The Canadian Mineralogist
Vol. 35, pp. 659-671 (1997)

RARE-EARTH ELEMENTS IN FLUORAPATITE, SEPARATION LAKE AREA, ONTARIO: EVIDENCE FOR S-TYPE GRANITE – RARE-ELEMENT PEGMATITE LINKAGE

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ABSTRACT

Fluorapatite is an important accessory and, locally, a major mineral in some extremely evolved, rare-element (Li, Rb, Cs, Be, Nb and Ta) pegmatites of the Separation Lake greenstone belt and the spatially associated S-type peraluminous granites of the Treelined Lake complex in the Umfreville – Conifer Lake granulite zone of the English River Subprovince, Ontario. The *REE* characteristics of fluorapatite in geochemically primitive pegmatites within the Treelined Lake complex are consistent with partitioning of *REE* with granitic melts that were derived from crustal sources. The chondrite-normalized *REE* patterns of fluorapatite in both primary and replacement units (mainly albite–mica pods) of the Separation Rapids rare-element pegmatites are characterized by marked discontinuities at Nd and Er. These unusual *REE* characteristics in fluorapatite have been modelled based on a granitic source (*i.e.*, the Treelined Lake complex) and the discontinuities at Nd and Er related to fractionation of monazite and garnet, respectively. Fluorapatite in the sodic wall-zones of the Separation Rapids rare-element pegmatites contains low abundances of *REE* and is characterized by chondrite-normalized *REE* patterns similar to those of the host amphibolites, supporting an infiltration of fluid from country rocks during the formation of pegmatites. The occurrence of fluorapatite with similar *REE* characteristics, including discontinuities at Nd and Er, in a miarolitic cavity within the Treelined Lake complex supports a genetic link between the Separation Rapids rare-element pegmatites and the S-type peraluminous granites associated with the granulite-facies metamorphism in the English River Subprovince.

Keywords: fluorapatite, S-type granite, rare-element pegmatite, discontinuities in *REE* plots, fractionation, monazite, garnet, Separation Lake Greenstone Belt, English River Subprovince, Ontario.

SOMMAIRE

La fluorapatite est un accessoire important et, ici et là, même un minéral majeur dans les venues pegmatitiques très fortement évoluées et enrichies en éléments rares (Li, Rb, Cs, Be, Nb et Ta) de la ceinture de roches vertes de Separation Lake et des granites archéens hyperalumineux de type S du complexe de Treelined Lake qui leurs sont associés, dans la zone de granulite dite de Umfreville – Conifer Lake, de la sous-province de English River, dans le secteur ouest de l'Ontario. Les teneurs en terres rares (TR) de la fluorapatite des pegmatites géochimiquement primitives du complexe de Treelined Lake concordent avec la répartition des TR dans un magma granitique dérivé d'une source dans la croûte. Les spectres des TR de la fluorapatite des pegmatites enrichies de Separation Rapids, d'origine primaire ou métasomatiques (provenant dans ce cas de lentilles à albite + micas), normalisés par rapport à une chondrite, montrent des inflexions importantes à Nd et Er. Ces caractéristiques, anomales pour la fluorapatite, ont été simulées en prenant comme point de départ une source granitique, par exemple le complexe de Treelined Lake. Les inflexions aux éléments Nd et Er seraient dues au fractionnement de la monazite et du grenat, respectivement. La fluorapatite des parois plutôt sodiques de ces massifs de pegmatite contiennent de faibles teneurs en TR et possèdent un spectre normalisé des TR semblable à celui des roches amphibolitiques encaissantes, ce qui nous pousse à proposer une infiltration de phase fluide venant de l'encaissant pendant la formation des pegmatites. La découverte de la fluorapatite d'une poche miarolitique du complexe de Treelined Lake ayant un spectre semblable, avec des inflexions aux mêmes points dans le spectre, semble indiquer un lien génétique entre les pegmatites enrichies de Separation Rapids et les granites hyperalumineux de type S associés au socle granulitique de la sous-province de English River.

(Traduit par la Rédaction)

Mots-clés: fluorapatite, granite de type S, pegmatite enrichie en éléments rares, inflexions, fractionnement, monazite, grenat, ceinture de roches vertes de Separation Lake, sous-province de English River, Ontario.

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INTRODUCTION

The Separation Rapids group of rare-element-enriched (Li, Rb, Cs, Be, Nb and Ta) granitic pegmatites, in the Separation Lake area, northwestern Ontario, represents the eastern limit of the Cat Lake – Winnipeg River pegmatite field, which includes the world-class Tanco deposit of Manitoba (Černý *et al.* 1981, Breaks *et al.* 1996). The pegmatite group includes the 2643 ± 2 Ma (Larbi *et al.* 1996) Separation Rapids pluton (a 4-km^2 S-type, peraluminous, pegmatitic granite), and is spatially associated with the Treelined Lake granitic complex centered in the Umfreville – Conifer Lake granulite zone of the English River Subprovince (Fig. 1; Breaks & Pan 1995, Breaks *et al.* 1996). A granulite-facies metasedimentary source has long been postulated in the literature for Archean fertile parent granites and their rare-element pegmatite haloes (*e.g.*, Breaks & Moore 1992), but is commonly difficult to confirm owing to the lack of definitive evidence in most pegmatite fields. The Separation Lake area therefore provides an opportunity to examine genetic relationships between rare-element pegmatites and S-type peraluminous granites associated with granulite-facies metamorphism.

The primary objective of this study is to document the rare-earth element (*REE*) characteristics of fluorapatite and coexisting minerals in the Separation Rapids rare-element pegmatites and the Treelined Lake granitic complex. The *REE* concentrations of fluorapatite apparently vary systematically from the Treelined Lake granitic complex to the Separation Rapids rare-element pegmatites and provide important insights into the petrogenesis of the S-type granites and the rare-element pegmatites.

