site vacancy (0.59–0.15 *pfu*) (Fig. 6), but only slightly

variable  $Fe_{tot}/(Fe_{tot} + Mg)$  (0.45–0.31). Tourmaline is also enriched in Ti, up to  $0.18$  *apfu*,  $1.46$  wt.% TiO<sub>2</sub>, except for the narrow outer rim, with ≤0.10 *apfu* Ti (Fig. 6c). Calcium has very similar behavior, with up to 0.21 *apfu* Ca in the core but only ≤0.15 *apfu* Ca in the outermost rim (Fig. 6a).

With his results of wet-chemical analyses, Povondra (1981) identified also "oxy-schorl" and "oxy-dravite" in the common pegmatites. However, the formula calculations normalized on  $V + W = OH + F = 4$  or on 3.5 *apfu*, as recommended by Novák *et al*. (2004a), yielded better results (*e.g.,* Si close to 6 *apfu*) for the normalization  $V + W = OH + F = 4$ . This suggests a low proportion of oxy-tourmaline components (Henry *et al*. 2011) in most tourmalines.

#### *Mössbauer spectroscopy*

The Mössbauer spectra acquired for two zoned crystals from pockets in pegmatites (locality Suky and Hodíškov) were fitted with four doublets corresponding to  $^{[6]}Fe^{2+}$  and one doublet corresponding to  $^{[6]}Fe^{3+}$  (all iron in a high-spin state; Fig. 7). The resulting hyperfine parameters of these doublets and their structural assignments (Table 2) are comparable to previously studied tourmalines (*cf.* Andreozzi *et al.* 2008, and references therein). Almost identical Mössbauer spectra collected from both core and intermediate zone of each single crystal of tourmaline indicates only slight variations in next-nearest-neighbor populations in our tourmaline crystals. The quantitative interpretation of Mössbauer spectra yielded nearly all iron as divalent  $\langle$  <3% Fe<sup>3+</sup>/  $Fe<sub>tot</sub>$ ) for both core and intermediate zone, respectively (Fig. 7). Consequently, we could exclude the role of  $Fe<sup>3+</sup>$  fluctuation in zoning or a different pleochroism among particular zones; this highlights the role of Ti in the zoning observed (as indicated above; Fig. 5). Rims of the tourmaline crystals were not examined owing to their small size (Fig. 5). The results of Mössbauer spectroscopy are only slightly different from the results of wet-chemical analyses of Povondra (1981), where higher but still low contents of Fe3+, less than 0.2 *apfu*  $\left($  <13% Fe<sup>3+</sup>/Fe<sub>tot</sub>), were only exceptionally revealed.

#### *The LA–ICP–MS data*

A single crystal of tourmaline from a pocket in common pegmatite at the locality Suky (Figs. 5, 8, Table 3) with a well-developed zoning, including a relatively thick outer rim, was studied using LA–ICP–MS. The results show a quite complicated behavior of some trace elements related to the zonal development observed with the optical microscope and in BSE images of the crystal. Titanium is the only element showing increasing concentrations from core (413–1586 ppm) through intermediate zone (1573–3288 ppm) to outer rim (3268–6008 ppm), invariably with sharp contacts

(Fig. 8, Table 3). These concentrations are generally equal with those measured by EMP. Along with low contents of  $Fe^{3+}$ , these results suggest Ti, possibly through an intervalence charge-transfer interaction involving Fe<sup>2+</sup> and Ti (Pezzotta & Laurs 2011), is very likely a chromophore that causes a brown color (and pleochroism) of these otherwise compositionally comparable zones. The other elements commonly demonstrate very similar concentrations in core and intermediate zone, but distinct levels in the outer rim; Mn, Ni, Zn, Sc and Ga decrease, and V, Cr, Co, Sr and REE increase in the rim, respectively (Fig. 8). Some elements do not show any evident variation, and their concentrations within core, intermediate zone and outer rim are rather constant (P, Cu, Sn, Pb; see Table 3 and Fig. 8). The additional elements sought (Be, As, Y, Zr, Ag, Cd, Sb, Cs, Ba, Hf, Ta, W, Bi, Th, U) were found to be below or mostly close to their detection limits. Lithium was found at a concentration of about the detection limit  $(\sim 50 \text{ ppm})$  in all zones.





