

Stable isotope and crystal chemistry of tourmaline across pegmatite - country rock boundaries at Black Mountain and Mount Mica, southwestern Maine, U.S.A.

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Abstract: Major element and stable isotope chemistry of tourmaline from two complexly-zoned rare element pegmatites has been studied to gain insights into the processes by which the pegmatites were formed. Two locations in the Oxford Pegmatite Field of western Maine, U.S.A., were chosen for this study: Black Mountain, an isolated body located in sillimanite zone, highly sulfidic metapelites and quartzite; and Mount Mica, which is bounded by schists and pegmatite and aplitic granite bodies commonly having gradational contacts with each other. At each locality, tourmaline was sampled from the surrounding country rocks into the contact and wall zones through to the pegmatite cores. Along these traverses, trends in major element crystal chemistry of tourmaline are similar for both localities; these include $\text{Li} + \text{Al} \leftrightarrow \text{Mg} + \text{Fe}^{2+}$, $\text{Na} + \text{Ca} + \text{K} \leftrightarrow \text{H}_3\text{O}^+$, and $\text{B} \rightarrow \text{Si}$ substitutions. Tourmaline compositions also reflect the parageneses in which they occur, especially Mg/Fe^{2+} , which increases as Fe^{2+} is taken up by pyrrhotite in the country rock at Black Mountain. Differences in the major element compositions of tourmaline between the two localities are readily understood in the context of parageneses. However, stable isotopes strongly suggest that two contrasting styles of pegmatite are involved. Black Mountain has tourmaline showing gradational isotope signatures between the pegmatite and surrounding country rocks. Mount Mica contains tourmaline that is clearly isotopically distinct from tourmaline in the surrounding country rock. One interpretation of this difference is that Black Mountain may have formed from partial melting of metasediments, in combination with precipitation from hydrothermal fluids related to the nearby batholiths, whereas Mount Mica formed as a fractionate of the nearby Sebago Batholith.

Key-words: tourmaline, stable isotopes, pegmatite, Maine, crystal chemistry, Black Mountain, Mount Mica.

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Introduction

The Oxford Pegmatite Field of southwestern Maine occupies a 50 km wide area (Wise & Francis, 1992) within the Kearsarge-Central Maine Synclinorium (Osberg *et al.*, 1985) and is associated with several granitic plutons produced by Middle Paleozoic Acadian orogenic events (Fig. 1). It is dominated by the Sebago batholith in the south and by several smaller plutons in the north. In keeping with the use of the term «field», these pegmatites have long been considered to have been generated within a common geological environment during a single magmatic event (Wise & Francis, 1992). This idea has been reinforced by extensive field observations and whole rock chemistry collected by M. Wise (unpublished data). However, this hypothesized genetic relationship between the pegmatites and the granites (*e.g.*, Černý *et al.*, 1991a and b) has not yet been rigorously tested by detailed mineralogical and

geochemical studies; in fact, some workers have recently proposed an anatectic origin for some of these pegmatites (Simmons *et al.*, 1995 & 1996).

Tourmaline (abbreviated Tur following Kretz, 1983) occurs in most rocks of the field area, and may record information bearing on interrelationships among the pegmatites, country rocks, and various igneous bodies. Hence, this study examines the changes in Tur chemistry from zones within two noted Maine pegmatites and from their proximal metamorphic country rocks. The goal of the study is comprehensive characterization of both the major element and stable isotope compositions of tourmaline, a notoriously difficult mineral to analyze, in order to gain insights into the processes by which the two pegmatites formed.

The sites chosen for this work are two pegmatites belonging to the complex type of the rare element class of granitic pegmatites (Černý, 1991c) located at Black Mountain (BMT) and Mount Mica (MM) (Fig. 1). Precise locations and

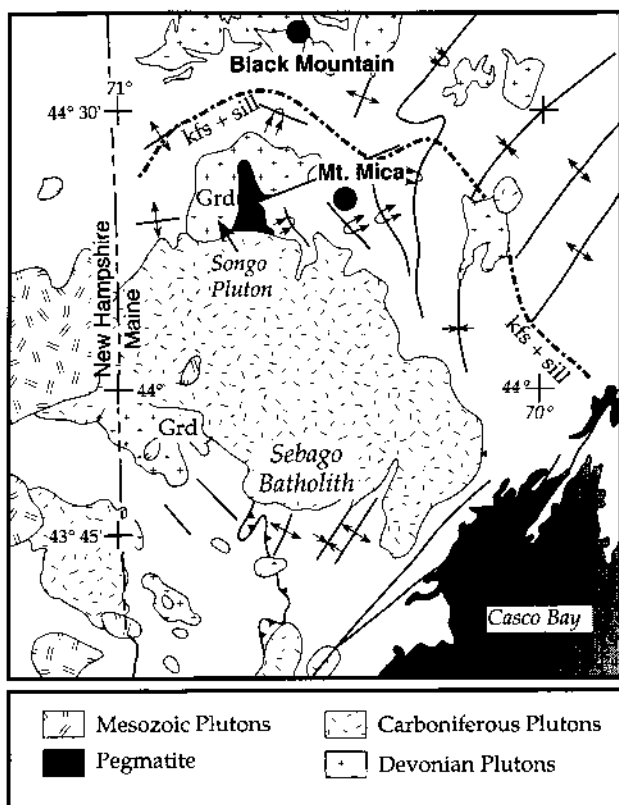


Fig. 1. Generalized geologic map of Maine, showing the location of the Mount Mica and Black Mountain localities with respect to nearby igneous bodies, based on Osberg *et al.* (1985). The Sil + Kfs isograd is from Guidotti (1985). Grd is granodiorite.

references to their mineralogy are given in Thompson *et al.* (1991). Both sites were chosen because tourmaline is present within the pegmatite and the country rocks. BMT is located in sillimanite zone, highly sulfidic metapelites and quartzites. In contrast, MM occurs in an area where at least 50 % of the rock exposures are pegmatite and aplitic granite bodies that commonly have gradational contacts with each other (Guidotti, 1963, 1965). The remainder of the exposed rocks are migmatitic, Sil - Kfs zone metapelites.

Geologic setting

The pegmatite at Mount Mica was the first gem tourmaline locality in North America and its geology is well studied (Bastin, 1911; Francis, 1985). It is located a few km northeast of the Sebago Batholith (Fig. 1) as a low-dipping, north-east-trending sill of undetermined thickness. It has a sharp, conformable contact with the overlying,

highly migmatitic Sil + Kfs zone metapelites. From the contact down, zones within the pegmatite were identified in keeping with the classification of Cameron *et al.* (1945): a very thin border zone, a thick wall zone, a cleavelandite + quartz pocket zone, and a garnet horizon (Table 1).