GEOLOGICAL SETTING AND THE SEPARATION RAPIDS RARE-ELEMENT PEGMATITES

The Separation Rapids rare-element pegmatite group is situated within the Late Archean Separation Lake greenstone belt at the English River – Winnipeg River subprovince boundary (Fig. 1a). The Separation Lake greenstone belt consists mainly of a sequence of highly deformed and metamorphosed mafic volcanic rocks and subordinate amounts of felsic volcanic and sedimentary rocks (banded iron formations, polymictic conglomerates, greywackes, *etc.*), and has been interpreted to represent an eastward extension of the 2.74 Ga greenstone–granite Bird River Subprovince of Manitoba (Blackburn & Young 1994). All supracrustal rocks of the Separation Lake greenstone belt have been subjected to medium-grade regional metamorphism [500–550°C and 3–4 kbar: Pan *et al.* (1996)].

The adjacent English River Subprovince consists of turbiditic metasedimentary rocks, deposited during the final stage of magmatic activity and tectonic accretion of the greenstone–granite Uchi Subprovince to the north, at about 2720–2710 Ma (Corfu *et al.* 1995). The sedimentary rocks have been intruded by multiple generations of plutonic rocks and subjected to a major regional deformation, metamorphism, and anatexis, which commenced with the emplacement of an extensive peraluminous granitic suite at 2691 Ma (Corfu *et al.* 1995). The English River Subprovince in the Separation Lake area is occupied largely by the Umfreville – Conifer Lake granulite zone, defined by an orthopyroxene-in isograd (Thurston & Breaks 1978, Breaks 1991, Pan *et al.* 1996). The Umfreville – Conifer Lake granulite zone is composed predominantly of granitic and metasedimentary rocks, minor amounts of mafic granulites and rare calc-silicates, and is characterized by signs of extensive partial melting in both metasedimentary rocks and mafic granulites. Metamorphic conditions in the Umfreville – Conifer Lake granulite zone have been estimated at 700–800°C and 5–6 kbar (Pan *et al.* 1996).

The Treelined Lake granitic complex of the English River Subprovince is a large (3–23 km × 63 km) S-type peraluminous granite containing cordierite – orthopyroxene – garnet – biotite (Breaks & Pan 1995, Pan *et al.* 1996; Fig. 1a). It exhibits a systematic variation in both mineralogy and texture from the Umfreville – Conifer Lake granulite zone toward the English River – Separation Lake boundary, *i.e.*, from a "messy" metasedimentary-enclave-rich, orthopyroxene – garnet – biotite granite (minor cordierite) in the northeast to an enclave-sparse, garnet–biotite granite (locally pegmatitic and commonly clotty) in the southwest (Breaks & Pan 1995). Accessory minerals include apatite, monazite and zircon and, locally, rutile, graftonite–sarcopside [$(Fe^{2+},Mn^{2+},Ca)_3(PO_4)_2 - (Fe^{2+},Mn^{2+},Mg)_3(PO_4)_2$] (Černý & Breaks, unpubl. data), gahnite and sillimanite. In addition, biotite-bearing potassic granitic pegmatites are not uncommon in the Treelined Lake granitic complex near the English River – Separation Lake boundary (Breaks *et al.* 1996). Rare-element-enriched muscovite-bearing granitic pegmatites containing local miarolitic cavities also are present within the southern parts of the Treelined Lake granitic complex. These pegmatites contain topaz, cassiterite, manganotantalite, gahnite, tourmaline and fluorapatite (Breaks *et al.* 1996).

The Separation Rapids rare-element pegmatite group belongs to the complex type, petalite subtype, according to the classification of Černý (1990), and has been divided by Breaks & Tindle (1994) into three zones on the basis of geological association and of mineral assemblages: interior beryl zone, exterior beryl zone, and petalite zone (Fig. 1b). The interior beryl zone lies within the parent Separation Rapids pluton and achieves maximum fractionation in local pods of albite ("cleavelandite"), lithian muscovite, quartz, and white beryl, as revealed by sparse blocky K-feldspar with up to 9,800 ppm Rb and 365 ppm Cs, and by accessory mineralogy [wodginite, stibiomicrolite, cassiterite, manganotantalite, fluorapatite and tourmaline; Tindle & Breaks (1996)]. The exterior beryl zone, with green to

