

Darrellhenryite, $\text{Na}(\text{LiAl}_2)\text{Al}_6(\text{BO}_3)_3\text{Si}_6\text{O}_{18}(\text{OH})_3\text{O}$, a new mineral from the tourmaline supergroup

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ABSTRACT

Darrellhenryite, $\text{Na}(\text{LiAl}_2)\text{Al}_6(\text{BO}_3)_3\text{Si}_6\text{O}_{18}(\text{OH})_3\text{O}$, a new member of the tourmaline supergroup (related to the alkali-subgroup 4), is a new Li-bearing tourmaline species, which is closely related to elbaite through the substitution ${}^{\text{I}}\text{Al}_{0.5}^{\text{IV}}\text{O}_1^{\text{I}}\text{Li}_{0.5}^{\text{IV}}(\text{OH})_{-1}$. It occurs in a complex (Li-bearing) petalite-subtype pegmatite with common *lepidolite*, Li-bearing tourmalines, and amblygonite at Nová Ves near Český Krumlov, southern Bohemia, Moldanubian Zone, Czech Republic. This zoned pegmatite dike cross-cuts a serpentinite body enclosed in leucocratic granulites. Pink darrellhenryite forms columnar crystals (sometimes in parallel arrangement) up to 3 cm long and up to 2 cm thick, associated with albite (var. *cleavelandite*), minor quartz, K-feldspar, petalite, rare polyolithionite, and locally rare pollucite. The optical properties and the single-crystal structure study ($R1 = 0.019$) of darrellhenryite are consistent with trigonal symmetry, $\omega = 1.636(2)$, $\epsilon = 1.619(2)$, birefringence: 0.017, space group $R3m$, $a = 15.809(2)$, $c = 7.089(1)$ Å, $V = 1534.4(4)$ Å³, and $Z = 3$. The chemical analysis, in combination with the results from the single-crystal structure refinement, gives the formula ${}^{\text{X}}(\text{Na}_{0.58}\text{Ca}_{0.01}\square_{0.41})_{1.00}{}^{\text{Y}}(\text{Li}_{1.05}\text{Al}_{1.95})_{3.00}{}^{\text{Z}}\text{Al}_6(\text{BO}_3)_3{}^{\text{W}}(\text{Si}_6\text{O}_{18})_3{}^{\text{V}}(\text{OH})_3{}^{\text{W}}(\text{O}_{0.66}\text{F}_{0.34})_{1.00}$, which can be simplified to an ideal formula of $\text{Na}(\text{LiAl}_2)\text{Al}_6(\text{BO}_3)_3\text{Si}_6\text{O}_{18}(\text{OH})_3\text{O}$. The strongest lines of the powder pattern [d in Å (I , hkl)] are 4.180 (39, 211), 3.952 (54, 220), 3.431 (73, 012), 2.925 (100, 122), 2.555 (90, 051), 2.326 (42, 511), 2.029 (42, 223), 2.021 (42, 152), 1.901 (50, 342), 1.643 (49, 603). The density is $D_{\text{meas}} = 3.03(3)$ g/cm³, $D_{\text{calc}} = 3.038$ g/cm³. Darrellhenryite is considered to have crystallized in Li- and B-rich but F-moderate environments in complex pegmatites; no influence of higher activity of O on the darrellhenryite formation is implied from its mineral assemblage. The name is for Darrell J. Henry, Professor of Geology at the Louisiana State University, Baton Rouge, U.S.A., an expert on the mineralogy, petrology, crystal chemistry, and nomenclature of tourmaline-supergroup minerals.

Keywords: Darrellhenryite, tourmaline group, chemical analyses, crystal structure, Nová Ves, Czech Republic

INTRODUCTION

Lithium-bearing tourmalines are of considerable scientific interest because they reflect compositional trends in granitic pegmatites (e.g., Selway et al. 1999, 2002), because of their complex zoning (e.g., Lussier et al. 2011) and because of their unknown stability field (e.g., London 2011). The current nomenclature of the tourmaline supergroup includes eight Li-bearing tourmalines (Henry et al. 2011), four of which are known to exist in nature: elbaite (Vernadsky 1913), fluor-liddicoatite (Dunn et al. 1977), rossmanite (Selway et al. 1998), and fluor-elbaite (Bosi et al. 2013) and four hypothetical species (Table 1). Darrellhenryite, $\text{Na}(\text{LiAl}_2)\text{Al}_6(\text{BO}_3)_3\text{Si}_6\text{O}_{18}(\text{OH})_3\text{O}$, represents a new Li-bearing tourmaline species, which belongs to the oxy-tourmaline series as do povondraite and chromo-alumino-povondraite (Henry et

al. 2011) and the more recently described minerals, oxy-schorl (Bačík et al. 2013) and oxy-dravite (Bosi and Skogby 2012). In this contribution, we present chemical analyses, the description of the crystal structure, spectroscopic data, and discuss paragenetic position of darrellhenryite in complex granitic pegmatites.

