

INTERNAL ZONATION AND CHEMICAL EVOLUTION OF THE BLACK MOUNTAIN GRANITIC PEGMATITE, MAINE

CATHLEEN D. BROWN[§] AND MICHAEL A. WISE

Department of Mineral Sciences, Smithsonian Institution, Washington, D.C. 20560, U.S.A.

ABSTRACT

The crystallization history of the Black Mountain granitic pegmatite, near Rumford, western Maine, is evaluated using compositions of rock-forming and accessory minerals. The pegmatite is strongly zoned; the zonation developed from the consolidation of a rare-element-enriched melt. Field and trace-element data show that the wall zone crystallized first from an initially B-enriched melt. Subsequent crystallization of three intermediate zones reflects changes in melt composition, which include a general decrease in levels of Mg, Ca, and Fe, and increases in levels of Be, Nb, Ta, Sn, and P. Significant enrichment in Li, Rb, Cs and F occurred during the latest stage of pegmatite crystallization prior to core development. Late albite-dominant units enriched in B, Nb>Ta, Sn and Zr replace primary zones; their chemical trends deviate from normal trends of fractionation (*e.g.*, late enrichment in Fe).

Keywords: granitic pegmatite, internal zonation, Black Mountain, Maine, tourmaline, spodumene, lepidolite, columbite, cassiterite.

SOMMAIRE

La cristallisation progressive de la pegmatite granitique de Black Mountain, près de Rumford, dans l'ouest du Maine, fait l'objet d'une évaluation fondée sur la composition des minéraux principaux et accessoires. Le massif de pegmatite est fortement zoné; cette zonation s'est développée par la consolidation d'un magma enrichi en éléments rares. Les relations de terrain et les concentrations des éléments traces montrent que c'est la zone externe, près de la paroi, qui a cristallisé d'abord à partir d'un bain fondu enrichi en bore. Par la suite, la cristallisation de trois zones intermédiaires témoigne de changements dans la composition du liquide, par exemple une diminution générale en concentration de Mg, Ca et Fe, et une augmentation en concentration de Be, Nb, Ta, Sn et P. Un enrichissement important en Li, Rb, Cs et F marque le stade final de cristallisation magmatique avant la formation du noyau de quartz. Des unités tardives à dominance d'albite enrichies en B, Nb>Ta, Sn et Zr remplacent ensuite les zones primaires; leurs tendances chimiques dévient du tracé évolutif normal d'un fractionnement magmatique, montrant par exemple un enrichissement tardif en fer.

(Traduit par la Rédaction)

Mots-clés: pegmatite granitique, zonation interne, Black Mountain, Maine, tourmaline, spodumène, lépidolite, columbite, cassitérite.

INTRODUCTION

The Black Mountain granitic pegmatite is located near the town of Rumford, Oxford County, western Maine. It hosts a diverse array of rare-element minerals including elbaite, beryl, spodumene, columbite, cassiterite, amblygonite–montebrasite and lepidolite. The workings, geology and mineralogy of the pegmatite and surrounding rocks were first described by Bastin (1911). Later studies (*e.g.*, Marble 1927, Bailey 1929, Verrow 1941, Hess *et al.* 1943, Maillot *et al.* 1949, Bjareby 1965, Gregory 1968) were generally simplistic descrip-

tions of the geology and mineralogy of the pegmatite. A detailed study by Cameron *et al.* (1954) resulted in a geological map of the pegmatite and a description of the internal zonation. Unfortunately, the zonal sequence described by these investigators is too simplified to be genetically meaningful. During our field examination of the pegmatite in the interval from 1991 to 1995, we were able to examine zones and units previously unexposed. In our study, we used textural features and mineral assemblages to define zones and units, which resulted in a resolution of major inconsistencies in previous mapping and a significant redefinition of the zonal

[§] E-mail address: brown.cathleen@nmnh.si.edu

sequence. These advances, in conjunction with compositional variations in tourmaline, muscovite, feldspar and columbite–tantanite from the various units, are used to evaluate the crystallization history of the pegmatite.

REGIONAL GEOLOGY

The Black Mountain pegmatite lies within the northern portion of the Oxford pegmatite field, close to the Mooselookmeguntic, Rumford, and Whitecap Mountain granitic plutons (Fig. 1). The Mooselookmeguntic and Rumford plutons are composed of several distinct units: 1) granodiorite, tonalite, and quartz diorite, 2) two-mica granite, and 3) two-mica granite with enclaves of gra-

nodiorite, tonalite and quartz diorite (Moench & Hildreth 1976). Pegmatitic granite occupies portions of the Rumford pluton and is described as a mixture of white granite, pegmatite and aplite (Moench & Hildreth 1976, Wise & Francis 1992). The Whitecap Mountain pluton also consists of fine-grained two-mica granite and coarse-grained pegmatitic leucogranite (Wise & Francis 1992).

Along the margins of the plutons are numerous bodies of granitic pegmatite. Mineralogically simple pegmatites lacking beryl or other rare minerals are composed of quartz, microcline (perthitic and graphic), sodic plagioclase, muscovite, biotite, schorl and garnet. In contrast, more complex bodies of pegmatite have well-