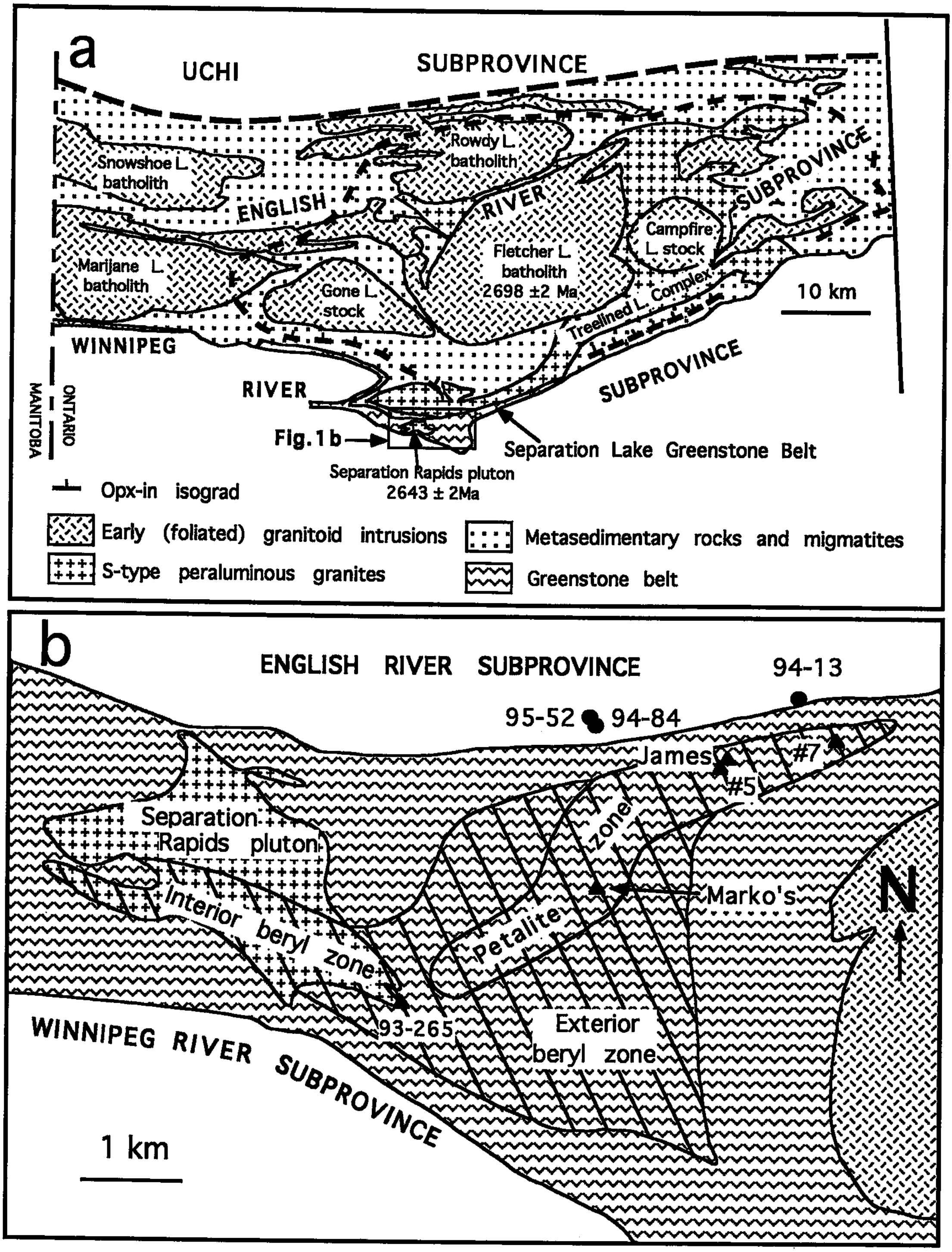
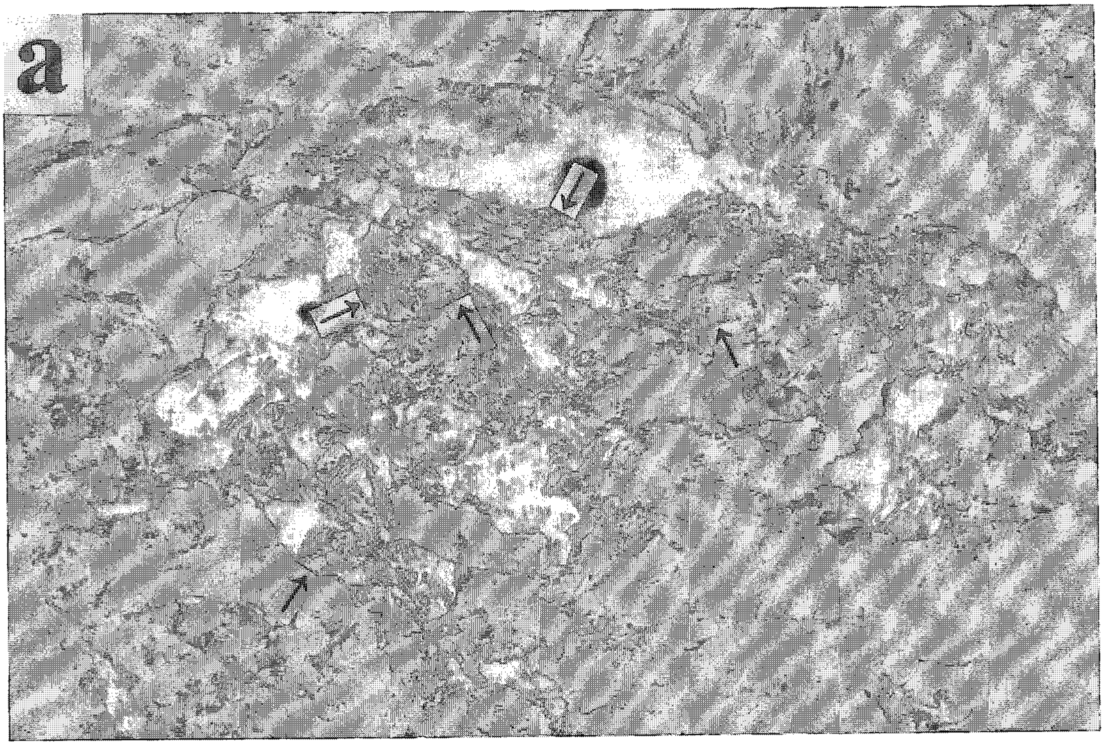


FIG. 1. Geological maps of Separation Lake area illustrating a) the Treelined Lake granitic complex and the Umfreville – Conifer Lake granulite zone in the English River Subprovince, and b) the Separation Rapids rare-element pegmatite group in the Separation Lake greenstone belt. Ear Falls is located to the east of the Umfreville – Conifer Lake granulite zone (Breaks 1991).



