

Original paper

# Secondary blue tourmaline after garnet from elbaite-subtype pegmatites; implications for source and behavior of Ca and Mg in fluids

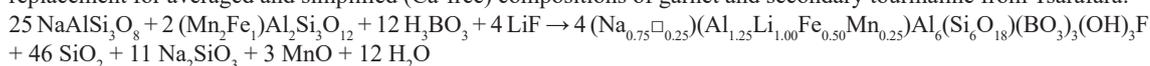
Zbyněk BUŘIVAL\*, Milan NOVÁK

Department of Geological Sciences, Masaryk University, Kotlářská 2, 611 37 Brno, Czech Republic; zbynek@burival.com

\* Corresponding author



Secondary blue tourmaline (schorl to Fe-rich fluor-elbaite to very rare Fe-rich fluor-liddicoatite) with quartz partially replace spessartine–almandine garnet and albite in elbaite-subtype pegmatites cutting pyroxene gneisses and calcite or dolomite marbles (at Tamponilapa and Tsarafara-Nord, Sahatany Valley, Madagascar) and paragneisses (at Ctidružice, Moldanubian Zone, Czech Republic). Only garnet from the albite adjacent to an unit with Li-bearing minerals (Li-micas, Li-tourmalines) underwent this alteration, whereas associated primary tourmaline (schorl to Mg-bearing schorl) remained unaltered. Textural relations and chemical composition of the individual minerals suggest the following equation of replacement for averaged and simplified (Ca-free) compositions of garnet and secondary tourmaline from Tsarafara:



Elevated contents of Al, Fe, Na and Mn in secondary tourmaline were likely sourced from the replaced garnet and albite, whereas the residual fluids supplied B, F, Li, and H<sub>2</sub>O. Garnet and associated primary tourmaline are rather Mg-rich but the secondary tourmaline is typically Mg-free. The contents of Ca are high in both tourmaline generations, but the secondary one is occasionally even more enriched compared to the associated primary tourmaline. Such a behavior of Mg and Ca suggests that no Mg was externally supplied during primary crystallization of primary tourmaline from the moment when pegmatite melt was sealed off the host rock. Negligible to none concentrations of Mg in secondary tourmaline show that the pegmatite system was closed to the host rocks during the hydrothermal alteration producing the secondary tourmaline generation. Evident absence of external Mg-contamination from host Ca, Mg-rich rock rules out also any contamination by Ca. High contents of Ca in primary tourmaline and garnet are related to originally Ca-enriched pegmatite melt contaminated before emplacement.

**Keywords:** tourmaline, garnet, subsolidus alterations, contamination, elbaite pegmatite

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## 1. Introduction

Hydrothermal replacement processes are common in many granitic pegmatites from primitive to highly fractionated ones (London 2008). They demonstrate exsolution of fluids from pegmatite melts; however, the timing of this fluid exsolution is still a matter of debate. The genetic hypotheses vary from coexistence of melt and fluid throughout pegmatite crystallization (e.g., Burnham and Nekvasil 1986; Veksler et al. 2002; Thomas et al. 2009) to exsolution of fluids at the end of primary magmatic crystallization, when pegmatite melt has been already completely crystallized (e.g., London 2008). Pseudographic replacement of coarse minerals by fine-grained assemblages evidently manifests subsolidus hydrothermal alteration. Products of alteration of primary minerals (e.g., Černý 2002; Roda-Robles et al. 2004; Gadas et al. 2016) as well as hydrothermal mineral assemblages in exocontact zones (e.g., Shearer et al. 1986; Morgan and London 1987; Novák et al. 1999) may provide valuable

information about chemical composition of fluids as well as P–T–X conditions of these alteration processes (London 2008).

Alteration processes in granitic pegmatites affect mainly less-refractory minerals such as spodumene (e.g., London and Burt 1982; Rao et al. 2012; Novák et al. 2015a; Cempírek et al. 2016), cordierite/sekaninaite (e.g. Schreyer et al. 1993; Jobin-Bevans and Černý 1998; Gadas et al. 2016), beryl (Černý 2002), andalusite (e.g., Ahn and Buseck 1988), and primary Li,Fe,Mn-phosphates (e.g., Dill 2009; Roda-Robles et al. 2012; Baijot et al. 2014). These primary coarse minerals are locally completely replaced by a fine-grained mineral assemblage or a single mineral.

Garnet is typically stable in granitic pegmatites. Its rare alterations as thin reaction rims composed of the assemblage tourmalines + micas + quartz are restricted solely to Li-bearing pegmatites (Němec 1982, 1983a, b, 1989; Felch et al. 2016). In this paper, we examine mineral assemblages and chemical composition of tourmaline replacing garnet from elbaite-subtype pegmatites

Tamponilapa and Tsarafara-Nord, Sahatany Valley, Madagascar and Ctidružice, Moldanubian Zone, Czech Republic cutting different host rocks. The main aim has been to reveal mechanism of replacement reactions, composition of residual fluids as well as a role of external contamination in the pegmatites.

## 2. Geological setting

### 2.1. Pegmatite fields of Central Madagascar

The pegmatites of the *Sahatany Valley pegmatite field* are located in the north-eastern part of the Itremo Region, Central Madagascar built by low- to medium-grade metamorphic rocks of the Itremo Complex (e.g., Cox et al. 1998). This region is characterized by numerous occurrences of Li-enriched gem-bearing pegmatites (e.g., Ranorosa 1986; Pezzotta 2005), occasionally with large pockets lined with attractive crystals of gemmy elbaite–liddicoatite (e.g., Estatoby, Tsarafara; Pezzotta and Praszker 2013). The mineral assemblages with elbaite–liddicoatite along with minor spodumene and/or lepidolite suggest dominance of elbaite-subtype pegmatites in this district (see Ranorosa 1986; Novák et al. 2015a). Mineral assemblages of most pegmatites hosted by calc-silicate rocks (pyroxene gneiss) or dolomite and calcite marbles also include common danburite, diopside–hedenbergite, axinite-group minerals, titanite, Mg-enriched tourmaline and Ca-enriched garnet. The presence of these minerals demonstrates external Ca, Mg contamination of the pegmatite melt (Novák 2007, 2013).