Darrellhenryite has been approved by the IMA-CNMNC (proposal no. 2012-026). Its chemical composition relates to the alkali-subgroup 4 (Table 1; Henry et al. 2011), which requires a new root name in this case, the name chosen is *darrellhenryite*, in honor of Darrell J. Henry (born 1951), Professor of Geology at the Louisiana State University, Baton Rouge, U.S.A., an expert on the mineralogy, petrology, crystal chemistry, and nomenclature of tourmaline-supergroup minerals. The type specimen is deposited with the Department of Mineralogy and Petrography of the Moravian Museum, Zelný trh 6, 659 37 Brno, Czech Republic, catalog number B10661. Holotype material (the crystal

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TABLE 1. Theoretical compositions of olenite, along with known and hypothetical Li-bearing tourmalines (wt%)

	1	2	3	4	5	6	7	8	9
SiO ₂	39.03	38.65	38.49	38.40	38.12	38.12	38.2	37.84	37.41
Al ₂ O ₃	44.15	46.46	40.82	40.73	43.12	37.73	37.81	40.13	47.61
B ₂ O ₃	11.30	11.20	11.15	11.12	11.04	11.04	11.06	10.96	10.84
Li ₂ O	1.62	0.80	2.39	2.39	1.58	3.16	3.17	2.35	–
Na ₂ O	–	–	3.31	3.30	3.28	–	–	–	3.22
CaO	–	–	–	–	–	5.93	5.94	5.89	–
H ₂ O	3.90	2.90	3.85	2.88	2.86	2.86	3.82	2.84	0.93
F	–	–	–	2.02	–	2.01	–	–	–
X site	□	□	Na	Na	Na	Ca	Ca	Ca	Na
Y site	LiAl ₂	Li _{0.5} Al _{2.5}	Li _{1.5} Al _{1.5}	Li _{1.5} Al _{1.5}	LiAl ₂	Li ₂ Al ₁	Li ₂ Al ₁	Li _{1.5} Al _{1.5}	Al ₃
Z site	Al ₆	Al ₆	Al ₆	Al ₆	Al ₆	Al ₆	Al ₆	Al ₆	Al ₆
B site	B ₃	B ₃	B ₃	B ₃	B ₃	B ₃	B ₃	B ₃	B ₃
T site	Si ₆	Si ₆	Si ₆	Si ₆	Si ₆	Si ₆	Si ₆	Si ₆	Si ₆
V site	(OH) ₃	(OH) ₃	(OH) ₃	(OH) ₃	(OH) ₃	(OH) ₃	(OH) ₃	(OH) ₃	O ₃
W site	OH	O	OH	F	O	F	OH	O	OH

Notes: 1 = rossmanite □(LiAl₂)Al₆Si₆O₁₈(BO₃)₃(OH)₃OH; 2 = □-Li-O root name □(Li_{0.5}Al_{2.5})Al₆Si₆O₁₈(BO₃)₃(OH)₃O; 3 = elbaite Na(Li_{1.5}Al_{1.5})Al₆Si₆O₁₈(BO₃)₃(OH)₃OH; 4 = fluor-elbaite Na(Li_{1.5}Al_{1.5})Al₆Si₆O₁₈(BO₃)₃(OH)₃F; 5 = darrellhenryite Na(LiAl₂)Al₆Si₆O₁₈(BO₃)₃(OH)₃O; 6 = fluor-liddicoatite Ca(Li₂Al)Al₆Si₆O₁₈(BO₃)₃(OH)₃F; 7 = liddicoatite Ca(Li₂Al)Al₆Si₆O₁₈(BO₃)₃(OH)₃OH; 8 = Ca-Li-O root name Ca(Li_{1.5}Al_{1.5})Al₆Si₆O₁₈(BO₃)₃(OH)₃O; 9 = olenite NaAl₃Al₆Si₆O₁₈(BO₃)₃(O)₃OH; all formulas from Henry et al. (2011).

chosen for structure refinement) is deposited at the Smithsonian Institution, National Museum of Natural History, Washington D.C., catalog number NMNH 175992, and co-type material is also deposited at this museum, catalog number NMNH 175993.

OCCURRENCE

Darrellhenryite was discovered in a complex (Li-bearing) pegmatite at Nová Ves near Český Krumlov, southern Bohemia, Czech Republic. The pegmatite belongs to the Moldanubian pegmatite province along with about 100 other lepidolite- and elbaite-subtype pegmatite bodies of the Variscan age (Novák and Povondra 1995; Novák 2005; Breiter et al. 2010; Ertl et al. 2004, 2012; Melleton et al. 2012). The pegmatite occurs as a symmetrically zoned dike, up to 8 m thick and about 100 m long, that cuts a serpentinite body (Teertstra et al. 1995; Novák and Černý 1998), which is enclosed in leucocratic granulites of the Blanský les Massif. From the contact inward, it consists of (1) an outermost granitic unit with biotite, (2) a coarse-grained albite unit locally with graphic intergrowths of Kfs + Qtz and aggregates of Ms + Qtz + Tur, (3) blocky K-feldspar and blocky petalite, and (4) an albite-lepidolite unit, the latter three units also containing Li-bearing tourmalines. No pockets have been found in the pegmatite. The Li-bearing primary minerals in the pegmatite are: petalite > lepidolite (trilithionite > polyolithionite) > Li-bearing tourmalines (darrellhenryite > fluor-elbaite) > amblygonite. Secondary spodumene + quartz aggregates after petalite and secondary montebrazite after primary amblygonite are also present. Accessory almandine-spessartine, fluorapatite, beryl, zircon, pollucite, cassiterite, columbite, and microlite-group minerals (fluornatromicrolite to fluorcalcimicrolite) were also found in the pegmatite. The pegmatite may be classified as a complex (Li)-type, petalite-subtype following the scheme proposed by Černý and Ercit (2005) but with substantial amounts of lepidolite, Li-bearing tourmalines, and amblygonite. The mineral assemblage and modal proportion of Li-bearing minerals are similar to much larger pegmatites of the petalite-subtype (e.g., Tanco, SE Manitoba, Canada: Stilling et al. 2006, and Utö,

Stockholm region, Sweden: Smeds and Černý 1989; Selway et al. 2002). However, the overall amount of Li-bearing minerals is much lower in the Nová Ves pegmatite than in these pegmatites.