The compositions were established with electron-microprobe analyses. The symbols are as defined in Figure 4.



Fig. 6. Left: Compositional evolution of tourmalines in a plot of Fe<sub>tot</sub>/(Fe<sub>tot</sub>+Mg) *versus X*-site vacancy. Individual dikes of pegmatite as well as muscovite-rich veins are distinguished by different symbols; grey-colored symbols represent central parts, and white-colored symbols, the outer rim of tourmaline crystals. Right: Triangular plots showing *X*-site compositions (a), Fe–Al–Mg compositions (b), and Ti/Al plot (c) of the individual types of tourmaline from pegmatite dikes and muscoviterich veins. Colored symbols are related to the localities shown in Figure 5 and Table 1: blue circles: Dolní Bory, yellow triangles: Bobrůvka, red squares: Suky.



FIG. 7. Room-temperature <sup>57</sup>Fe Mössbauer spectrum of tourmaline sample from common pegmatite with pockets (locality Suky, Table 2): a) core, b) intermediate zone. White doublets:  $Fe^{2+}$ , grey:  $Fe^{3+}$  (see the text for more details).

#### **Discussion**

## *Compositional trends in zoned crystals of tourmaline*

Tourmaline crystals from common pegmatites with pockets exhibit rather primitive compositions, with low Li, Mn and F contents (Tables 1, 3; see also Povondra 1981, Novák *et al.* 2004a), but a highly variable Fe<sub>tot</sub>/  $(Fe<sub>tot</sub> + Mg)$  value, low to high Al, and low to moderate contents of Ti and Ca (Table 1, Fig. 6). Similar primitive compositions are typical of Al-rich metapelites (Henry & Dutrow 1996), and only rarely have they been described from granitic pegmatites (*e.g.,* Roda *et al*. 2005, Roda-Robles *et al*. 2011, Hezel *et al*. 2011, Novák *et al*. 2011). The core (foitite – schorl – magnesiofoitite) and intermediate zone (schorl–dravite), being volumetrically dominant in most crystals from pockets (Fig. 5), are compositionally similar, commonly showing a slight decrease in *X*-site vacancy and Al, but a significant increase of Ti, from the core to the intermediate zone. The  $Fe<sub>tot</sub>$  (Fe<sub>tot</sub> + Mg) value commonly decreases (only rarely increases) from the core to the intermediate zone (Fig. 6). However, the core compositions at the individual localities vary significantly, from foitite–schorl with  $Fe_{tot}/(Fe_{tot} + Mg) = 0.99$  at Hodíškov to dravite – magnesio-foitite with  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$  = 0.43 and 0.41 in the pegmatite Pikárec (Figs. 6a, b). The crystals, which evidently grew continuously from surrounding massive pegmatite units into pockets, have commonly a higher  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$  value, whereas crystals growing solely in pockets are enriched in Mg.

TABLE 2. ROOM-TEMPERATURE <sup>57</sup>Fe MÖSSBAUER HYPERFINE PARAMETERS FOR THE CORE AND INTERMEDIATE ZONE OF THE TOURMALINE CRYSTAL FROM POCKETS AT SUKY AND HODÍŠKOV \_



See Figure 7 and the text for more details). Notation: ä: isomer shift (±0.02 mm s<sup>-1</sup>),  $\Delta E_0$ : quadrupole splitting (±0.02 mm s<sup>-1</sup>),  $\Gamma$ : peak width (±0.02 mm  $s^{-1}$ ), RA: relative area in the spectrum ( $\pm 3\%$ ), and int.: intermediate.