The *Tamponilapa and Tsarafara-Nord pegmatites* form subhorizontal dikes, c. 2–3 m thick, cutting calc-silicate rocks with intercalations of quartzite. Pegmatite fragments from dumps of the Tsarafara-Nord mine show common Li-mica along with Li-tourmaline. In the well-exposed Tamponilapa pegmatite (Fig. 1) the following textural-paragenetic units were identified: medium- to coarse-grained outer granitic unit with biotite, graphic unit (Kfs + Qz) locally with tourmaline and quartz graphic intergrowths, blocky K-feldspar, medium- to coarse-grained albite/cleavelandite unit and rare pockets with Li-tourmaline and hambergite crystals. All units except for the granitic unit, as well as enclaves of the host rock, are rather randomly distributed in the pegmatite (Fig. 1). In the current outcrop, Li-tourmalines were found only scarcely as narrow green to pink rims around black tourmaline. Micas are rare except for biotite partly replaced by fibrous dark blue ferroholmquistite. Reaction rim with black dravite and violet holmquistite is locally developed at the exocontact with carbonate and calc-silicate rock. Accessory minerals include garnet, beryl, danburite, Li-mica, spodumene, axinite, zircon, columbite, microlite, cassiterite, topaz, fluorapatite, magnetite, and rutile (Ranorosa 1986; unpublished data of the authors).

### 2.2. Moldanubian pegmatite field

The Moldanubian pegmatite field is characterized by the occurrence of numerous granitic pegmatites of different origin and mineralogy (e.g. Novák et al. 2013, 2015b). The pegmatites of the rare-element class are the most abundant and exhibit high variability in size, textural differentiation, degree of fractionation and mineralogy from small, simple barren to large, highly fractionated pegmatites typically with LCT signature. Lepidolite-subtype pegmatites predominate over elbaite-subtype pegmatites (Novák and Povondra 1995).

Pegmatite from *Ctidružice* (dated at  $323 \pm 5$  Ma; U–Pb columbite; Melleton et al. 2012) forms subvertical, symmetrically zoned dike, about 2–3 m thick, cutting biotite paragneiss (Čech 1962). From the contact to the core, following units are developed: medium- to coarse-grained granitic unit with biotite, volu-

**Fig. 1** Pegmatite outcrop at the Tamponilapa, Sahatany Valley, Madagascar. A hammer shaft is 35 cm long.

metrically dominant graphic unit (Kfs+Qz±Bt or Tur), coarse-grained albite unit (Ab+Qz±Ms, Tur) locally with blocks of brownish K-feldspar and fine-grained greenish saccharoidal albite unit locally penetrating other ones. Prismatic crystals of pink Li-tourmaline and coarse flakes of colorless to violet Li-mica occur in the most evolved parts of the albite unit and typically are associated with brownish graphic and blocky K-feldspar. Scarcely, pink elbaite occurs also in the saccharoidal albite unit. Accessory minerals include garnet, beryl, zircon, columbite, cassiterite, stokesite, F-rich hambergite, and fluorapatite (Čech 1962; Novák et al. 1998). Němec (1983a) described reaction rims of masutomilite around garnet at this locality.

### 3. Experimental

#### 3.1. Chemical analyses

Chemical composition of all minerals was studied using the Cameca SX-100 electron microprobe (Laboratory of Electron Microscopy and Microanalysis, the joint facility of the Masaryk University and Czech Geological Survey in Brno; operator P. Gadas). Following analytical conditions were applied: accelerating voltage 15 kV, beam diameter 1–5 µm, beam current 10–20 nA. Following standards were used for tourmaline and garnet ( $K_{\alpha}$  lines if not specified otherwise): albite (Na), sanidine (Si, Al, K), pyrope (Mg), titanite (Ti), ScVO<sub>4</sub> (Sc, V), chromite (Cr), vanadinite (Cl), fluorapatite (P), wollastonite (Ca), almandine (Fe), spessartine (Mn), gahnite (Zn), topaz (F). For garnet, some standards were added ( $L_{\alpha}$  lines): YPO<sub>4</sub> (Y), zircon (Zr). Raw data were reduced using the PAP correction (Pouchou and Pichoir 1985). Some of the electron-microprobe analyses from Ctídrůžice were performed on the Cameca Camebax SX-50 electron microprobe, University of Manitoba, Winnipeg with a beam diameter of 4–5 µm and an accelerating voltage of 15 kV (for more details see e.g. Selway et al. 1999).

#### 3.2. Mineral formula calculation

Crystal-chemical formulae of tourmaline were calculated assuming the general formula

$XY_3Z_6T_6O_{18}(BO_3)_3V_3W$ , where X = Na, Ca, K, vacancies; Y = Fe, Mg, Mn, Ti, Al, Zn; Z = Al, Fe, Mg; T = Si; B = B; V + W = OH + F + O = 4 (Henry et al. 2011). In case of Li-free tourmaline, we used calculation on the basis of  $\Sigma T + Z + Y = 15$ ; ratio of Fe<sub>2</sub>O<sub>3</sub> and FeO was adjusted to fit an electroneutral formula. Tourmaline formulae for Li-bearing samples were calculated on the basis of 31 anions (O, OH, F), assuming B = 3 *apfu*, and Li = 15 – (Y + Z + T). Crystal-chemical formulae of garnets were calculated on the basis of 12 anions and 8 cations with ratio of Fe<sub>2</sub>O<sub>3</sub> and FeO calculated to fit an electroneutral formula. All mineral abbreviations in the text, tables and figures were taken from Whitney and Evans (2010).

