1 **Revision 2**

2 **Apatite as an archive of pegmatite-forming processes: an example**

3 **from the Berry-Havey pegmatite (Maine, USA)**

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14 **ABSTRACT**

15 Apatite is an accessory phase in all the units of the internally zoned Berry-Havey

16 complex pegmatite. This body presents a highly fractionated core zone, enriched in Li, F, B,

17 Be and P, which hosts three different types of pockets, some of them often containing tens to

18 hundreds of gemmy euhedral Li-rich tourmaline crystals, together with other mineral phases

19 such as lepidolite. Processes involved in the complex internal evolution of pegmatitic melts

20 that give rise to zoned bodies containing pockets are not completely understood. In order to

21 shed light on these processes, apatite from all the different units of the Berry-Havey

22 pegmatite (wall zone, intermediate zone, core margin and core zone pods) and from the

23 three pocket types (Li-poor, Li-rich and apatite seams) has been characterized

24 petrographically and later analyzed for major (electronic microprobe) and trace elements (LA-

25 ICP-MS). Results indicate that apatite chemistry changed significantly during the

26 crystallization of the Berry-Havey pegmatite, reflecting the conditions at each stage and

27 mainly depending on the fractionation degree, fO_2, and paragenetic association.

28   Fluorapatite is found in all the units except the core margin, the Li-poor pockets and the

29   seams, where Mn-bearing fluorapatite is present. A gradual increase of the Mn content in

30   apatite from the pegmatite border (wall zone) inwards, up to the formation of subrounded

31   masses of Mn-Fe phosphate in the core zone pods, parallels the increasing fractionation of

32   the melt. Phosphate crystallization would deplete the residual melt in Mn, probably causing

33   the significant Mn-decrease observed in apatite from the core zone pods and Li-rich pockets.

34   The late depletion of Mn could also be related to an increase of fO_2 in the melt during the

35   later stages of its evolution.

36   Main trace element variations in apatite at both, pegmatite and crystal scales,

37   correspond to REE, Y and Sr. Yttrium and REE behave in a very similar way, decreasing

38   inwards, i.e. with fractionation of the pegmatitic melt (ΣREE from 1796 ppm in the apatite

39   from the wall zone to 0 ppm in the core zone; and Y from 1503 ppm in the apatite from the

40   wall zone to 0 ppm in the core zone); which could be due to early crystallization of REE-

41   bearing phosphates such as monazite and xenotime. Strontium shows a more complex

42   trend, with an initial depletion in apatite from the wall zone (52 ppm) to the intermediate zone

43   (3 ppm) and a pronounced increase from the core margin (23-87 ppm) up to the core zone

44   and pockets (up to 2.87 wt%). This increase of Sr at the latest fractionation stages of the

45   pegmatite is interpreted to be associated with a late incompatible character of this element in

46   highly fractionated melts, related to the composition of feldspars from the core margin

47   (mainly pure albite). The lack of Ca in feldspars would decrease affinity for Sr incorporation

48   into their structure and, consequently, Sr would go preferentially to apatite in the core zone

49   pods and, more markedly, in the pockets.

50   Apatite also records changes in the redox conditions during crystallization, with the

51   highest fO_2 at the end of the crystallization, mainly reflected in the Eu and Ce anomalies. The

52   chemistry of apatite also reflects the evolution of the pegmatitic melt during crystallization

53   regarding the fluids saturation and pockets generation. Accordingly, at least two exsolution

54   events took place during the Berry-Havey crystallization history: (i) at the beginning of the

55   core zone crystallization, giving rise to the Li-poor pockets; and (ii) after the crystallization of

56   the Li-rich pods of the core zone, resulting in the Li-rich pockets. The apatite-rich seams may

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57   have crystallized between these two exsolution events, or later, at a subsolidus stage, after a

58   Na-autometasomatism episode.

59   This study shows how a detailed petrographic and chemical characterization of apatite

60   associated with different units of a highly fractionated, internally zoned pegmatite may help

61   understand the crystallization history of pegmatitic melts. It is also evidenced that during the

62   internal evolution of pegmatites, apatite chemistry records variations in the fO_2, elemental

63   fractionation, interaction with competing mineral phases, fluids activity and exsolution events.

64   In addition, it is shown how apatite chemistry may be useful as an exploration tool for

65   pegmatites.

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67   *Keywords: Berry-Havey pegmatite, Maine, apatite, internal fractionation, pockets*

68   *formation*

69

70   **INTRODUCTION**

71   The Oxford pegmatite field is characterized by the occurrence of some complexly zoned,

72   highly evolved granitic pegmatites. The Berry-Havey is one of the best examples of these

73   bodies that are enriched in incompatible elements (namely Li, F, B, Be and P) in their most

74   fractionated units. This pegmatite presents a well-developed internal zoning, with textural

75   and chemical changes from the contact with the country rocks inward. In addition, numerous

76   miarolitic cavities (pockets) that contain abundant euhedral crystals of gemmy tourmaline,

77   among other phases, are common in the inner zones of this pegmatite. The crystallization

78   history of the Berry-Havey is, therefore, presumed to be complex. Actually, the internal

79   evolution of pegmatitic melts mainly in heterogeneous zoned bodies is not completely

80   understood. Numerous studies on pegmatite fields have been carried out worldwide, as well

81   as experimental work (e.g. Jahns and Burnham, 1969; Veksler et al. 2002; London, 2005), to

82   understand and decipher the characteristics and formation mechanisms of complex

83   pegmatites. In this regard, micas, feldspars, tourmaline and, to a lesser extent, garnet, Fe-

84   Mn-phosphates, Nb-Ta oxides and quartz have been used as petrogenetic indicators to

85   unravel the internal evolution of pegmatitic bodies (e.g., Černý, 1982; London, 2008; Roda-

86   Robles et al. 2012, 2015a, b; Hernández-Filiberto et al. 2021; Müller et al. 2021). In the

87   Berry-Havey pegmatite, textural and chemical variations observed in tourmaline and mica

88   occurring in all the zones of the body are indicative of the crystallization of a strongly

89   fractionated melt from the borders inward (Roda-Robles et al. 2015a, b). Chemical data from

90   apatite presented in this study are consistent with this model. However, chemical variations

91   in this phosphate are more complex and difficult to interpret in comparison to those of

92   tourmaline and mica, probably due to the stronger influence of parameters such as fO_2 or

93   melt polymerization on apatite composition. Thus, a detailed study of the apatite chemistry

94   may be key to understanding the complex crystallization history of this pegmatite. In this

95   paper we present the results of the petrographic and chemical study of apatite associated

96   with the different zones of the Berry-Havey pegmatite and discuss their implications for its

97   petrogenesis. Likewise, the usefulness of apatite as an archive of pegmatite-forming

98   processes and as an exploration tool for fractionated pegmatites is highlighted.

99

GEOLOGICAL SETTING

101   The Berry-Havey pegmatite belongs to the Oxford pegmatite field, which is located SW

102   of the state of Maine (USA). This field outcrops in the middle-south area of the "Central

103   Maine Belt" (CMB), a large NE–SW trending synclinorium (Reusch and Staal, 2011) that

104   constitutes the main tectonostratigraphic unit of the northern Appalachians. The CMB is

105   bordered by the Norumbega Shear Zone System to the SE, which puts the CMB in contact

106   with the Neoproterozoic to Silurian rocks of the Avalon Composite Terrain; whereas to the

107   NW this belt is bordered by the Ordovician rocks of the Bronson Hill Belt (Guidotti, 1989;

108   Solar and Tomascak, 2001) (Fig. 1). The CMB is composed of a Lower Paleozoic

109   sedimentary succession together with igneous rocks (e. g. Osberg 1978; Solar and Brown

110   2001). During the Acadian orogeny the CMB was deformed and metamorphosed

111   (greenschist in the NE to upper amphibolite facies and migmatite to the SW, Guidotti, 1989;

112   1993) (Fig. 1), and also intruded by Devonian to Permian plutons (e.g. Moench et al. 1995;

113   Bradley et al. 1998; Solar and Brown, 2001, Solar and Tomascak, 2016).

114   Most pegmatites from the Oxford field concentrate on the northern and eastern margins

115   of the so-called "Migmatite-Granite complex" (MGC) (previously known as Sebago Migmatite

116   Domain (Solar and Tomascak, 2016)). The MGC, located in the core of the CMB, presents a

117   high metamorphic set of facies (amphibolite and migmatite). The main rocks of the MGC are

118   metapelitic or metapsammitic migmatite and diatexite with subordinate centimeter- to meter-

119   scale bodies of granite (medium-grained two-mica granite to pegmatite) (Tomascak et al.

120   1996a, b). Migmatitic rocks commonly present well-distributed penetrative fabrics, interpreted

121   as deformation, which continued after migmatite formation and at subsolidus conditions

122   (Solar and Tomascak, 2016). An age of 376 ± 14 Ma has been obtained for migmatites from

123   the MGC (Solar and Tomascak, 2016). The MGC acts as country rock to the Sebago pluton,

124   located to the south. This pluton is quite homogeneous texturally and chemically, mainly

125   corresponding to a fine- to medium-grained, two-mica granite (Solar and Tomascak, 2016). It

126   is younger than the surrounding migmatites, with ages of 293 Ma (U-Pb TIMS in monazite,

127   Tomascak et al. 1996b). This important age difference between migmatites from the MGD

128   and the Sebago pluton rules out a direct relationship between both lithologies. According to

129   Solar and Tomascak (2009, 2016) migmatite-forming processes in this area had finished

130   before the formation of the Sebago pluton.

131   Pegmatites from the Oxford field present different degrees of evolution, from barren

132   bodies to highly evolved ones that show well-developed internal zoning (Roda-Robles et al.

133   2015b). They may intrude in migmatites, amphibolites and mica schists, either concordant to

134   the host rock foliation or as irregular and discordant bodies (Simmons et al. 2017). There is

135   not a general consensus on the origin of these pegmatites. According to Wise and Brown

136   (2010), they are related to the Sebago pluton by fractional crystallization processes.

137   However, pegmatites from this field have been dated in a range of ≈ 250–270 Ma (Bradley et

138   al. 2016), which means that they are >20 Ma younger than the Sebago pluton. This important

139   difference in the age of the two lithologies makes it difficult to link the origin of the pegmatites

140   to the Sebago pluton. More recent studies (Simmons et al. 2016; Webber et al. 2019)

141   propose a direct anatectic origin for these pegmatites, with the hosting migmatites and

142   metasedimentary rocks as the source for the pegmatitic melts.

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GENERAL GEOLOGY OF THE BERRY-HAVEY PEGMATITE

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145   The Berry-Havey pegmatite is a tabular to irregular body, hosted by a hornblende-rich

146   amphibolite, with minor diopside or biotite in places (Roda-Robles et al. 2015b). The lower

147   contact with the country rock is not observed, whereas the upper contact only outcrops to the

148   SW of the quarry. There, the pegmatite is mainly conformable to the country rock, dipping ~

149   40º SSE, whereas it gets more horizontal to the north (Fig. 2). Locally, the pegmatite

150   presents some thin (<50 cm thick) dike-like branches that intrude discordantly into the

151   country rock. No metasomatism effects have been observed in the hosting amphibolite close

152   to the contact with the pegmatite. According to the present level of exposure, the thickness of

153   the Berry-Havey pegmatitic body is estimated to be over 30 m. Nevertheless, the lack of

154   exposure of the footwall makes this estimation difficult. The pegmatite is cross-cut by three

155   sub-vertical mafic dykes with a NNE-SSW strike (Fig. 2). The Berry-Havey presents a well-

156   developed complex internal structure, where four different zones may be distinguished

157   according to their spatial distribution, textural features, paragenesis, and chemical

158   composition (Roda-Robles et al. 2015b) (Fig. 2; Table 1). These include a wall zone,

159   intermediate zone, core margin, and core zone. These zones are subparallel and may show

160   irregular limits among them, especially in the transition from the core margin to the core

161   zone. From the contact inward, the following units may be distinguished (Roda-Robles et al.