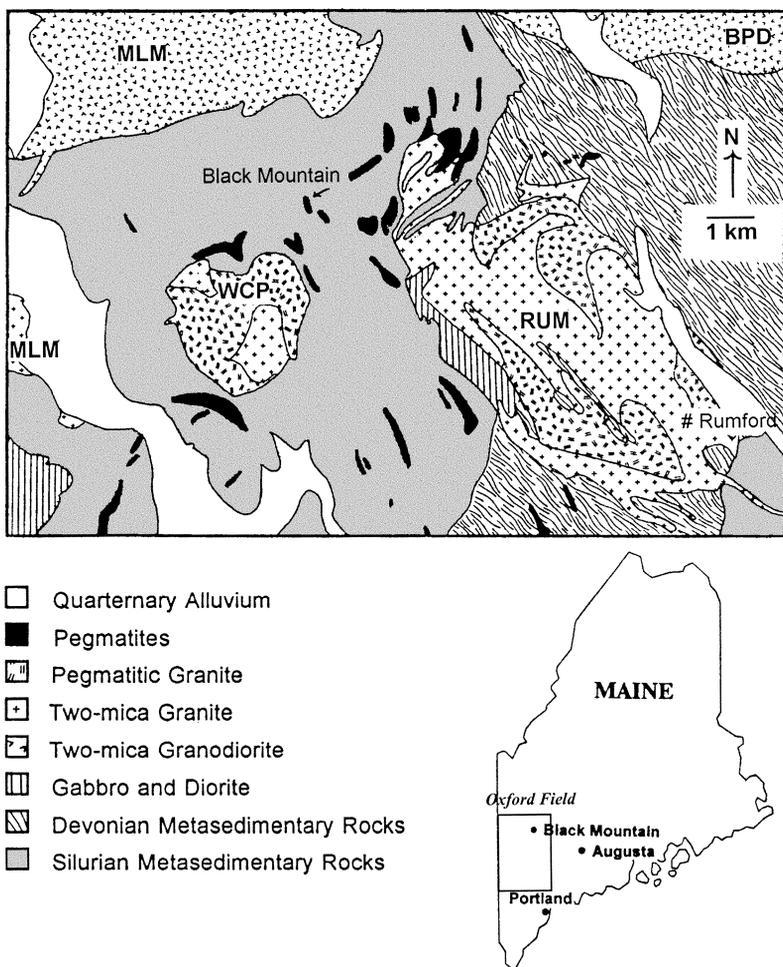


FIG. 1. Geological map of the Rumford area showing location of the Black Mountain pegmatite. General geology modified after Moench & Hildreth (1976). Symbols: BPD: Bunker Pond pluton, MLM: Mooselookmeguntic pluton, RUM: Rumford pluton, WCP: Whitecap Mountain pluton.

developed internal zonation and may contain beryl, columbite–tantalite and spodumene (Cameron *et al.* 1954).

INTERNAL ZONATION OF THE BLACK MOUNTAIN PEGMATITE

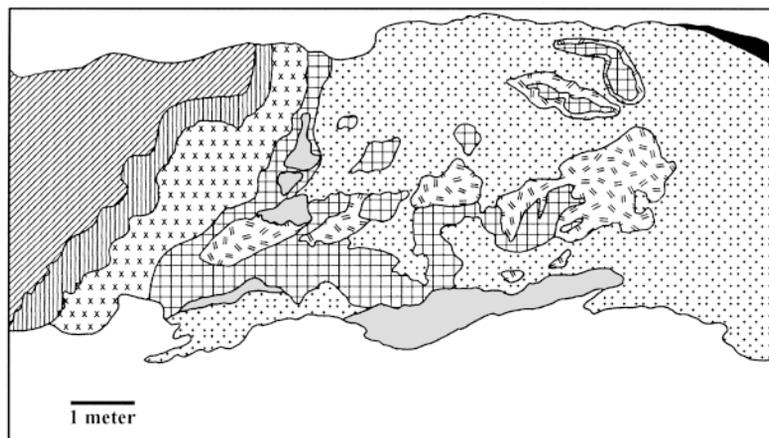
The Black Mountain pegmatite intrudes interlayered sulfide-bearing biotite schist, two-mica schist and impure quartzite of the Small Falls Formation (Maillot *et al.* 1949, Moench & Hildreth 1976). The thickness of the pegmatite (0.61 to 12.2 m) was determined from drill-core data (Maillot *et al.* 1949). Recent mining of the pegmatite has provided excellent exposures of the internal structure. Observed differences in mineralogy and texture were used to define five zones within the pegmatite (Fig. 2). Smaller units showing layered, aplitic, replacement or pseudo-granitic textures occur primarily in the innermost intermediate zones.

Muscovite, together with subhedral gray–white albite and anhedral gray quartz form the main constituents of the wall zone. Subordinate blue–black tourmaline, subhedral reddish crystals of almandine and rare

zircon, beryl and apatite also are present. A tourmalinized exocontact was locally observed between the wall zone and the biotite schist country-rock. Tourmaline is generally oriented parallel to the pegmatite – country-rock contact. Locally, aplite occurs as the first unit in contact with the country rock, followed by the wall zone. The aplite is a fine-grained mixture of quartz, albite, muscovite, black tourmaline and apatite. The tourmaline in the aplite is disseminated throughout and locally concentrated into bands.

The first intermediate zone consists primarily of medium- to coarse-grained muscovite (up to 75 cm long) and platy albite (“cleavelandite”). Flattened crystals of green and rarely blue–black tourmaline are commonly found between sheets of muscovite. Occasional pale yellow crystals of beryl and rare columbite, cassiterite and zircon are hosted by “cleavelandite”.

The second intermediate zone mainly consists of gray, massive, anhedral quartz and white to buff-colored, blocky, perthitic potassium feldspar. White amblygonite–montebrasite (up to 15 cm in diameter) and rare albite, pink tourmaline, zircon and columbite occur locally.



- ▨ Metasedimentary rocks
- ▤ Wall zone
- ⊗ First Intermediate zone
- ▣ Second Intermediate zone
- ⊙ Third Intermediate zone - QSK
- Third Intermediate zone - LEP
- Core
- ⊕ Replacement unit

FIG. 2. Schematic cross-section of the eastern exposure of the Black Mountain granitic pegmatite.

The third intermediate zone is characterized by four texturally distinct units (Fig. 2) whose contacts are transitional: 1) quartz – spodumene – microcline unit (QSK), 2) lepidolite pods (LEP), 3) tourmaline – lepidolite – albite unit (TLA), and 4) lepidolite – albite unit (LA).

Crystals of spodumene in the QSK unit occur as white elongate blades and range from approximately 3.5 to 34 cm in length. Blocky, white, buff or gray microcline occurs in crystals up to 8 cm in size. This unit also contains interstitial albite, accessory green and colorized (pink to green) tourmaline, and rare lepidolite.

Fine-grained lepidolite with minor albite and quartz occurs as pods and may contain crystals of predominantly pink tourmaline and rare columbite. Pale blue tourmaline is rarely found in these pods. The smaller pods of lepidolite are up to 30 cm in length and are in cases surrounded by K-feldspar. A large pod of lepidolite located near the first intermediate zone is approximately 3 m in length and ranges in color from violet to pale green to silver. These pods are small in comparison to the giant exposures (up to 3.6×5.8 m) of lepidolite bodies observed at Black Mountain by Hess *et al.* (1943) and Marble & Morrill (1945).