### 4. Results

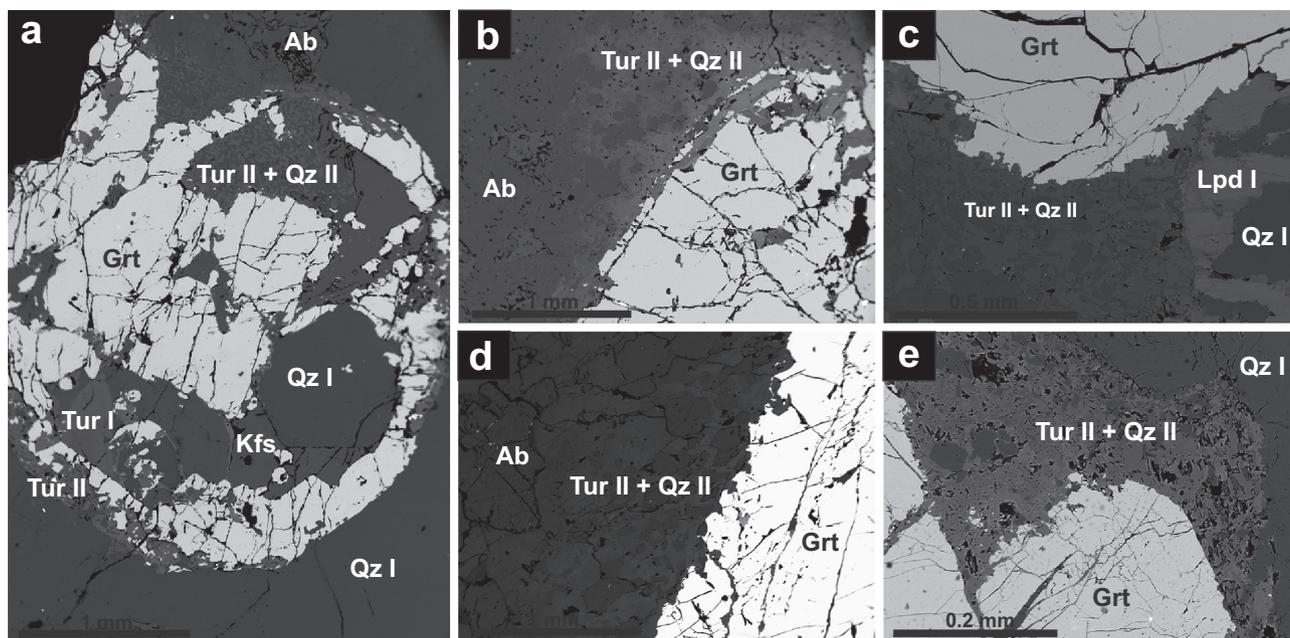
#### 4.1. Paragenetic position of garnet and its chemical composition

Garnet-bearing primary mineral assemblages in the studied pegmatites include albite, quartz, spessartine-dominant garnet (Figs 2–3), black primary tourmaline (schorl to Mg-rich schorl), K-feldspar and locally either minor Li-mica, Mn-rich fluorapatite, and columbite (Ctídrůžice) or fluorapatite, spodumene, and danburite (Tamponilapa).

Only in the Ctídrůžice pegmatite, rare coarse flakes of violet Li-mica and prismatic grains of pink Li-tourmaline are associated with red to orange-red spessartine (Fig. 2) that forms rather rare equidimensional subhedral grains and scarcely also quartz–garnet graphic intergrowths, up to 1–2 cm in size, typically in Li-bearing, albite-rich units. This garnet is not hydrothermally altered.

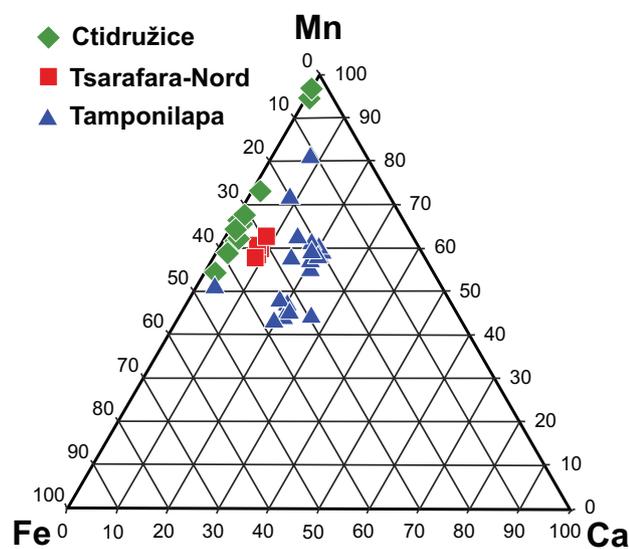


**Fig. 2** Garnet in albite, rimmed by secondary blue tourmaline from Tamponilapa.



**Fig. 3** Back-scattered electron images of replacement textures after garnet built by fine-grained assemblage secondary tourmaline + quartz on the contact with albite. **a** – Skeletal garnet from Tamponilapa; note none to weak replacement near primary quartz or primary tourmaline; **b** – Tamponilapa; **c** – Ctídužice; **d** – Tšarafara-Nord; note absence of replacement on the contact with primary quartz and lepidolite; **e** – Ctídužice.

The individual garnet crystals from Madagascar localities are rather heterogeneous (Fig. 4) and garnet from Tamponilapa is typically Ca-enriched (up to 0.78 *apfu*), whereas garnets from Tšarafara-Nord and especially from Ctídužice are Ca-poor ( $\leq 0.07$  *apfu*). The contents of Mg are negligible in Tšarafara-Nord and Ctídužice, whereas the Tamponilapa garnet is slightly Mg-enriched ( $\leq 0.09$  *apfu*). Concentrations of P, Na, Y and Sc are below or close to the detection limits of electron microprobe analyses.

