

### Tourmaline from the rare-element Berry-Havey pegmatite (Oxford County, Maine, USA): paragenesis and petrography

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The Berry-Havey pegmatite (Poland, Androscoggin County, Maine, USA), belongs to the Oxford pegmatite field, which is proximal to the Sebago granitic batholith. The Berry-Havey is a highly evolved rare-element pegmatite, is enriched in Li, F, B, Be and P, and contains gem-bearing cavities. The pegmatite has a complex internal structure, with five different zones identified: border zone, two intermediate zones, core margin and core zone. Tourmaline is present in all of them, with textural and compositional differences among the different zones.

In the border zone tourmaline is scarce, occurring as very fine-grained prismatic black crystals, together with quartz, feldspars, muscovite, garnet and biotite. The mineralogy of the first intermediate zone is quite similar whereas the average grain size is larger. Graphic intergrowths of quartz and K-feldspar are very common in this zone. Locally, schorl may also be very abundant, appearing as black prismatic crystals up to 6 X 40 cm. In the second intermediate zone the quartz-K-feldspar graphic intergrowths are not so common. In this zone tourmaline is abundant and displays a characteristic texture. Many crystals exhibit a tapered black prism growing perpendicular to the pegmatite contacts, that increases in width in the direction of the core of the pegmatite. The thick end of the crystal is often "crowned" by black ± green tourmaline + quartz ± K-feldspar. The crystals, that show a sharp boundary between the black schorl and the greenish elbaite, are frequently broken, giving rise to a pull-apart structure. The next zone is the core margin, which is the most complex zone, not only because of its mineralogy, but also because of its textures. The main minerals of this zone are quartz, feldspars, micas from the muscovite-lepidolite series and tourmaline. The core margin also contains rounded Fe-Mn phosphate pods, montebasite, beryl and greenish, fine-to-medium-grained elbaite crystals associated with medium-to-coarse book muscovite crystals. Lepidolite occurs as fine-grained irregular to rounded purple masses, close to the muscovite + green elbaite-containing areas. Small pinkish elbaite crystals have been observed inside the lepidolite masses. Prismatic crystals of watermelon tourmaline have also been observed in this zone, but most are replaced by clay minerals. Moreover, radial prisms of multicoloured tourmaline (elbaite) embedded in feldspars and quartz are also abundant. Finally, the quartz-K-feldspar core represents the innermost zone of the pegmatite. Along the boundary between the core margin and core, pockets are common and may contain multicoloured gem-quality tourmaline, quartz, hydroxylherderite, cassiterite, cookeite, lepidolite, and beryl, among others.

The textural and chemical variations observed in tourmaline from the pegmatite, from the border zone to the core suggest an inward crystal fractionation model. The development of the quartz-K-feldspar and quartz-tourmaline graphic intergrowths could indicate undercooling of the pegmatite melt. The development of quartz-tourmaline crowns around elongated tourmaline crystals frequently showing pull-apart structures could be related to a sudden change in P conditions. Finally, the pockets located along the core margin and core zone of the pegmatite suggest elevated fluid activity during the last stages of crystallization of this pegmatite.

### Hydrothermal synthesis, crystal chemistry, and stability of fillowite-type phosphates in the Na – Mn – Fe<sup>2+</sup> – Mg – PO<sub>4</sub> system

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Fillowite [Na<sub>2</sub>CaMn<sub>7</sub>(PO<sub>4</sub>)<sub>6</sub>], johnsomervilleite [Na<sub>2</sub>Ca(Fe<sup>2+</sup>)<sub>7</sub>(PO<sub>4</sub>)<sub>6</sub>], and chladniite [Na<sub>2</sub>CaMg<sub>7</sub>(PO<sub>4</sub>)<sub>6</sub>] are Na-, Mn-, Fe- and Mg-bearing phosphate minerals which occur as primary phases in granitic pegmatites, in metamorphic rocks and in meteorites. Their crystal structure (space group R-3, a = 15.28 Å, c = 43.51 Å, Z = 18) is characterized by a hexagonal packing of three types of rods parallel to the c axis [1]. The rods are constituted by an alternation of five- to seven-coordinated sites containing Fe, Mn, and Mg, of six- to nine-coordinated sites containing Na and Ca, of PO<sub>4</sub> tetrahedra, and of cation vacancies.