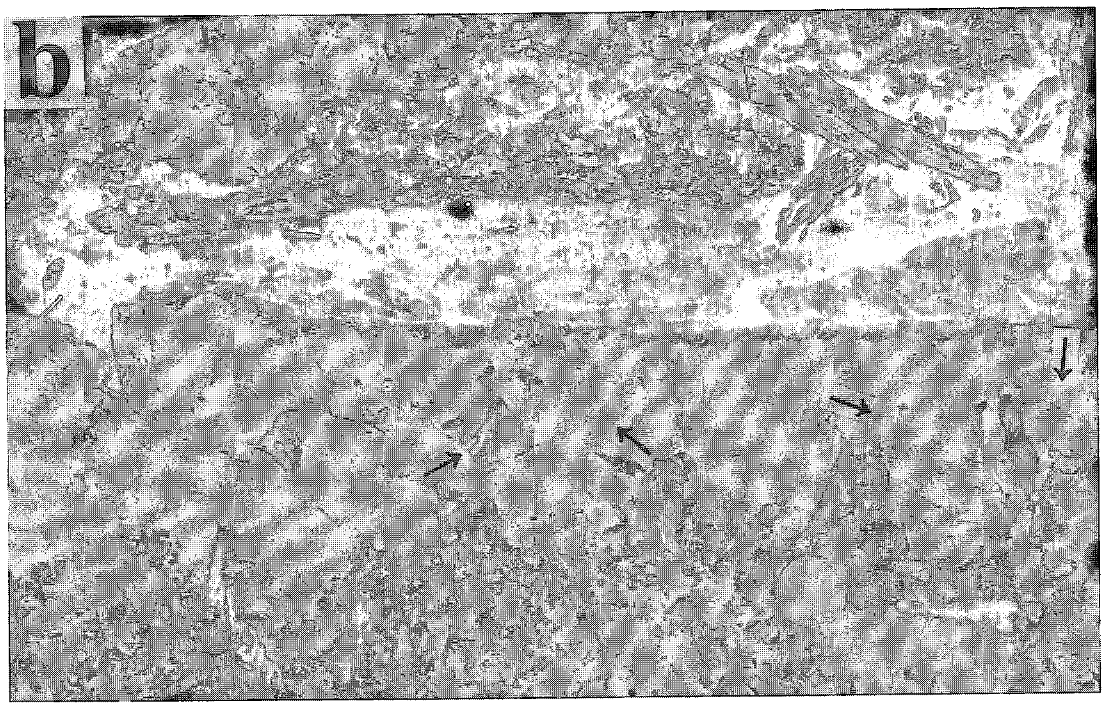


FIG. 2. Photographs illustrating a) abundant apatite (arrows) in an albite–mica pod in the petalite-rich core zone of Pegmatite #5, and b) aggregates of apatite (arrows) in the wall zone of James Pegmatite in contact with mafic metavolcanic rocks. The $1 coin is 2.5 cm in diameter.

white beryl, manganotantalite, ferrocolumbite and manganocolumbite, cassiterite and gahnite, is characterized by anomalously high F contents, indicated by topaz megacrysts in aplite dykes and topaz + fluorite in quartz-rich pegmatite units and secondary topaz + fluorite in biotite-rich metasomatic selvages [bulk F up to 8.5 wt%; Breaks *et al.* (1996)]. The petalite zone comprises eleven bodies of petalite- and wodginite-bearing pegmatite. Accessory minerals include cassiterite, microlite, löllingite, rare primary spodumene, tourmaline, nigerite, scheelite and several rare species of wodginite (Breaks *et al.* 1996).

Internal zonation within individual pegmatites is best displayed by Marko's Pegmatite (Fig. 1b), the largest (8 × 130 m) petalite-bearing pegmatite in the Separation Rapids pegmatite group. Here, a petalite-free, quartz-rich wall zone, which contains the assemblage beryl – muscovite – albite – quartz, envelops a core zone rich in petalite (80–95%) and subordinate blocky K-feldspar and quartz. Pods of radial albite and muscovite aggregates are particularly abundant in the core zones of several petalite-bearing pegmatites, and locally contain abundant fluorapatite (up to 40%; Fig. 2a).

Sample Descriptions and Analytical Procedures

Fourteen samples from the English River Subprovince and the Separation Rapids rare-element pegmatite group have been examined in this study. Details of all samples are given in Table 1. Briefly, the biotite-bearing potassic pegmatites associated with the Treelined Lake granitic complex are geochemically primitive (Breaks *et al.* 1996) and are characterized by abundant apatite (*e.g.*, up to 5 vol.% in sample 94–13). The miarolitic cavities in the Treelined Lake granitic complex contain an assemblage of K-feldspar, quartz, albite, and apatite. At Ear Falls, apatite has been found in concentrations up to 5% in a pegmatitic leucosome and its associated biotite-rich selvage in low-leucosome-fraction layered migmatites, which represent an example of incipient anatexis of metasedimentary rocks in the English River Subprovince (Breaks 1991, Pan *et al.* 1996). In the Separation Rapids rare-element pegmatite group, apatite is present in various primary and replacement units, including the beryl – lithian muscovite – K-feldspar – quartz – albite pods in the parent Separation Rapids pluton. It is particularly abundant in the petalite-bearing pegmatites, where it occurs locally as massive aggregates in the wall zones (Fig. 2b) and attains a maximum abundance in the albite–mica pods of Pegmatite #5 (Fig. 2a).

Chemical compositions of apatite and coexisting minerals were determined by a combination of electron-microprobe analysis and inductively coupled plasma – mass spectrometry techniques. Electron-microprobe analyses were carried out using wavelength-dispersion spectrometry on a JEOL JXA–8600 Superprobe at the University of Saskatchewan. Operating conditions included an accelerating voltage of 15 kV, a beam current

TABLE 1. DETAILS OF APATITE SAMPLES EXAMINED

English River Subprovince (source region)

Number	Location	Description	Mineral association
94-13	Treelined Lake complex	biotite-bearing potassic pegmatite	Qtz, Kfs, Pl, Bt, Grt
94-85	Treelined Lake complex	biotite-bearing potassic pegmatite	Qtz, Kfs, Pl, Bt, Grt
94-84	Treelined Lake complex	miarolitic cavity	Qtz, Mus, Ab
EF-1	Ear Falls	low-leucosome-fraction, layered migmatite	Qtz, Pl, Bt

Separation Rapids rare-element pegmatites

Number	Location	Description	Paragenesis	Mineral association
93-265	Separation Rapids pluton	parent granite	primary	Kfs, Qtz, Mus, beryl, Ab
95-117b	Pegmatite #5	albite-rich wall zone	primary	Ab, Mus
94-310	Pegmatite #7	albite-rich wall zone	primary	Ab
P7-1	Pegmatite #7	quartz-rich wall zone	primary	Qtz, Mus, beryl, Ab
JP-1	James Pegmatite	quartz-rich wall zone	primary	Qtz, Mus, beryl, Ab
95-109a	Marko's Pegmatite	albite-mica pod	replacement	Ab, Qtz, Kfs, Mus
94-44r	Marko's Pegmatite	albite-mica pod	replacement	Ab, Qtz, Mus
MP-1	Marko's Pegmatite	albite-mica pod	replacement	Ab, Qtz, Mus
95-109e	Marko's Pegmatite	albite-mica pod	replacement	Ab, Mus
94-80g	Pegmatite #5	in petalite megacrysts	replacement	petalite, Grt, Mus, Ab

Mineral abbreviations after Kretz (1983).