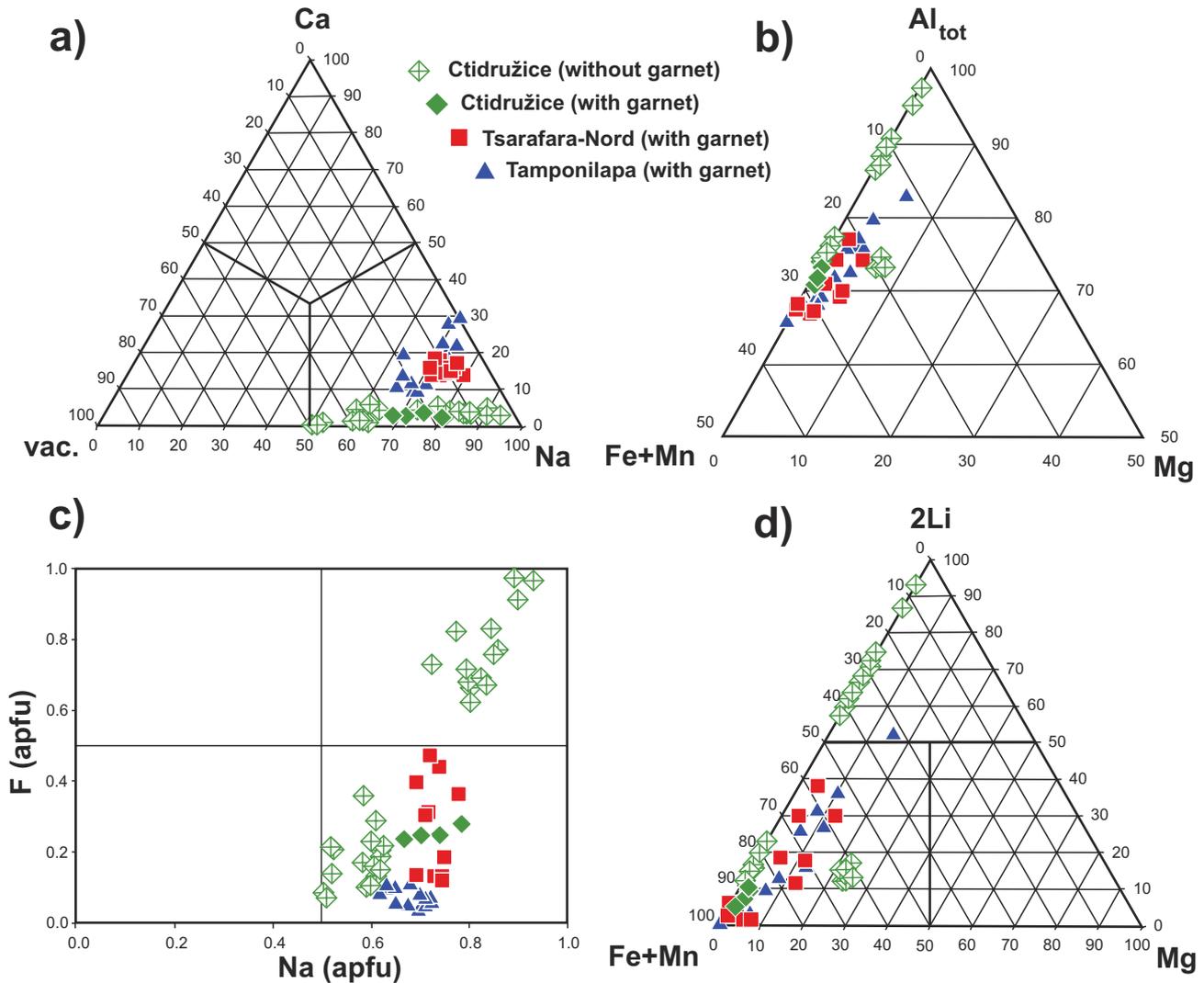


**Fig. 4** Ternary plots of the Fe, Mn and Ca contents in garnets (*apfu*).

#### 4.2. Primary tourmaline and its chemical composition

Chemical composition of primary tourmaline from all textural–paragenetic pegmatite units was studied only in the Ctídužice pegmatite (Fig. 5). This pegmatite contains black tourmalines (Mg-schorl to schorl) in the graphic and intermediate albite units and mostly pink to red and rare green tourmaline (fluor-elbaite) in the albite unit associated with common Li-mica (Mn-rich polyolithionite; Zahradníček and Novák 2012). Such evolution is typical of elbaite-subtype pegmatites (see Novák and Povondra 1995) with rather low Mg ( $\leq 0.69$  *apfu*) in tourmaline from outermost unit and high Mn ( $\leq 0.95$  *apfu*) in pink fluor-elbaites from inner albite unit (Tab. 1, Fig. 5). Low Ca content and vacancy in the X-site are also characteristic as is variable, but frequently high, F (0.11–0.96 *apfu*; Fig. 5c).

Prismatic crystals and their aggregates of primary black tourmaline scarcely associated with the altered garnet are locally in a direct sharp contact (Fig. 3a). No replacement textures were observed; hence, equilibrium between both minerals is very likely. Their compositions given in Fig. 5 show heterogeneous schorl with variable contents of Mg, Ca and F; low Ca but rather elevated Mg in Ctídužice ( $\leq 0.06$  *apfu*;  $\leq 0.60$  *apfu*) in contrast with moderate Ca but low Mg in Tamponilapa (0.09–0.29 *apfu*; 0.19–0.54 *apfu*) and in Tšarafara-Nord (0.14–0.18 *apfu*; 0.13–0.46 *apfu*). The localities differ in concentrations of F – very low in Tamponilapa (0.04–0.11 *apfu*) but high to moderate in Tšarafara-Nord and Ctídužice (0.13–0.48 *apfu*; 0.11–0.29 *apfu*).



**Fig. 5** Plots of the chemical composition of primary tourmalines. **a** – Vacancy–Ca–Na, in position X; **b** – Fe + Mn–Al–Mg in positions Y and Z; **c** – Na in position X vs. F in position W; **d** – Fe + Mn–2Li (calc.)–Mg in positions Y and Z (*apfu*).