A fine- to medium-grained equigranular, hypidiomorphic mixture of lepidolite, albite, quartz and pink tourmaline with occasional blades of spodumene occurs as a prominent textural unit of the third intermediate zone. This unit commonly displays segregated layers of lepidolite plus pink tourmaline and albite, imparting a banded texture. This unit has been referred to by Bastin (1911) as a tourmaline granite; throughout this paper, this rock will be referred to as the tourmaline – lepidolite – albite unit (TLA). This unit is locally altered, consisting of friable, fine-grained albite, silver lepidolite, pink to green (\pm colorless or bicolored) tourmaline and minor quartz, zircon and Nb–Ta oxides. Typically, the amount of pink tourmaline in the TLA unit diminishes to the point where it is no longer present, resulting in a gray, granitic-textured lepidolite – albite unit (LA). This unit becomes coarsely layered, and consists of alternating 1-cm-thick bands of white–gray “cleavelandite” and silver lepidolite. The layered portion of the LA unit is considered to have been produced by the replacement of a lepidolite pod by “cleavelandite” along subparallel fractures. Also occurring in the LA unit are rare amounts of amblygonite–montebrasite, columbite, cassiterite and quartz.

Locally, the QSK unit becomes progressively depleted in microcline and more enriched in quartz and spodumene, and ultimately grades into what is interpreted as a quartz core located near the center of the exposure. Medium- to coarse-grained “cleavelandite” hosting rare books of silver-colored muscovite is common. Rare, minute grains of cassiterite and columbite were also observed in this zone.

A replacement unit consisting of coarse white–gray “cleavelandite” hosts large dark green–blue tourmaline,

bladed black–brown columbite, and brown–black massive crystals of cassiterite up to 3 cm across. Gray quartz occasionally occurs within the “cleavelandite”, whereas medium- to coarse-grained muscovite and blades of spodumene are rare. Zircon and green apatite is locally common, generally associated with columbite or cassiterite.

Saccharoidal albite occurs in two places at the Black Mountain pegmatite. In the wall zone, interstitial saccharoidal albite contains small (<1 mm in diameter) disseminated blue–black tourmaline and reddish garnet. Saccharoidal albite in the third intermediate zone is present as small lenses that contain disseminated minute grains of columbite, cassiterite and blue tourmaline.

EXPERIMENTAL METHODS

K-feldspar, albite, Nb–Ta oxides, tourmaline and mica-group minerals were collected from various zones of the pegmatite (Table 1) and subsequently analyzed using an ARL–SEMQ electron microprobe at the National Museum of Natural History, Smithsonian Institution (accelerating voltage 15 kV, sample current 20 nA, and counting time 10 seconds). The following standards were used: hornblende (Si, Al, Na, K, Ca, Fe, Mg), gahnite (Zn), ilmenite (Ti) and apatite (P) and synthetic MnNb_2O_6 (Mn, Nb), $\text{CaTa}_4\text{O}_{11}$ (Ta), ScTiO_5 (Sc), and SnO_2 (Sn). Data reduction was done using the Bence & Albee (1968) procedure.

Unaltered and inclusion-free muscovite, lepidolite and feldspar were hand-separated from pegmatite samples for trace-element analysis. After separation, samples were pulverized and sieved to 200 mesh; 1.6 grams of sample was combined with 0.4 grams of cellulose, shaken for 5 minutes in a glass jar to homogenize the mixture, and pressed into a pellet for X-ray fluorescence analysis (on an automated Philips PW1480 X-ray spectrometer). Mica-group minerals were analyzed for As, Ba, Cs, Ga, Ni, Nb, Pb, Rb, Sn, Sr, Ta, Tl, Zn and Zr, whereas K-feldspar and albite were analyzed for Ba, Cs, Ga, Pb, Rb, Sn, Sr and Tl. Lithium analyses were performed by flame-photometry with a Perkin Elmer atomic absorption spectrophotometer (Model 3030). Sample digestion was performed by a standard hydrofluoric–perchloric acid method. Analytical standards were prepared from acid-soluble compounds.

MINERAL CHEMISTRY

Columbite-group minerals

Columbite at Black Mountain shows an increase in Ta and Mn, whereas Nb and Fe decrease, from the replacement unit through the various units of the third intermediate zone (Table 2). Sn and Ti values generally remain low (<0.5 wt.% of the oxide), but are slightly higher in the replacement unit. Ca amounts to less than 1.0 wt.% in columbite-group minerals in all zones.

TABLE 1. DISTRIBUTION OF TOURMALINE, COLUMBITE, MICROCLINE AND MICA-GROUP MINERALS IN THE BLACK MOUNTAIN PEGMATITE

Pegmatite zone/unit	Mineral	Description
Exocontact	dravite biotite-muscovite	<1.5-cm-long dark brown crystals fine- to medium-grained flakes
Aplite	schorl muscovite	1- to 1.5-cm-long black crystals fine-grained pale green flakes
Wall	schorl muscovite	25- to 30-cm-long black-blue crystals yellowish books up to 50 cm in length
First Intermediate	elbaite muscovite manganocolumbite	a) <5-cm-long flattened green crystals b) green overgrowths on dark green- blue crystals yellowish books up to 75 cm in length small blades (≤ 2 mm) enclosed in quartz
Second Intermediate	elbaite lepidolite manganocolumbite microcline	rare, small (<1 cm) pink crystals rare, fine- to medium-grained masses small (≤ 2 mm) blades enclosed in quartz white, coarse-grained crystals up to 1.5 m long
Third Intermediate	elbaite/rossmanite schorl lithian muscovite lepidolite manganocolumbite microcline	a) green crystals with pale green or pink rims occur in the QSK portion of the pegmatite; crystals are typically <1.5 cm in diameter b) < 7-cm-long pink or rare bicolored (green/pink, pink/colorless) crystals (TLA unit) c) 3- to 20-cm-long sprays of pink crystals (LEP pods) d) rare < 1.5-cm-long pale blue crystals (LEP pods) <1 mm blue-black crystals in saccharoidal albite fine-grained silver, green and purple flakes fine- to medium-grained silver to purple flakes brown-black crystals <3 cm long buff, white or gray crystals up to 8 cm long
Replacement	schorl mica manganocolumbite	black-green crystals up to 4 cm in diameter and 16 cm long, may have light green rim of elbaite medium- to coarse-grained books up to 12 cm long brownish black, long thin blades up to 6 cm long