Approximately 9 km to the north of MM, the BMT pegmatite is an isolated body in close proximity to the Rumford and Whitecap Mountain plutons, which are diorite to granodiorite and a mixture of leucogranite and pegmatite granite, respectively. The BMT pegmatite occurs as six lenticular, subparallel bodies (Francis *et al.*, 1993) striking northwest within Sil zone metapelites plus minor quartzites of the highly sulfidic Smalls Falls formation (Moench & Hildreth, 1976). Nine units have been identified in the main exposure of the pegmatite (Brown & Wise, 1991 and Francis *et al.*, 1993): border zone, wall zone, first and second intermediate zones, quartz-cleavelandite zone (core), cleavelandite-tourmaline unit, aplite, K-feldspar + quartz unit, and «rubellite» granite (Table 1).

Table 1. Tourmaline Samples from Black Mountain (BMT and BMCR) and Mount Mica (MM).

Sample	Distance*	Description of location (zones after Brown and Wise, 1991)	Assemblage
BMT-A	-18.0	core	Qtz + Clv (+ Ms + Elb)
BMT-19	-10.0	lepidolite pods, 2nd intermediate	Spd + Qtz + Kfs (+ Lpd + Elb + Col)
BMT-17	-10.0	lepidolite pods, 2nd intermediate	Spd + Qtz + Kfs (+ Lpd + Elb + Col)
BMT-49	-10.0	2nd intermediate	Spd + Qtz + Kfs (+ Lpd + Elb + Col)
BMT-65	-10.0	2nd intermediate	Spd + Qtz + Kfs (+ Lpd + Elb + Col)
BMT-B	-5.0	1st intermediate	Ms + Clv (+ Elb)
BLKMTN-33	-5.0	1st intermediate	Ms + Clv (+ Drv)
BMT-20	-5.0	cleavelandite - 1st intermediate	Clv + Elb + Col (+ Ms + Cst + Zrn)
BMT-11	-5.0	cleavelandite - 1st intermediate	Clv + Elb + Col (+ Ms + Cst + Zrn)
BMT-75	-1.5	wall	Ab + Qtz + Ms + Srl (+ Grt + ?Ap)
BMT-3	-1.5	wall	Ab + Qtz + Ms + Srl (+ Grt + ?Ap)
BMT-47 (green)	-1.5	wall	Ab + Qtz + Ms + Elb (+ Grt + ?Ap)
BMT-47 (black)	-0.5	border zone	Qtz + Ms + Pl (+ Srl + Py)
BMCR-2	-0.1	border zone	Qtz + Kfs + Grt + Pl + Srl
BMCR-1	0.1	(at the contact)	Qtz + Ms + Mc + Pl + Po + (Rt + Ap + Drv)
BMCR-3	1.0	upper sillimanite	Qtz + Bt + Ms + Pl + Po + (Rt + Ap + Drv)
BMCR-4	2.0	upper sillimanite	Qtz + Bt + Ms + Pl + Po + (Rt + Drv + Sil)
BMCR-5	3.5	upper sillimanite	Qtz + Bt + Ms + Pl + Sil + Po + (Rt + Drv)
BMCR-6	4.5	upper sillimanite	Qtz + Bt + Ms + Pl + Po + (Rt + Ap + Drv + Ttn)
MM-I-G	-12.0?	core/pocket zone	Clv + Qtz + Elb
MM-I-P	-12.0?	core/pocket zone	Clv + Qtz + Elb
MM-H-B	-12.0?	core/pocket zone	Clv + Qtz + Elb
MM-F	-6.0	wall zone	Qtz + Fsp + Srl
MM-D	-2.5	wall zone	Qtz + Pl + Ms + Srl
MM-E1	-2.0	wall zone	Qtz + Pl + Ms + Ap + Srl
MM-E2	-2.0	border zone	Qtz + Pl + Ms + Grt + Ap + Srl
MM-A	0.1	metasomatic zone at the contact	Qtz + Fsp + Ms + Bt + Drv
MM-B	0.5	in pegmatitic pod in country rock	Qtz + Fsp + Srl
MM-C	2.5	mica schist	Qtz + Sil + Bt + Ms + Pl + Kfs + Grt + (Ilm + Tur + Ap)
MM-G	15.0	mica schist	Qtz + Sil + Bt + Ms + Pl + Kfs + (Ilm + Ap) (rare Drv)

*Distance from pegmatite-country-rock contact. (accessory minerals given in parentheses) (Clv = cleavelandite; Col = columbite; other symbols after Kretz, 1983)

Methods

Most previous studies of tourmaline using microbeam techniques have assumed B and H contents for tourmaline and avoided the issue of Fe³⁺ altogether, but by doing so, they may have obscured subtleties in its crystal chemical variation. Therefore, tourmaline compositions in this study were characterized by multiple analytical techniques in order to obtain the best possible (*i.e.*, complete except for oxygen) chemical compositions for each sample. Preference was given to use of microbeam methods where possible, in order to avoid the averaging effects resulting from the use of bulk samples.

Electron microprobe

Tourmaline compositions were determined on

BMT country rock samples and all MM samples using the JEOL automated electron microprobe (EPMA) at Rutgers University and at the University of Maine using their ARL-SEM-Q (Tables 2 and 3). Methodology at Rutgers is described in detail in Delaney *et al.* (1996); those operating conditions were: 15 kV accelerating voltage, 20 nA beam current, 30 s count times, and a focussed beam. Natural and synthetic mineral standards were used and matrix correction was done by the Noran ZAF routine. At the University of Maine, EPMA analyses were done on the ARL-SEM-Q microprobe automated with an Advanced Microbeam Micro-3WD system using 15 kV accelerating voltage, 10 nA beam current, 10 s count times, a focussed beam, and Phi-Rho-Z matrix corrections (Donovan *et al.*, 1992). Analytical errors on all analyses are $\pm 1-2$ relative

Table 2. Chemical compositions of tourmaline, Mount Mica samples.