#### 4.3. Textural relations of garnet hydrothermal replacement

Replacement reactions were observed only in garnet grains located close to the contact of the host Li-free/poor albite unit with the pegmatite unit enriched in Li (minor to common Li-mica and/or Li-tourmaline); otherwise, garnet remains unaffected. The replacement takes place mainly along the direct contact of garnet with albite and scarcely also with K-feldspar (Fig. 3); none to weak reactions were observed next to quartz (Fig. 3a). Secondary assemblages replacing garnet and albite consist dominantly of a fine-grained aggregate of mostly dark blue secondary tourmaline and quartz. In the Ctidružice pegmatite, replacement of garnet by Li-mica (masutomilite) was found (Němec 1983a; unpublished data of the authors). Primary tourmaline locally associated with altered garnet was not affected by alteration (Fig. 3a).

#### 4.4. Chemical composition of secondary tourmaline

Secondary, dark blue to locally black secondary tourmaline (schorl to Fe-rich fluor-elbaite to very rare Fe-rich fluor-liddicoatite; Tab. 2, Fig. 6) replacing garnet and albite (Fig. 3) is characterized by low to negligible Mg and Ti, moderate but variable Fe, and relatively elevated but variable Al, Mn, Li and F compared to the primary tourmaline associated with the garnet (Figs 5–6).

Only Ca and Na exhibit evidently different behavior in the individual localities (Fig. 6a). Calcium is low (0.02–0.06 *apfu*) in Ctidružice but Madagascar tourmalines all show significantly higher contents (0.07–0.43 *apfu* and 0.09–0.60 *apfu* in Tamponilapa and Tsarafara-Nord, respectively). These values are slightly (Ctidružice) to significantly (Tsarafara-Nord) higher relative to the primary tourmaline associated with the garnet.

**Tab. 1** Electron-microprobe analyses of selected primary tourmalines associated with altered garnets (wt. % and *apfu*)

Locality Sample	Tamponilapa			Tsarafara-Nord			Ctidružice		
	MDG5/53	MDG4/19	Mean (13)	MDG1/61	MDG1/65	Mean (12)	CT6/4	CT6/22	Mean (4)
SiO <sub>2</sub>	33.32	36.31	34.49	34.53	36.88	34.87	34.46	34.63	34.73
TiO <sub>2</sub>	0.24	1.17	0.46	0.88	0.57	0.78	0.53	0.19	0.42
B <sub>2</sub> O <sub>3</sub> *	9.76	10.70	10.15	10.02	10.78	10.16	10.15	10.32	10.26
Al <sub>2</sub> O <sub>3</sub>	26.83	35.42	30.99	28.24	36.45	30.28	31.19	33.18	32.27
FeO	20.27	5.95	14.55	18.18	3.72	14.94	17.07	16.61	16.59
MnO	0.84	1.18	1.09	0.42	1.71	0.80	0.80	0.66	0.73
ZnO	0.00	0.00	0.06	0.00	0.00	0.04	0.00	0.15	0.04
MgO	0.97	1.84	1.11	1.55	0.39	0.97	0.22	0.11	0.16
Li <sub>2</sub> O*	0.00	1.18	0.36	0.02	1.89	0.46	0.15	0.11	0.17
CaO	1.44	0.67	0.92	0.84	1.97	0.92	0.14	0.15	0.15
Na <sub>2</sub> O	1.97	2.17	2.08	2.06	1.86	2.16	2.37	2.16	2.21
K <sub>2</sub> O	0.05	0.00	0.02	0.06	0.00	0.05	0.04	0.05	0.05
F	0.09	0.22	0.13	0.25	1.33	0.58	0.52	0.47	0.48
H <sub>2</sub> O*	3.32	3.59	3.44	3.34	3.09	3.23	3.26	3.34	3.31
(O=F)	-0.04	-0.09	-0.05	-0.11	-0.56	-0.24	-0.22	-0.20	-0.20
Total	99.06	100.30	99.79	100.32	100.07	100.00	100.74	101.90	101.41
T Si	5.933	5.895	5.907	5.992	5.948	5.967	5.903	5.834	5.882
T Al	0.067	0.105	0.093	0.008	0.052	0.033	0.097	0.166	0.118
B	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Z Al	5.564	6.000	5.899	5.767	6.000	5.925	6.000	6.000	6.000
Z Mg	0.258	0.000	0.080	0.233	0.000	0.075	0.000	0.000	0.000
Z Fe <sup>3+</sup>	0.177	0.000	0.022	0.000	0.000	0.000	0.000	0.000	0.000
Z Fe <sup>2+</sup>	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000
Y Ti	0.032	0.143	0.059	0.115	0.069	0.101	0.068	0.024	0.054
Y Al	0.000	0.672	0.253	0.000	0.877	0.142	0.200	0.423	0.323
Y Fe <sup>3+</sup>	0.199	0.000	0.027	0.000	0.000	0.007	0.000	0.000	0.000
Y Fe <sup>2+</sup>	2.820	0.807	2.073	2.638	0.502	2.146	2.445	2.340	2.351
Y Mn	0.127	0.162	0.158	0.062	0.234	0.115	0.117	0.094	0.105
Y Zn	0.000	0.000	0.008	0.000	0.000	0.006	0.000	0.019	0.005
Y Mg	0.000	0.446	0.203	0.168	0.093	0.173	0.057	0.028	0.041
Y Li	0.000	0.770	0.240	0.017	1.226	0.311	0.107	0.073	0.117
X Ca	0.274	0.117	0.170	0.156	0.341	0.168	0.026	0.026	0.028
X Na	0.680	0.683	0.690	0.694	0.581	0.719	0.787	0.705	0.727
X K	0.012	0.000	0.005	0.014	0.000	0.010	0.009	0.011	0.011
X vac	0.034	0.200	0.136	0.137	0.078	0.104	0.177	0.258	0.234
V OH	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
V O	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
W F	0.052	0.110	0.070	0.139	0.676	0.308	0.281	0.249	0.256
W OH	0.948	0.890	0.930	0.861	0.324	0.692	0.719	0.751	0.744
W O	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000

\*P, V, Cr, Cl below their respective detection limits

Secondary tourmalines from the individual localities also differ in the concentrations of F (Fig. 6c) from very low in Tamponilapa ( $\leq 0.20$  *apfu*) to high or very high in Tsarafara-Nord and Ctidružice (0.48–0.82 *apfu*; 0.55–1.00 *apfu*). In Tsarafara-Nord, secondary tourmaline is strongly F-enriched relative to primary tourmaline (Figs 5–6).