162   2015b) (Fig. 2; Table 1):

163   (1) The Wall Zone only outcrops at the SW area of the quarry, in direct contact with the

164   hosting amphibolite in the hanging wall. The wall zone most commonly presents a fine to

165   medium-sized pegmatitic texture. A gneissic texture may be locally observed. Main minerals

166   include quartz, plagioclase, K-feldspar, biotite, and muscovite, with tourmaline and garnet as

167   common minor phases, and accessory apatite.

168   (2) The Intermediate Zone volumetrically is the most important zone, as it corresponds to

169   ≈ 58 % of the outcrop. Its minerals include quartz and feldspars (K-feldspar and plagioclase)

170   which appear graphically intergrown in > 85 % of this zone. Other common phases are

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171 biotite, muscovite, garnet, and black tourmaline, which occur scattered as fine- to medium-

172 sized crystals. Apatite occurs as an accessory phase. The intermediate zone appears over

173 and under the core zone/core margin. The upper intermediate zone is tabular, with a

174 thickness of ~5 m; whereas the lower intermediate zone, more important volumetrically, is

175 more irregular in shape and thickness (Fig. 2). To the N of the quarry the mineralogy and

176 texture of the lower intermediate zone change locally (Fig. 2). There mineralogy is simpler,

177 with quartz, K-feldspar and black tourmaline; whereas texturally the graphic intergrowth is not

178 observed and the grain size is coarser, locally with blocky K-feldspar and quartz, and with

179 black tourmaline prismatic crystals up to 40 cm in length.

180 (3) The Core Margin corresponds to ≈ 32% of the pegmatite outcrop, occurring between

181 the upper and lower intermediate zone units, closer to the hanging wall than to the footwall

182 (Fig. 2). It is mainly composed of "vuggy" albite, quartz, muscovite, schorl, and accessory

183 green tourmaline and apatite. The lower limit between the core margin and the intermediate

184 zone is commonly marked by a discontinuous garnet-rich layer. This layer is used as a guide

185 for the miners in this region, as pockets are not usually found below this level. Over the

186 garnet layer, a quite continuous layer constituted by tourmaline prisms occurs in the core

187 margin (Fig. 2). The core margin zone envelops the different pods that constitute the core

188 zone of the pegmatite.

189 (4) The Core Zone is the innermost and most complex zone of the pegmatite. It is not a

190 continuous unit, but it is formed by a number of more or less irregular pods, sometimes

191 interconnected, of different sizes (from ≈ 2 m up to 10 m across) that are enclosed in the

192 core margin (Fig. 2). Main mineral phases of the core zone pods include blocky feldspars

193 and quartz, that host irregular masses of fine-grained, usually purple, lepidolite, book

194 muscovite ± light pinkish lepidolite, albite, greenish and pinkish tourmaline, and sub-rounded

195 nodules of phosphates (with primary lithiophilite, triplite, dickinsonite, and/or montebrasite);

196 with minor Cs-rich beryl and accessory cassiterite, Nb-Ta oxides and apatite.

197 (5) Pockets: the pods of the core zone may contain isolated or interconnected miarolitic

198 cavities (pockets), with very variable sizes, from a few cm^3 to ≈ 0.7 m^3. Two different mineral

199 associations may be observed inside these pockets. Some of them are Li-poor and may

200 include euhedral quartz crystals (often smoky), dark "etched" tourmaline, and zoned apatite

201 lenses. Most commonly, pockets are Li-rich, main minerals including quartz, blocky K-

202 feldspar, bladed albite, lepidolite, cassiterite, Cs-rich beryl, green and "watermelon" gemmy

203 tourmaline, and purple or bluish apatite. In addition, in both the core margin and the core

204 zone, locally some elongated "seams" of ≤ 50 cm long and ≤ 20cm wide, which may be filled

205 with apatite, can be observed. These seams are hosted by blocky feldspar in the core zone

206 and/or by platy albite in the core margin.

Textural features of the apatite

208 Apatite is ubiquitous in all the units distinguished in the Berry-Havey pegmatite although

209 in different proportions. In the wall zone and intermediate zone, apatite is an accessory

210 phase, appearing as grayish, fine-grained anhedral to euhedral crystals (< 5 mm) (Table 1,

211 Fig. 3a). It is more abundant in the core margin, occurring as fine to medium-sized (< 3 cm),

212 bluish prismatic to anhedral crystals. In addition, some anhedral crystals occur intergrown

213 with garnet in the garnet layer, between the intermediate zone and the core margin (Fig. 3b).

214 In the core zone apatite is very scarce in the pods, where it occurs as subhedral to euhedral

215 fine-grained prisms (< 3 mm) showing concentric zoning under the petrographic microscope

216 (Fig. 3c). In contrast, apatite is abundant in many of the pockets, where it can even be the

217 main phase in the seams, which can be filled up with strongly colored apatite (Fig. 3d). In

218 general, apatite from the pockets is subhedral to euhedral, often occurring as short

219 hexagonal prisms or lenses (< 1 cm Ø) with different colors in hand sample. In the Li-poor

220 pockets euhedral apatite crystals coat or grow over euhedral quartz crystals (Fig. 3e), also

221 coexisting with etched dark tourmaline. These euhedral crystals correspond to hexagonal

222 lenses that show concentric sharp zoning, with a grayish to clear blue core and a narrow

223 pure whitish rim (Fig. 3e). In Li-rich pockets, apatite short prisms show a deep purple or

224 bluish color without concentric chromatic zoning (Fig. 3f). The anhedral to prismatic apatite

225 that fills the elongated "seams" also presents a common deep purple color (Fig. 3d).

226 Occasionally, it is also possible to observe in those cavities some euhedral short hexagonal

227 apatite prisms that present a purple core and a deep green rim.

228

SAMPLING AND ANALYTICAL METHODS

230   Apatite samples were taken from all the zones of the pegmatite. Those from the core

231   zone, where apatite is especially abundant in some of the pockets and in some elongated

232   "seams", are the best-represented samples. Characterization of apatite was carried out via

233   the study of its texture in hand sample and thin section, and of its chemical composition by

234   electron-microprobe and LA-ICP-MS techniques.

235   Electron-microprobe quantitative analyses were performed at the University of Paul

236   Sabatier (Toulouse, France) using a Cameca SXFive (Raimond Castaing Center). The

237   operating conditions included an accelerating voltage of 15 kV; a beam current of 10 nA, and

238   a beam diameter of 2 μm. A low current was used due to the sensitivity of apatite under the

239   beam. Analyzed surfaces were chosen in the same way to reduce damage. The time of

240   analyses was 10s for each element and 5s on the background; except for F, with 20s and

241   10s on the background. The following standards were used: albite (Na), periclase (Mg),

242   corundum (Al), wollastonite (Ca, Si), hematite (Fe), $MnTiO_3$ (Mn), tugtupite (Cl), Durango

243   apatite (F, P), $LaPO_4$ (La), $CePO_4$ (Ce), and celestine (Sr). Data were reduced using the

244   procedure of Pouchou and Pichoir (1985). Analytical errors are estimated to be on the order

245   of ±1–2%. Measuring of F and Cl by EMPA may involve difficulties that may be solved by

246   doing the analyses having crystals oriented with their *c*-axes perpendicular to the incident

247   electron beam (Goldoff et al. 2012). In the present study only analyses made on the apatite

248   crystals from the Li-poor, Li-rich pockets and seams could be oriented this way.

249   Trace element analyses in apatite were performed by laser-ablation inductively-coupled

250   plasma mass-spectrometry (LA-ICP-MS) at the Geochronology and Isotope Geochemistry-

251   SGIker facility of the University of the Basque Country (Spain). More than 100 analyses were

252   performed by the ablation of apatite crystals in ca. 100 μm thick petrographic sections or on

253   crystals mounted in epoxy using a UP213 Nd:YAG laser ablation system (New Wave)

254   coupled to a Thermo Fisher Scientific X Series 2 quadrupole ICP-MS instrument with

255   sensitivity enhanced through a dual pumping system. Spot diameters of ca. 100 μm were

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256   used for analyses, associated with repetition rates of 10 Hz and laser fluence at the target of

257   ca. 5.5 J/cm^2. The ablated material was carried in He and then mixed with Ar. The NIST SRM

258   612 reference glass was used to perform tuning and mass calibration, by inspecting the

259   signal of 238 U to obtain ca. 14,000,000 cps/ppm, and by minimizing the ThO$^+$/Th$^+$ ratio to

260   ca. 1%. Raw data were processed using Iolite 2.3 (Paton et al. 2011; Paul et al. 2012) and

261   Ca as an internal standard (data obtained by electron microprobe). Apatite from Cerro

262   Mercado (Durango, Mexico) was used to optimize the procedure and as a secondary

263   standard to control the quality of the results in each analytical session.

264

265   APATITE MINERAL CHEMISTRY

266   **Major elements**

267   Apatite from the Berry-Havey pegmatite presents chemical differences for some major

268   elements depending on the unit where it occurs (Table 2, Fig. 4). In addition, some crystals

269   show marked concentric zoning for certain elements. All the analyzed apatite crystals are

270   fluorapatite. The highest F contents correspond to apatite from the pockets, with values

271   ranging from 5 to 6.14 wt% for the purple prisms, 4.99 to 5.91 wt% for the zoned gray-white

272   lenses, and 2.96 to 3.81 wt% for the zoned purple-green crystals of the seams (Table 2, Fig.

273   4a). The F content of apatite from the intermediate zone is as well > 4 wt%. Wall zone apatite

274   has 2.84 to 3.10 wt% F. The lowest F values correspond to apatite from the core margin, in

275   the range 1.28 – 2.98 wt% F (Table 2, Fig. 4a). The OH content in the halogen site is low in

276   general, ranging from 0 apfu in the apatite from the intermediate zone, Li-poor and Li-rich

277   pockets; up to > 0.5 apfu in the apatite from the core margin; with intermediate values in

278   apatite from the wall zone, seams, and core zone pods (Table 2). All the analyzed apatite

279   crystals have extremely low Cl contents, the highest value (0.13 wt%) corresponding to a

280   zoned gray-white crystal from a pocket (Table 2).

281   The most significant differences for the major elements on the pegmatite and single

282   crystal scales correspond to Mn (Table 2, Figs. 4b, 5a,b, 6a). The lowest Mn values are

283   found in apatite from the wall zone (0.07-0.20 wt% MnO), from the pods of the core zone

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284   (0.06-0.55 wt% MnO), and in the purple crystals from the Li-rich pockets (0.08-0.48 wt%

285   MnO). In contrast, apatite crystals from the Intermediate zone and core margin show higher

286   Mn values in the range of 3.10-9.97 wt% MnO. Zoned gray-white apatite crystals from the Li-

287   poor pockets present marked Mn variations across the crystals, from 4.65 wt% MnO in the

288   grayish cores to 0.21 wt% MnO in the white rims (Table 2, Fig. 6a). In the case of the purple-

289   green crystals from the seams, the variation trend is not so clear but, overall, Mn values

290   increase from the purple core (0.93-3.26 wt% MnO) to the greenish rim (3.67-4.23 wt% MnO)

291   (Fig. 6a).