Geochemical fractionation, documented mainly by an evident increase in Fe/(Fe+Mg), was indicated only in tourmaline from the contact zone of the blocky K-feldspar in the pegmatite Znětínek and in a zoned crystal of tourmaline from a pocket at the locality Bobrůvka (Figs. 5, 6a, b, c), where the  $Fe/(Fe + Mg)$ value increases significantly from core to intermediate zone. Most tourmaline crystals from pockets in common pegmatites and all crystals from muscovite-rich veins are more or less Mg-enriched, and such reverse compositional evolution in  $Fe<sub>tot</sub>/(Fe<sub>tot</sub> + Mg)$  distinct from those found in granitic pegmatites is striking (Fig. 6).

A late tourmaline (outer rims of crystals or fibrous aggregates) from granitic pegmatites with a dominant dravite component was found at some localities, *e.g.,* the contaminated elbaite-subtype pegmatite Bližná I (Novák 1998, Novák *et al*. 1999a, 2012) and an intragranitic NYF euxenite-type pegmatite Kožichovice III from the Třebíč pluton (Novák *et al*. 2010). The early tourmaline crystals at these localities (Mn-rich elbaite–liddicoatite at Bližná I and Ti-enriched dravite – schorl – uvite at Kožichovice III) are compositionally very distinct (*e.g.,* 0.33 and 1.25–1.41 *apfu* Mg, respectively). It seems that a Ca- and Ti-poor dravitic tourmaline with low to moderate  ${}^{Y}$ Al<sub>tot</sub> (~0.0–0.3 *apfu*), but high and variable *X*-site vacancy (0.2–0.5 *pfu*) is a stable composition at low-T conditions (see *e.g.,* Sperlich *et al*. 1996, Henry & Dutrow 1996). The outermost rim of tourmaline crystals from muscovite-rich veins,

TABLE 3. CONCENTRATION OF TRACE ELEMENTS (MOVING AVERAGE) FOR CORE, INTERMEDIATE ZONE AND OUTER RIM OF TOURMALINE FROM SUKY \_

| ele-<br>ment | core               |     |                | intermediate zone |     |                | rim                 |                |                | d.l.           |
|--------------|--------------------|-----|----------------|-------------------|-----|----------------|---------------------|----------------|----------------|----------------|
|              | range              | δ   | $\overline{x}$ | range             | δ   | $\overline{x}$ | range               | δ              | $\overline{x}$ |                |
| P            | 314<br>$150 -$     | 28  | 217            | $162 -$<br>370    | 33  | 225            | $160 -$<br>434      | 49             | 222            | 3              |
| Sc           | 251<br>$82 -$      | 27  | 133            | $80 -$<br>174     | 27  | 115            | 45<br>113<br>$\sim$ | 15             | 84             | 9              |
| Τi           | $413 - 1586$       | 217 | 988            | 1573 - 3288       | 217 | 2455           | $3268 -$<br>6008    | 650            | 4299           | 4              |
| v            | b.d.l.             |     |                | b.d.l.            |     |                | $277 -$<br>607      | 68             | 430            | $\mathbf 0$    |
| Cr           | 6<br>$1 -$         | 1   | 2              | 9<br>$1 -$        | 1   | 3              | $85 -$<br>270       | 37             | 149            | 1              |
| Mn           | $970 - 3482$       | 380 | 1864           | 1066 - 3535       | 380 | 2312           | 292<br>$129 -$      | 36             | 204            | 2              |
| Ni           | 884<br>$374 -$     | 107 | 579            | $350 - 888$       | 107 | 623            | $209 -$<br>384      | 46             | 286            | 0.3            |
| Co           | b.d.l.             | ٠   |                | b.d.l.            | ۰   |                | 17<br>$7 -$         | 2.6            | 11.6           | 0.1            |
| Cu           | 40<br>$14 -$       | 5   | 20             | $14 -$<br>50      | 4   | 22             | 34<br>$12 -$        | $\overline{4}$ | 18             | 4              |
| Zn           | 821<br>$286 -$     | 90  | 465            | 927<br>$306 -$    | 90  | 546            | $72 -$<br>194       | 27             | 119            | 3              |
| Ga           | $23 -$<br>56       | 8   | 39             | $62 - 203$        | 23  | 136            | $70 -$<br>212       | 24             | 124            | $\overline{2}$ |
| Sr           | b.d.l.             | ٠   |                | b.d.l.            | ٠   |                | $33 -$<br>98        | 13             | 58             | 0.2            |
| Sn           | 181<br>$70 -$      | 19  | 103            | 170<br>$77 -$     | 19  | 109            | 131<br>$57 -$       | 15             | 93             | $\overline{4}$ |
| Pb           | 0.76<br>$b.d.l. -$ | 0.1 | 0.26           | $0.2 -$<br>1.4    | 0.1 | 0.5            | $0.8 -$<br>3        | 0.5            | 1.6            | 0.1            |
| Nb           | $0.3 -$<br>2.1     | 0.3 | 1              | $0.8 -$<br>4.7    | 0.3 | 1.8            | 0.5<br>$b.d.l. -$   | 0.1            | 0.2            | 0.1            |
| ΣREE         | $0.2 -$<br>1.8     | 0.3 | 0.7            | $0.4 -$<br>4      | 0.3 | 2.1            | $3.6 -$<br>11.4     | $\overline{2}$ | 6.8            | 0.1            |