	Samples from within the pegmatite							Samples from country rock		
	I-G	I-P	H-B	F	D	E1	E2	A	B	G
SiO ₂	37.98	37.94	36.41	34.53	34.64	34.19	35.89	35.16	35.15	35.35
Al ₂ O ₃	42.16	43.38	39.11	33.44	33.59	34.16	35.37	33.46	34.87	33.81
TiO ₂	0.03	0.08	0.06	0.30	0.43	0.42	.26	0.80	0.38	0.85
FeO	0.94	0.00	6.24	13.42	12.32	13.39	11.05	7.18	8.81	6.71
Fe ₂ O ₃	0.00	0.00	0.00	0.37	0.87	0.00	0.51	0.00	0.00	0.00
MgO	0.01	0.02	0.01	0.93	1.27	1.19	0.73	5.88	4.19	5.85
MnO	0.22	0.23	1.16	0.23	0.24	0.22	0.41	0.05	0.11	0.03
CaO	0.54	0.48	0.04	0.08	0.09	0.08	0.08	0.30	0.27	0.72
Na ₂ O	1.85	1.77	2.10	1.74	1.83	1.82	2.04	2.04	1.79	1.92
K ₂ O	0.01	0.01	0.01	0.05	0.05	0.06	0.05	0.06	0.03	0.05
B ₂ O ₃	11.77	11.51	11.27	10.04	10.37	10.12	10.51	10.47	10.46	10.44
ZnO	0.02	0.01	0.48	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.02
Cr ₂ O ₃	0.01	0.01	0.02	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.06
Li ₂ O	2.46	2.54	1.30	0.05	0.07	0.04	0.08	0.01	0.01	0.01
F	1.32	1.35	0.97	0.28	0.28	0.40	0.62	0.22	0.08	0.21
H ₂ O	3.19	3.27	3.05	2.67	2.87	3.22	2.92	3.02	3.01	3.01
-O=F	0.66	0.68	0.49	0.14	0.14	0.20	0.31	0.11	0.04	0.11
Total	102.51	102.59	102.25	98.22	99.03	99.44	100.68	98.75	99.25	99.04
Si	5.826	5.791	5.811	5.972	5.908	5.814	5.969	5.875	5.869	5.883
Al	7.622	7.804	7.356	6.816	6.752	6.846	6.933	6.589	6.861	6.631
Ti	0.003	0.009	0.007	0.039	0.055	0.054	0.033	0.101	0.048	0.106
Fe ³⁺	0.121	0.000	0.833	1.941	1.758	1.904	1.537	1.003	1.230	0.934
Fe ²⁺	0.000	0.000	0.000	0.048	0.112	0.000	0.064	0.000	0.000	0.000
Mg	0.002	0.005	0.000	0.240	0.323	0.302	0.181	1.465	1.043	1.451
Mn	0.029	0.030	0.157	0.034	0.035	0.032	0.058	0.007	0.016	0.004
Ca	0.089	0.078	0.007	0.014	0.016	0.015	0.014	0.054	0.048	0.128
Na	0.550	0.524	0.650	0.582	0.605	0.600	0.658	0.661	0.579	0.619
K	0.002	0.002	0.008	0.010	0.011	0.013	0.011	0.013	0.006	0.011
B	3.116	3.032	3.106	2.997	3.052	2.970	3.018	3.019	3.015	2.998
Zn	0.002	0.001	0.057	0.000	0.000	0.000	0.000	0.000	0.000	0.002
Cr	0.001	0.001	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.008
Li	1.520	1.557	0.834	0.033	0.045	0.030	0.055	0.007	0.007	0.006
F	0.640	0.652	0.490	0.215	0.210	0.282	0.406	0.171	0.087	0.111
H	3.264	3.324	3.247	3.080	3.265	3.652	3.239	3.366	3.352	3.347

*No tourmaline grains could be extracted for analysis from sample MM-C. Only a single grain of tourmaline was found in MM-G, so H extraction was impossible; therefore the average of H₂O from MM-A and B was used.

Table 3. Chemical compositions of tourmaline from Black Mountain country rock samples.*

	BMCR-2 ¹	BMCR-1	BMCR-3	BMCR-4	BMCR-5
SiO ₂	34.91	34.89	35.75	35.95	36.12
Al ₂ O ₃	34.14	32.61	32.10	33.69	34.67
TiO ₂	0.13	0.83	0.85	0.68	0.56
FeO	11.84	7.24	4.82	1.95	1.08
Fe ₂ O ₃	0.00	0.00	0.00	0.00	0.27
MgO	2.10	6.27	8.57	10.05	9.92
MnO	0.26	0.28	0.04	0.04	0.03
CaO	0.04	0.87	1.06	1.31	1.25
Na ₂ O	1.82	1.89	1.97	1.62	1.47
K ₂ O	0.03	0.04	0.00	0.04	0.03
B ₂ O ₃	10.11	9.97	10.67	10.85	10.94
ZnO	n.a.	n.a.	n.a.	n.a.	n.a.
Cr ₂ O ₃	n.a.	n.a.	n.a.	n.a.	n.a.
Li ₂ O	0.10	0.11	0.06	0.04	0.04
F	0.30	0.48	0.21	0.31	0.21
H ₂ O	3.16	2.44	2.55	2.80	2.91
-O=F	0.15	0.24	0.11	0.16	0.11
Total	99.05	98.06	98.77	99.45	99.59
Si	5.914	5.935	5.952	5.856	5.839
Al	6.816	6.537	6.298	6.467	6.605
Ti	0.017	0.106	0.106	0.083	0.068
Fe ²⁺	1.677	1.030	0.671	0.266	0.145
Fe ³⁺	0.000	0.000	0.000	0.000	0.033
Mg	0.530	1.590	2.127	2.441	2.391
Mn	0.037	0.040	0.006	0.006	0.004
Ca	0.007	0.159	0.189	0.229	0.216
Na	0.598	0.623	0.636	0.512	0.461
K	0.006	0.009	0.000	0.008	0.006
B	2.954	2.926	3.073	3.052	3.053
Zn	0.000	0.000	0.000	0.000	0.000
Cr	0.000	0.000	0.000	0.000	0.000
Li	0.069	0.078	0.041	0.023	0.025
F	0.222	0.331	0.162	0.221	0.158
H	3.570	2.768	2.832	3.042	3.138

*Sample BMCR-6 contained no tourmaline in the thin section used for analyses. ¹This sample was collected as part of the country rock suite; however, on the basis of its location and assemblage it is grouped with the pegmatitic samples throughout this ms.

percent for major elements, \pm 5-10 relative percent for minor elements. For tourmaline from within the BMT pegmatite, analyses are given in Dyar *et al.* (1998); those EMPA data are averages of analyses done as part of a round robin, interlaboratory microprobe comparison study (Dyar *et al.*, in preparation). Up to 41 different analyses of each sample were used, representing analyses done at the University of Houston, Rice University, USGS Reston, National Museum of Natural History, plus Maine and Rutgers as noted above.

Particle-Induced (γ -Ray Emission (PIGE))

PIGE analyses of Li, B, and F were performed by David Robertson at the University of Kentucky

accelerator laboratory using the following nuclear reactions: $^7\text{Li}(p,p')^7\text{Li}$, $^{10}\text{B}(p,p')^{10}\text{B}$, $^{10}\text{B}(p,\alpha)^7\text{Be}$ and $^{19}\text{F}(p,p')^{19}\text{F}$. The application of the method is described in detail in Robertson & Dyar (1996). Relative standard deviations on these values are estimated at 5-15 %. The PIGE method uses bulk (100 mg) samples, so it was difficult to correlate those B and Li results with microbeam SIMS data on the same elements. Because microscale measurements were preferred for consistency with the electron microprobe analyses, it was decided to use the SIMS data for B and Li and the PIGE data for F only for the samples from within the BMT pegmatite itself.