of 10 nA, a 2- to 5-μm beam, and 30 s counting time; mineral standards were used. Apatite and the coexisting feldspars and micas were separated by hand-picking from a 60–70 mesh (0.0212–0.025 cm) fraction under a binocular, and were subsequently checked by back-scattered electron imaging. Analyses of apatite, feldspars and micas for the rare-earth elements were performed on a Perkin–Elmer Sciex® Elan 5000 inductively coupled plasma – mass spectrometer (ICP–MS) at the University of Saskatchewan, according to the standard protocol of sample preparation of Xie *et al.* (1994) and solution-ICP–MS procedure of Jenner *et al.* (1990). All feldspar and mica separates and most of the apatite separates were dissolved readily in $HF–HNO_3$ solutions within 48 hours, except for several separates of Mn-rich apatite, which required repeated $HF–HNO_3$ and additional perchlorate treatments to ensure complete dissolution. Two samples of fluorapatite from Durango, Mexico [one from the reference collection of the Department of Geological Sciences, University of Saskatchewan, and the other used previously as a standard for *REE* analysis in apatite by secondary-ion mass spectrometry: Pan *et al.* (1993)], were also included in this ICP–MS study. Our ICP–MS results of the Durango fluorapatite are in excellent agreement with data in the literature (Table 2). Difficulties were also encountered during the dissolution of one of the Durango apatite samples, but this apparently did not affect the *REE* values (Table 2).

RESULTS

All samples of apatite examined in this study are homogeneous with respect to major and minor elements, as revealed by electron-microprobe analysis. They invariably consist of fluorapatite (Table 2). In particular, cerium in several samples of apatite occurs at levels detectable with an electron microprobe, and does not exhibit any significant zonation. Also, exsolution lamellae or inclusions of *REE*-rich minerals (Pan *et al.* 1993) were not observed in any grains of apatite examined in this study.

English River Subprovince and the Treelined Lake granitic complex

Fluorapatite in the two bodies of geochemically primitive pegmatite has variable F contents (2.5–3.78 wt% F), and a low Mn content (0.47–0.57 wt% MnO; Table 2); it is characterized by a convex-upward chondrite-normalized *REE* pattern with a negative Eu anomaly (Eu_N/Eu_N^* = 0.06; Fig. 3a). Fluorapatite in the miarolitic cavity (94–84) of the Treelined Lake granitic complex has 3.41 wt% F and 4.54 wt% MnO, and is characterized by a chondrite-normalized *REE* pattern with prominent discontinuities at Nd and Er, and a negative Eu anomaly (Fig. 3a). Fluorapatite in the low-leucosome-fraction layered migmatites at Ear Falls

TABLE 2. COMPOSITIONS OF FLUORAPATITE FROM THE SEPARATION LAKE AREA

	English River Subprovince				Separation Rapids rare-element pegmatite group										Durango, Mexico		
Sample	94-13	94-85	94-84	EF-1	93-265	95-117b	94-310	P7-1	JP-1	95-109a	94-44r	MP-1	95-109e	94-80g	1	2	3
P_2O_5 (wt%)	42.4	42.2	42.9	41.2	42.3	40.9	41.5	41.4	41.2	41.7	41.9	40.9	41.4	41.8	40.6(0.35)		40.78
SiO_2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.30(0.08)		0.34
Fe_2O_3	0.12	0.46	0.15	0.12	0.06	0.11	0.06	0.16	0.20	0.05	0.04	0.11	0.07	0.04	0.05(0.02)		0.06
MnO	0.57	0.47	4.54	0.10	4.61	1.69	1.42	1.10	2.33	4.73	3.89	3.93	5.68	0.78	0.01(0.01)		0.01
CaO	54.5	54.3	48.8	56.3	51.3	54.3	54.1	54.5	54.0	49.9	51.8	52.0	49.3	55.0	54.3(0.26)		54.02
Na_2O	0.12	0.12	0.01	0	0.02	0	0	0.0	0.02	0.16	0.08	0.07	0.13	0	0.25(0.04)		0.23
F	3.78	2.50	3.41	3.42	3.01	3.33	3.79	3.52	3.24	3.63	3.45	3.24	3.19	3.18	3.50(0.12)		3.53
Cl	0.02	0.09	0	0	0	0	0	0	0.01	0	0	0	0	0	0.46(0.08)		0.41
O≡F,Cl	1.71	1.15	1.54	1.44	1.36	1.50	1.71	1.59	1.46	1.64	1.55	1.46	1.44	1.43	1.57		1.58
Total	99.8	99.0	98.3	99.7	99.9	98.8	99.2	99.1	99.5	98.5	99.3	98.8	98.3	99.4	97.9		99.94
La (ppm)	257	153	86	89	148	19	29.5	36	128	89	207	152	142	4.86	3287	2929	3213
Ce	718	502	240	267	370	44	69	99	532	374	811	704	687	7.97	3982	3884	4183
Pr	119	78	27	42	40	5.29	9.09	13.5	81	56	121	115	112	0.77	350	350	
Nd	643	438	93	229	87	27	43	52	267	236	452	465	512	2.76	1185	1154	1185
Sm	232	161	107	82	73	7.85	14.9	61	374	286	518	564	1175	0.81	130	146	133
Eu	5.46	3.54	1.57	9.02	0.98	4.75	6.5	4.9	1.39	1.82	4.23	3.77	4.40	3.53	16	16	14.9
Gd	292	190	110	99	57	9.75	18.9	60.	462	328	585	605	1377	0.89	117	131	
Tb	60	41	29	19	19	1.73	3.47	13.6	111	76	133	129	321	0.16	12	13	
Dy	408	297	116	129	95	11.2	23.7	56	426	275	527	520	883	1.06	67	77	75.7
Ho	75	60	10	23	7.8	2.44	4.76	5.1	27	23	43	41	51	0.23	13	14	
Er	197	175	20	54	17	7.03	14.1	11.8	45	42	83	79	72	0.67	35	39	
Tm	24	24	3.81	6.25	3.52	1.11	2.4	2.2	6.5	6.32	13	11.6	9.56	0.13	4.66	5.14	
Yb	131	21	32	30	28	7.91	16.8	20	40	41	77	68	55	1.01	26	27	34.7
Lu	15	17	3.73	3.49	2.53	1.18	2.58	2.6	3.5	4.22	7.3	6.7	4.72	0.16	3.43	3.64	3.6
La_N/Yb_N	1.33	4.92	1.82	2.0	3.57	1.62	1.19	1.22	2.16	1.47	1.82	1.51	1.74	3.25			
Eu_N/Eu_N^*	0.06	0.06	0.04	0.35	0.05	1.27	1.19	0.25	0.01	0.02	0.02	0.02	0.01	12.7			

wt.%, weight percent; ppm, parts per million; La_N/Yb_N, chondrite-normalized La/Yb value; $Eu_N^* = (Sm_N \times Gd_N)^{0.5}$. Data on the Durango fluorapatite are included for comparison.; data on major elements in column 1 represent an average result of 10 electron-microprobe analyses; REE concentrations in columns 1 and 2 were obtained by dissolution ICP-MS as part of this study; the data on major elements in column 3 are taken from Jarosewich et al. (1980), whereas the data on the REE are taken from Roelandts (1988).