In order to understand the stability of fillowite-type phosphates, the Na(Mn<sub>1-x</sub>Fe<sup>2+</sup>)<sub>x</sub>(PO<sub>4</sub>)<sub>3</sub> system (x = 0, 0.25, 0.5, 0.75, 1.0) [2], as well as compounds with the Na(Mn<sub>0.33</sub>Fe<sup>2+</sup><sub>0.33</sub>Mg<sub>0.33</sub>)<sub>4</sub>(PO<sub>4</sub>)<sub>3</sub>, Na(Mn<sub>0.6</sub>Fe<sup>2+</sup><sub>0.2</sub>Mg<sub>0.2</sub>)<sub>4</sub>(PO<sub>4</sub>)<sub>3</sub>, Na(Mn<sub>0.2</sub>Fe<sup>2+</sup><sub>0.6</sub>Mg<sub>0.2</sub>)<sub>4</sub>(PO<sub>4</sub>)<sub>3</sub>, Na(Mn<sub>0.2</sub>Fe<sup>2+</sup><sub>0.2</sub>Mg<sub>0.6</sub>)<sub>4</sub>(PO<sub>4</sub>)<sub>3</sub> starting compositions, were synthesized under hydrothermal conditions, between 400 and 700°C, at 1 kbar. Single-phase fillowite is obtained only for the NaMn<sub>4</sub>(PO<sub>4</sub>)<sub>3</sub> starting composition, between 400 and 700°C. The incorporation of iron and magnesium immediately provokes the crystallization of alluaudite [Na<sub>2</sub>(Mn,Fe<sup>2+</sup>,Mg)<sub>2</sub>Fe<sup>3+</sup>(PO<sub>4</sub>)<sub>3</sub>] and of sarcopside [(Fe<sup>2+</sup>,Mn,Mg)<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>], in association with fillowite in the Mn-rich part of the Mn-Mg-Fe diagram. In the Mg-rich part of the system, an assemblage alluaudite + hydrated alluaudite [□Na(Fe,Mn)Mg<sub>2</sub>(PO<sub>4</sub>)(HPO<sub>4</sub>)<sub>2</sub>] + (Mg,Mn,Fe)<sub>2</sub>P<sub>2</sub>O<sub>7</sub> occurs at 400 and 500°C, and an assemblage alluaudite + (Mg,Mn,Fe)<sub>2</sub>P<sub>2</sub>O<sub>7</sub> occurs at 600 and 700°C. Finally, in the Fe-rich and central parts of this diagram occurs the alluaudite stability field.

The electron-microprobe analyses of fillowite-type phosphates show an decrease of the Mg/(Fe<sub>tot</sub>+Mn+Mg) ratio, from 0.25 at 400°C, to 0.20 at 600°C, followed by an increase to 0.28 at 700°C. At 800°C, the Mg-richest fillowite-type phosphate has been observed, with a composition Na<sub>2.14</sub>(Mn<sub>4.90</sub>Mg<sub>2.15</sub>Fe<sup>2+</sup><sub>0.33</sub>Fe<sup>3+</sup><sub>0.37</sub>)(PO<sub>4</sub>)<sub>6</sub>.

The Mg/(Fe<sub>tot</sub>+Mn+Mg) ratio of sarcopside increases from 0.25 at 400°C, to 0.39 at 500°C, and then decreases to 0.14 at 700°C. The composition of the Mg-richest sarcopside is (Mg<sub>1.17</sub>Fe<sup>2+</sup><sub>1.62</sub>Mn<sub>0.21</sub>)(PO<sub>4</sub>)<sub>2</sub>.

At high temperatures, the proportions of alluaudite-type phosphates significantly increase compared to experiments at lower temperatures. This feature probably results in an oxidation of iron, favoured by the increase of hydrogen diffusion through the gold capsule at high temperatures.

[1] Araki, T. & Moore, P.B. (1981) *Am. Mineral.*, **66**, 827-842.

[2] Rondeux, M. & Hatert, F. (2006) *Ber. Dtsch. Min. Ges., Beih. z. Eur. J. Mineral.*, **18**(1), 111.