Mössbauer analyses

Mössbauer analyses were performed by M.D.D. and Marjorie Taylor in the Mineral Spectroscopy Laboratory at West Chester University and by Yao-min Xia at the University of Illinois. Sources of 50-20 mCi ^{57}Co in Pd were used on two constant acceleration spectrometers. Results were calibrated against an α -Fe foil of 6 μm thickness and 99 % purity. Spectra were fitted using a version of the program STONE modified to run on IBM and compatible personal computers. The program uses a nonlinear regression procedure with a facility for constraining any set of parameters or linear combination of parameters. Lorentzian line shapes were used for resolving peaks, as there was no statistical justification for the addition of a Gaussian component to the peak shapes. A statistical best fit was obtained for each model for each spectrum using the χ^2 and Misfit parameters; practical application of these parameters is discussed by Dyar (1984). For samples with < 1 wt% total Fe as FeO, the errors on $\text{Fe}^{3+}/\Sigma\text{Fe}$ are estimated at \pm 3 % for doublet areas; for samples with > 1 wt% total Fe as FeO, $\text{Fe}^{3+}/\Sigma\text{Fe}$ errors are 1-3 %. Fitting methods for tourmaline spectra are discussed in Taylor (1996) and Dyar *et al.* (1998).

Secondary Ion Mass Spectrometry (SIMS)

Analyses of B and Li were performed by Grant Fowler and Michael Wiedenbeck at the University of New Mexico/Sandia National Laboratory SIMS facility using a Cameca IMS 4f ion microprobe and the same grain mounts used for electron microprobe study. A primary O-plasma accelerated to 12.5 kV was used, and the 20 nA beam was

focused to a diameter of $\sim 25 \mu\text{m}$. Secondary ions were accelerated using a nominally 4.5 kV potential to which a 50 V offset energy filter was applied using a 25 V full width energy window. The secondary ion beam was mass separated using a 90° sector magnet operated in low mass resolution mode ($M/dM = \sim 360$). Ions were detected using an ETP electron multiplier operated in pulse counting mode; count rates were kept below ~ 20 MHz in order to minimize the impact of detector dead-time corrections.

Li and B concentration analyses involved magnet peak-stepping that included four mass stations: 6.5 background (2 s), ${}^7\text{Li}$ (2 s), ${}^{11}\text{B}$ (3 s) and ${}^{30}\text{Si}$ (2 s); all data were normalized to the observed ${}^{30}\text{Si}$ intensity. Mineral standards analyzed by nuclear techniques and wet chemistry were used (Francis *et al.*, 1994). All samples were analyzed twice and the results were averaged to obtain the results given in Tables 2 and 3. Standard deviations were generally less than 0.02 wt% B_2O_3 and Li_2O . Additional information on the method is given in Hervig (1996).

Hydrogen analysis

Hydrogen contents were determined by means of a procedure for collecting and measuring all the structural H using a volumetric measurement of water vapor extracted from silicates (see Bigeleisen *et al.*, 1952 and Holdaway *et al.*, 1986 for details of the technique) by M.D.D. and Kurt Ferguson at Southern Methodist University. For this study, every fifth sample was run twice; distilled H_2O was used for calibration. All data are corrected for the blank of 2.5 torr (roughly 0.5–2% of the total wt% H_2O , depending on sample size and volume of H_2O vapor). Errors are estimated at ≤ 0.1 wt% H_2O based on replications. The hydrogen samples were then analyzed for their isotopic compositions on a Finnigan MAT 252 mass spectrometer. Results are given in Table 4; the precision of those analyses is $\pm 2\%$.

Hydrogen (and oxygen) isotope values are reported in this paper in units of per mil (‰), and mineral pair fractionations are expressed as Δ_{A-B} where

$$\Delta_{A-B} = \delta_A - \delta_B \sim 1000 \ln \alpha_{A-B}$$

Table 4. Stable isotope data (given in ‰) and calculated temperatures.

Sample	δD Tur	$\delta^{18}\text{O}$ Tur	$\delta^{18}\text{O}$ Ms	$\delta^{18}\text{O}$ Qtz	Qtz-Tur			Qtz-Ms			
					Kotzer ¹	K&K ²	Zheng ³	B'92 ⁴	B&J ⁵	Eslinger ⁶	Zheng ³
BMT-A	-75.7	12.52	12.30	14.01	680	707	757	660	703	622	829
BMT-19	-58.1	14.57									
BMT-17	-65.9	12.61									
BMT-49	-71.4	12.02									
BMT-65	-68.9	12.80		14.60	569	586	650	611			
BMT-B	-68.2	13.04	11.98								
BLKMTN-33	-77.1	12.36	11.64	14.52	478	490	554	557	522	370	541
BMT-20	-72.0	13.20									
BMT-11	-81.7	13.56									
BMT-75	-80.0	12.60									
BMT-3	-81.0	13.46	11.78	14.64	852	899	894	722	524	373	545
BMT-47 (green)	-70.9										
BMT-47 (black)	-74.8	12.60	11.74	14.75	481	492	556	466	508	353	519
BMCr-2	-84.7	12.36	12.10	14.54	474	485	549	567	578	438	628
BMCr-1	-82.9	12.61	11.47	13.50	1141	1244	1065	793	641	525	730
BMCr-3	-96.4	12.28		12.78	*	*	*	907			
BMCr-4	-100.6	12.54	11.57	13.55	997	1068	988	762	650	539	744
BMCr-5	-80.4	12.67	11.82	12.72	*	*	*	1109	938	1238	1215
MM-I-G	-113.1	14.18									
MM-I-P	-111.1	13.77									
MM-H	-130.8	14.24									
MM-F	-105.4	13.22		15.28	501	513	578	575			
MM-D	-110.1	13.54	13.22	15.35	565	581	646	609	625	501	703
MM-E1	-112.9	13.41									
MM-E2	-116.4	13.49	13.31	15.25	581	599	663	617	658	549	756
MM-A	-87.2	14.00	13.50	16.54	409	417	473	518	504	350	514
MM-B	-90.6	14.13	13.91	16.65	412	420	477	520	538	389	567

¹Kotzer *et al.* (1993). ²King & Kotzer, in preparation, cited in King (1990). ³Zheng (1993). ⁴Blamart *et al.* (1992).

⁵Bottinga & Javoy (1975). ⁶Eslinger (1971). * indicates samples with absurdly high calculated temperatures.