Darrellhenryite (holotype) forms subhedral, short, columnar crystals and parallel aggregates, up to 3 cm long and up to 2 cm thick, which occur exclusively in the *cleavelandite*-rich portions of the albite-lepidolite unit in the central part of the dike. The associated minerals also include minor quartz, relics of pale brown, blocky K-feldspar and colorless blocky petalite, rare colorless flakes of polyolithionite and locally rare altered pollucite. Very rare inclusions of late muscovite, visible only in BSE images, occur within the aggregates of darrellhenryite.

METHOD

Chemical composition

Chemical analyses of darrellhenryite were carried out by various methods. These included a complete wet chemical analysis (Povondra et al. 1985); 13 EMP analyses (using WDS) with a Cameca SX 100 electron microprobe, LA-ICP-MS (Li; eight analyses); and a combination of wet chemical, near-IR, and thermogravimetric analysis (TGA; 3 analyses) for H₂O. To determine the OH content of darrellhenryite, ~15 mg of the material was used for each analysis, performed on a Mettler-Toledo TGA/SDTA 851 (University of Vienna). The powder was heated from 25 to 1100 °C (5 °C/min) in N₂ gas (gas flow: 25 mL/min). Analytical data of darrellhenryite, including standards for EMPA, are given in Table 2. A synthetic multi-layered pseudocrystal (Si/W layers, *d* = 60 Å, PC1 in terms of Cameca) was used as reflector for the fluorine analysis. It provides a much higher count rate than the more commonly used TAP (thallium acid phthalate) crystal. The substantial overlap of the FKα position with the shoulder of the FeLα,β peak on the pseudocrystal (Witter and Kuehner 2004) is not significant due to very low Fe content in darrellhenryite. The PHA settings of the detector were used in differential mode (baseline 750 mV and window 3500 mV) to reduce the “Bremsstrahlung” intensity and the influence of the second order of the AlKα peak on the low background position. We chose a natural lepidolite with well-defined F content (8.45 wt% F) as a standard. Prior to conducting the analysis, care was taken to determination precise background positions and the peak maximum of the FKα line. The positions of the FKα peak in darrellhenryite and lepidolite are identical. The fluorine peak counting time was 60 and 30 s on each of the background positions, and the exponential fit of background was used. Raw EMPA data and B, H, and Li content determined by Povondra et al. (1985), were corrected using the X-Phi (Merlet 1994) matrix procedure.

Concentration of Li and trace elements and were investigated by LA-ICP-MS (Department Chemistry, Masaryk University, Brno) using an Agilent 7500ce (Agilent Technologies, Santa Clara, California, U.S.A.) quadrupole ICP-MS with an attached UP 213 laser ablation system (New Wave Research, Inc., Fremont, California, U.S.A.). The samples were placed into a SuperCell (New Wave Research, Inc., Fremont, California, U.S.A.) having a volume of 33 cm³ and ablated using a commercial Q-switched Nd:YAG laser operated at a wavelength of 213 nm (pulse duration 4.2 ns). Ablated material was transported from the sample chamber using helium carrier gas (1 L/min) and mixed with argon (0.6 L/min) prior to the

TABLE 2. Analytical data for darrellhenryite

Constituent	wt%*	wt%†	Range†	S.D.†	Probe standard
SiO ₂	37.94	38.38	38.69–38.12	0.17	almandine
Al ₂ O ₃	42.77	43.49	43.75–43.10	0.16	grossular
B ₂ O ₃	11.01	11.01*			
FeO	0.17				almandine
MnO	0.02	0.02	0.11–0.00	0.04	spessartine
CaO	0.07	0.05	0.09–0.00	0.03	fluorapatite
Li ₂ O	1.88	1.63	1.87–1.37	0.21	NIST 610
Na ₂ O	1.81	1.92	2.03–1.78	0.07	albite
K ₂ O	0.12				sanidine
H ₂ O	2.86	2.86*			
F	0.64	0.71	0.78–0.61	0.07	lepidolite
O=F	–0.27	–0.30			
Total	99.02	99.77			

* Wet chemical analysis from Povondra et al. (1985).

† Electron microprobe, LA-ICP-MS (Li₂O), TGA: 2.9(1) wt% H₂O, near-infrared spectroscopy: 2.87 wt% H₂O.

torch. Optimization of LA-ICP-MS parameters (gas flow rates, sampling depth, voltage of ion optics) was performed using glass reference material NIST SRM 612 to maximize the S/N ratio. Potential polyatomic interferences were minimized by a collision reaction cell in He mode (2.5 mL/min).

Crystal structure

The tourmaline of darrellhenryite was studied on a Bruker AXS Kappa APEX II CCD diffractometer equipped with a monocapillary optics collimator and graphite-monochromatized MoK α radiation. Single-crystal X-ray diffraction data were collected at room temperature (out to $\sim 80^\circ 2\theta$), integrated and corrected for Lorentz and polarization factors and absorption correction by evaluation of partial multiscans. The structure was refined (using a starting model; Ertl et al. 2010) with SHELXL-97 (Sheldrick 1997) using scattering factors for neutral atoms. The H atom bonded to the O3 atom was located from a difference-Fourier map and subsequently refined. Refinement was performed with anisotropic displacement parameters for all non-hydrogen atoms. Table 3 provides crystal data and details of the structure refinement. Site occupancies were refined according to well-known characteristics of the tourmaline structure (B, O1–O8, and H3 were constrained to fully occupy their respective sites, Na was refined at the X site, Al and Li were refined at the Y site; for other details see Table 4). The refinements converged at $R_1(F)$ values of $\sim 1.95\%$ (Table 3). The atomic parameters and equivalent isotropic displacement parameters are given in Table 4. In Table 5 we present selected interatomic distances. (CIF¹ file is available on deposit.)