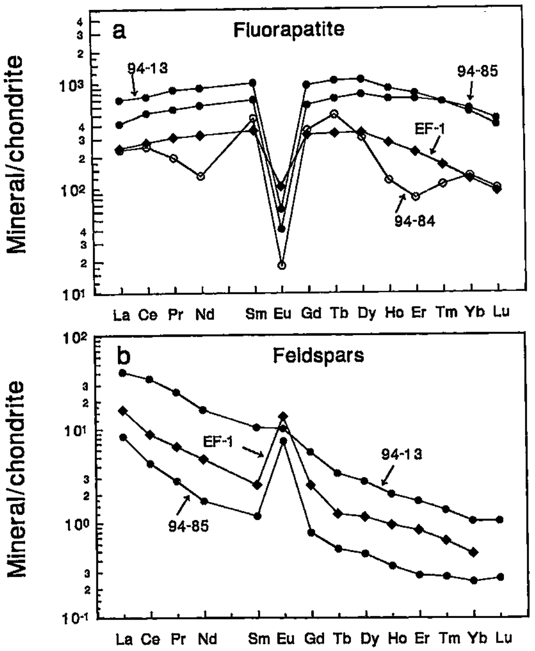


FIG. 3. Chondrite-normalized *REE* patterns of minerals from the English River Subprovince: a) fluorapatite; b) feldspars.

contains 3.42 wt% F and 0.1% MnO, and is also characterized by a convex-upward chondrite-normalized *REE* pattern with a negative Eu anomaly (Fig. 3a).

The K-feldspar in the geochemically primitive pegmatites contains less than 570 ppm Rb and 9 ppm Cs (Table 3), and is characterized by smooth chondrite-normalized *REE* patterns with a positive Eu anomaly ($1.33 < Eu_N/Eu_N^* < 7.69$; Fig. 3b). The K-feldspar in contact with the miarolitic cavity is characterized by high concentrations of Rb and Cs (Table 3). Plagioclase in sample EF–1 is oligoclase (An_{21}; Table 3) and is characterized by a smooth chondrite-normalized *REE* pattern with a positive Eu anomaly ($Eu_N/Eu_N^* = 5.4$; Fig. 3b). Biotite in sample EF–1 contains only 403 ppm Rb and 25 ppm Cs (Table 3). Its high *REE* content is related to difficulties in separating pure biotite owing to its intergrowth with very fine-grained fluorapatite.

Separation Rapids rare-element pegmatite group

Fluorapatite in the Separation Rapids pegmatite group varies significantly in Mn content from 0.78 to 5.68 wt.% MnO (Table 2). There is also an apparent correlation between the MnO content and the *REE* abundance in fluorapatite, which attains maximum values in the albite–mica pods of the petalite-bearing pegmatites (Table 2). The most striking feature of fluorapatite in both primary and replacement units of the Separation Rapids rare-earth-enriched pegmatites is the presence of a marked discontinuity at Nd and Er in chondrite-normalized *REE* patterns (Fig. 4), which are also characterized by a pronounced negative Eu anomaly ($0.01 < Eu_N/Eu_N^* < 0.25$; Table 2). Fluorapatite in the albite-rich wall zones of the petalite-bearing pegmatites (94–310, 95–117b), however, is clearly distinguished by its low Mn and *REE* contents (<1.69 wt.% MnO, <69 ppm Ce; Table 2). It is also characterized by flat chondrite-normalized *REE* patterns ($1.2 < La_N/Yb_N < 3.3$), with a positive Eu anomaly ($1.19 < Eu_N/Eu_N^* < 1.27$; Fig. 4b), similar in shape to those of the whole-rock samples of the host amphibolites (*i.e.*, mafic metavolcanic rocks; Fig. 4b). The fluorapatite found as inclusions in petalite megacrysts (94–80g) is unusual in that it has the lowest Mn and *REE* contents (Table 2) and is characterized by a chondrite-normalized *REE* pattern with a pronounced positive Eu anomaly (Fig. 4c).

The K-feldspar in the Separation Rapids rare-element pegmatite group is characterized by high Rb and Cs contents [up to 29,000 ppm Rb and 1050 ppm Cs in the petalite zone; Breaks *et al.* (1996)], which approach values in the extremely fractionated pegmatites found at Cross Lake, Manitoba (Černý *et al.* 1985). Albite coexisting with fluorapatite in the albite–mica pods is much poorer in Rb and Cs than the K-feldspar (Table 3). The chondrite-normalized *REE* patterns of feldspars, despite exceedingly low absolute abundances of the *REE*, mimic those of fluorapatite, with discontinuities at Nd and Er, and a negative Eu anomaly (Fig. 4d).

Mica flakes in the albite–mica pods of the Separation Rapids pegmatites vary from muscovite to lithian muscovite and are characterized by high F contents (up to 8 wt.% F; Table 3). They also contain elevated contents of Li (up to 7,600 ppm), Rb (24,000 ppm) and Cs (1700 ppm; Table 3). ICP–MS analyses show that micas in the albite– mica pods of the Separation Rapids rare-element pegmatites have very low contents of *REE* (Table 3).