Lastly, the individual localities differ in concentrations of Fe + Mn that tend to be slightly higher in Ctidružice (Fig. 6d).

## 5. Discussion

### 5.1. Primary and secondary mineral assemblages

Primary mineral assemblages with spessartine-dominant garnet (Fig. 4) contain albite, quartz and black primary tourmaline (schorl to Mg-rich schorl; Fig. 5). The host pegmatite unit mostly does not contain Li-bearing minerals and shows moderate degree of fractionation within the

**Tab. 2** Electron-microprobe analyses of selected secondary tourmalines replacing garnets (wt. % and *apfu*)

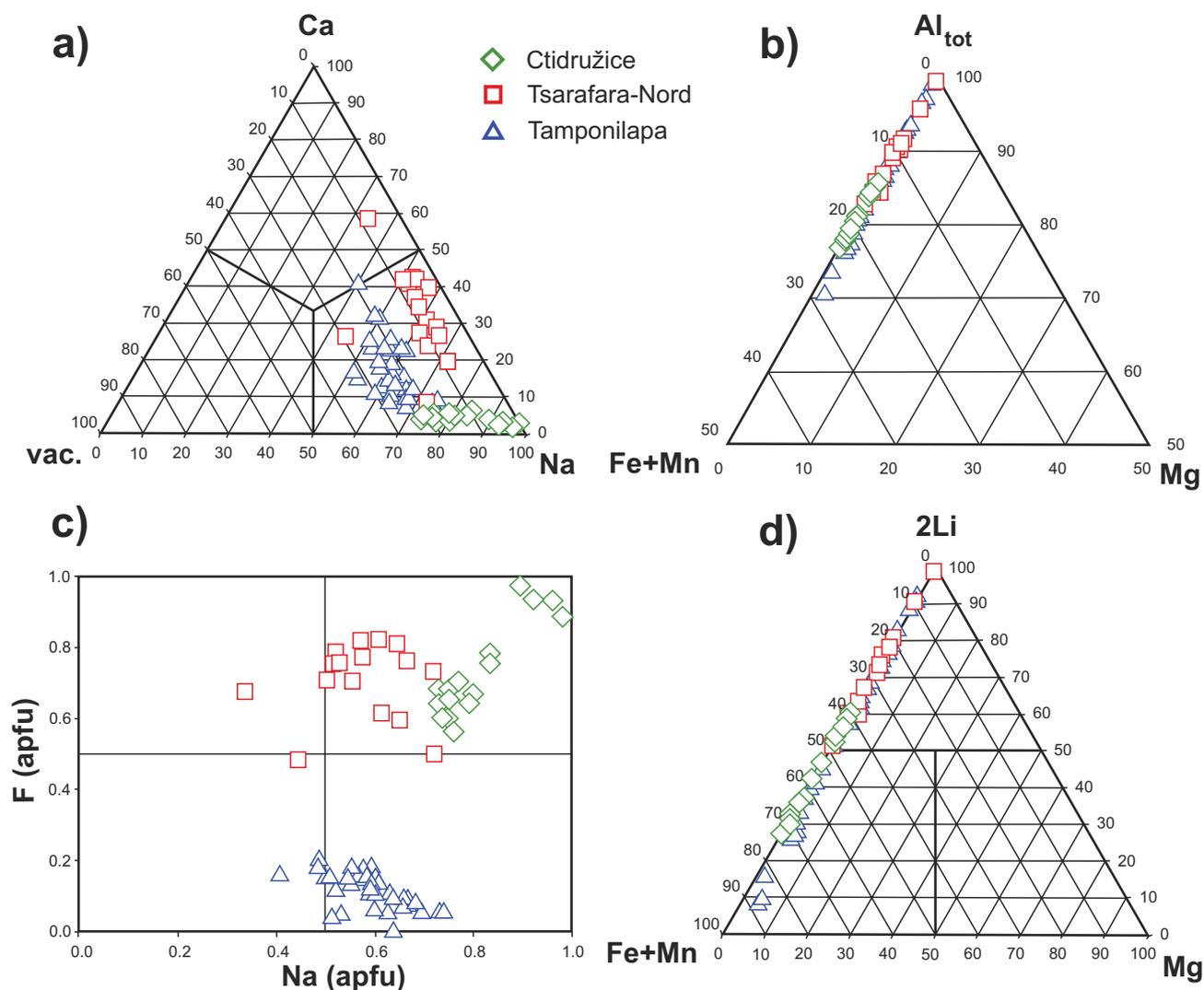
Locality Sample	Tamponilapa			Tsarafara-Nord			Ctidružice		
	MDG5/18	MDG4/84	Mean (43)	MDG1/59	MDG1/58	Mean (16)	GM3/30	CT6/2	Mean (18)
SiO <sub>2</sub>	34.91	34.70	35.74	38.13	36.54	36.77	35.39	35.62	35.51
TiO <sub>2</sub>	0.00	0.10	0.02	0.00	0.00	0.03	0.00	0.11	0.09
B <sub>2</sub> O <sub>3</sub> *	10.82	10.13	10.59	11.07	10.59	10.75	10.47	10.49	10.51
Al <sub>2</sub> O <sub>3</sub>	41.54	31.14	36.86	40.60	35.06	36.92	36.33	34.23	35.38
FeO	0.29	15.81	6.53	0.00	8.36	3.86	4.64	9.72	7.33
MnO	1.46	1.48	1.99	0.24	2.34	2.21	3.95	4.75	4.40
ZnO	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.50	0.10	0.00	0.00	0.02	0.00	0.00	0.02
Li <sub>2</sub> O*	2.00	0.18	1.22	2.32	1.13	1.82	1.23	0.58	0.87
CaO	2.35	0.44	0.96	1.57	0.49	1.91	0.13	0.30	0.22
Na <sub>2</sub> O	1.31	2.20	1.88	1.46	2.26	1.82	2.88	2.32	2.54
K <sub>2</sub> O	0.00	0.04	0.02	0.00	0.04	0.01	0.00	0.06	0.05
F	0.31	0.10	0.21	0.98	0.98	1.38	1.91	1.15	1.41
H <sub>2</sub> O*	3.59	3.45	3.55	3.36	3.19	3.05	2.71	3.08	2.95
(O=F)	-0.13	-0.04	-0.09	-0.41	-0.41	-0.58	-0.80	-0.48	-0.60
Total	98.45	100.23	99.60	99.29	100.55	99.98	98.83	101.92	100.70
T Si	5.607	5.952	5.865	5.989	6.000	5.946	5.873	5.903	5.874
T Al	0.393	0.048	0.135	0.011	0.000	0.054	0.127	0.097	0.126
B	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Z Al	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000
Z Mg	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Z Fe <sup>3+</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Z Fe <sup>2+</sup>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Y Ti	0.000	0.013	0.002	0.000	0.000	0.004	0.000	0.014	0.012
Y Al	1.469	0.248	0.984	1.505	0.785	0.982	0.978	0.588	0.772
Y Fe <sup>3+</sup>	0.000	0.000	0.000	0.000	0.060	0.006	0.000	0.000	0.000
Y Fe <sup>2+</sup>	0.039	2.269	0.914	0.000	1.087	0.520	0.644	1.346	1.016
Y Mn	0.198	0.214	0.277	0.031	0.325	0.303	0.556	0.666	0.616
Y Zn	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000
Y Mg	0.000	0.129	0.025	0.000	0.000	0.005	0.000	0.000	0.004
Y Li	1.293	0.128	0.795	1.464	0.743	1.180	0.823	0.385	0.578
X Ca	0.405	0.081	0.168	0.264	0.086	0.331	0.024	0.053	0.040
X Na	0.407	0.732	0.599	0.443	0.719	0.573	0.925	0.747	0.816
X K	0.000	0.009	0.003	0.000	0.008	0.002	0.000	0.014	0.010
X vac	0.188	0.178	0.230	0.293	0.188	0.095	0.051	0.187	0.135
V OH	3.000	3.000	3.000	3.000	3.000	3.000	2.999	3.000	3.000
V O	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000
W F	0.156	0.052	0.109	0.484	0.509	0.708	1.001	0.601	0.739
W OH	0.844	0.948	0.890	0.516	0.491	0.292	0.000	0.399	0.261
W O	0.000	0.000	0.000	0.000	0.000	0.000	-0.001	0.000	0.000