The X-ray powder diffraction (XRD) data for darrellhenryite (pinkish white powder) were collected on a PW 3710 Philips diffractometer using CuK α radiation (40 kV, 40 mA), equipped with a secondary graphite monochromator. The irradiated sample area was constantly $12 \times 12 \text{ mm}^2$ (automatic divergence slits gave a constant area for each 2θ angle). A sample of darrellhenryite was prepared on a round zero-background silicon holder, dispersed in a few drops of acetone and allowed to dry. The XRD data were collected [at 295(1) K] over the 2θ range of

TABLE 3. Crystallographic data and refinement details for darrellhenryite

a , c (Å)	15.809(2), 7.089(1)
V (Å ³)	1534.4(7)
Crystal dimensions (mm)	$0.15 \times 0.15 \times 0.10$
Collection mode, $2\theta_{\text{max}}$ (°)	full sphere, 79.95
h, k, l ranges	$-28/28, -28/27, -12/12$
Number of frames	620
Total reflections measured	19944
Unique reflections	2249
$R_1^a(F)$, $wR_2^b(F^2)$, R_{int}^c (%)	1.95%, 4.32%, 3.50%
Flack x parameter	0.016(59)
"Observed" reffs. [$F_o > 4\sigma(F_o)$]	2169
Extinct. coefficient	0.00517(26)
No. of refined parameters	95
Goodness-of-fit ^d	1.065
$\Delta\sigma_{\text{min}}$ $\Delta\sigma_{\text{max}}$ (e/Å ³)	$-0.57, 0.64$

Note: X-ray radiation: MoK α ($\lambda = 0.71073$ Å); $Z = 3$; space group is $R3m$ (no. 160); multi-scan absorption correction; refinement on F^2 . Frame width, scan time, detector distance: 3° , 15 s, 35 mm. Scan mode: sets of ω and θ scans.

^a $R_1 = \sum ||F_o| - |F_c|| / \sum |F_o|$.

^b $wR_2 = [\sum [w(F_o^2 - F_c^2)]^2 / \sum [w(F_o^2)]^2]^{1/2}$, $w = 1/[\sigma^2(F_o^2) + (aP)^2 + bP]$, $P = [2F_c^2 + \text{Max}(F_o, 0)]/3$.

^c $R_{\text{int}} = \sum |F_o^2 - F_o^2(\text{mean})| / \sum |F_o^2|$.

^d $\text{Goof} = S = [\sum [w(F_o^2 - F_c^2)]^2 / (n - p)]^{1/2}$.

TABLE 4. Table of atomic parameters in darrellhenryite

Site	x	y	z	U_{eq}	Occ.
X	0	0	0.7509(3)	0.0199(6)	Na _{0.56(1)}
Y	0.87800(4)	0.93900(2)	0.34846(7)	0.0068(2)	Al _{0.651(3)} Li _{0.349}
Z	0.70347(2)	0.74017(2)	0.37588(3)	0.00539(5)	A _{1.00}
B	0.89096(4)	0.78193(9)	0.5307(2)	0.0057(2)	B _{1.00}
T	0.80823(1)	0.81027(2)	0.98406(3)	0.00450(4)	Si _{1.00}
H3	0.740(2)	0.870(1)	0.600(4)	0.051(8)	H _{1.00}
O1	0	0	0.2079(2)	0.0252(6)	O _{0.66(4)} F _{0.34}
O2	0.93962(3)	0.87924(7)	0.4938(1)	0.0125(2)	O _{1.00}
O3	0.73727(8)	0.86864(4)	0.4772(1)	0.0124(2)	O _{1.00}
O4	0.90589(3)	0.81179(7)	0.9106(1)	0.0085(1)	O _{1.00}
O5	0.81242(7)	0.90621(4)	0.8883(1)	0.0091(1)	O _{1.00}
O6	0.80533(4)	0.81595(4)	0.21009(8)	0.0070(1)	O _{1.00}
O7	0.71341(4)	0.71375(4)	0.90703(7)	0.00641(9)	O _{1.00}
O8	0.79052(4)	0.72987(4)	0.54631(8)	0.0071(1)	O _{1.00}

Note: For the definition of U_{eq} see Fischer and Tillmanns (1988).

TABLE 5. Selected interatomic distances (Å) in darrellhenryite

X-O2 $\times 3$	2.461(2)
X-O5 $\times 3$	2.747(1)
X-O4 $\times 3$	2.815(1)
Mean	2.674(1)
Y-O1	1.9450(11)
Y-O2 $\times 2$	1.9552(7)
Y-O6 $\times 2$	1.9574(7)
Y-O3	2.1320(12)
Mean	1.9837(9)
Z-O6	1.8656(6)
Z-O7	1.8815(6)
Z-O8	1.8850(6)
Z-O8'	1.8985(7)
Z-O7'	1.9425(6)
Z-O3	1.9598(5)
Mean	1.9055(6)
T-O7	1.6069(7)
T-O6	1.6081(6)
T-O4	1.6180(4)
T-O5	1.6326(4)
Mean	1.6164(5)
B-O2	1.358(2)
B-O8 ($\times 2$)	1.3799(9)
Mean	1.373(1)

Note: Standard deviation in parentheses.

$5\text{--}62^\circ$ using 0.01° steps and a counting time of 1 s/step. Because of the very limited amount of material with a composition close to the end-member of darrellhenryite, the collection of high-quality data was limited to 2θ less than 65° because of the low quality of data at higher angles. Silicon SRM 640b was used as internal standard. The powder diffraction data (indexing is based on the structure refinement and only reflections with $I_{\text{calc}} \geq 1$ are listed) are listed in Table 6.