The fractionation trend of columbite-group minerals from Black Mountain is illustrated in the columbite quadrilateral (Fig. 3). This trend agrees well with the general trend observed in F-enriched pegmatites, as presented by Černý *et al.* (1986). Whereas no columbite-group mineral from the first intermediate zone was available for analysis, that from the second intermediate zone show little variation in Mn/(Mn + Fe) and Ta/(Ta + Nb). The third intermediate zone contains manganocolumbite, with Mn/(Mn + Fe) ranging from

0.80 to 0.97 and Ta/(Ta + Nb) values from 0.10 to 0.48. Manganocolumbite from the LA unit appears to contain the most chemically evolved compositions. Columbite from the replacement unit varies little in its Ta/(Ta + Nb) value (0.10 to 0.12), whereas the Mn/(Mn + Fe) values vary over a broad range, from 0.38 to 0.92.

Micas

The composition of the mica-group minerals is relatively constant throughout the pegmatite. The proportion of K varies from 10.3 to 12.6 wt.% K₂O for all zones. The Mg and Fe contents are typically low (<0.5 wt.% MgO and <2.0 wt.% FeO) in muscovite and lepidolite. Average Li contents range from ~1.0 wt.% Li₂O in muscovite to ~3.5 wt.% Li₂O in lithian muscovite to ~4.2 wt.% Li₂O in lepidolite.

The trace-element concentrations of muscovite, lithian muscovite and lepidolite are, on the other hand, considerably more variable than the major elements (Table 3). The ratio K/Rb, which serves as an effective indicator of fractionation (*cf.* Černý *et al.* 1981, 1985, Shearer *et al.* 1985, Tomascak 1991), ranges from 58.3 to 47.4 for the wall zone, from 39.3 to 22.1 for the first intermediate zone, and from 35.2 to 24.4 for the replacement-unit muscovite. In lepidolite from the third intermediate zone, K/Rb varies from 7.9 to 7.5. Figure 4 shows the relationship of Cs, Sn and Zn enrichment in muscovite, lithian muscovite and lepidolite with respect to K/Rb fractionation. These elements generally increase in concentration with decreasing K/Rb. For example, Zn values for the wall-zone muscovite range from 120 to 168 ppm, whereas muscovite from the first intermediate zone and replacement unit range from 110 to 395 ppm and 210 to 411 ppm, respectively. Zn levels in lepidolite range from 236 to 298 ppm. Ga remain relatively constant (~120–145 ppm) in muscovite throughout the pegmatite.

Tourmaline-group minerals

Electron-microprobe analyses of tourmaline in the Black Mountain suite show the presence of dravite, schorl, elbaite and rossmanite. Rare rossmanite occurs as a rim on elbaite crystals in lepidolite pods, the layered tourmaline-lepidolite unit and the spodumene-bearing portion of the third intermediate zone.

Chemical heterogeneity is ubiquitous in all crystals analyzed. Much of the variation in tourmaline composition is due to changes in Mg, Fe, Al, and Mn; Si, Ca, and K vary little among all tourmaline species regardless of their position in the pegmatite, whereas variations in Na seem erratic. The general chemical trend is toward decreasing Mg and Fe with increasing Al, Mn and presumably Li from the country rock – pegmatite contact to the innermost zones. Mg contents are greatest in the exocontact dravite. The change in tourmaline composition from the exocontact to the inner zones of

TABLE 2. VARIATION IN COMPOSITION OF COLUMBITE-GROUP MINERALS, BLACK MOUNTAIN GRANITIC PEGMATITE, MAINE

	Second Intermediate		Third Intermediate			Replacement	
	QSK	SAC	TLA	LEP	LA		
FeO wt. %	1.21–2.58	1.57–4.09	1.97–3.87	1.06–3.80	0.34–1.11	0.00–3.34	0.84–12.57
MnO	16.39–16.76	15.42–16.68	15.48–17.59	15.23–18.17	18.03–18.44	15.27–18.78	7.63–18.76
Fe ₂ O ₃	0.76–2.08	0.00–2.19	0.00–0.68	0.00	0.00–0.84	0.00–2.07	0.00–2.09
Sc ₂ O ₃	0.46–0.47	0.00–0.43	0	0	0.14–0.16	0.00–0.39	0.00–0.42
TiO ₂	0.04–0.06	0.03–0.09	0.03–0.06	0.01–0.09	0.00–0.02	0.00–0.08	0.02–0.20
SnO ₂	0.11–0.14	0.04–0.44	0.09–0.16	0.10–0.17	0.08–0.12	0.01–0.14	0.02–0.19
Nb ₂ O ₅	61.98–63.69	61.92–66.58	54.09–65.73	61.83–65.93	58.61–59.12	32.24–56.10	64.58–72.66
Ta ₂ O ₅	14.64–16.06	13.69–17.27	12.56–26.65	13.95–18.77	21.68–23.08	25.61–50.05	6.48–14.05
Mn/(Mn + Fe)	0.84–0.85	0.79–0.84	0.8–0.9	0.80–0.94	0.94–0.96	0.83–0.99	0.38–0.92
Ta/(Ta + Nb)	0.13–0.14	0.11–0.14	0.10–0.23	0.11–0.15	0.18–0.19	0.22–0.48	0.05–0.12

QSK: quartz – spodumene – K-feldspar unit, SAC: saccharoidal albite unit, TLA: tourmaline – lepidolite – albite unit, LEP: lepidolite pod, LA: lepidolite–albite unit. Analysis by electron microprobe. The proportion of Fe₂O₃ is calculated.