\*P, V, Cr, Cl below their respective detection limits

pegmatite body. Garnet grains that underwent alteration are typically adjacent to the neighboring Li-bearing unit (with Li-mica and/or pink Li-tourmaline) and the distance between a Li-rich mineral and the altered garnet ranges from several mm (Fig. 2) to ~10 cm. These textural relations are very similar to those described from the Mt. Mica pegmatite (Felch et al. 2016); however, a narrow zone of black schorl developed between replaced garnet and dominant blue tourmaline rim was not found at our localities. In lepidolite-subtype pegmatites of the Moldanubian Zone, garnets from albite unit are also replaced

by zinnwaldite/masutomilite mica plus minor blue tourmaline (Němec 1983a; unpublished data of the authors); otherwise, textural features of replacement are similar to the elbaite-subtype pegmatites.

The textural features described above suggest that the fluids promoting replacement of garnet were very likely generated in the adjacent Li-bearing albite unit and that they were active only up to the distance of ~10 cm. In the studied pegmatites, as well as in the Mt. Mica pegmatite (Felch et al. 2016) and other Moldanubian Zone pegmatites (Němec 1982, 1983a, b), most garnet was

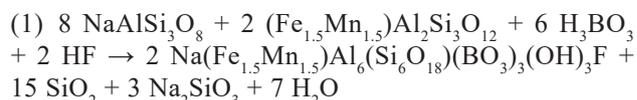


**Fig. 6** Plots of the chemical composition of secondary tourmalines. **a** – Vacancy–Ca–Na, in position X; **b** – Fe + Mn–Al–Mg in positions Y and Z; **c** – Na in position X vs. F in position W; **d** – Fe + Mn–2Li (calc.)–Mg in positions Y and Z (apfu).

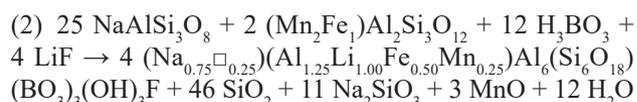
not affected by any alteration and its replacement in less evolved pegmatites without Li-bearing minerals has not been described. Hence, only highly evolved pegmatites with Li-bearing minerals may generate fluids aggressive enough to replace garnet. Timing of the fluid exsolution was likely immediately after, or simultaneously with, crystallization of rare Li-bearing minerals from the albite unit and from the adjacent Li-rich unit.

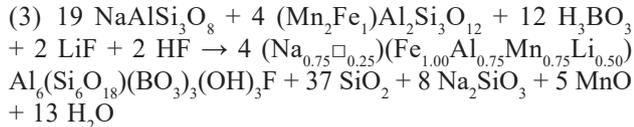
## 5.2. Replacement reactions and composition of fluids

The textural relations showing replacement of garnet and albite by tourmaline and quartz suggest the following simplified reaction. It was derived assuming immobile Al, idealized Ca-free mineral compositions, fixed Fe/Mn ratio and simple fluids enriched only in H<sub>2</sub>O, B and F. For Li-free compositions of secondary tourmaline the equation is:



However, the X-site in secondary tourmalines is often partly vacant, contains Ca, and Y, Z-sites are always enriched in Li and Al (Fig. 6a, d). Iron is preferred in the tourmaline structure (Fig. 6), whereas garnet is enriched in Mn (Fig. 4). Based on the average compositions of garnet and Ca-free secondary tourmaline, simplified reactions can be derived for two distinct compositions of the secondary Li-bearing tourmaline (2 – Tsarafara; 3 – Ctidružice):





We suggest a direct reaction of residual fluids with primary garnet and albite; consequently, the elevated contents of Al, Fe, Na and Mn in secondary tourmaline and Si in secondary quartz were very likely sourced from primary garnet and albite. The residual fluids supplied B, F, Li, and H<sub>2</sub>O; however, presence of Al, Fe, Na and Mn in the reacting fluids is not excluded. The excess of Na and Mn in the reactions (2) and (3) may be dissolved in fluids and transported away (in e.g., soluble hydrous sodium silicates) because no secondary Mn-bearing minerals occur close to the replaced garnet. However, the basic assumption for the equations that Al is immobile may not necessarily be correct.