292   Iron contents are low in all the analyzed apatite, i.e. always ≤ 0.32 wt% FeO (Table 2,

293   Figs. 5b, c). A clear relation between the pegmatite unit and the Fe content has not been

294   determined. However, a certain positive correlation between the Mn and Fe contents is

295   observed (Fig. 5b). In the case of the zoned crystals, in the gray-white lenses there is a slight

296   Fe decrease from core (0.31 wt% FeO) to rim (0.00 wt% FeO) (Fig. 6b). In contrast, like Mn,

297   in the purple-green zoned crystals the Fe tends to increase from the purple core (0.07-0.15

298   wt% FeO) to the greenish rim (0.17-0.18 wt% FeO) (Fig. 6b).

299   Calcium values also show some differences. In general, as expected, there is a good

300   negative correlation between Ca and Mn (Figs. 4c, 5a, 6a, c), with the lowest contents

301   (41.72-43.39 wt% CaO) in the Mn-richest crystals from the core margin, and the highest Ca

302   contents (55.00 to 55.99 wt% CaO) in the Mn-poorest apatite from the wall zone and the core

303   zone. Although it may display a clear chromatic concentric zoning under the microscope (Fig.

304   3c), no chemical zoning of major elements has been observed in apatite crystals from the

305   core zone pods.

306

307   **Trace elements**

308   Like major elements, some trace elements in apatite show differences depending on the

309   pegmatitic unit and across zoned crystals. Main variations correspond to REE, Y, and Sr

310   (Fig. 7a-e). Yttrium and REE show similar trends, with a continuous decrease from the wall

311   zone to the core zone (Table 3, Figs. 7a-d, 8a). Values for Y and ΣREE for apatite from the

312   wall zone are in the ranges 1155 to 1503 ppm and 1796 to 2178 ppm, respectively. In the

313   intermediate zone, these values decrease to 225 to 261 ppm Y and 1219 to 1280 ppm

314   ΣREE. Apatite from the core margin is even poorer, with values of 22.7 to 26.3 ppm Y and

315   309 to 355 ppm ΣREE. In the core zone, there are significant differences in the Y and ΣREE

316   contents when comparing apatite from the pods and that of the different types of pockets.

317   Apatite from the pods is the poorest in these elements, with Y contents below the detection

318   limit and ΣREE values < 0.14 ppm (Table 3, Figs. 7, 8a). Purple apatite of the Li-rich pockets

319   also shows extremely low contents of Y (0.14-0.54 ppm) and ΣREE (0.11-0.37 ppm). Zoned

320   gray-white apatite crystals from the Li-poor pockets are richer in these elements, with Y and

321   ΣREE values varying respectively in the range of 979 to 31.7 ppm, and 921 to 41.3 ppm from

322   grayish core to white rim (Table 3, Figs. 7b, c, d, 8a, b, 9a-d). The zoned purple-green

323   apatite from the seams has intermediate Y and ΣREE values, with Y and ΣREE contents that

324   vary respectively in the ranges of 2 to 23 ppm and 0 to 5 ppm, and 53.2 to 210 ppm and 0.0

325   to 42.2, for the purple cores and greenish rims, respectively (Table 3, Figs. 7a-d, 8a, b, c, 9a-

326   d).

327   Overall, Sr presents a more complex trend, with a rough negative correlation with ΣREE

328   and Y (Figs. 7a, b, e, 8b, c; 9e). On the pegmatite scale, there is a Sr decrease in apatite

329   from the wall zone to the intermediate zone, and a final marked increase in the pods of the

330   core zone and pockets (wall zone: 50-52.5 ppm; intermediate zone: 2.9-16.1; core margin:

331   2.6-23.4 ppm; core zone pods: 18-1614: Li-poor pockets: 507-28710 ppm; Li-rich pockets:

332   5910-19800 ppm; seams: 17-3640 ppm) (Table 3, Figs. 7e, 8b, c). Apatite from the pods of

333   the core zone, as well as the zoned crystals from the Li-poor pockets and seams (gray-white

334   and purple-green respectively), show significant Sr variations on the crystal scale (Table 3,

335   Fig. 9e). The greatest variations correspond to the gray-white apatite crystals, where the Sr

336   content changes in the range from 759 ppm in the core to 28710 ppm in the rim, with a

337   continuous increase outwards (Fig. 9e). The Sr content of the purple-green zoned crystals

338   from the seams also increases outwards, from 17 to 207 ppm Sr in the purple core to 93 to

339   3640 ppm in the green rim (Table 3, Fig. 9e). Some of the small euhedral apatite crystals

340   from the core zone pods (Fig. 3c) also present an increasing Sr-trend from the core (27 ppm)

341   to rim (662 ppm) (Table 3).

342   Uranium, Th, and Pb concentrations also show differences on the pegmatite and mineral

343   scales (Table 3, Figs. 7f, g, h, 8d, e, f). Uranium values are higher than the Pb contents in

344   apatite from the wall zone, intermediate zone, and core margin. Thorium is only higher than

345   Pb in apatite from the intermediate zone and core margin (Fig. 8d, e). Likewise, the rims of

346   gray-white zoned crystals from the Li-poor pockets and those of zoned crystals from the core

347   zone pods are richer in U than in Pb. The highest U and Th contents correspond to apatite

348   from the core margin (153-590 ppm U and 154-190 ppm Th), whereas the lowest contents

349   are those from the purple crystals from the Li-rich pockets (0.04-0.82 ppm U and Th below

350   detection limits) and from the pods of the core zone (0.01-49.2 ppm U and <0.2 ppm Th)

351   (Table 3, Figs. 7f, g, h, 8d, e, f). On the crystal scale, U, Pb, and Th elements behave

352   similarly in the purple-green crystals, with an initial increase and a later decrease from core

353   to rim (Fig. 9f, g, h); whereas in the gray-white crystals, Pb and Th behave similarly, with a

354   decrease from the core to the rim, whereas U presents the opposite trend (Fig. 9f, g, h).

355   The pairs of elements Y-Ho and, to a lesser extent, Zr-Hf show good positive

356   correlations (Fig. 8g, h). There is a continuous decrease in the Y and Ho contents from the

357   wall zone, through the intermediate zone, followed by the core margin and the core zone. On

358   the crystal scale, the gray-white and purple-green zoned crystals show a clear depletion in Y

359   and Ho from the core to the rim (Fig. 9a, b, d), whereas in apatite from the core zone pods

360   their content is below the LA-ICP-MS detection limit. In contrast to Y-Ho, a clear relationship

361   between pegmatite units and Zr-Hf contents has not been identified. The highest contents in

362   both elements correspond to the zoned apatite crystals from the core zone pods and to the

363   rims of the purple-green prisms from the seams (Fig. 8h).

364

365   **REE patterns**

366   Chondrite-normalized REE patterns for apatite from different zones of the pegmatite

367   show significant differences, with a continuous decrease inwards. The wall zone apatite

368   crystals present nearly sub-horizontal well-defined patterns, a marked negative Eu anomaly

369   (0.08-0.09) and slight tetrad effects (Figs. 10a, 11). Apatite from the intermediate zone is

370   poorer in REE and shows a quite different pattern, with a pronounced negative slope for the

13

371 HREE, very marked Eu negative anomaly (0.01-0.02) and strong tetrad effects (Figs. 10a,

372 11). The lanthanide pattern for apatite from the core margin is similar to that of the

373 intermediate zone, but with lower concentrations. It also presents a marked negative slope

374 for the HREE, strong but less marked Eu negative anomaly (0.12-0.15), and important tetrad

375 effects (Figs. 10a, 11). Apatite of pods in the core zone is extremely impoverished in REE,

376 with no or incomplete/erratic patterns. This is also the case for the purple apatite crystals

377 from the Li-rich pockets (Figs. 10a, 11).

378   The zoned apatite crystals from the Li-poor pockets and seams show significant

379 variations in their lanthanide patterns with an overall decrease from core to rim. The patterns

380 for the gray-white crystals are well-defined. The core and intermediate areas of the crystals

381 show sub-horizontal patterns, whereas in the rims the patterns slope upwards from LREE to

382 HREE (Fig. 10b). The Eu negative anomaly decreases from core to rim (Figs. 10b, 11). In the

383 whole crystals, the tetrad effects are equally important, but less pronounced than in apatite

384 from the intermediate zone and core margin (Figs. 10b, 11).

385   The purple-green zoned crystals in the seams also show conspicuous lanthanide zoning.

386 As in the gray-white lenses, the REE contents decrease outwards, in tandem with a

387 decrease in the Eu anomaly. However, in contrast with the gray-white lenses, the Eu

388 anomaly becomes positive in many rim analyses (0.52-2.97). The patterns tend to slope

389 downwards from LREE to HREE and the tetrad effects are less pronounced (Figs. 10c, d,

390 11).

391

392   **DISCUSSION**

393 **Variations in Mn**

394   The analyzed apatite crystals from the Berry-Havey pegmatite show a negative Ca-Mn

395 correlation (Fig. 5a), with the highest Mn values in apatite from the core margin and

396 intermediate zone, and the lowest in the wall zone and core zone (Fig. 4). According to some

397 authors (e.g. Miles et al. 2013; Belousova et al. 2002), reducing conditions could favor the

398 incorporation of Mn in apatite as the size (0.66Å) and charge of Mn^{2+} are more similar to

399 those of Ca^{2+} (1.00Å) than those of Mn^{3+} (0.58Å) and Mn^{4+} (0.39Å). The evolution of the fO_2

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400   during pegmatitic crystallization is usually difficult to determine. The composition of biotite

401   from the wall zone and intermediate zone (unpublished data) is consistent with the NNO

402   buffer (calculated according to the diagram of Wones and Eugster, 1965). Also, the presence

403   of $(Fe+Mn)^{2+}$-rich primary phosphates such as lithiophilite ($LiMn^{2+}PO_4$), triplite ($Mn^{2+}_2(PO_4)F$)

404   and dickinsonite ($(KNa)(Mn^{2+}\square)Ca(Na_2Na)Mn^{2+}_{13}Al(PO_4)_{11}(PO_4)(OH)_2$) in the pods of the core

405   zone indicates that the fO_2 was still reduced to predominantly Fe^{2+} near NNO (London, pers.

406   comm) at least until the beginning of the crystallization of the core zone. In contrast, the

407   chemical composition of Li-tourmaline (elbaite-darrellhenryite) coexisting with purple apatite

408   in the Li-rich pockets from the core zone reveals a final increase in the fO_2 that enables the

409   elbaite-darrellhenryite exchange vector ($^YAl^WO_2{}^YLi_{-1}{}^W(OH)_{-2}$) to operate during the

410   crystallization of the gemmy tourmaline found in the Li-rich pockets (Roda-Robles et al.

411   2015b). Moreover, the Eu and Ce anomalies observed in apatite from the pockets mainly

412   correspond to oxidizing conditions (see below). Accordingly, redox conditions during the wall

413   zone, intermediate zone, and core margin crystallization were likely reduced, shifting toward

414   more oxidizing conditions during the crystallization of the core zone pods. Finally, the

415   pockets most probably crystallized under oxidizing conditions (Fig. 12a). Thus, the

416   incorporation of Mn into apatite of the intermediate zone and core margin could have been

417   favored by the relatively low fO_2 of the pegmatitic melt, near the NNO buffer. However,

418   factors other than redox conditions should have contributed to the high Mn content in apatite

419   of some pockets, as well as to the low Mn content in apatite of the wall zone. Even though

420   crystallization of the wall zone most probably proceeded under low fO_2 conditions, the Mn

421   content in apatite from this zone is very low. The most feasible reason may be related to low

422   availability of Mn in the pegmatitic melt at the beginning of crystallization. This would be

423   supported by the garnet chemistry in the wall zone, which corresponds to almandine

424   ($Alm_{76.2}Sp_{19.5}Pyr_{4.3}$, Roda-Robles et al. 2017). The spessartine component increases from

425   core to rims in single garnet crystals and from the wall zone up to the core margin as a result

426   of fractionation (Roda-Robles et al. 2017), a behavior commonly observed in many

427   pegmatites (e.g. Baldwin and von Knorring, 1983; Černý et al. 1985; Sokolov and Khlestov,

428   1990; Hernández-Filiberto et al. 2021). Therefore, at the beginning of pegmatite

429 crystallization, Mn concentration in the melt would be relatively low, which is reflected not

430 only in the garnet but also in the apatite composition of the wall zone.