TABLE 3. VARIATION IN CONCENTRATION OF SELECTED TRACE ELEMENTS IN MICA-GROUP MINERALS, BLACK MOUNTAIN GRANITIC PEGMATITE, MAINE

	Wall	First Intermediate	Third Intermediate	Replacement
	Ms	Ms	Li-Ms/Lpd	Ms
Li ppm	1080–2233	1040–1456	17703–24652	1074–1212
Cs	0–117	4–386	2339–7911	46–185
Ga	120–145	129–145	64–81	135–140
Nb	167–226	188–244	148–233	199–243
Zn	120–168	110–395	236–298	210–411
Rb	1665–2098	2534–4570	11207–12915	2883–4130
Sn	170–237	310–678	191–360	421–700
Tl	42–50	50–65	87–93	54–61
K/Rb	47.4–58.3	22.1–39.3	7.5–7.9	24.4–35.2
K/Cs	870.9–19880	261.7–25363.3	12.3–42.0	548.5–2203.6

Ms: muscovite, Li-Ms: lithian muscovite, Lpd: lepidolite. Trace-element analysis by X-ray-fluorescence spectroscopy; concentration of Li obtained by atomic absorption spectroscopy.

the pegmatite is roughly reflected by the color of the tourmaline.

Schorl from the wall and replacement unit has the highest concentration of Fe, up to 13 and 9 wt.% FeO, respectively. Green elbaite has lower values of Fe than schorl, but with higher values of Mn (0.2–1.5 wt.% MnO). Pink to colorless elbaite and rossmanite contain little to no Fe, Mg, Ca and K. Zn and Mn rarely attain concentrations greater than 1.0 wt.% of the oxide. Light blue–gray elbaite contained 0.8 to 1.3 wt.% Fe and higher Zn (1.0–1.9 wt.% ZnO) and Mn (0.5–2.5 wt.% MnO) compared to coeval pink elbaite.

Variations in tourmaline composition with color and zonal position in the pegmatite are summarized in Figure 5, which clearly shows a change in tourmaline color with decreasing ΣR^{2+}_Y ($\Sigma R^{2+}_Y = \text{Mg} + \text{Fe}^{2+} + \text{Mn} + \text{Zn}$

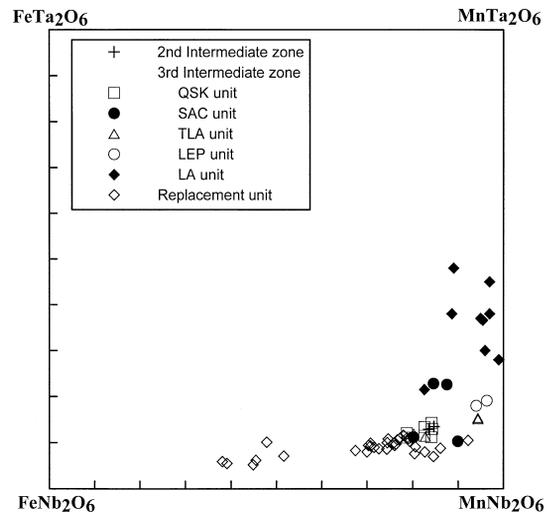


FIG. 3. Columbite quadrilateral showing the evolution of columbite compositions. Symbols: QSK: quartz – spodumene – microcline unit, SAC: saccharoidal albite, TLA: tourmaline – lepidolite – albite unit, LEP: lepidolite pods, LA: lepidolite – albite unit.

in the Y site). The color of tourmaline is strongly influenced by the presence of Fe or Mn, with black and green mainly attributed to the presence of Fe²⁺ or Fe³⁺, and pink coloration attributed to minor amounts of Mn with low concentrations or lack of Fe or other chromophores. Figure 5 also suggests that tourmaline compositions evolve from schorl – dravite to elbaite – rossmanite with progressive fractionation within the pegmatite.

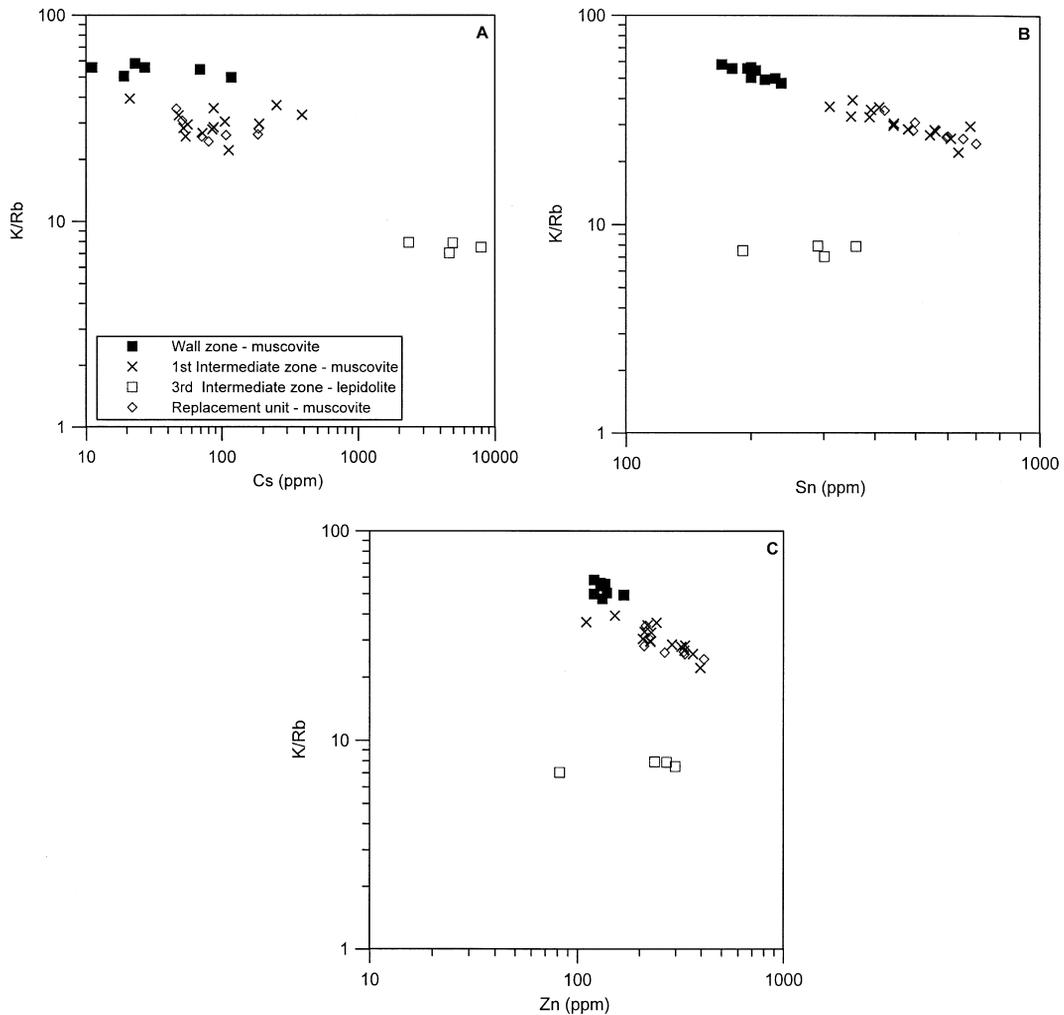


FIG. 4. Plots of K/Rb versus A) Cs, B) Sn and C) Zn for muscovite and lepidolite from the Black Mountain pegmatite.