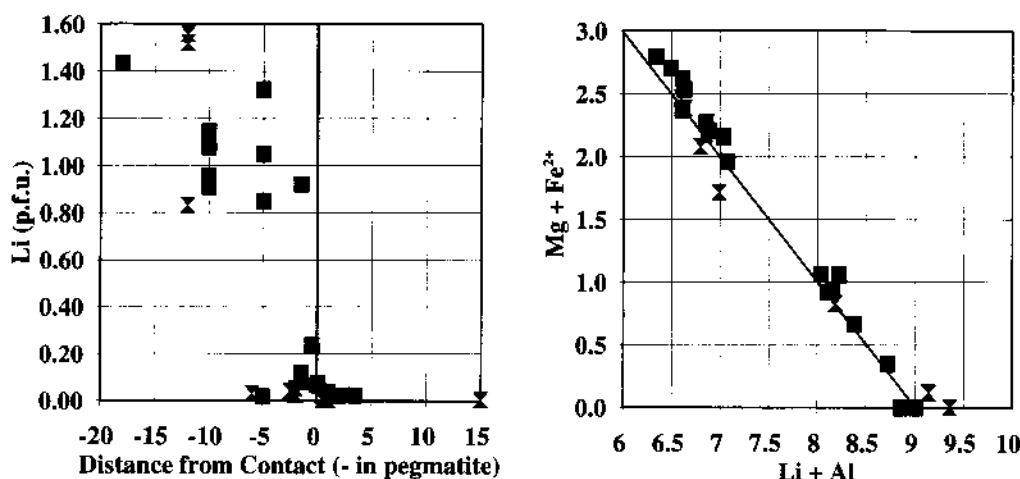


Fig. 2. (A) Li contents of Tur in each of the two pegmatites are plotted here as a function of distance (expressed in m) into the pegmatite. Squares represent samples from Black Mountain and hourglasses represent Mount Mica. (B) $(\text{Li} + \text{Al}) \leftrightarrow (\text{Mg} + \text{Fe}^{2+})$ substitution is represented in the suite analyzed here, with a gap between the schorl-dravite and elbaite ends of the continuum. The line shown in (B) has a slope of -1 which would represent 1:1 substitution; a regression line fit to these data would have a slightly gentler slope suggesting the effects of other minor substitutions such as those involving Mn. Overall, these trends reflect the decreasing concentration of Li out from the core of both pegmatites. Note that this relationship changes systematically from core to contact and from contact into country rock at both localities.

Oxygen isotope analysis

The oxygen isotopic composition of silicate minerals was determined using the method of Clayton & Mayeda (1963). Approximately 10 mg of sample were placed in a nickel reaction vessel with approximately 170 torr of BrF_5 and reacted at 400°C for at least 14 hours. The liberated O_2 was then converted to CO_2 by heating it with a graphite rod. The CO_2 was collected and analyzed on a Finnigan MAT Delta E mass spectrometer. The data are reported as δ values with respect to Standard Mean Ocean Water. Replicate analyses of the standard NBS-28 give an average value of 9.69 ‰ with a standard deviation of 0.12 ‰ ($n = 10$). The expected value of NBS-28 is 9.65 ‰. The reproducibility of oxygen isotope data for the analyzed samples is about 0.2 ‰.

Summary of analytical results

The process of obtaining high quality, complete compositional data for tourmaline is not a simple matter. In this study it involves two microbeam techniques (EMPA for all reasonably expected major elements in the context of the petrologic set-

ting involved and SIMS for the light elements B and Li), and three techniques requiring the use of highly purified bulk sample separates (Mössbauer for Fe^{3+} , PIGE for F, and uranium extraction for determination of structural H_2O). Thus, the results of five variably disparate approaches were amalgamated to achieve an analytical sum of $100 \pm 2.5\%$.

Chemical formulae

All tourmaline compositions are normalized to 31 oxygen totals, minus the applicable number of halogen ions, following the method of Deer *et al.* (1992) for clinohumite analysis. This procedure subtracts the oxygen equivalent of fluorine from the total oxide percentages, and also subtracts half the fluorine contents from the atom proportion total. Details are given in Appendix 1 of Deer *et al.* (1992). This procedure was necessary in order to have formulas that charge balance to neutrality, because the more commonly used method of simply subtracting twice the wt% F from the number of normalizing oxygens does not yield electronically neutral formulas. However, in some samples this method results in stoichiometries in which the sum of cations in the sites that are known to be full

from XRD data (*i.e.*, the Si, B, Y, and Z sites) is greater than the stoichiometric 18 cations. Dyar *et al.* (1998) have shown that such "nonstoichiometry" cannot be correlated with any particular analytical variables, and it is therefore the result of random analytical errors propagated from the oxide measurements. Accordingly, the recalculated data presented in Tables 2 and 3 are given with the caveat that they should not be used for comparisons where site filling is required; for such comparisons, a more valid approach is to normalize the oxide data so that the sum of cations in the Si + B + Z + Y sites is equal to exactly 18. Both methods are equally valid, and each has its advantages. In the discussion below, cations p.f.u. for the 31 O recalculation method will be used.

Trends in major element chemistry

Major element variations of tourmaline in various zones of pegmatites have been discussed elsewhere (*e.g.*, Dietrich, 1985; Jolliff *et al.*, 1986) and need not be repeated in detail here. However, these studies lacked complete chemical analyses, and therefore a few additional comments can be made here regarding chemical changes between pegmatite zones. The most dramatic change is the shift from schorl to elbaite compositions toward the cores of both pegmatites. At MM, only the core tourmaline contains significant Li; at BMT, samples from the core into and including the cleavelandite-1st intermediate zone contain varying but significant amounts of Li. Only at BMT do the country rocks contain significant (*i.e.*, > 0.01 wt% Li₂O) (Fig. 2a). The combined data sets show a 1:1 substitution of Mg + Fe²⁺ for Li - Al with a gap in the middle of the series (Fig. 2b). Similarly, Dietrich's (1985) larger compilation of literature data (his Fig. 4-2) also shows a paucity of data between 30% and 50% along the elbaite-schorl join, but data from the Bob Ingersoll pegmatite in the Black Hills (Jolliff *et al.*, 1986) do not. The ratio of Li + Al / Mg + Fe²⁺ increases from country rock to contact and into the core at both localities (Tables 2 and 3).

The change in B (p.f.u.) in tourmaline across the contact from pegmatite core into country rock is not significant, suggesting B saturation throughout the system. What is significant, however, is the fact that almost all our analyses show B in excess of 3 atoms p.f.u., implying ubiquitous substitution of B into the Si site as suggested earlier by Dyar *et al.* (1994). This observation is consistent with preliminary conclusions of single crystal XRD stud-

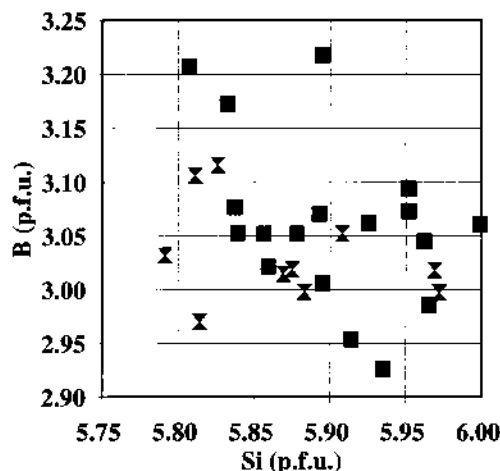


Fig. 3. In many samples, B contents of tourmaline are greater than the 3 atoms of B p.f.u. that would be predicted if B occupies only the B site. The suggestion of an inverse relationship between Si and B, shown here, may imply that B is substituting for Si, especially at BMT. Squares represent samples from Black Mountain and hourglasses represent Mount Mica.

ies on in progress at Miami University of Ohio that suggest slightly low Si site occupancies in tourmaline (E. Bloodaxe, personal communication, 1997). The data on Si and B have probably the largest error bars of any of the cations analyzed, yet the plot of Si vs. B shown in Fig. 3 is not inconsistent with Si ↔ B substitution, especially for the BMT samples. The data shown here neither support nor rule out this hypothesis, probably because of large error bars on measurements of both Si and B, and because those errors are propagated by the fact that highly charged cations have a disproportionate effect on normalized mineral compositions. There is no systematic difference between metamorphic and pegmatitic samples with regard to B contents.