Notations: d.l.: detection limit, b.d.l.: below detection limit,  $\delta$ : standard deviation, x: average. Concentrations are quoted in ppm.



which are the latest and evidently crystallized from hydrothermal fluids, have mostly a very similar compositional signature (Fig. 6), except for elevated contents of Ti. Hence, only the very narrow outer rim is Ti-poor (≤0.10 *apfu*) and fits perfectly the compositions of the late dravite (Novák 1998, Novák *et al*. 1999a, 2010). Trace-element profiles from our LA–ICP–MS study of the sample from the locality Suky reveal distinct behavior within the individual zones (Fig. 8). However, except for Ti, all other elements have very similar to almost equal concentrations in the core and intermediate zone, whereas the rim is distinct (Table 3, Fig. 8).

On the basis of the compositional data (Table 1, Fig. 6) and the textural development of zoned crystals from pockets (Fig. 5), the crystallization of a moderately evolved melt in a closed system resulted in the core zone of the tourmaline crystals. The striking change in the zonal texture but only a weak change in chemical composition from the core to intermediate zone (except for Ti; Fig. 8, Table 3) indicate some change in a parent medium (*e.g.,* transition from a magmatic to a hydrothermal stage). Both core and intermediate zone crystallized in a somewhat closed system related to a primary process of growth. The crystallization of the outer rim of crystals (and whole crystals of tourmaline from muscovite-rich veins) proceeded in hydrothermal conditions in an open system, with the fluids of rather constant composition corresponding to a compositional uniformity of the country rocks. Also, fine oscillatory zoning in tourmaline, common mainly in the intermediate zone and less abundant in the outer rim (Fig. 5), is a feature found in many examples of hydrothermal tourmaline from magmatic–hydrothermal systems (*e.g.,* London & Manning 1995, Buriánek & Novák 2004); the associated pegmatites were the likely source of fluids. However, these results are only preliminary, and a more thorough chemical and isotopic study of fluid and solid inclusions is required to reveal the process of crystallization.

## *Geological position of common pegmatites and remarks on their classification*

The regional distribution of common pegmatites with pockets is not strictly consistent with all areas of regional migmatization within the Moldanubian Zone. They are evidently concentrated in the Strážek Unit (Fig. 1), where discordant rare-element-enriched pegmatites also are very abundant (Breiter *et al*. 2010, Cempírek *et al*. 2010). Unfortunately, a lack of suitable outcrops rule out a systematic structural and petrological study of the common pegmatites and their host rocks. Such an investigation would be required to determine the compositional-textural relations and relevant magmatic or metamorphic or anatectic or deformation event that may have generated these pegmatites. Nevertheless, relative to other regional units of the Moldanubian Zone, a fertility of the Strážek