Alkali feldspars

The dominant feldspar of the Black Mountain pegmatite is albite, which occurs in every zone of the pegmatite. Although several generations of albite exist within the pegmatite, the albite shows essentially no compositional variation.

K/Rb values of microcline range from 16.6 to 14.8 for the second intermediate zone and from 15.9 to 15.0 for the third intermediate zone. Rb, Cs and Tl contents are high in microcline from the second intermediate zone (Rb \leq 7423 ppm, Cs \leq 921 ppm, Tl \leq 68 ppm) and third intermediate zone (Rb \leq 7563 ppm, Cs \leq 1135 ppm, Tl \leq 70 ppm).

DISCUSSION

The composition of the microcline, mica-, tourmaline-, and columbite-group minerals, in conjunction with textural and paragenetic features, have been used to constrain the crystallization history of the Black Mountain pegmatite. Textural characteristics suggest that the bulk of the pegmatite formed as a result of primary crystallization from a melt in the order aplite \rightarrow wall zone \rightarrow first, second, third intermediate zones \rightarrow core. The various units of the third intermediate zone are transitional to one another, although textural relationships suggest that the order of crystallization proceeded in the sequence QSK \rightarrow TLA \rightarrow LA. Crystallization of the

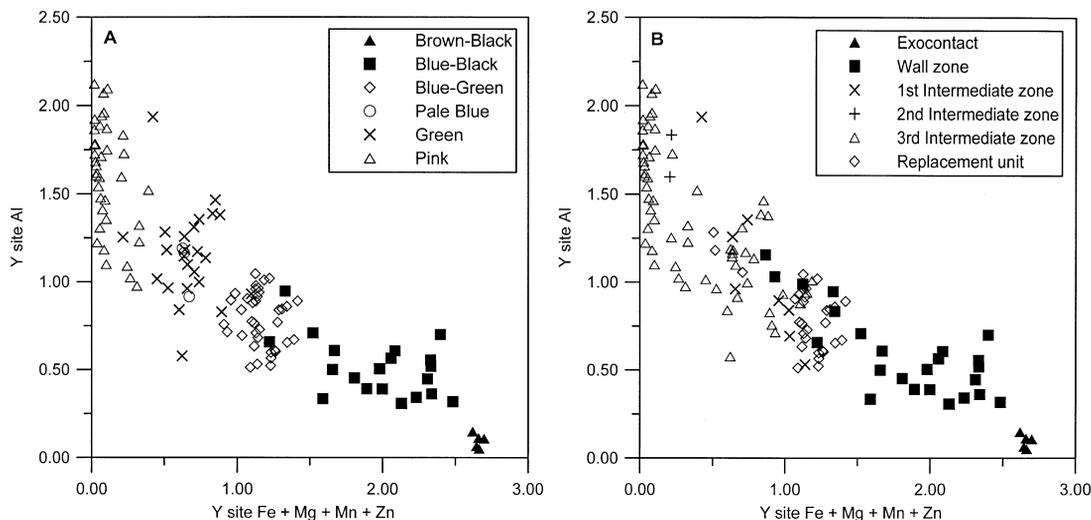


FIG. 5. Plots of Y-site Al versus the sum of Y-site R^{2+} from tourmaline-group minerals in the Black Mountain pegmatite. Note the relationship of tourmaline color (A) to position in the pegmatite (B). Each symbol represents an average result of five analyses per grain.

lepidolite pods and lenses may have been coeval with any one or all of the above units. Most of the chemical data obtained from the suite of minerals studied corroborate the zonal sequence determined by field examination. However, prominent inconsistencies in chemical trends were noted regarding the replacement unit and are discussed further in this paper.

Chemical evolution of primary zones

The melt that ultimately produced the Black Mountain granitic pegmatite was relatively Fe-rich in its initial stage, as indicated by the crystallization of copious amounts of schorl and almandine. With progressive crystallization, Fe-rich phases became increasingly less common. Both tourmaline and columbite, which crystallized after wall-zone consolidation, became increasingly Fe-poor, but Mn-enriched. Tourmaline compositions evolved from Fe- and Mg-dominant species to species enriched in Li and Al, with subordinate Mn. Similarly, columbite-group minerals evolved from ferroan manganocolumbite to near-end-member manganocolumbite.

Beryllium mineralization in the Black Mountain pegmatite is extremely limited and restricted to the early stages of pegmatite development. The Be content of the melt probably increased fairly rapidly during the crystallization of the wall and first intermediate zones until the melt achieved saturation with respect to beryl. The lack of additional Be minerals in subsequent zones suggests that the pegmatite-forming melt became severely depleted in Be following the crystallization of beryl.

The accumulation of the high-field-strength elements Zr, Sn, Nb, and Ta began relatively early in the development of the Black Mountain pegmatite. The first appearance of zircon and cassiterite, in the wall zone and first intermediate zone, respectively, suggests that the pegmatite-forming melt achieved saturation with respect to these minerals very early in its evolution. The overall abundance of Zr and Sn in the melt may not have been very high, as zircon and cassiterite occur only in minor quantities throughout much of the primary units of the pegmatite.

The Nb/Ta value of the melt was generally high throughout the evolution of the Black Mountain pegmatite. Although melt composition ultimately evolved toward Ta enrichment, Nb remained dominant over Ta. The melt achieved columbite saturation during consolidation of the first intermediate zone. Early-formed columbite show Ta/(Ta+Nb) values similar to those from the earliest units of the third intermediate zone, and only columbite formed during the late stages of the third intermediate zone shows any significant enrichment in Ta.