Variation in the H and F contents of tourmaline is shown in Fig. 4a. No systematic trend is evident in the tourmaline from MM. However, at BMT the country rock samples are generally lower than pegmatite samples as might be expected for metamorphic vs. igneous samples. These data show variation greater than the analytical errors, and far greater than the amount of Fe³⁺ present in any of these samples. Thus the variation cannot be explained by oxy substitution. Foit & Rosenberg (1977) examined the substitution of (OH) ↔ R¹ + O²⁻ in their much larger data set of naturally-

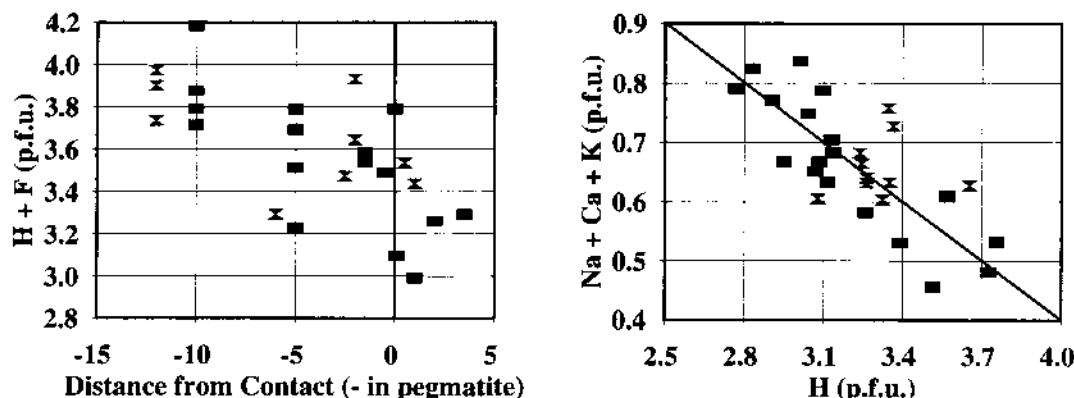


Fig. 4. (A) H + F contents decrease from the core to country rock (expressed in m) at both localities studied. Squares represent samples from Black Mountain and hourglasses represent Mount Mica. (B) All of the variation in H contents can be explained by the substitution of H_3O^+ into apparent vacancies in the X site, as suggested by the line with a slope of $-1/3$ shown here. This trend is best demonstrated by the Black Mountain data.

occurring tourmaline and found only a "relatively insignificant" correlation. However, in the current study, H is strongly inversely correlated with X site occupancy in a ratio of $3\text{H} : 1\text{X}$ cation (Fig. 4b). This strongly suggests that some contribution from H_3O^+ must be filling the apparent vacancies in the X site. This would imply that X cations control the amount of H in the tourmaline from both localities.

Variation of Al content (Fig. 5) is probably explained by the decrease of elbaite (*i.e.*, Li) substitution. At BMT, there is a steady decrease in Al within the pegmatite from the contact going toward the core; the change within MM is more bimodal but still shows a decrease from core to contact. At MM there is only a small change in Al across the contact, perhaps because the samples are all Al-saturated (as indicated by the presence of Sil in the assemblage). It is interesting to note that the only BMT country rock sample that contains Sil has a similar (to MM) Al content in its tourmaline.

In addition to the crystal chemical trends evident in all the tourmaline samples studied here, some systematic differences are observed between pegmatite and country rock tourmaline at the two localities studied. Most likely these differences are related to variations in bulk composition and mineral assemblage, which would include considerations such as: 1) the presence of biotite in the country rock at both localities; 2) the change in modal feldspar composition from plagioclase toward K-feldspar (see Table 1); 3) the addition of

oxide phases in the country rock at both locations: rutile at BMT and ilmenite at MM; and 4) the addition of pyrrhotite to the assemblage at BMT only.

Of these, the most significant differences between the two pegmatites involve Mg/Fe²⁺.

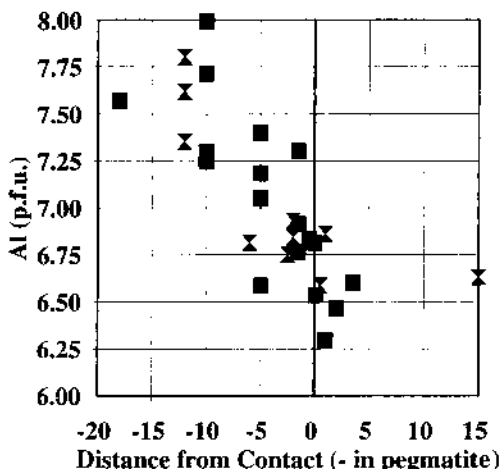


Fig. 5. Al contents of tourmaline decrease from pegmatite cores into surrounding country rocks at both localities studied. Squares represent samples from Black Mountain and hourglasses represent Mount Mica. In general, Black Mountain is a more complex pegmatite, with continuous crystallization trends, while Mount Mica has essentially bimodal tourmaline chemistry with wall zone schorl and core elbaite.

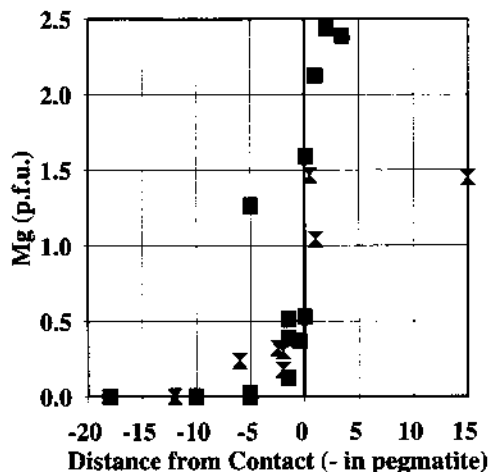


Fig. 6. Mg content of tourmaline is negligible at the cores of both pegmatites studied, but increases sharply near the contact into country rock. Squares represent samples from Black Mountain and hourglasses represent Mount Mica.