Spectroscopic investigations

Near-infrared spectra were obtained with a Nicolet Magna 860 FTIR, a silica beam splitter, and a tungsten-halogen source. Polarized spectra were obtained with a LiIO₃ crystal polarizer. The darrellhenryite sample was prepared as a doubly polished 1.132 mm thick slab, in which the c axis is parallel to the section plane. A small, clear area was located in the otherwise cracked sample through which the spectrum could be obtained. The OH content was determined from the integrated area of the OH overtone bands in the 6350 to 7300 cm^{-1} region. Because a calibration of this method specific to this species does not exist, the calibration used in Ertl et al. (2010) for elbaite-schorl series tourmalines was chosen for this study. Optical spectra in the visible to near-infrared region were obtained with a home-built microspectrometer based on diode-array technology, as described in Taran and Rossman (2001).

RESULTS

Physical and optical properties

Darrellhenryite has vitreous luster and a pinkish white streak. The crystals are translucent to transparent with an intense to pale pink color. The mineral is brittle with conchoidal fracture. Mohs hardness is ~ 7 . The measured density reported by Povondra et al. (1985) is 3.03(3) g/cm^3 . The calculated density is 3.041 g/cm^3 using the empirical formula given by Povondra et al. (1985) and the present unit cell (single-crystal) data, whereas the calculated density is 3.038 g/cm^3 using the present empirical formula and unit-cell data (single-crystal data). Darrellhenryite is non-fluorescent under long-, medium-, or short-wave UV light. It is uniaxial (–), $\omega = 1.636(2)$ (1.637), $\epsilon = 1.619(2)$ (1.621) (590 nm); birefringence: 0.017 (0.016) (data in parentheses from Povondra et al. 1985). In thin section, it is colorless. Fragments

¹ Deposit item AM-13-1008, CIF. Deposit items are stored on the MSA web site and available via the *American Mineralogist* Table of Contents. Find the article in the table of contents (at GeoScienceWorld or www.minsocam.org), and then click on the deposit link.

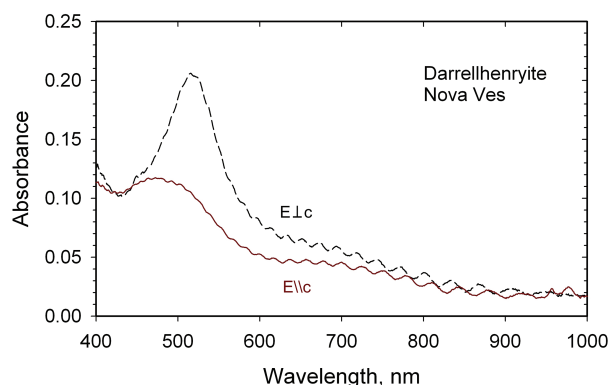
TABLE 6. X-ray powder diffraction data for darrellhenryite

d_{meas} (Å)	d_{calc} (Å)	I_{meas}	$h k l$
7.906	7.911	5	110
6.295	6.299	15	101
4.924	4.927	18	021
4.575	4.578	7	300
4.180	4.183	39	211
3.952	3.954	54	220
3.431	3.434	73	012
3.353	3.355	18	131
3.084	3.086	8	401
2.989	2.991	14	410
2.925	2.927	100	122
2.873	2.875	4	321
2.592	2.594	11	312
2.555	2.557	90	051
2.433	2.435	1	241
2.364	2.366	38	003
2.355	2.356	31	232
2.326	2.328	42	511
2.284	2.286	3	600
2.266	2.267	2	113
2.168	2.170	13	502
2.147	2.148	21	431
2.100	2.101	25	303
2.090	2.091	14	422
2.029	2.030	42	223
2.021	2.023	42	152
2.004	2.005	10	161
1.978	1.979	4	440
1.901	1.902	50	342
1.887	1.888	6	351
1.854	1.855	12	413
1.835	1.837	9	621
1.816	1.817	3	710
1.802	1.803	1	612
1.759	1.760	15	104
1.718	1.719	5	024
1.674	1.675	6	262
1.643	1.644	49	603
1.629	1.630	27	271
1.608	1.609	2	523
1.582	1.583	20	550
1.576	1.577	9	404
1.565	1.566	4	811
1.544	1.545	6	324
1.534	1.535	8	461
1.523	1.524	9	900
1.514	1.515	11	722
1.508	1.508	3	731

Note: d -values in angstroms. The unit-cell data parameters refined from the powder data (CuK α radiation) are $a = 15.820(2)$, $c = 7.093(1)$ Å. The 10 reflections with the highest intensity are in bold. Standard deviations for measured d -values are $\leq 0.1\%$.

about 1 mm thick show pleochroism that varies from colorless ($\parallel c$) to pale pink ($\wedge c$).

The color is caused primarily by an absorption band centered at ~ 520 nm in the polarization direction perpendicular to c (Fig. 1). Weak, sharper features occur at 449 and 457 nm, and a hint of a broad and weak band appears centered at ~ 700 nm. All of these features and the band in the $E \parallel c$ direction are characteristic of most pink tourmalines, which owe their color to Mn^{2+} believed to be produced by the natural irradiation of Mn^{2+} in the tourmaline (Reinitz and Rossman 1988). The sample is full of pervasive internal fractures, which cause the wave-like appearance in the spectrum (Fig. 1) due to interference fringes. A 10.4 μm thick air gap in this particular sample would produce the fringes seen in Figure 1. Spectra obtained from different areas in the sample had significant differences in the intensity of the 520 nm band, attributed to Mn, indicating an inhomogeneous distribution of this chromophore.