431 In contrast, the highest Mn values in apatite are found in the core margin. Garnet

432 coexisting with this apatite corresponds to spessartine ($Alm_{42.6}Sp_{57.3}Pyr_{0.1}$, Roda-Robles, et

433 al. 2017), which means a higher availability of Mn in the melt during the formation of core

434 margin than during the formation of wall zone. The previous crystallization in the wall zone

435 and intermediate zone of tourmaline (schorl/foitite) and, to a lesser extent, biotite would allow

436 the MnO/FeO ratio of the pegmatitic melt to increase, as Mn is incompatible or presents a

437 low compatibility with these two minerals (London et al. 2001a; García-Serrano et al. 2017;

438 Maner et al. 2019).

439 The Mn content of apatite in pods of the core zone becomes low again (Figs. 4a, 5a),

440 even though the fO_2 was still reduced during their crystallization (as supported by the

441 presence of nodules of Mn^{2+}-rich primary phosphates). An explanation for this Mn depletion

442 in apatite may be found in the consumption of most of the Mn by these Mn-rich phosphate

443 nodules in the core zone pods, which would drastically reduce the concentration of Mn in the

444 remaining melt.

445 Finally, apatite from the pockets and seams shows important differences in the Mn

446 contents, which may account for different time/conditions in the formation of miarolitic

447 cavities, even if they are found in relatively close proximity to other cavities, mainly in the

448 core zone. Like the apatite from core zone pods, the purple apatite of Li-rich pockets inside

449 those pods is Mn-poor. Miarolitic cavities usually are found in the innermost part of zoned

450 pegmatites and are considered to represent the transition from an igneous to a hydrothermal

451 stage, hosting minerals that first formed from a silicate melt and, later, others that crystallized

452 from an aqueous solution (Jahns, 1982; London et al. 2020). Actually, the temperatures

453 estimated for pocket formation are relatively low, in the range of 435-355°C (London et al.

454 2020). In the case of the Berry-Havey pegmatites, the pegmatite crystallized from the

455 borders inwards and, according to the mineralogical association, mineral chemistry, and

456 spatial distribution, the Li-rich pockets were formed during the latest crystallization stages, at

457 the lowest temperatures and most oxidizing conditions (Roda-Robles et al. 2015b). There are

458   several lines of evidences to support this. First, these pockets are usually hosted by the most

459   fractionated pods of the core zone, with significant amounts of Li-F±B±P-rich phases (e.g.

460   lepidolite, F-elbaite, montebrasite (LiAl(PO$_4$)(OH, F)) and lithiophilite). The K/Rb ratio in K-

461   feldspar and mica, which has been often used as an indicator of the fractionation degree of

462   granitic pegmatites (e.g. Roda-Robles et al. 2006; London et al. 2012; Brown et al. 2017;

463   Garate-Olave et al. 2018), decreases progressively from the wall zone (308-425 in K-feldspar

464   and 116-125 in Al-mica), through the intermediate zone (98-131 in K-feldspar and 41-101 in

465   Al-mica) and the core margin (10-44 in Al-mica), to the core zone, where this ratio is as low

466   as 15 to 20 for K-feldspar and 6 to 18 for Al-mica (unpublished data). Moreover, the Li

467   content in tourmaline (Roda-Robles et al. 2015b) and in mica increases along the same path.

468   The inward increase in the F content from the beginning of crystallization is evidenced by the

469   Fluorine Intercept Value in biotite, calculated according to Munoz (1984), which decreases

470   from 4.22 in the wall zone, to 3.91 in the intermediate zone below the core margin, and to 3.5

471   in the intermediate zone over the core margin. An even higher F-content in the melt at the

472   final crystallization stages is attested by the common occurrence of the F-rich tourmaline in

473   the core margin and core zone (Roda-Robles et al. 2015b) as well as by the presence of

474   subrounded montebrasite pods in the core zone with F values in the range 3.1-5.6 wt%

475   (unpublished data). According to the correlation between F in montebrasite and F in the melt

476   determined by London et al. (2001b), the [F] in the melt during the core zone crystallization

477   would be in the range 0.83-1.51 wt%. Therefore, the Li-rich pockets would have formed at

478   the latest stages of crystallization of the Berry-Havey pegmatite and, by then, most of the Mn

479   could have been consumed. This, in conjunction with the high fO_2 and high concentration of

480   fluxes, arguably contributed to the low Mn content in the purple apatite of the Li-rich pockets.

481

482   **Strontium and Y variations**

483   Opposite to Mn and some REE (see below), the Sr and Y contents in apatite are

484   believed to be independent of the fO_2 (Yu et al. 2021). Feldspars can accommodate Sr

485   substituting for Ca and/or K (Smith, 1974), and its compatible behavior causes its

486   concentration to typically decrease in apatite with fractionation (e.g., Belousova et al. 2002;

487 Chu et al. 2009; Cao et al. 2012; 2013; Pan et al. 2016; Azadbakht et al. 2018).

488 Consequently, apatite associated with fractionated granites and pegmatites often has the

489 lowest Sr values, in some cases < 100 ppm (Belousova et al, 2002). In the Berry-Havey

490 pegmatite, in the first stages of crystallization Sr behaves accordingly, its content in apatite

491 decreasing from the wall zone to the intermediate zone. However, the Sr trend exhibits an

492 inflection point in the intermediate zone, and apatite becomes richer from the intermediate

493 zone, through the core margin, to the core zone, with the highest Sr-contents in apatite from

494 pockets and seams, sometimes close to 3 wt% (Figs. 7e, 8b, c; Table 3). Similar behavior

495 has been recently described and discussed in apatite occurring in the most fractionated

496 pegmatites and leucogranites from the Central Iberian Zone (Spain and Portugal) (Roda-

497 Robles et al. 2022). In the Berry-Havey pegmatite, plagioclase composition changes

498 progressively from the contact with the country rock inward, with a dramatic decrease in the

499 CaO and Sr contents from the wall zone (up to 13 wt% and 1044 ppm, respectively) to the

500 intermediate zone (< 1.14 wt% and < 4 ppm, respectively), and even lower in plagioclase

501 from the core margin and the core zone (< 0.1 wt% and < 2 ppm, respectively) (unpublished

502 data). According to the experimental works of Icenhower and London (1996), and the data

503 compiled by Smith (1974), plagioclase with an Na content corresponding to the andesine

504 composition may be richer in Sr than other more anorthitic feldspars. However, chemical

505 variations observed in plagioclase from the Berry-Havey pegmatite clearly indicate that the

506 most albitic plagioclase is the poorest in Sr, as may also be seen in the data compiled by

507 Smith (1974) for pegmatitic feldspars. Accordingly, we propose that the extremely Na-rich

508 composition of plagioclase from the high levels of magmatic fractionation could have made it

509 difficult for Sr to enter into the structure of the very pure albite. Consequently, Sr could have

510 started behaving as an incompatible element from the crystallization of the intermediate

511 zone, with the concomitant Sr increase in apatite from the inner zones of the pegmatite,

512 which at that point would have been the only sink for Sr in that highly evolved paragenesis.

513 This increasing trend for Sr is also observed in the individual zoned crystals, with Sr poorer

514 cores and richer rims (Fig. 9e). This is especially observed in gray-white lenses, whose Sr-

18

515   values in the rims (19,800-28,710 ppm) are nearly 40 times higher than those of the cores

516   (507-759 ppm).

517   Yttrium behavior in apatite is commonly opposite to that of Sr. The Y-richest apatite (>1

518   wt%) is associated with granitic pegmatites (Belousova et al. 2002), as this element usually

519   behaves as incompatible, increasing with magmatic fractionation (e.g., Belousova et al.

520   2002; Chu et al. 2009; Cao et al. 2012; Bromiley, 2021). However, like the REE, Y tends to

521   decrease in LCT granitic pegmatites (those enriched in Li, Cs and Ta) with fractionation. This

522   could be related to the relatively high availability of P in these pegmatitic melts, as attested to

523   by the common occurrence of phosphates in these rocks. Under such conditions, early

524   crystallization of Y(+REE)-bearing phosphates, such as xenotime-Y and/or monazite-Ce,

525   even in very low proportions, may lead Y and REE to behave as compatible, hence depleting

526   their concentration in melts during fractionation (Jolliff et al. 1989). In the Berry-Havey

527   pegmatite, Fe-Mn- and Al-phosphates are common in the inner zone, which indicates that P

528   concentration in the melt was high enough to attain phosphate saturation. Monazite-Ce and

529   xenotime-Y are accessory minerals that are found in the intermediate zone and in the garnet

530   layer between the intermediate zone and the core margin. Therefore, it would be possible

531   that early crystallization of these phases could be responsible for the Y and REE depletion in

532   apatite during fractionation of the Berry-Havey pegmatite, with a decrease for apatite from

533   the wall zone, through the intermediate zone and the core margin; up to the core zone.

534   Zoned apatite crystals also show a decreasing Y-content from core to rim (Fig. 9d).

535

536   **REE patterns**

537   At the Berry-Havey pegmatite, the REE content in apatite decreases from the wall zone

538   inwards, i.e., with magmatic evolution, as it has been previously reported in other localities

539   (e.g., Jolliff et al. 1989; Bromiley, 2021; Roda-Robles et al. 2022). Incorporation of REE into

540   apatite occurs via two main substitution mechanisms that mainly involve Ca and, to a lesser

541   extent, P, Si and Na: $REE^{3+}_1 Na^+_1 Ca^{2+}_{-2}$ and $REE^{3+}_1 Si^{4+}_1 P^{5+}_{-1} Ca^{2+}_{-1}$ (Sha and Chapell, 1999;

542   Pan and Fleet, 2002; Webster and Piccoli, 2015; Harlow, 2015). The low REE content in the

19

543   studied apatite impedes the determination of the influence of the two exchange vectors in the

544   incorporation of REE in the studied apatites.

545   Nearly all the studied apatite crystals show more or less pronounced tetrad effects, with

546   values of $TE_{1,3}$ >1.1 (calculated according to Irber, 1999). Except for apatite from the wall

547   zone that presents intermediate $TE_{1,3}$ values, the tetrad effects become less marked in

548   apatite from intermediate zone to core zone, parallel to a decrease in the Y content. (Fig.

549   11b). According to some authors, strong tetrad effects in apatite may be related to high fluid

550   activity in the melt (Kawabe, 1995; Bau, 1996; Irber, 1999; Monecke et al. 2002; Zhao et al.

551   2002). However, aqueous fluid activity in the pegmatitic melt during the crystallization of the

552   inner zones of the Berry-Havey pegmatite (core margin, core zone) would probably be much

553   higher than during the crystallization of the intermediate zone. Even more so, apatite crystals

554   associated with the pockets, which are likely related to the activity of aqueous fluids exsolved

555   from the pegmatitic melt, have much lower $TE_{1,3}$ values than apatite from the intermediate

556   zone, the core margin and, in some cases, the wall zone. Consequently, tetrad effects in the

557   apatite from the Berry-Havey pegmatite appear to be unrelated to the aqueous fluid activity

558   during their crystallization.