Unit favoring the formation of more evolved and large pegmatites, commonly with pockets lined with crystals of feldspars, quartz, muscovite and various tourmalines (schorl, elbaite, liddicoatite, foitite), is evident. It is manifested by the occurrences of famous pegmatites, *e.g.,* the sekaninaite-bearing pegmatites with pockets at Dolní Bory (Černý *et al*. 1997), the pocket-bearing elbaite-subtype pegmatite from Řečice (Novák *et al*. 1999b, Novák 1999), lepidolite-subtype pegmatites from Rožná (Černý *et al*. 1995, Selway *et al*. 1998, Breiter *et al*. 2010), from Dobrá Voda (Selway *et al*. 1999, Novák & Taylor 2000) and from Laštovičky (Selway *et al*. 1999, Novák *et al*. 2004b).

The distribution of common pegmatites with pockets (Fig. 1), their geological position, as well as their commonly concordant character and transitional contacts with host rocks (Figs. 2a, 2b, 3), suggest that they may have originated during regional anatexis [authigenic pegmatites in the sense of London (2008)]. The transport of anatectic melts was likely insignificant to absent, especially in the case of small concordant pegmatite bodies with diffusive contacts. Also a relatively large, zoned pegmatite body from the locality Znětínek (this study) is concordant to foliation of the host migmatized biotite gneiss (Fig. 3), although relatively sharp contacts indicate transport of the pegmatite-forming melt. The source of B in the melt is not known, and its examination is beyond the scope of this paper. The presence of accessory tourmaline in the host migmatized gneisses and granulites indicates that its breakdown during anatectic melting may have supplied B (Kawakami 2001, van Hinsberg *et al.* 2011). Nevertheless, a pluton-derived origin of the common pegmatites cannot be excluded, although their regional distribution (Fig. 1) does not indicate any relation to potential parental plutons.

The prevailing concordant relationships of common pegmatites with pockets to host rocks (Figs. 2a, b), their zoned internal structure, the typical pegmatite textures (*e.g.,* graphic intergrowths of K-feldspar + quartz, blocky K-feldspar, quartz core; Fig. 3), and overall mineral assemblages suggest that the massive parts of common pegmatites crystallized from a melt. In contrast, the textural relations and mineral assemblages of most tourmaline crystals in pockets and chiefly from muscovite-rich veins indicate that they rather crystallized from hydrothermal fluids at a lower T. The mineral assemblages and overall geological and petrographic features indicate a wide range of P–T conditions at T  $\leq 500-650$ °C and P less than  $\sim 3-4$  kbar for massive units (with andalusite  $+$  cordierite) and lower T, down to  $\sim$ 400–200 $\degree$ C in the mineral assemblages of pockets characterized by primary muscovite I and common replacement of K-feldspar by the assemblage muscovite  $II +$  smoky quartz II. Abundant muscovite and the absence of K-feldspar in muscovite-rich veins imply lower P–T conditions, but these are not elucidated in detail.

The common pegmatites with pockets from the Moldanubian Zone have been rarely found as outcrops (Figs. 2a, b); these were mostly destroyed by mineral collectors. This makes their structural, geochemical and petrological study and discussion of origin as well as classification uncertain. They exhibit a simple zonal internal structure, textural–paragenetic units and mineral assemblages typical for common granitic pegmatites. Their spatial, compositional, mineralogical and genetic relations to anatexis (migmatization of gneisses and granulites) indicate that they may belong to the abyssal class of pegmatites (Černý & Ercit 2005, London 2008, Simmons & Webber 2008). However, the main textural difference that they exhibit relative to ordinary abyssal (anatectic) pegmatites is the presence of abundant pockets. The combination of evidently concordant and transitional (diffusive) contacts to host rocks and primitive chemical composition of minerals as typical features of abyssal pegmatites, and of common large pockets lined with large crystals of smoky quartz, feldspars, micas, and tourmaline as typical features of miarolitic pegmatites, does not fit the current classification of granitic pegmatites. However, it documents the extremely high variability of granitic pegmatites and their origin (London 2008).

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