Finely, disseminated crystals of apatite in the aplite and wall zone, nodules of apatite and triphylite in the first intermediate zone, and subhedral crystals of ambygonite-montebasite in the second and third intermediate zone all indicate that the concentration of P in the melt was relatively high early in the pegmatite's development and remained high until the very late stages of pegmatite consolidation.

In the Black Mountain pegmatite, Rb enrichment is best documented by the composition of the mica. In the first intermediate zone, muscovite has a higher Rb content than wall-zone muscovite. Rb enrichment is highest in lepidolite of the third intermediate zone (Table 3). Cs concentration in the micas and microcline tends to increase with progressive crystallization; Cs is higher in microcline from the third intermediate zone than in that of the second intermediate zone. Early generations of muscovite have a lower Cs content than later generations. Extreme enrichment in Cs culminates in the crystallization of minor yet small masses of pollucite, which occur with spodumene, lepidolite, tourmaline, albite and quartz of the third intermediate zone (Foote 1896). The high Cs contents, coupled with extremely low K/Rb values found in lepidolite, suggest that the lepidolite-bearing units were some of the latest units to crystallize in the entire pegmatite. This assemblage is very typical of the third intermediate zone. We can therefore postulate that the melt became enriched in Cs only late in the evolution of the pegmatite.

Li behaved incompatibly throughout the early stages of pegmatite consolidation, becoming only mildly concentrated in micas and tourmaline. The appearance of amblygonite–montebrasite in the second intermediate zone marks the first indication of significant Li and F enrichment in the pegmatite-forming melt. Crystallization of the third intermediate zone reflects a continued increase in the activity of Li, K and F, such that spodumene + microcline assemblages were stable. In addition, F contents increased to levels that stabilized lepidolite, but not topaz or fluorite.

The pegmatite-forming melt that generated the Black Mountain pegmatite was initially saturated with respect to schorl. Restricted tourmalinization near the wallrock–pegmatite interface provides clear evidence of B loss to the surrounding host-rocks and may explain the general decrease in the amount of tourmaline in the first and second intermediate zones. However, abundant tourmaline continued to crystallize during the later stages of pegmatite consolidation. We conclude that only a small proportion of B managed to escape the confines of the solidifying body and that much of the boron in the original melt was conserved.

Development of replacement features

Volumetrically, the bulk of the late-stage replacement is represented by a unit containing “cleavelandite” + tourmaline. Textural evidence indicates that consolidation of this unit took place at some time following the solidification of the quartz core. “Cleavelandite” nucleated on the surface of the quartz core and proceeded to grow radially into the adjacent QSK portion of the third intermediate zone. “Cleavelandite” also grew along fractures that developed in the core, effectively creating pods and lenses of quartz rimmed by “cleavelandite”.

Extensive replacement of the third intermediate zone is not readily obvious in the current exposure, but evidence of incipient alteration can be found. Some crystals of spodumene that are completely surrounded by “cleavelandite” or in contact with the albite front show definite signs of alteration, whereas spodumene crystals not in direct contact with the “cleavelandite” unit remained unaffected. These textural features show that replacement was likely the last event and involved the final residual fluids.

One of the most intriguing aspects of the replacement unit is the chemical evolution it displays relative to the primary zones. The composition of the replacement unit cannot be considered to be highly fractionated, on the basis of the fairly primitive composition of the muscovite. K/Rb and Cs values generally overlap with those corresponding to the first intermediate zone. Examination of the replacement unit reveals that a significant portion of B, Nb, Zr and Sn mineralization occurred during its development. The composition of tourmaline is notably Fe-rich and similar to those found in the wall and first intermediate zones. With respect to their Mn/(Mn + Fe) values, columbite from the replacement unit is also Fe-rich and represent the least fractionated of all columbite-group minerals in the pegmatite. Relative to the primary zones, the accessory mineralogy of the replacement unit suggests that a late-stage reversal of the normal trend of fractionation of increasing Mn occurred with progressive evolution of the pegmatite. This apparent reversal in composition alludes to some process of Fe enrichment with progressive crystallization of the pegmatite. Late Fe enrichment in granitic pegmatites is not unexpected. So-called NYF-type granitic pegmatites typically contain Fe-rich phases (*e.g.*, hematite, siderite, magnetite, goethite) in late miarolitic cavities or internal zones. LCT-type granitic pegmatites containing miarolitic cavities may host green to pink elbaite capped by a late Fe-rich tourmaline.

Late enrichment in Fe in the replacement unit at Black Mountain may have been achieved through several processes. Assimilation of Fe-enriched country-rock by the pegmatite-forming melt during solidification of the replacement unit could potentially contaminate and enrich the melt with Fe. However, relict blocks of altered country-rock, or schlieren that might represent partially digested country-rock, were not observed.

A metasomatic influx of Fe-rich fluid might have introduced Fe into the pegmatite-forming melt, yet no evidence for this style of Fe enrichment is present at Black Mountain. London (1990) argued that during the early stages of crystallization, a pegmatite may be open to the infiltration of ferromagnesian components from the wall rock. He cited the deposition of tourmaline in the wallrock and the development of comb-structured tourmaline in the border zone of the pegmatite as evidence for the migration of fluids between the pegmatite and host rock. The tourmaline that crystallized as a re-

sult of fluid infiltration is typically dravite and schorl. In the case of the Black Mountain pegmatite, B-rich fluids clearly migrated into the wallrock, as indicated by the tourmaline-bearing exocontact. The absence of comb-structured tourmaline in the wall zone suggests that there was little, if any, influx of fluids into the pegmatite. At the time of crystallization of the final residual fluid, the pegmatite essentially was a closed system, and fluid migration into the pegmatite from an outside source, which could have led to late enrichment in Fe, probably did not occur.