559   The abundance of aqueous fluids has also been proposed to be associated with non-

560   CHARAC (CHArge-and-Radius-Controled) Y/Ho ratios (Bau, 1996; Peretyazhko and Savina,

561   2010; Cao et al. 2013). Most of the studied apatite crystals from the Berry-Havey pegmatite

562   plot out of the Y/Ho CHARAC range (24-34) (Fig. 12b). Only Y/Ho ratios from apatite from

563   the wall zone and some values corresponding to the purple cores of zoned crystals from the

564   seams plot in or close to those values. It should be noted that Y/Ho values increase

565   progressively for apatite from the wall zone, the intermediate zone, and the core margin.

566   Similarly, in the zoned crystals from the pockets and seams, the Y/Ho value increases

567   outwards, i.e. during crystallization. Purple apatite from the Li-bearing pockets also shows

568   some of the highest Y/Ho values. In short, the apatite of the Berry-Havey pegmatite shows a

569   non-CHARAC behavior that is characteristic of magmatic-pegmatitic systems with high SiO_2

570   contents enriched in H_2O, Li, B, F, P, and/or Cl, as suggested by Bau, (1996); Peretyazhko

571   and Savina, (2010) and Cao et al. (2013).

572

Europium and Ce anomalies

Most of the REE present just a single trivalent valence state and are readily incorporated into apatite via the two REE coupled substitutions noted above. In contrast, Eu and Ce may occur with two different oxidation states (Eu^{2+} - Eu^{3+} and Ce^{3+} - Ce^{4+}, respectively) (e.g., Burnham and Berry, 2014). In both cases, the trivalent Eu and Ce cations are more easily incorporated into the apatite structure thanks to the higher similarity of their ionic size to that of Ca^{2+} (Cao et al. 2013). As a result, Eu and Ce can present anomalous contents in apatite compared to the other trivalent REE (Bromiley, 2021). Such anomalies have been often related to fO_2 conditions during the crystallization of melts, with strong Eu-negative anomalies (high Eu^{2+}/Eu^{3+} ratios) in apatite formed under low fO_2, and no or positive Eu anomalies under relatively oxidizing conditions (e.g. Sha and Chappell, 1999; Pan and Fleet, 2002; Belousova et al. 2002; Chu et al. 2009; Cao et al. 2012; Miles et al. 2014; Azadbakht et al. 2018; Bromiley, 2021). In addition, the Eu anomaly may be conditioned by the crystallization of other mineral phases that compete for this element, namely feldspars, where Eu^{2+} is highly compatible (especially in K-feldspar) (Kontak and Martin, 1997; Belousova et al. 2002). Consequently, if apatite crystallizes after or during the formation of feldspar, it would show a negative Eu anomaly (Drake and Weill 1975; Sha and Chappell, 1999; Belousova et al. 2002; Cao et al. 2013; Miles et al. 2014; Abdullin et al. 2016). The Ce anomaly is usually less pronounced than the Eu anomaly (Sha and Chappell, 1999; Belousova et al. 2002; Chu et al. 2009; Ding et al. 2015), and it does not seem to be affected by the presence of other minerals except for the REE-bearing phases such as zircon, monazite, or xenotime. (Bromiley, 2021). Compared to the Eu anomaly, the Ce anomaly is considered to be less appropriate to assess the redox conditions during apatite crystallization (Cao et al. 2012), but a combination of both anomalies seems to be more useful for that purpose (Cao et al. 2012; Pan et al. 2016; Azadbakht et al. 2018).

Apatite from the Berry-Havey pegmatite presents significant differences in Eu and Ce anomalies regarding its position inside the body, with a negative correlation for both values (Fig. 12b). The strongest Eu negative and Ce positive anomalies correspond to apatite of the

21

601 intermediate zone, which suggests reducing conditions and would be in agreement with the

602 high Mn contents of apatite from this zone. However, the abundance of feldspars in the

603 intermediate zone (> 85 vol%) could also have influenced the availability of Eu for the

604 coexisting apatite, enhancing its negative Eu anomaly in the intermediate zone. On the other

605 hand, the less marked Eu and Ce anomalies in the wall zone apatite compared to the

606 intermediate zone (Fig. 12a) could be related to the early character of this phosphate in the

607 wall zone, inferred from textural characteristics (Fig. 3a). In the core margin the apatite

608 presents a less marked Eu anomaly, plotting in the "moderate oxidized" field (Fig. 12b),

609 which is in agreement with an increase of fO_2 inwards during pegmatite crystallization.

610 Moreover, except for garnet, no other phases competing for Eu coexist with apatite in this

611 zone. The garnet intergrown with bluish apatite in the core margin (Fig. 3b) is extremely

612 depleted in REE, which shows an erratic pattern (unpublished data). This indicates that

613 apatite controls the lanthanide distribution in the core margin. The extremely low REE

614 concentration in apatite from the core zone pods and the Li-rich pockets prohibits evaluation

615 of their Eu or Ce anomalies.

616 The zoned apatite crystals from the Li-poor pockets and seams show significant

617 differences from core to rim. In the first case, all the values plot in the "moderate oxidized"

618 field (Fig. 12a). The decrease in Eu and Ce anomalies from the gray core to the whitish rim

619 could well correspond to an increasing fO_2 during crystallization of the Li-poor pockets, as it

620 is also supported by the progressive decrease in Mn from core to rim in those lenses. Zoned

621 apatite crystals from the seams present a much broader range for the Eu anomaly than that

622 of Ce. While the purple cores of these crystals plot mainly in the "moderate oxidized" field,

623 many of the greenish rims show a positive Eu anomaly falling in the "oxidized" field (Fig.

624 12a). Accordingly, the fO_2 could have increased dramatically during crystallization of the

625 seams. Nevertheless, lower or positive Eu anomalies in apatite from the pockets and the

626 seams could also be related to the higher solubility of Eu in F- and Cl-rich H_2O-fluids

627 exsolved from the melt, in comparison with the other REEs (Abramov, 2001).

628

22

629 **Chemistry of apatite as a recorder of the crystallization history of the Berry-Havey**

630 **pegmatite**

631   Evidence for the exsolution of H_2O-rich fluids during the last stages of crystallization of

632 the Berry-Havey pegmatite is supported by the occurrence of pockets in the innermost parts

633 of the body. Based on the apatite composition and the mineral association of the different

634 types of pockets (Li-rich, Li-poor, and seams), the exsolution of fluids presumably proceeded

635 during different crystallization stages with, at least, three generations of pockets. The earliest

636 one would correspond to the Li-poor pockets where gray-white zoned apatite lenses occur.

637 Exsolution of the fluids would take place at the end of the crystallization of the core margin,

638 when the availability of Fe, Mn, and REE was still high enough for the crystallization of Fe-

639 rich tourmaline (schorl) and Mn+REE-rich apatite. Exsolution processes could be related to a

640 second boiling prompted by massive crystallization of tourmaline from the tourmaline layer in

641 the boundary between the core margin and the core zone. The sudden depletion in B

642 experienced by the melt could decrease its capability to keep H_2O dissolved (London, 1990)

643 with part of it exsolving in discrete H_2O-rich batches related to these pockets that present an

644 intermediate paragenesis with no Li-bearing minerals.

645   As indicated above, the Li-rich pockets including purple apatite crystals are interpreted to

646 correspond to the latest pocket generation. These pockets are hosted by the most

647 fractionated pods of the core zone, which are constituted by Li-F±B±P-rich phases. The

648 dramatic depletion of B, Li, F, and P fluxes in the residual melt could again induce the

649 exsolution of H_2O-rich fluids, which would be quite enriched in Li and other incompatible

650 elements, and strongly depleted in Mn and REE at that stage of crystallization. In these

651 pockets, the only sink for Sr would be apatite, which shows some of the highest

652 concentrations (Figs. 7e, 8b, c).

653   Finally, since apatite is the only mineral phase identified in the seams, this makes it

654 more difficult to determine their timing. In this case, apatite presents intermediate Fe, Mn,

655 and REE values between those of the apatite from the Li-poor and Li-rich pockets. Taking

656 into account that REE decrease with fractionation, these seams could have formed at some

657 point between the formation of the Li-poor and Li-rich pockets. However, the presence of

23

658 apatite as their only phase would involve the exsolution of a fluid phase just containing P and

659 Ca as main elements, which seems difficult to explain for such relatively high degrees of

660 fractionation. Another possibility could be that the formation of these seams was related to

661 the activity of late hydrothermal fluids that autometasomatized some portions of the

662 pegmatite, mainly the core margin and the core zone, causing albitization and greisenization.

663 During albitization, Ca and P (+Sr, Y and REE) would have been leached from plagioclase.

664 Those fluids could have then precipitated apatite in fractures and vugs, giving rise to the

665 apatite-rich seams, which always occur hosted by vuggy albite from the core margin or

666 blocky feldspar from the core zone. Nevertheless, the origin of the seams is not completely

667 understood at present.

668 The chemistry of apatite may also be a good indicator of the fractionation attained by the

669 pegmatites. This has been previously described for apatite associated with pegmatites and

670 leucogranites from the Central Iberian Zone (Spain and Portugal) showing different evolution

671 degrees (Roda-Robles et al. 2022). In the U-Th-P and Y-REE-Sr ternary plots (Fig. 13)

672 apatite from the most fractionated units plots significantly closer to the Pb and Sr vertex.

673

674 **IMPLICATIONS**

675 This study shows how a detailed petrographic and chemical characterization of apatite

676 associated with the different units of a highly fractionated, internally zoned pegmatite, may

677 help understand the crystallization history of pegmatitc melts. It is evidenced that during the

678 internal evolution of pegmatites, the apatite chemistry records variations in the fO_2, elemental

679 fractionation, interaction with competing mineral phases, fluids activity, and exsolution

680 events. The main chemical elements in apatite that provide petrogenetic information in

681 pegmatitic rocks include Mn, Sr, the REE, and Y.

682 In this work it is also shown how the chemistry of apatite may be useful in determining

683 the degree of fractionation attained by a pegmatite. Therefore, the usefulness of this

684 phosphate as an exploration tool for pegmatites is obvious. In this sense, the two ternary

685 diagrams Y-ΣREE-Sr and U-Th-Pb may help to discriminate between apatite crystals

24

686   associated with pegmatitic facies showing different fractionation degrees, with apatite from

687   the most evolved facies plotting closer to the Sr and Pb vertex.

688

689   **ACKNOWLEDGMENTS**

700

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946 **FIGURE CAPTIONS:**

947 **Fig. 1.** (a) Map showing the location of the Migmatite-Granite-Complex in New England

948 and (b) the principal geological features of the area and location of the Berry-Havey

949 pegmatite.

950

951 **Fig. 2.** Schematic map with two cross sections of the Berry-Havey pegmatite.

952

953 **Fig. 3**. (a) Microphotograph of a subhedral apatite crystal from the Wall Zone (under

954 plane-polarized light). (b) Dark bluish apatite crystals intergrowth with garnet from the garnet

955 layer, in the limit between the Intermediate Zone and the Core Margin. (c) Microphotograph

956 of some subhedral apatite crystals from a pod of the Core Zone (under crossed-polarized

957 light). (d) Seam filled by purple apatite hosted by blocky feldspar in the Core Zone. (e)

958 Strongly zoned grayish-white apatite euhedral lenses over quartz from a Li-poor pocket. (f)

959 Euhedral purple prism of apatite over a mass of albite and pinkish lepidolite from a Li-rich

960 pocket.