DISCUSSION

In the present study, we show a systematic variation of *REE* content of fluorapatite from the Separation Rapids rare-element pegmatite group and its spatially associated Treelined Lake granitic complex. In particular, we document unusual *REE* characteristics (*i.e.*, the marked discontinuities at Nd and Er) of fluorapatite from various primary and replacement units of the Separation Rapids pegmatites and in the miarolitic cavity of the Treelined Lake granitic complex.

Cerium and Eu anomalies in chondrite-normalized *REE* patterns are not uncommon in mineral or whole-rock samples. They are attributable to net removal in the form of Ce^{4+} and Eu^{2+}. However, other rare-earth elements, including Nd and Er, are not known to exhibit fractionation from their neighboring elements during geological processes. Fleet & Pan (1995a, b) demonstrated experimentally that the uptake of *REE* in fluorapatite is controlled largely by differences between ionic radii of individual *REE* and the sizes of the Ca sites in this mineral, whereas other factors, such as crystal-field stabilization energy, are of minor importance. Therefore, fluorapatite itself is incapable of fractionating Nd or Er from the neighboring rare-earth elements during its crystallization. Indeed, the chondrite-normalized *REE* patterns of fluorapatite from various geological environments in the literature are smooth, without any discontinuities other than the Ce and Eu anomalies (*e.g.*, Fleischer & Altschuler 1986, Pan *et al.* 1993). The only exception is fluorapatite from rare-element pegmatites of Black Hills, South Dakota, which is also characterized by discontinuities at Nd and Er (Walker *et al.* 1986, Jolliff *et al.* 1989). It is also noteworthy that the *REE* contents in fluorapatite of the Black Hills rare-element pegmatites were determined by two independent methods [isotope dilution: Walker *et al.* (1986); secondary-ion mass spectrometry: Jolliff *et al.* (1989)]. Therefore, the observed discontinuities at Nd and Er in these rare-element pegmatites are not analytical artifacts. Moreover, this exclusive association suggests that the discontinuities at Nd and Er in fluorapatite must reflect the petrogenesis of the host rare-element pegmatites and parent granites (see below).

Treelined Lake granitic complex

A detailed investigation of the petrogenesis of the Treelined Lake granitic complex is currently under way. Preliminary results from field observations and mineralogical and geochemical analyses (including whole-rock *REE* characteristics) suggest an origin involving mainly partial melting of the clastic sedimentary rocks of the English River Subprovince during granulite-facies metamorphism (*cf.* Vielzeuf *et al.* 1990, Icenhower & London 1995), with input from the mantle (Pan *et al.* 1996). This is supported by preliminary Sm–Nd isotopic results of Larbi *et al.* (1996; ε_{Nd} values from –2 to +1), indicative of mixed sources of crustal materials and juvenile components. The *REE* characteristics of fluorapatite in the geochemically primitive pegmatites of the Treelined Lake granitic complex are similar to those of

TABLE 3. COMPOSITIONS OF FELDSPARS AND MICAS FROM THE SEPARATION LAKE AREA

	Feldspars							Micas					
Sample	94-13	94-85	94-84	EF-1	93-265	P7-1	MP-1	EF-1	93-265	93-265	94-310	P7-1	JP-1
SiO_2 (wt%)	64.8	65.6	63.1	63.1	69.1	68.8	68.1	36.4	46.3	49.4	45.1	45.1	45.9
P_2O_5	0	0.18	0	0.10	0	0.02	0	nd	nd	nd	nd	nd	nd
TiO2	nd	nd	nd	nd	nd	nd	nd	1.73	0	0	0.42	0.09	0.07
Al_2O_3	18.1	18.1	18.1	22.3	18.9	19.4	19.9	17.7	34.8	22.8	32.2	32.8	33.8
Fe_2O_3	0.01	0	0.09	0.02	0	0.05	0.01	19.2	1.57	4.75	3.95	4.44	3.60
MgO	nd	nd	nd	nd	nd	nd	nd	11.1	0.0	0.05	1.75	1.30	0.74
MnO	nd	nd	nd	nd	nd	nd	nd	0.17	0.44	1.64	0.07	0.13	0.21
CaO	0	0.05	0	4.44	0.04	0.64	0.29	0	0	0	0.04	0.01	0
K_2O	16.1	14.3	16.1	8.67	0.07	10.9	0.07	9.20	10.5	9.1	8.88	9.41	8.25
Na_2O	0.50	1.87	0.23	0.30	11.3	0.04	10.9	0.10	0.33	0.1	0.23	0.18	0.20
F	nd	nd	nd	nd	nd	nd	nd	0.48	1.82	8.05	0.81	0.25	0.68
Cl	nd	nd	nd	nd	nd	nd	nd	0	0.0	0.0	0.0	0.02	0.0
O≡F,Cl								0.20	0.77	3.39	0.34	0.11	0.29
Total*	99.5	100.1	98.6	99.2	99.4	99.8	99.3	96.1	95.9	95.5	95.2	96.2	96.6
Li(ppm)	0	7.4	39	2.96	46	31	18	134	88	7600	3065	2330	4345
Rb	570	285	7340	83	570	236	71	403	7980	12900	13800	19382	23960
Sr	83	230	13	712	12	34	6	4.63	21	12	375	420	66
Cs	8.9	2.98	1170	1.48	1040	4.69	238	25	201	1164	705	1709	890
La	15	3.09	0.11	6	0.14	0.48	0.05	19	0.12	0.33	0.14	2.25	0.18
Ce	34	4.13	0.15	8.65	0.34	1.09	0.1	38	0.14	0.62	0.32	8.42	0.25
Pr	3.45	0.38	0.01	0.89	0.04	0.16	0.01	3.83	0.03	0.07	0.07	1.2	0.05
Nd	12	1.21	0.02	3.4	0.19	0.59	0	14	0.06	0.16	0.22	4.39	0.14
Sm	2.41	0.27	0.01	0.58	0.12	0.47	0	2.29	0.05	0	0.06	2.84	0
Eu	0.88	0.64	0	1.18	0	0.07	0	0.23	0	0	0	0	0
Gd	1.71	0.24	0	0.76	0.19	0.44	0	2.01	0.03	0	0.04	1.45	0.08
Tb	0.19	0.03	0	0.07	0.04	0.06	0	0.3	0.01	0	0.01	0.21	0.01
Dy	1.04	0.18	0	0.44	0.21	0.22	0.02	2.06	0.02	0.02	0.07	0.66	0.35
Ho	0.17	0.03	0	0.08	0.03	0.02	0	0.38	0	0	0.01	0.04	0.1
Er	0.42	0.07	0	0.21	0.1	0.04	0	1.1	0	0.02	0.05	0.09	0.33
Tm	0.05	0.01	0	0.04	0.01	0.01	0	0.15	0	0	0.02	0.03	0.07
Yb	0.26	0.06	0	0.12	0.11	0.06	0	0.89	0	0.02	0.03	0.08	0.48
Lu	0.04	0.01	0	0	0.01	0	0	0.13	0	0	0	0	0.06
La_N/Yb_N	39	35		34	0.86	5.4		14.4		11.0	3.83	19	0.25
Eu_N/Eu_N*	1.33	7.69		5.43		0.47		0.33					