**FIGURE 1.** Optical absorption spectrum of darrellhenryite normalized to 1.0 mm thickness. (Color online.)

Chemical composition

The empirical formulas are calculated on the basis of 31 (O,OH,F): $(Na_{0.55}K_{0.02}Ca_{0.01}\square_{0.42})_{\Sigma 1.00}(Li_{1.19}Fe_{0.02}Al_{1.96})_{\Sigma 3.17}Al_{6.00}(BO_3)_{3.00}(Si_{5.99}Al_{0.01})O_{18}(OH)_{3.00}(O_{0.67}F_{0.32}OH_{0.01})$ from Povondra et al. (1985) and $(Na_{0.58}Ca_{0.01}\square_{0.41})_{\Sigma 1.00}(Li_{1.03}Al_{2.02})_{\Sigma 3.05}Al_{6.00}(BO_3)_{2.98}Si_{6.01}O_{18}(OH)_{3.00}(O_{0.65}F_{0.35})$, a combination of EMPA, LA-ICP-MS (Li), TGA (H_2O), and B_2O_3 taken from Povondra et al. (1985). The ideal formula for darrellhenryite is $Na_{1.00}(Li_{1.00}Al_{2.00})_{\Sigma 3.00}Al_{6.00}(BO_3)_{3.00}Si_{6.00}O_{18}(OH)_{3.00}O_{1.00}$. Very low concentrations of other elements (Fe, Mn, K, Ca) are typical, as well as moderate contents of F (Table 2). Analyses obtained using LA-ICP-MS indicate that most trace elements are present at or below the detection limits (0.1–15 ppm) except for low contents (+ detection limits in parentheses) of Be (≤ 17 ; 0.1), Ni (≤ 11 ; 0.3), Cu (≤ 13 ; 1), Zn (≤ 10 ; 3), and Sn (≤ 77 ; 2) (all in ppm) and high contents of Ga (134–405 ppm; 2). The Ga content is comparable to that of Cu-enriched Li-bearing tourmalines from pegmatites in Brazil, Nigeria, and Mozambique (Peretti et al. 2009), but higher than that of black tourmaline from NYF-type pegmatites of the Třebíč Pluton, Czech Republic, with Ga = 78–160 ppm (Novák et al. 2011), and black tourmaline from common pegmatites of the Strážek Moldanubicum, Czech Republic, with Ga = 2–212 ppm (Gadas et al. 2012).

Near-IR spectrum

Water content can be determined from the intensity of the OH overtone absorptions in the 7000 cm^{-1} region of the near-infrared spectrum (Fig. 2). The total integrated area of the spectrum ($\parallel c + 2 \times \perp c$) in Figure 2 is 1092.5 per cm^2 (normalized to 1 cm thickness). Using the density of 3.038 g/cm^3 and dividing by the factor 113 (taken from Ertl et al. 2010), an H_2O concentration of 2.87 wt% is determined. This is in remarkably close agreement with the value in Table 2, 2.86 wt%, determined by classical wet chemical methods and with the value determined by thermogravimetric analysis (TGA) [2.9(1) wt%; Table 2]. This value is also consistent with the proposed darrellhenryite formula.

Crystallography

Single-crystal X-ray studies gave the following data: trigonal symmetry, space group: $R3m$, $a = 15.809(2)$, $c = 7.089(1)$

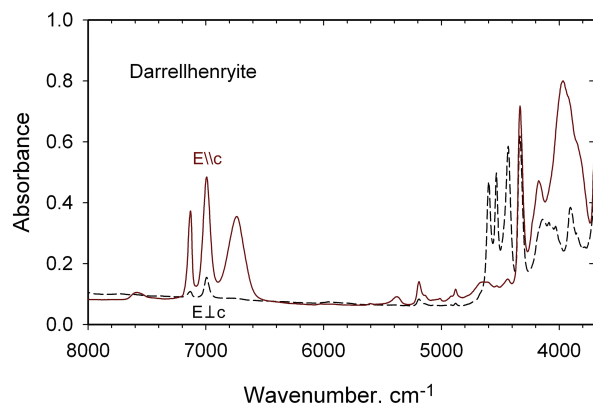


FIGURE 2. Near-IR spectrum of darrellhenryite (sample gr3075). (Color online.)

\AA , $V = 1534.4(4) \text{ \AA}^3$, and $Z = 3$. Darrellhenryite is isotypic with elbaite and other rhombohedral ($R3m$) members of the tourmaline supergroup. The refined formula for darrellhenryite, $^{x}(\text{Na}_{0.56}\square_{0.44})^{y}(\text{Li}_{1.05}\text{Al}_{1.95})_{\Sigma 3.00}^{z}\text{Al}_6^{t}(\text{Si}_6\text{O}_{18})(\text{BO}_3)_3^{v}(\text{OH})_3^{w}(\text{O}_{0.66}\text{F}_{0.34})$, is in reasonably good agreement with the empirical formula $^{x}(\text{Na}_{0.58}\text{Ca}_{0.01}\square_{0.41})^{y}(\text{Li}_{1.03}\text{Al}_{2.02})_{\Sigma 3.05}^{z}\text{Al}_6(\text{BO}_3)_{2.98}^{t}(\text{Si}_6\text{O}_{18})^{v}(\text{OH})_3^{w}(\text{O}_{0.65}\text{F}_{0.35})$; slight differences are within the limits of the refinement and may also reflect minor chemical zoning confirmed by chemical analyses and different ways of formula elucidation.

An H atom (H3) at the site associated with O3 was easily located in this refinement. Ertl et al. (2002) showed that the bond-angle distortion (σ_{oct}^2) of the ZO_6 octahedron in a tourmaline is largely a function of the $\langle Y\text{-O} \rangle$ distance of that tourmaline, although the occupant of the O(3) site (V position in the general formula) also affects that distortion. The covariance, r , of $\langle Y\text{-O} \rangle$ and σ_{oct}^2 of the ZO_6 octahedron is -0.99 (Fig. 2 in Ertl et al. 2005) for all investigated tourmalines whose V site is occupied by 3 (OH) groups. Darrellhenryite (with $^z\sigma_{\text{oct}}^2 = 52.85$ and $\langle Y\text{-O} \rangle \approx 1.984$; Table 5) lies exactly on the V site = 3 (OH) line. Hence, the V site of darrellhenryite is completely occupied by (OH). The refinement shows that the W site is occupied by $(\text{O}_{0.66}\text{F}_{0.34})$ (Table 4, Fig. 3). Hence, this site is clearly dominated by oxygen, considering also the chemical data (Table 2).