Replacement of a poorly fractionated primary zone, such as the wall or first intermediate zone, might allow existing Fe-bearing phases to break down and release Fe that could recombine with the components of the melt to form new Fe-bearing phases. However, at Black Mountain, there is no evidence of extensive replacement of these zones. Furthermore, replacement of these primitive zones could not account for the enrichment in Nb, Zr and Sn observed in the replacement unit. These elements were thus not depleted from the melt during crystallization of the primary zones, but instead were concentrated into the final residual fluids. On the whole, the observed chemical features suggest that during the process of pegmatite evolution, the melt achieved a high degree of fractionation until the final stages of consolidation, at which stage the composition of the residual fluids shifted toward a more primitive, less fractionated medium.

The textural relationship between the replacement unit and adjacent primary zones clearly establishes its temporal position within the pegmatite. The explanation for the unusual chemical trends, however, warrants further investigation.

SUMMARY

1. Field evidence and chemical trends indicate that the Black Mountain granitic pegmatite crystallized from the margins inward in the following sequence: aplite → wall zone → first, second, third intermediate zones → core.

2. The pegmatite was generated from a B-enriched silicate melt that became increasingly enriched in rare alkalis (*e.g.*, Li, Rb, Cs), high-field-strength elements (*e.g.*, Nb, Ta, Zr, Sn) and volatile components (*e.g.*, F) during its crystallization. Enrichment in Be, P and Li promoted the formation of beryl, primary phosphates and lithium aluminosilicates, respectively.

3. Late-stage enrichment of Fe, as documented by the mineralogy of the late-developing replacement unit, is apparently contrary to the "normal" fractionation trends exhibited by most rare-element granitic pegmatites. The late enrichment in Fe is apparently not related to external factors affecting the crystallizing melt, and therefore must be directly related to the process of melt evolution.

ACKNOWLEDGEMENTS

The authors are indebted to Joe Martin for providing unlimited access to the Black Mountain property, to Woody Thompson of the Maine Geological Survey for logistical support, and to Tim Rose and Paul Tomascak for their assistance in the field. We thankfully acknowledge Carl Francis of the Harvard Mineralogical Museum for providing additional samples of the Black Mountain suite for study. Reviews by Lance Kearns and Bob Martin greatly improved the manuscript. This research was supported by the Sprague Funds of the Smithsonian Institution.

REFERENCES

- BAILEY, E.M. (1929): A catalog of minerals found at Rumford, Maine. *Rocks Minerals* **4**, 121.
- BASTIN, E.S. (1911): Geology of the pegmatites and associated rocks of Maine. *U.S. Geol. Surv., Bull.* **445**.
- BENCE, A.E. & ALBEE, A.L. (1968): Empirical correction factors for the electron microanalysis of silicates and oxides. *J. Geol.* **76**, 382-403.
- BJAREBY, G. (1965): Fifty years of mineral collecting. *Rocks Minerals* **40**, 645-647.
- CAMERON, E.N., LARABEE, D.M., MCNAIR, A.H., PAGE, J.J., STEWART, G.W. & SHAININ, V.E. (1954): Pegmatite investigations, 1942-1945, in New England. *U.S. Geol. Surv., Prof. Pap.* **255**.
- ČERNÝ, P., ERCIT, T.S. & WISE, M.A. (1986): Mineralogy, paragenesis and geochemical evolution of Nb- and Ta-oxide minerals in granitic pegmatites, *Int. Mineral. Assoc., 14th Gen. Meeting (Stanford), Abstr. Programs*, 71.
- _____, MEINTZER, R.E. & ANDERSON, A.J. (1985): Extreme fractionation in rare-element granitic pegmatites: selected examples of data and mechanisms. *Can. Mineral.* **23**, 381-421.
- _____, TRUEMAN, D.L., ZIEHLKE, D.V., GOAD, B.E. & PAUL, B.J. (1981): The Cat Lake - Winnipeg River and Wekusko Lake pegmatite fields, Manitoba. *Manitoba Mineral Res. Div., Econ. Geol. Rep.* **ER80-1**.
- FOOTE, H.W. (1896): On the occurrence of pollucite, manganocolumbite and microlite at Rumford, Maine. *Am. J. Sci.* **151**, 457-461.
- GREGORY, G.E. (1968): Black Mountain quarries of Rumford, Maine. *Gems Minerals* **366**, 32-34.
- HESS, F.L., WHITNEY, R., TREFETHEN, J. & SLAVIN, M. (1943): The rare alkalis of New England. *U.S. Bur. Mines, Inf. Circ.* **7232**.
- LONDON, D. (1990): Internal differentiation of rare-element pegmatites: a synthesis of recent research. *In Ore-Bearing*

- Granite Systems; Petrogenesis and Mineralization Processes (H.J. Stein & J.L. Hannah, eds.). *Geol. Soc. Am., Spec. Pap.* **246**, 35-50.
- MAILLOT, E.E., BOOS, M.F. & MOSIER, M. (1949): Investigation of Black Mountain beryl deposit, Oxford County, Maine. *U.S. Dep. Int., Bur. Mines RI 4412*, 1-10.
- MARBLE, C.F. (1927): The Black Mountain mica mine – Maine. *Rocks Minerals* **2**, 6.
- _____ & MORRILL, P. (1945): Report on two Maine mines. *Rocks Minerals* **20**, 58-59.
- MOENCH, R.H. & HILDRETH, C.T. (1976): Geologic map of the Rumford Quadrangle, Oxford and Franklin Counties, Maine. *U.S. Geol. Surv., Map GQ-1272*.
- SHEARER, C.K., PAPIKE, J.J. & LAUL, J.C. (1985): Chemistry of potassium feldspars from three zoned pegmatites, Black Hills, South Dakota: implications concerning pegmatite evolution. *Geochim. Cosmochim. Acta* **49**, 663-673.
- TOMASCAK, P.B. (1991): *Granites and Rare-Element Pegmatites of the Aylmer Lake Pegmatite Field, Slave Structural Province, N.W.T.: Petrochemistry, Mineralogy and Exploration Guidelines*. M.Sc. thesis, Univ. of Manitoba, Winnipeg, Manitoba.
- VERROW, H.J. (1941): The pegmatite minerals of Black Mountain, Rumford, Maine. *The Mineralogist* **9**, 119-120, 143.
- WISE, M.A. & FRANCIS, C.A. (1992): Distribution, classification and geologic setting of granitic pegmatites in Maine. *Northeastern Geol.* **14**, 82-93.

Received June 13, 2000, revised manuscript accepted December 28, 2000.