961

962 **Fig. 4.** Major element variations in apatite from the different units distinguished in the

963 Berry-Havey pegmatite: (a) F; (b) Mn^{2+}; and (c) Ca. (All values in a.p.f.u.).

964

965 **Fig. 5.** Binary plots for apatite from the different units of the Berry-Havey pegmatite (all

966 the values are in a.p.f.u.): (a) Ca versus Mn^{2+}; (b) Fe^{2+} versus Mn^{2+}; and (c) Ca versus Fe^{2+}.

967

968 **Fig. 6.** Representative chemical zoning patterns for the major elements in zoned apatite

969 crystals (left: grayish-white crystal from a Li-poor pocket; and right: purple-green crystal from

970 a seam). (**a**) Mn^{2+}, (**b**) Fe^{2+}, and (**c**) Ca. (All values are in a.p.f.u.).

971

972 **Fig. 7.** Variations in the main trace elements in apatite from the different units

973 distinguished in the Berry-Havey pegmatite. (All values in ppm) (Legend as in Fig. 4).

974

975 **Fig. 8.** Binary plots of main trace elements in apatite (all values in ppm): (a) ΣREE

976 versus Y; (b) Sr versus ΣREE; (c) Sr versus Y; (d) Pb versus Th; (e) Th versus U; (f) Pb

977 versus U; (g) Ho versus Y; and (h) Hf versus Zr.

978

979 **Fig. 9.** Representative chemical zoning patterns for main trace elements in zoned

980 apatite crystals (left: grayish-white crystal from a Li-poor pocket; and right: purple-green

981 crystal from a seam). (All values in ppm).

982

983 **Fig. 10.** Chondrite-normalized (McDonough and Sun, 1995) lanthanide patterns for: (a)

984 apatite from the wall zone, the intermediate zone, the core margin, and the purple apatite

985 from the Li-rich pockets in the core zone; (b) zoned grayish-white apatite lenses from Li-poor

986 pockets in the core zone; and, (c) and (d) zoned purple-green apatite crystals from the

987 seams. (Legend as in Fig. 4).

988

989 **Fig. 11.** Binary plots of (a) the Eu anomaly, (defined as Eu/Eu* = $Eu_n/(Sm_n \times Gd_n)0.5$)

990 and the $TE_{1,3}$ value for the tetrad effects calculated according to the procedure of Irber

991 (1999); and (b) Y (ppm) versus $TE_{1,3}$. (Legend as in Fig. 4).

992

993   **Fig. 12.** Binary plots of (a) the Ce anomaly (defined as $Ce/Ce^* = Eu_n/(La_n \times Pr_n)0.5$)

994   versus the Eu anomaly (defined as $Eu/Eu^* = Eu_n/(Sm_n \times Gd_n)0.5$), and (b) Y/Ho versus Y (in

995   ppm). The grayish area represents the region where the Y/Ho ratios correspond to CHARAC

996   values. (Legend as in Fig. 4).

997

998   **Fig. 13.** Triangular plots for apatite: (a) Pb-U-Th; and, (b) Sr-Y-ΣREE, for the different

999   units distinguished in the Berry-Havey pegmatite. (Legend as in Fig. 4).

1000

1001   **TABLE CAPTIONS:**

1002   **Table 1.** Main characteristics of the Berry-Havey pegmatite zones and the associated

1003   apatite.

1004   **Table 2.** Representative microprobe analyses with major element composition (wt%)

1005   and unit formula (a.p.f.u.) calculated on the basis of 26 (O,F,Cl,OH), H_2O^* estimated, for

1006   apatite associated with the different units distinguished in the Berry-Havey pegmatite.

1007   **Table 3.** Representative trace element analyses (values in ppm) for apatite associated

1008   with the different units distinguished in the Berry-Havey pegmatite.

32

ZONE		MINERALS*	GENERAL TEXTURES	APATITE TEXTURE	APATITE CHEMISTRY (ppm)
WALL ZONE		Qz, Kfs, Pl, Bt, Ms ± Grt ± Srl ± Ap	Homogeneous, very fine to medium grained facies. Locally gneissic facies	Fine grained whitish subhedral crystals < 5 mm	Fluorapatite ΣREE: 1796-2178 Y: 1155-1503 Sr: 49.9-52.5
INTERMEDIATE ZONE		Qz, Kfs, Pl ± Grt ± Bt ± Srl ± Ap	Qz-Kfs graphic intergrowths (> 85% volume)	Fine grained whitish anhedral crystals < 5 mm	Fluorapatite ΣREE: 1219-1286 Y: 225-261 Sr: 2.9-16.1
CORE MARGIN		Ab, Qz, Srl ± green-Elb ± Grt ± Ap	Matrix of tabular crystals of clevelandite ("vuggy" Ab), where coarse tourmaline tapered prismatic crystals occur. Grt layer	Medium sized bluish-greenish prismatic or anhedral crystals < 5 cm Intergrowth with Grt in the Grt layer	Mn-rich fluorapatite ΣREE: 309-355 Y: 22.7-26.3 Sr: 87-23.4
CORE ZONE pods		Qz, Kfs, Ab, Lpd, Ms, Li-Ms, Mbs, Brl, Fe-Mn Phosphates, Ross-Elb, ± Cst ± Clb-Ttl ± Ap	Blocky Qz, Kfs, and Ab. Purple fine-grained or book Lpd. Coarse book Ms rimmed by Lpd. Multicoloured tourmaline prisms. Fe-Mn-phosphates or Mbs sub-rounded pods	Fine-sized prismatic crystals with a concentric optical zoning	Fluorapatite ΣREE: 0-0.14 Y: 0 Sr: 27-1614
CORE ZONE Pockets	Li-poor pockets	Qz, Kfs, Ab, Srl, Ap	Blocky Qz and Kfs, "vuggy" Ab. Sub to euhedral smoky Qz, Etched dark tourmaline	Hexagonal zoned lenses with a greyish-light blue core and a white rim (≈1 cm Ø)	Mn-rich fluorapatite ΣREE: 9.55-983 Y: 31.7-979 Sr: 759-28710
	Li-rich pockets	Qz, Kfs, Ab , Lpd, Li-Ms, Mbs, Brl, Fe-Mn Phosphates, Rsm-Elb ± Ap	Blocky Qz and Kfs, "vuggy" Ab. Multicoloured tourmaline, Lpd "pencils"	Deep purple or bluish short hexagonal prisms (≈1 cm Ø)	Fluorapatite SREE: 0.09-0.37 Y: 0.14-0.54 Sr: 5910-19800
	Seams	Ap	Elongated cavities (≤ 50 cm long and ≤ 20 cm wide) completely or partially filled with Ap	Anhedral purple masses or hexagonal prisms. Zoned prisms with purple core and greenish rims	Mn-rich fluorapatite ΣREE: 0.629-210 Y: 0-23 Sr: 17-3640

*Mineral symbols according to Warr (2021): Ap=apatite, Qz=quartz, Kfs= K-feldspar, Pl=plagioclase, Ms=muscovite, Grt= garnet, Srl=Schorl, Bt=biotite, Elb= elbaite, Lpd= lepidolite, Mbs= Montebrasite, Brl= beryl, Rsm= rossmanite, Cst= cassiterite, Clb-Ttl= columbite-tantalite. Grain size: very fine = <6 mm; fine = 6 mm to 2.5 cm; medium = 2.5 cm to 10 cm.

Table 1

Nº anal.	1	2	3	4	5	6	7	8	9	10	11	12	13
Zone	Wall Zone		Intermediate Zone		Core Margin		Core Zone (pods)		Core Zone (Li-poor Pocket)		Core Zone (Li-rich Pocket)	Core Zone (seam)	
weight(%)									grayish core	withish rim	purple	purple core	greenish rim
P_2O_5	43.29	42.08	41.15	41.57	42.34	42.13	41.95	41.61	42.25	40.43	41.42	40.96	41.76
SiO_2	0.03	0.05	-	-	0.04	0.03	0.01	0.02	-	-	-	b.d.l.	-
Al_2O_3	0.02	b.d.l.	0.01	0.01	b.d.l.	b.d.l.	b.d.l.	0.01	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.
La_2O_3	0.07	0.05	-	-	0.03	0.03	-	-	-	-	-	-	-
Ce_2O_3	0.01	0.22	-	-	0.05	0.06	-	-	-	-	-	-	-
MgO	b.d.l.	b.d.l.	0.03	0.04	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.02	0.03	0.03	b.d.l.	b.d.l.
CaO	55.00	55.86	48.84	48.86	42.34	42.13	55.72	55.11	50.62	53.69	54.36	53.46	56.25
MnO	0.07	0.38	6.38	6.43	9.70	9.55	0.21	0.28	4.65	0.21	0.10	3.91	0.93
FeO	0.03	0.01	0.32	0.31	0.21	0.15	b.d.l.	0.03	0.31	b.d.l.	b.d.l.	0.17	0.10
SrO	0.07	b.d.l.	-	-	b.d.l.	b.d.l.	-	-	0.09	3.41	2.35	-	-
Na_2O	0.02	0.13	b.d.l.	b.d.l.	0.02	0.04	0.04	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.01	0.00
F	3.10	3.24	4.28	4.34	2.62	2.66	3.51	3.34	5.91	5.34	5.00	3.17	3.17
Cl	b.d.l.	0.01	0.01	b.d.l.	b.d.l.	b.d.l.	0.01	b.d.l.	0.08	b.d.l.	b.d.l.	0.08	0.03
H_2O*	0.34	0.26	b.d.l.	b.d.l.	0.48	0.45	0.12	0.19	b.d.l.	b.d.l.	b.d.l.	0.25	0.29
O=F	1.31	1.37	1.80	1.83	1.10	1.12	1.48	1.41	2.50	2.25	2.11	1.35	1.34
TOTAL	100.74	100.92	99.20	99.74	96.73	96.11	100.08	99.18	101.43	100.86	101.17	100.66	101.18

Structural formulae on the basis of 26 (O,F,Cl,OH) (a.p.f.u.)