The T site is completely occupied by Si. A refinement of $\text{Si} \leftrightarrow \text{B}$ at the T site gives no clear indication for significant amounts of $^{[4]}\text{B}$ (>0.10 apfu). Hence, in the final refinement the T site was refined only with Si (Table 4). Recently, Lussier et al. (2011) investigated liddicoatite samples from Anjanaboina, Madagascar, which contain essentially no $^{[4]}\text{B}$. Most of these Al- and Li-rich tourmalines have a $\langle T\text{-O} \rangle$ distance of $1.617(1) \text{ \AA}$, which is in good agreement with the $\langle T\text{-O} \rangle$ distance of $\sim 1.616(1) \text{ \AA}$ in darrellhenryite (Table 5).

The X-ray powder diffraction data for darrellhenryite is presented in Table 6. Unit-cell parameters refined from the powder data (Table 6), obtained by a least-squares refinement of the setting angles of all reflections, are as follows: $a = 15.820(2)$, $c = 7.093(1) \text{ \AA}$, $V = 1537.4(6) \text{ \AA}^3$, very similar to those from the single-crystal X-ray study and data of Povondra et al. (1985).

Compositional evolution of tourmalines from the Nová Ves pegmatite and paragenetic position of darrellhenryite

Tourmaline is a common accessory mineral in complex pegmatites of the Moldanubian Zone including the Nová Ves pegmatite (Povondra et al. 1985; Novák and Povondra 1995; Novák et al. 2004; Breiter et al. 2010). Its chemical composition evolves during pegmatite crystallization: black Mg-poor oxy-schorl from the outer albite unit \rightarrow black oxy-schorl to blue or green Fe-rich fluor-elbaite in muscovite + quartz aggregates from the albite unit \rightarrow green Fe-rich fluor-elbaite to pink fluor-elbaite to darrellhenryite from outer parts of the albite-lepidolite unit \rightarrow pink to pinkish darrellhenryite in albite (*cleavelandite*) from the inner part of the albite-lepidolite unit (Fig. 3). The compositional trends (behavior of Na, Al, Fe, and F) are similar to ordinary lepidolite-subtype pegmatites from the Moldanubian Zone (Selway et al. 1999; Novák 2000); however, most tourmalines from Nová Ves belong to the oxy series (Fig. 3). Based on a large set of EMP analyses, rossmanite, a common accessory mineral in the lepidolite-subtype pegmatites from the Moldanubian Zone, Czech Republic (Selway et al. 1998, 1999), has not been identified from the Nová Ves locality.

Darrellhenryite is evidently the least common Li-bearing tourmaline, and it is known only from the Nová Ves locality. The mineral assemblage of darrellhenryite (holotype + other samples) is characterized by abundant albite + minor quartz \pm K-feldspar \pm petalite (or secondary spodumene + quartz) \pm rare polyolithionite \pm pollucite. It differs from other pink Li-bearing tourmalines in complex pegmatites of lepidolite-subtype and elbaite-subtype from the Moldanubian Zone. In lepidolite-subtype pegmatites, pink Li-bearing tourmaline (fluor-elbaite $>$ rossmanite $>$ elbaite; Povondra et al. 1985; Selway et al. 1998, 1999; Novák 2000) is typically associated with common Li-rich micas (trilithionite $>$ polyolithionite; Černý et al. 1995) + albite \pm quartz, and this assemblage indicates high activity of F. In elbaite-subtype pegmatites, red to pink Li-bearing tourmaline (fluor-elbaite $>$ fluor-liddicoatite; Povondra et al. 1985; Novák et al. 1999a, 2012) is commonly the only Li-bearing mineral, Li-rich micas (polyolithionite; Novák and Povondra 1995; Novák et al. 1999b; Zahradníček and Novák 2012) being absent or very rare; hence, F enters almost exclusively tourmaline. Consequently, composition of associated Li-rich micas and their abundance play a crucial role in the formation of darrellhenryite because they control activities of Li and F in the system. Darrellhenryite is likely related to Li- and B-rich but F-moderate environments characterized by the assemblage albite \pm quartz $>$ Li-rich micas. No influence of higher activity of O on the darrellhenryite formation is inferred from the mineral assemblage, similar to the case of oxy-schorl (Bačík et al. 2012).

Due to a low number of complete chemical analyses of Li-bearing tourmalines and because we did not find any chemical analysis of Li-bearing tourmaline corresponding to darrellhenryite, we checked also electron microprobe analyses of Ca-poor Li-bearing tourmalines published to date, where Li and H were calculated using stoichiometric constraints: $\text{Li (apfu)} = 15 - \Sigma (\text{Y} + \text{Z} + \text{T})$ and $(\text{OH} + \text{F}) = 4$ (e.g., Selway et al. 1999). Only a few analyses of pink Li-bearing tourmalines show a combination of Na, Al, and F contents ($\text{Na} > 0.5$ apfu, $\text{Al} > 7.5$ apfu, $\text{F} <$