Boron, F and Li are major components of exsolved fluids in granitic pegmatites along with H<sub>2</sub>O (see also London 2008). Presence of these volatiles in exsolved fluids has been considered by many authors (e.g., Veksler et al. 2002; Simmons and Webber 2008; London 2008; Thomas et al. 2009). Indeed, fluids with similar volatiles likely facilitated hydrothermal alterations in exocontact in some large granitic pegmatites such as Tanco, Manitoba (Morgan and London 1987).

### 5.3. Contamination and behavior of Mg and Ca

The Tamponilapa and Tsarafara-Nord pegmatites cut Ca, Mg-rich rocks and also the host Itremo Complex contains abundant calc-silicate rocks as well as calcite and dolomite marbles (Cox et al. 1998). Consequently, external contamination of pegmatite melts was feasible as documented by the chemical composition of tourmaline and garnet (Figs 4–5) and by the occurrence of danburite, diopside-hedenbergite and axinite (Ranoroosa 1986; Novák 2007, 2013). Primary tourmaline associated with garnet is only moderately enriched in Mg but elevated contents of Ca are evident in primary tourmaline and also in garnet (Fig. 4) except for the Ctidružice pegmatite. In contrast, secondary tourmaline is almost Mg- and Ti-free but occasionally even more enriched in Ca (Fig. 6a). Consequently, Mg and Ca behaved in rather different way in primary and secondary tourmalines and they are discussed separately.

Magnesium is usually exhausted during crystallization of early minerals (biotite, tourmaline) in outer pegmatite units. In moderately fractionated pegmatite units (e.g., albite units), Mg is depleted (e.g., Jolliff et al. 1986; Selway et al. 1999; Novák et al. 2017). Pyrope component in garnet is generally confined to high pressures. Therefore, in the more evolved granitic

pegmatites that crystallized in low P conditions (< 2–4 kbars), Mg content in garnet is typically very low even if this element is present in associated minerals (e.g., Baldwin and von Knorring 1983; London 2008; Müller et al. 2012; Hönig et al. 2014). At the studied localities, primary tourmaline is typically Mg-poor and in the most evolved portions of albite unit even Mg-free. Consequently, no Mg was supplied during primary crystallization of tourmaline from the moment when pegmatite melt was sealed off the host rock.

Negligible contents of Mg in secondary tourmaline suggest that this element was not present in the replacing fluids and that the pegmatite system was still closed during later hydrothermal alteration. Because tourmaline accommodates Mg in a very wide range of P–T conditions from diagenetic (Henry and Dutrow 2012) to UHP terrains (van Hinsberg et al. 2011; Lussier et al. 2016), absence of Mg in tourmaline is very reliable evidence for lack of this element in the system and, consequently, lack of external contamination in this particular process.

Calcium may stay available to later stages of primary crystallization (e.g., apatite, liddicoatite, graftonite; e.g., Teertstra et al. 1999; Novák et al. 1999; Martin and de Vito 2014 and references therein). However, late enrichment of Ca in pegmatites, although it was observed on numerous localities worldwide (e.g. Novák et al. 1999; Tindle et al. 2002, 2005), is still not fully understood. It could be caused by originally Ca-enriched melt (Novák and Gadas 2010; Gadas et al. 2012) or the melt contaminated by Ca in early stage of crystallization (post-emplacment stage). Also Ca–F complexing (Weidner and Martin 1987) may prevent the Ca consumption by early feldspars and thus this element remains in fluids to a very late stage.

Recently, Martin and de Vito (2014) proposed late Ca-miniflood from host rocks as a source of calcium. This process, albeit theoretically possible, is hardly applicable to the studied localities. If Ca was supplied from the host rocks, in our case calc-silicate rocks and marble, a simultaneous influx of Mg would be inevitable. It was indeed documented by the composition of late tourmalines elsewhere, e.g., dravite overgrowths or veining in early tourmaline (e.g., Němec 1989; Novák et al. 1999; Macek and Novák 2012). Moreover, the amount of Ca in studied tourmalines increases from the primary to the secondary stage (Figs 5a, 6a). Hence, the source of Ca was melt contamination at pre-emplacment stage (Novák et al. 2012) and during, or slightly after, emplacment to the site of crystallization (post-emplacment stage; Novák et al. 2013, 2017). This is consistent with the results of Teertstra et al. (1999) and Novák et al. (1999). Clearly, evolution of Ca in granitic pegmatites during solidus and subsolidus conditions requires detailed studies of further appropriate localities.

## 6. Conclusions

Mixture of secondary blue tourmaline (schorl to Fe-rich fluor-elbaite to very rare Fe-rich fluor-liddicoatite) with quartz partially replace spessartine–almandine garnet and albite in elbaite-subtype pegmatites at Tamponilapa and Tsarafara-Nord, Sahatany Valley, Madagascar and at Ctídrůžice, Moldanubian Zone, Czech Republic. Only garnet from the albite adjacent to an unit with Li-bearing minerals (Li-micas, Li-tourmalines) underwent this alteration. Composition of secondary tourmalines reflects the reaction of hydrothermal fluids with primary albite and garnet.

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