Within the pegmatites, the Mg/Fe²⁺ variation is probably responding to the amount of Li and Li + Al elbaite substitution as noted earlier (Fig. 2b). The Mg/Fe²⁺ covariation *across contacts* is very likely a reflection of sulfidation at BMT vs. no sulfidation at MM. This difference is best illustrated by the plot of Mg (p.f.u.) vs. distance across the contact shown in Fig. 6. At BMT, Mg contents continue to increase sharply into the country rock relative to the pegmatite; at MM the three country rock samples have comparable Mg contents. A possible explanation for this change is that the Mg in the outer zones of both pegmatites is increased due to chemical exchange with the country rock.

Stable isotope geochemistry

Hydrogen isotopes

As has been noted in the stable isotope literature (*e.g.*, King, 1990), tourmaline is a useful geochemical indicator due to its wide stability range and relative insensitivity to subsequent thermal and fluid events that might overprint isotopic signatures. Therefore the stable isotopes in the two different pegmatites might be expected to preserve and record differences in conditions of formation if major element compositional effects can be ruled out. The most likely cations to influence

hydrogen partitioning are Fe²⁺ and Mg, as is the case for biotite and other micas (Suzuoki & Epstein, 1976); however, the isotopic values measured in this study are independent of Fe²⁺/Fe³⁺ + Mg. Consistent with the conclusions reached by King (1990) for hydrothermal Tur and Blamart *et al.* (1992) for metapelite Tur, wt% H₂O contents do not correlate with δD either. Therefore, the observed variations can be interpreted to reflect differences in the temperatures and isotopic compositions of the fluids that formed them.

At BMT, there is a general trend of decreasing δD_{Tur} from 2nd intermediate zone toward and into country rock (Fig. 7), with the exception of the core sample BMT-A and sample BMCR-5. There is no distinct break in δD_{Tur} values between tourmaline from the pegmatite and tourmaline from the country rock; in fact, the contact appears to be isotopically gradational. The existence of this gradient implies some exchange of fluids between pegmatite and country rock. Perhaps the original primary magmatic isotopic signature of the pegmatite itself may be preserved in its core.

The δD values of Tur in the MM pegmatite are significantly lower than those in the BMT pegmatite or in the country rock at either locality (Fig. 7). The contact between the pegmatite and the country rock is sharp rather than gradational, and it is clear that the δD is not equilibrated across the contact. Very little exchange of H could have taken place between the pegmatite and the country

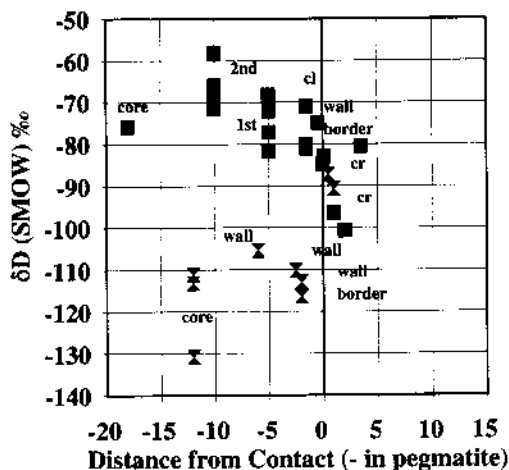


Fig. 7. At BMT (squares), δD decreases from the 2nd intermediate zone across the contact and into the country rock. A different trend is observed at MM (hourglasses), where $\delta D_{\text{country rock}} \gg \delta D_{\text{pegmatite}}$.

rock. However, the very low (-105‰ to -131‰) δD values observed within the pegmatite suggest interaction of the primary magma with meteoric water. Two possible scenarios might give rise to the light values: 1) pre- or syn-emplacement interaction between the primary pegmatitic magma and the meteoric waters, or 2) structure-controlled infiltration of regional fluid flow, possibly channeled parallel to the foliation of the country rock schist, resulting in contact between the meteoric waters and the pegmatite but not the country rock.

In summary, the δD ranges of the BMT pegmatite (-80‰ to -60‰) and the MM samples are so different that it is difficult to come up with a mechanism that would allow them to have the same primary magma. Either there were different fluids in the magmas from which the pegmatites crystallized, or the two locations experienced significantly different retrograde histories. If the core sample of BMT is indeed a primary δD value, and if the core samples from MM are also primary, then it appears that the former hypothesis is more likely.

Oxygen isotopes

Oxygen isotope values were measured for tourmaline and coexisting quartz (Qtz) and muscovite (Ms) from as many samples as feasible. Where possible, temperatures were calculated from the $\delta^{18}O$ values for coexisting Qtz-Ms and Qtz-Tur pairs (Table 4) using both empirical and theoretical calibrations (Eslinger, 1971; Bottinga & Javoy, 1975; King & Kotzer, in preparation, cited in King, 1990; Blamart *et al.*, 1992; Zheng, 1993; Kotzer *et al.*, 1993).

At BMT, no significant distinctions between pegmatite and country rock are seen for $\delta^{18}O_{Tur}$ and $\delta^{18}O_{Ms}$; however, $\delta^{18}O_{Qtz}$ is significantly lower in the country rock than in the pegmatite (Fig. 8). This suggests that oxygen in tourmaline and muscovite equilibrated across the contact. However, quartz underwent substantial subsolidus post-crystallization re-equilibration with a lighter fluid, as evidenced by the low Qtz-mineral temperatures in the pegmatite, especially in the immediate area of the contact between the pegmatite and the country rock. The country rock samples give extremely high, disequilibrium temperatures for Qtz-Tur, suggesting that either the fractionations are out of the appropriate temperature ranges for the geothermometers or that quartz is out of equilibrium with the rest of the system as noted above.

At MM, $\delta^{18}O_{Tur}$ is highest in the core, lowest

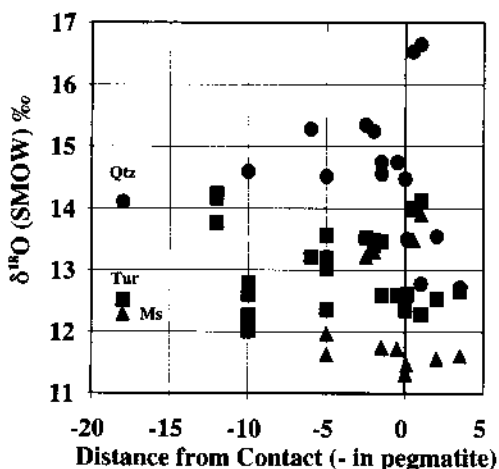


Fig. 8. Differences in $\delta^{18}O$ are shown for tourmaline (squares), muscovite (triangles), and quartz (circles) as a function of distance (expressed as m) from the contact between pegmatite and country rock. Black symbols represent BMT samples, and gray symbols represent MM.

at the wall, and then high again in the country rock, while the four data points for $\delta^{18}O_{Ms}$ show a small increase from wall to exocontact into country rock. Values for $\delta^{18}O_{Qtz}$ show dramatic differences between pegmatite ($\sim 15.3\text{‰}$) and country rock ($\sim 16.5\text{‰}$), and these result in two apparent groups of temperatures: $\sim 600^\circ\text{C}$ inside the pegmatite and $\sim 450^\circ\text{C}$ in the country rock. The magnitude of this difference is greater than the error bars, suggesting that re-equilibration was definitely more marked in the country rocks. This might suggest that once the coarse-grained pegmatites crystallized, they effectively underwent closure, and lost contact with fluids that might have allowed them to re-equilibrate. The finer-grained country rock samples, on the other hand, continued to exchange oxygen during cooling (A. Matthews, personal communication, 1998).