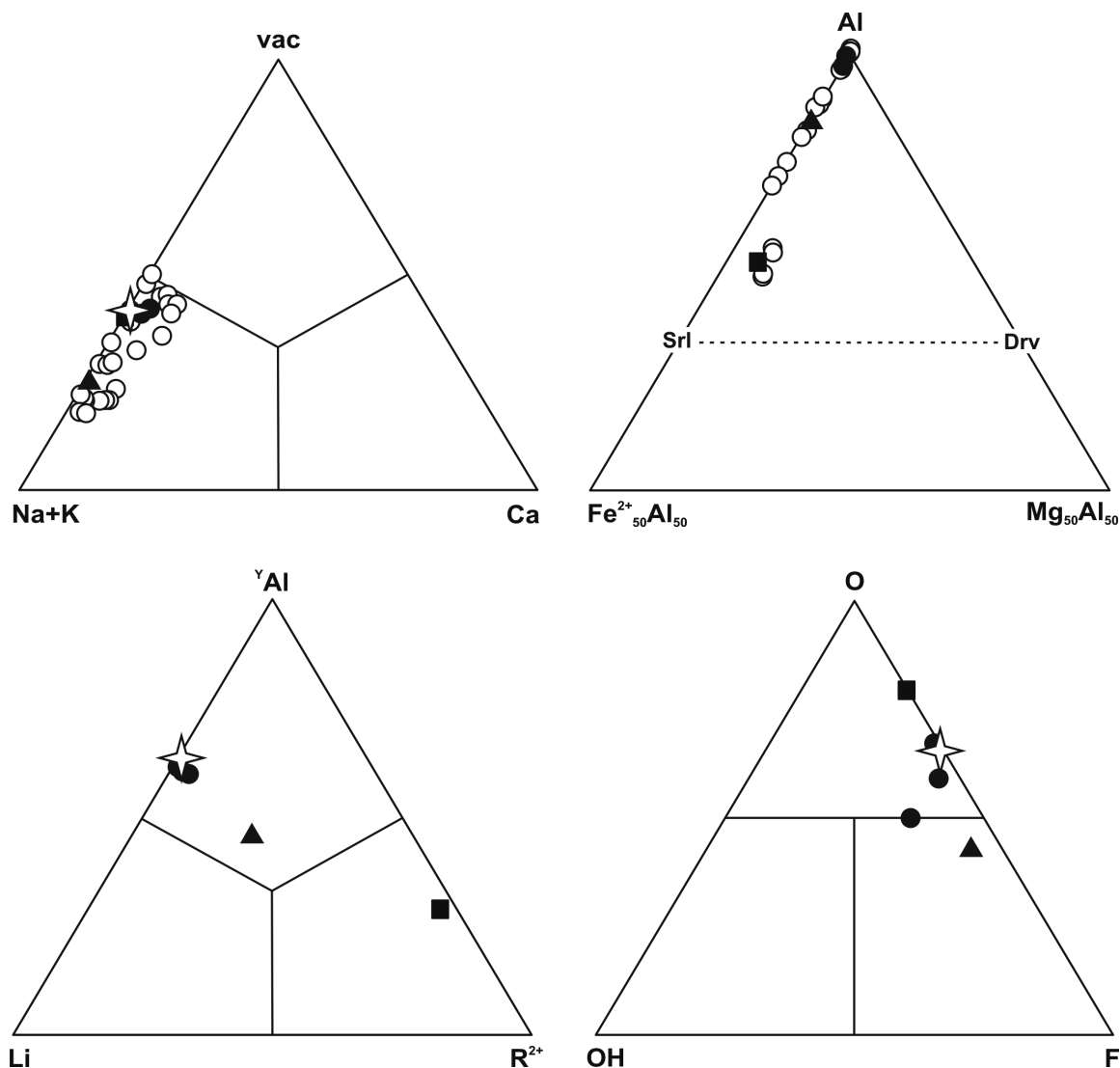


FIGURE 3. Chemical composition of tourmalines from the Nová Ves pegmatite. Open circles = EMPA data; solid symbols = wet chemical analyses (Povondra et al. 1985); square = black oxy-schorl; triangle = green Fe-rich fluor-elbaite; circles = pink darrellhenryite to darrellhenryite = fluor-elbaite; star = type material data (Table 2, analysis no. 2).

0.5 apfu), which are indicative of darrellhenryite. The samples come from Dobrá Voda, Czech Republic (Table 1, analysis no. 9; Selway et al. 1999); Tanco, SE Manitoba, Canada (Table 4, analysis no. 5; Selway et al. 2000); Utö, Sweden (Table 2, analysis no. 7; Selway et al. 2002); and Bennet Mine, southern Maine (Table 8, analysis no. 8; Wise and Brown 2010). They typically show high vacancies at the *X* site (~0.43–0.48 pfu) and low to moderate F (0.23–0.39 apfu), so they are compositionally close to rossmanite (Selway et al. 1998). Consequently, a detailed study (ideally including EMPA, single-crystal X ray diffraction, LA-ICP-MS, SIMS, and spectroscopic methods) is required to distinguish the Ca-poor Li-bearing tourmalines (Table 1)—elbaite, fluor-elbaite, rossmanite, darrellhenryite, and the hypothetical □-Li-O phase (vacant subgroup 4; Henry et al. 2011)—from each other.

ACKNOWLEDGMENTS

We sincerely thank the reviewers A. McDonald and F. Colombo for their careful reviews of the manuscript as well as the comments of the members of the CNMNC of the IMA, which improved this manuscript significantly. This work was funded by GAP 210/10/0743 (M.N., P.G., and R.Š.) and by the Austrian Science Fund (FWF) project no. P23012-N19 (A.E.) with contributions from NSF grant EAR-0947956 (G.R.R.). M.V.G. acknowledges the European Regional Development Fund project “CEITEC” (CZ.1.05/1.1.00/02.0068).

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MANUSCRIPT RECEIVED NOVEMBER 26, 2012

MANUSCRIPT ACCEPTED MAY 27, 2013

MANUSCRIPT HANDLED BY FERNANDO COLOMBO