	1	2	3	4	5	6	7	8	9	10	11	12	13
P	6.074	5.953	5.962	5.981	6.244	6.250	5.971	5.974	5.910	5.789	5.870	5.880	5.911
Si	0.005	0.008	-	-	0.007	0.005	0.002	0.003	-	-	-	b.d.l.	-
Al	0.005	b.d.l.	0.001	0.001	b.d.l.	b.d.l.	b.d.l.	0.002	b.d.l.	b.d.l.	b.d.l.	b.d.l.	b.d.l.
La	0.004	0.003	-	-	0.002	0.002	-	-	-	-	-	-	-
Ce^{3+}	0.001	0.013	-	-	0.003	0.004	-	-	-	-	-	-	-
Mg	b.d.l.	b.d.l.	0.008	0.011	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.006	0.008	0.008	b.d.l.	b.d.l.
Ca	9.768	10.000	8.955	8.897	7.902	7.910	10.035	10.013	8.962	9.728	9.749	9.712	10.076
Mn^{2+}	0.010	0.054	0.924	0.926	1.430	1.417	0.029	0.041	0.650	0.030	0.014	0.561	0.132
Fe^{2+}	0.004	0.001	0.046	0.044	0.030	0.021	b.d.l.	0.005	0.043	b.d.l.	b.d.l.	0.025	0.014
Sr	0.006	b.d.l.	-	-	b.d.l.	b.d.l.	-	-	0.009	0.334	0.228	-	-
Na	0.005	0.042	b.d.l.	b.d.l.	0.007	0.014	0.012	b.d.l.	b.d.l.	b.d.l.	b.d.l.	0.003	0.000
F	1.625	1.712	2.315	2.334	1.445	1.474	1.864	1.789	3.087	2.854	2.648	1.700	1.673
Cl	b.d.l.	0.003	0.003	b.d.l.	b.d.l.	b.d.l.	0.002	b.d.l.	0.021	b.d.l.	0.000	0.022	0.008
OH*	0.375	0.285	0.000	0.000	0.555	0.525	0.134	0.211	0.000	0.000	0.000	0.278	0.319
(F+Cl+OH)	2.000	2.000	2.317	2.335	2.000	2.000	2.000	2.000	3.108	2.854	2.649	2.000	2.000

nº anal.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Zone	Wall Zone		Intermediate Zone		Core Margin		Core Zone (pods)			Core Zone (Li-poor Pocket)		Core Zone (Li-rich Pocket)		Core Zone (Seam)	
(All values ppm)										grayish-core	whitish-rim	purple		purple core	greenish rim
Li	11.19	15.88	11.2	15.6	21.3	28.1	0	0	0	8.9	-	-	1.8	1.74	54
Be	-	0.003	0.33	6.2	6.1	2.39	-	0.85	1.71			-	0.86	-	6.6
Si	-	280	-	-	1010	1370	1480	1030	0	-	-	-	-	2250	3550
K	-	1.94	10	25	7.9	179	0	0	0	-	-	-	1.4	-	26
Sc	0.65	1.12	1.09	1.27	1.35	1.336	0.77	1.49		-	1.23	-	-	-	-
Ti	62.9	77	80.3	101	46.2	53.9	179.3	180.2	186.8	180	191	104	91	177.4	178
V	-	-	-	-	-	0.099	0	0	0	-	-	-	-	-	-
W	0.077	0.086		0.04	0.161	0.24	1.52	0.0126	1.25	0.0245	0.0027	-	0.0076	0.022	0.01
Ga	-	-	0.18	0.25	-	1.29	0.342	0.268	0.351	-	0.156	-	-	0.477	0.66
Ni	0.265	-	-	-	0.206	-	0.76	0.64	0.87	-	-	-	-	0.61	0.83
Mo	-	-	0.43	0.485	0.531	0.557	0	0.191	0	0.34	-	-	-	0.078	0.092
Zn	1.13	0.82	2.71	2.35	15.7	7.54	0	0	0.94	5.2	-	2.34	-	-	1.15
Rb	0.144	0.107	-	0.14	0.22	19.5	0	0	0	-	-	-	-	-	1.08
Sr	50.8	52.5	2.94	16.1	23.4	8.7	34.3	1614	662	759	28710	19800	9470	32.5	2094
Y	1155	1503	225	230	23.69	25.27	0	0.104	0	979	31.7	0.536	0.46	8	0.023
Zr	-	0.05	0.16	4.4	0.105	0.155	62	7.35	32.2	0.076	0.065	0.139	0.374	-	37
Nb	-	-	-	1.15	0.9	1.37	0.231	0	0.51	-	-	-	-	-	0.027
Sn	0.21	0.14	7.54	8.1	2.15	4.53	0.063	0	0	2.47	0.204	0.09	0.092	0.062	0.222
Cs	0.0331	0.0308	-	0.102	0.271	2.94	0.014	0	0	-	-	-	-	-	0.39
Ba	0.33	0.068	0.2	3.4	0.53	0.23	0.11	0	0	0.056	0.59	1.88	0.111	-	0.283
La	134	173	277	289	153.4	168	-	0.0087	-	94.3	0.28	0	0.062	12.8	0.105
Ce	413	486	628	655	138.7	162.2	-	-	0.055	268	0.462	0.011	0.0617	24.1	0.192
Pr	65.1	73.3	47.6	50.1	6.23	6.94	-	-	0.0116	28.4	0.044	0	0.0088	2.2	0.0186
Nd	279	313	77.2	79.6	5.88	6.48	-	0.0011	0.027	72.2	0.106	0	0.033	5.57	0.044
Sm	96.3	113.4	56.8	58.9	2.91	3.1	-	-	0.022	47.4	0.123	0.0073	0.0204	2.67	0.016
Eu	3.263	3.414	0.301	0.307	0.115	0.138	-	-	-	1.815	0.0283		0	0.553	-
Gd	128.5	160	52.7	53.8	2.48	2.7	-	-	0.021	71.9	0.252	0.006	0.024	1.63	0.0047
Tb	34.2	42.1	15.63	16.21	0.782	0.842	-	-	-	24.3	0.1452	0.0041	0.0065	0.338	0.002
Dy	273	340	58.3	59.3	3.52	3.79	-	0.0024	-	177.9	1.649	0.0167	0.046	1.76	0.004
Ho	51.9	65	2.67	2.7	0.181	0.1948	-	-	-	23.5	0.298	0.0037	0.0083	0.191	-
Er	143	183	2.28	2.29	0.243	0.271	-	0.0038	-	46.7	0.99	0.0169	0.0288	0.459	-
Tm	21.53	27.81	0.191	0.175	0.0314	0.0341	-	-	-	8.3	0.351	0.0038	0.0055	0.095	-
Yb	137	177	0.685	0.748	0.186	0.248	-	0.0023		50.9	4.24	0.033	0.055	0.728	-
Lu	16.6	21.63	0.0631	0.0597	0.02	0.0261	-			5.26	0.582	0.0077	0.0097	0.114	-
Hf	0.086	0.111	0.0222	0.29	0.0034	0.0115	2.72	0.725	1.31	0.058	0.0076	0.0107	0.0099	-	6.62
Ta	0.0204	0.0274	0.0013	0.0102	0.0422	0.137	0.0125	0.0016	0.0111	0.0085	-	-	-	0.0021	0.066
Tl	-	-	-	-	0.0148	0.126	-	-	-	-	-	-	-	-	0.0034
Pb	4.66	5.57	38.3	36.6	25.55	23.42	4.65	12.17	0.941	193	4.93	341	9.33	6.4	17.5
Th	0.3	0.073	52.6	56.8	166.7	190	0.0079	-	-	13.5	0.0208	-	-	0.94	0.07
U	6.07	11.15	152.5	162.5	590	153.6	0.129	0.16	49.2	26.4	108.4	3.31	0.51	0.93	3.31
ΣLREE	987	1159	1086	1133	307.00	346.72	0.00	0.01	0.12	510.00	1.02	0.02	0.19	47.34	0.38
ΣHREE	809	1020	132.8	135.6	7.56	8.24	0.00	0.01	0.02	411.00	8.54	0.09	0.18	5.87	0.01

-Below detection limit

Igneous Rocks

Sebago Pluton
Migmatite-Granite-Complex
Granodiorites and tonalites
Syenites and quartz monzonites
Gabbros
Granites

Metamorphic and Sedimentary Rocks

Schists and metasedimentary rocks (Devonian)
Shales, mudstones, quartzites, limesones and migmatites (Silurian-Devonian)
Sandstones (Silurian)
Marbles and metasedimentary rocks (Silurian)
Slates and calc-silicate rocks (Silurian)
Sandstones (Ordovician-Silurian)
Metavolcanic rocks (Precambrian-Ordovician)
Slates (Precambrian-Ordovician)

BHB Bronson Hill Belt
CMB Central Maine Belt
NSZ Norumbega Shear Zone System
NT Nashoba Terrane
ACT Avalon Composite Terrane
SB Sebago Batholith

ME, Maine; NY, New York; VT, Vermont; NH,
New Hampshise; MA, Massachusetts; CT,
Connecticut; RI, Rhode Island.

A: Modified from Brown and Solar (1998)

B: Modified from Osberg et al. (1985), Nicholson et al (2006), Wise and Brown (2010)

Fig. 1

LEGEND

Dumps

Water

Core Zone: Pockets, multicolored tourmaline + Lep + Clay minerals
Pods, blocky Qz + blocky Kfs + book Ms + lepidolite masses + phosphate pods (Mbs + Fe-Mn) ± Brl (morg.) ± Cst ± Clb-Ttl

Core margin, Qz + Clevelandite + schorl suns with crown of graphic intergrowth (black ± green Tur)

Intermediate-II, prismatic coarse schorl crystals inside blocky Kfs and Qz

Intermediate-I, Qz + Kfs graphic intergrowth ± Grt ± Bt ± Tur (schorl)

Wall Zone, fine medium grained facies Qz + Bt + Kfs + Ms ± Grt ± Tur

Metamorphic country rock (amphibolites)

Basalt dyke

Garnel layer

Tourmaline layer

N

0   10   20m

B   B'   A   A'

A —————— A'   B —————————————— B'
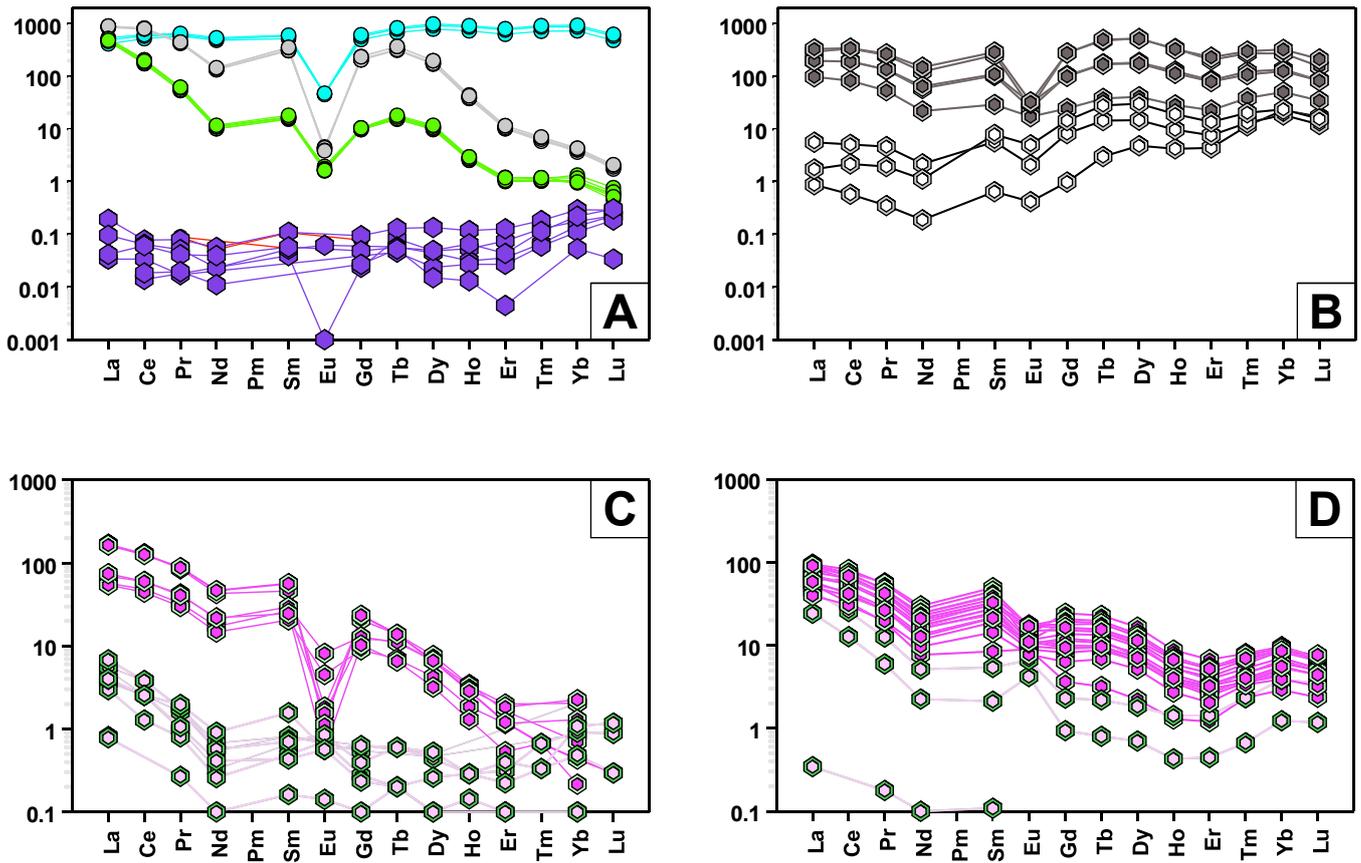
?   ?   ?   ?   ?