On the δ - δ plot shown in Fig. 9, the differences between the two pegmatites are clearly shown. At BMT, it is apparent that the Qtz is undergoing most of the retrograde $\delta^{18}O$ change, because Tur and Ms remain relatively constant while Qtz varies over 2.5‰ . It is interesting that the core sample is closest to the country rocks on the δ - δ plot; this may imply that the core formed in equilibrium with the surrounding rocks. At MM, there is little evidence for any kind of interaction between the pegmatite and the country rock. Each

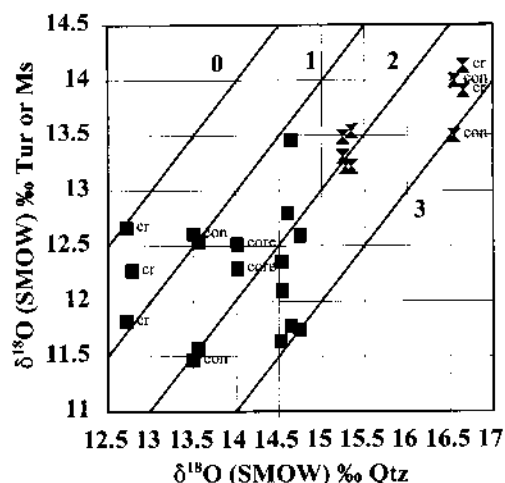


Fig. 9. Fractionation of $\delta^{18}\text{O}$ between Qtz-Tur (black squares for BMT and hourglasses for MM) and Qtz-Ms (gray symbols). Core, contact (con) and country rock (cr) data are labeled; unlabeled points correspond to

appears to have equilibrated and cooled distinctly, and it is even possible that the country rock at MM was never heated to the temperatures experienced by the pegmatite.

Discussion and conclusions

Major element crystal chemistry of tourmaline from Black Mountain and Mount Mica is similar and typical for Li-rich rare element pegmatites. The well-known trend of $\text{Li} + \text{Al} \leftrightarrow \text{Mg} + \text{Fe}^{2+}$ substitution appears discontinuous. The strong negative correlation of Hl with $\text{Na} + \text{Ca} + \text{K}$ indicates that vacancies are not present as has been previously assumed; instead, H_2O^- fills the X sites. B contents of up to 3.22 B p.f.u. are observed, and substitution of the excess B for Si cannot be ruled out. Chemical composition of tourmaline also reflects mineral assemblage of the rocks in which they are found, especially Mg/Fe^{2+} , which increases as Fe^{2+} is taken up by pyrrhotite in the country rock at BMT. Aside from modest differences in the Mg content of tourmaline in the country rock at the two localities, there is little in the major element compositions of tourmaline at the two localities to distinguish them.

However, stable isotopes strongly suggest that two contrasting styles of pegmatite are represented here: BMT, in which the chemistry changes

gradually from pegmatite into surrounding country rocks, and MM, which contains tourmaline, quartz, and muscovite that are clearly isotopically distinct from tourmaline in the surrounding country rock. There are obvious differences in the characteristics of the regional geology. BM is an isolated body that occurs in sillimanite zone, highly sulfidic metapelites and quartzites. In contrast, MM occurs in an area where at least 50% of the rocks outcropping are pegmatite and aplitic granite bodies that commonly have gradational contacts with each other. The remainder of the rocks there are migmatitic Sil-Kfs zone metapelites.

Can the differences in tourmaline chemistries at the two localities be attributed to different formation processes for the two occurrences? Older ideas for pegmatite genesis (*i.e.*, before Jahns & Burnham, 1969) suggested that pegmatites could have anatectic origins. Rare earth element geochemical data recently obtained by Simmons and coworkers (Falster *et al.*, 1997; Simmons *et al.*, 1995 and 1996) tentatively support this conclusion for pegmatites occurrences both in Wisconsin (Hoskin Lake) and in western Maine. Their model suggests that intrusive granites supply heat that causes partial melting of metasediments, resulting in pegmatites with geochemical signatures very similar to those of the surrounding country rock. The model implies that some pegmatites are not the result of fractional crystallization of batholiths. This anatectic model might predict no change or gradational changes in the stable isotopes (*e.g.*, as seen in Fig. 7 and 8) and, potentially, in some major elements (*e.g.*, Li, H + F and Mg, as seen in Fig. 2a, 4a and 5) across the contact between pegmatite and country rock. This is, in fact, what is observed at BMT. Perhaps the presence of Li in the country rock at BMT might have facilitated anatexis by acting as a flux to further melting.

In contrast, crystal chemistry (for example, the change in Mg shown in Fig. 6) and stable isotope chemistry (Fig. 7 and 8) of tourmaline at MM show sharp contrasts between pegmatite and country rock, consistent with a magmatic fractionation origin probably related to the nearby Sebago Batholith. Further REE (like those of Simmons *et al.*, 1995 and 1996) and stable isotope studies of tourmaline from the Sebago Batholith itself and other plutonic bodies in western Maine (currently in progress by these authors) should serve to characterize their isotopic signatures and facilitate clarification of the interrelationships between the bodies.

Acknowledgments: This research was supported by NSF grants EAR-9527020 to M.D.D. and EAR-9526403 to C.V.G. Additional support for D.C., K.W., K.J., M.D.D., and J.B.B. was provided by NSF grant EAR-9500026 and the W.M. Keck Foundation through the Keck Geology Consortium. We thank Ian Richards and Kurt Ferguson of the Stable Isotope Laboratory, Southern Methodist University, for their help with stable isotope analyses; Jeremy Delaney of Rutgers University and Martin Yates of the University of Maine for electron microprobe data; and Yao-min Xia and Jun Wu of the Soil Science Department, University of Illinois for assistance with Mössbauer analyses. The ion microprobe data presented here were measured at the University of New Mexico/Sandia National Laboratories' SIMS facility, a national multi-user facility supported in part by NSF EAR 95-06611, and we thank Michael Wiedenbeck and Grant Fowler for the analyses. This manuscript was greatly improved by discussions with Steve Dunn and by the comments of Edward Grew, Alan Matthews, and Stephan Hoernes, to whom we are very grateful.

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Received 24 October 1997

Accepted 9 September 1998