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7
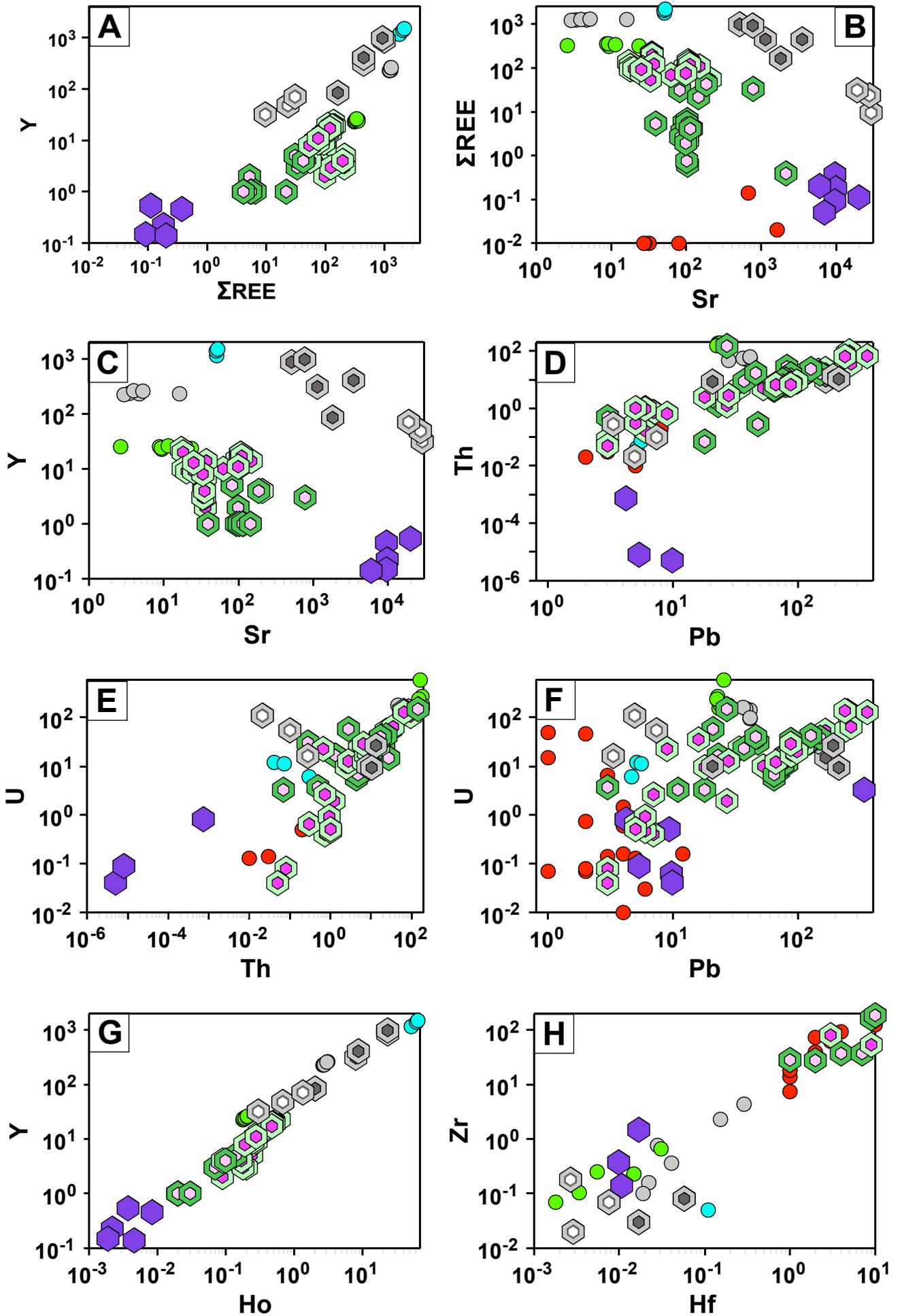
Fig. 8
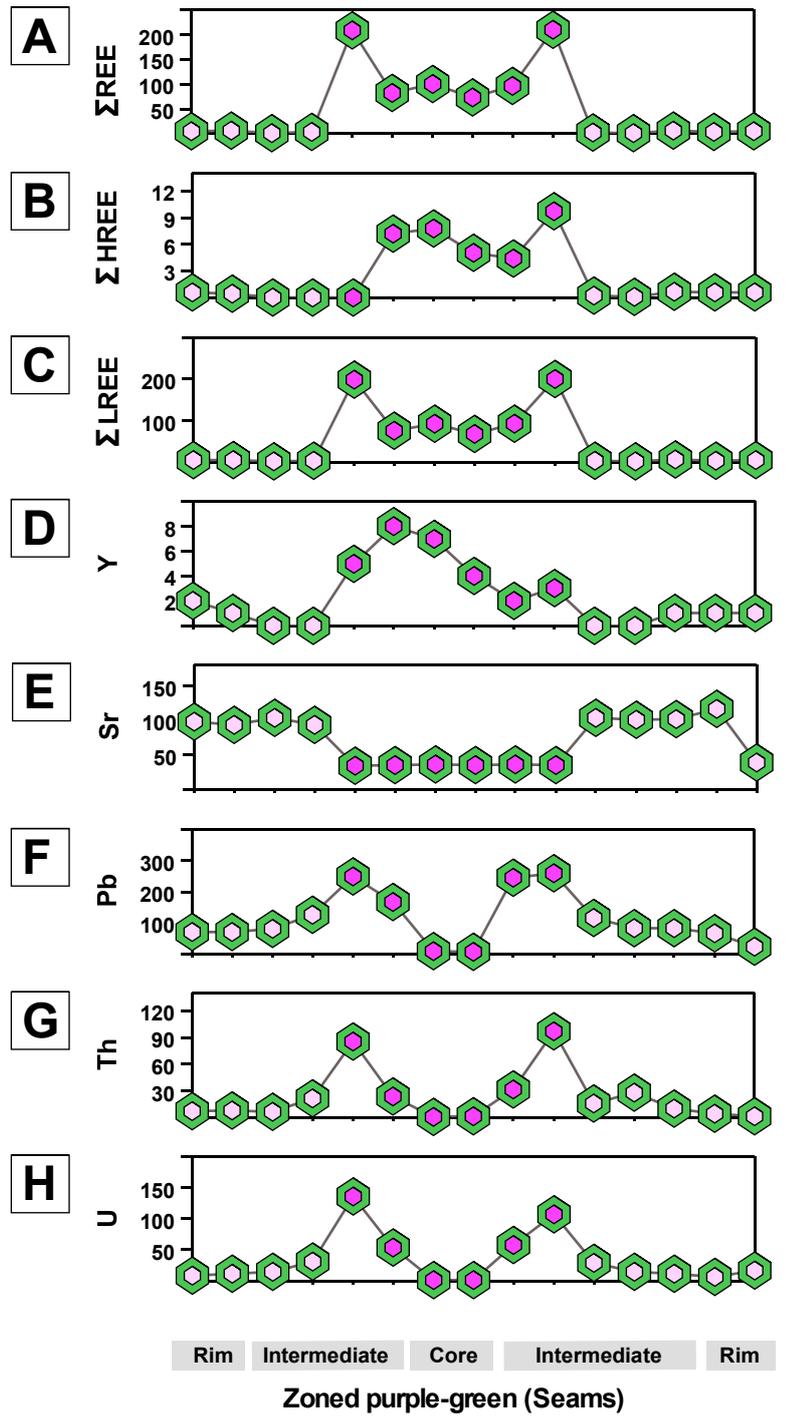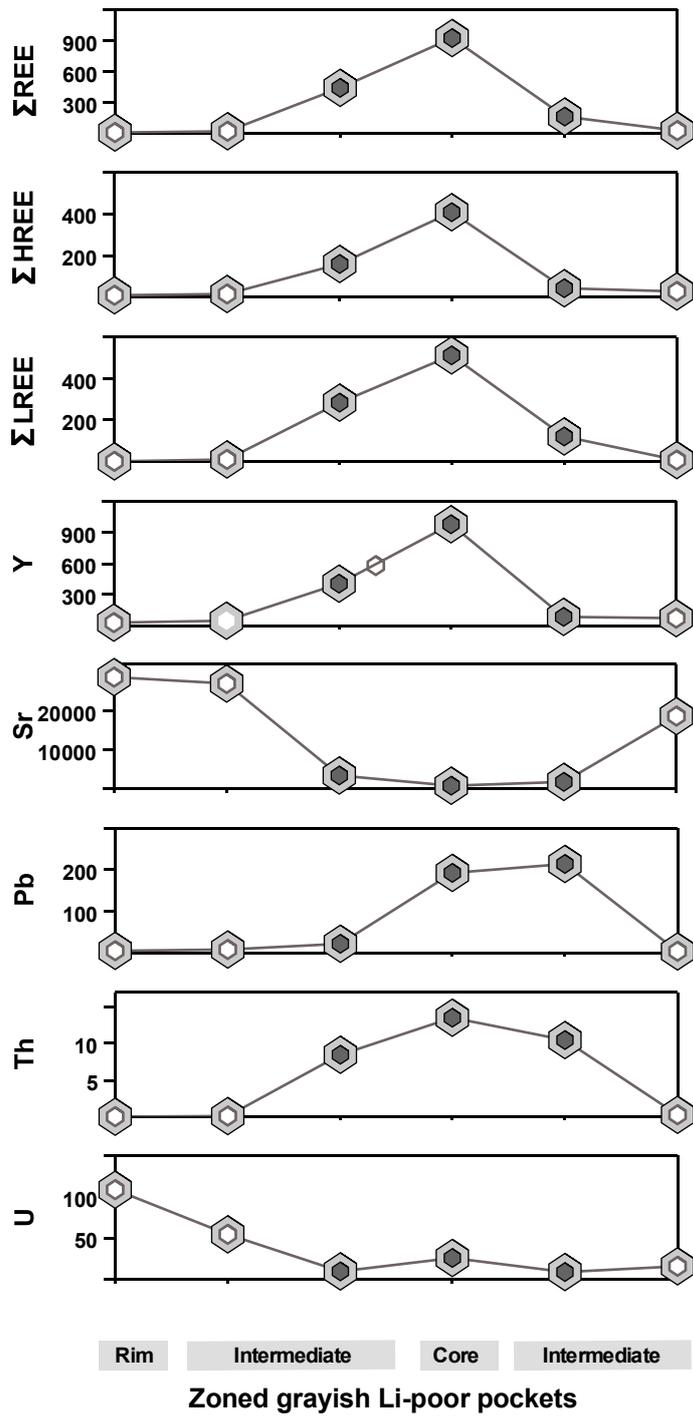
Fig. 9

Fig. 10

Fig. 11

Fig. 13

Fig. 12