nd, not detected; *, total includes Li_2O and Rb_2O by ICP-MS.

fluorapatite from the low-leucosome-fraction layered migmatites at Ear Falls (Fig. 3a), and are indicative of crystallization from granitic melts that have undergone limited degree of differentiation.

Separation Rapids rare-element pegmatites

The rare-element-enriched albite–mica pods in granitic pegmatites are generally considered to represent the latest-stage, but primary, crystallization from a residual melt (London *et al.* 1989, Černý 1990, London 1992), although some of the albite–mica assemblages may also form metasomatically as a replacement of K-feldspar or lithium aluminosilicates (London 1992). London *et al.* (1988) demonstrated experimentally that many elements, including Li, Rb, Cs and the *REE*, either show no preference for vapor over melt or are strongly partitioned into the melt. Moreover, there is only very limited fractionation among the *REE* during the separation of a vapor phase from the silicate melt (London *et al.* 1988). Therefore, metasomatic processes, even if they contributed to the formation of some of the albite–mica pods, cannot be directly responsible for the unusual *REE* characteristics of the fluorapatite documented in this study. The restrictive occurrences of fluorapatite in assemblages of the petalite-bearing pegmatites and the chemically most evolved part of the Separation Rapids pluton suggest a genetic link with extremely differentiated, residual melts. In the following section, we present a two-stage model based on fractional crystallization of a granitic melt represented by the Treelined Lake complex to explain the unusual *REE* characteristics of the fluorapatite in the Separation Rapids rare-element pegmatites.

The first stage of fractional crystallization corresponds largely to that of the Treelined Lake granitic complex. Following the study of Yurimoto *et al.* (1990), we have used the Rayleigh fractionation model and taken sample 95–52 (representative of the Treelined Lake granitic complex; Pan *et al.* 1996) as the composition of the parent melt (Table 4). The bulk distribution-coefficients of *REE* (*D*) were calculated on the basis of published mineral/melt partition coefficients and the average modal abundances of minerals, estimated from point counting of ten samples of the Treelined Lake granitic complex (Table 4). The minor amount of biotite and muscovite, which are present in all samples examined but do not

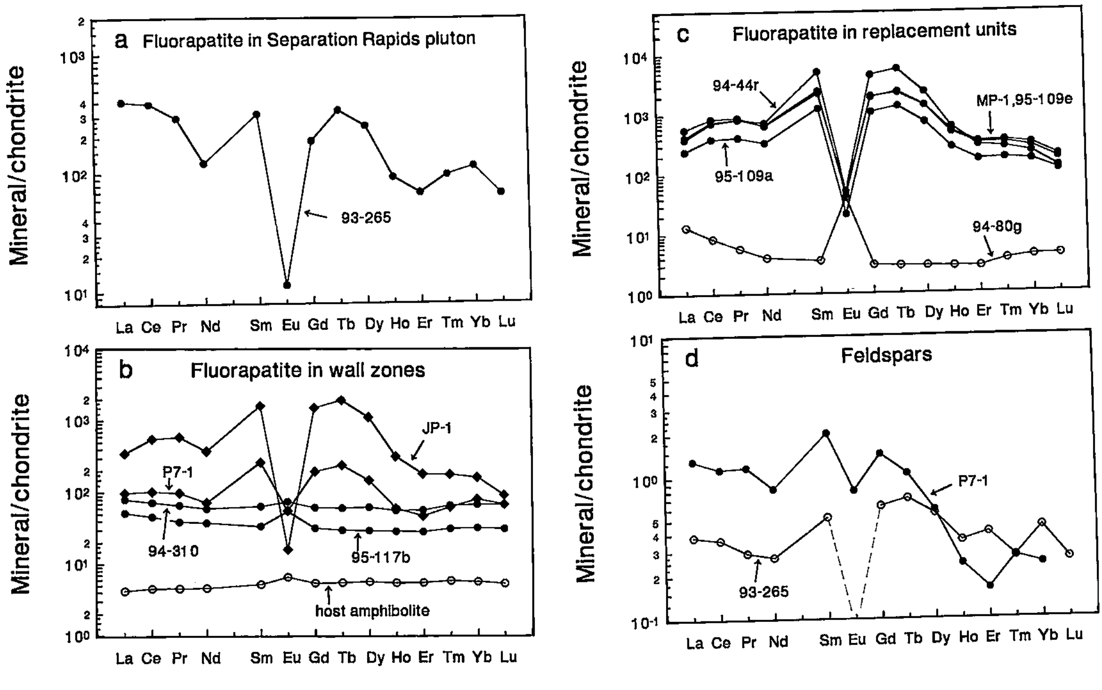


FIG. 4. Chondrite-normalized *REE* patterns of fluorapatite and coexisting minerals of the Separation Rapids rare-element pegmatite group. a) Fluorapatite in the Separation Rapids pluton (93–265). b) Fluorapatite in the primary wall-zones of the petalite-bearing pegmatites. Symbols: solid diamonds represent fluorapatite in the quartz-rich variety, solid circles, fluorapatite in albite-rich variety; a whole-rock sample of the host amphibolite (open circles) is shown for comparison. c) Fluorapatite in replacement units of the petalite-bearing pegmatites. Symbols: solid circles represent fluorapatite in albite–mica pods, and open circles, fluorapatite in petalite megacrysts (94–80g). d) The feldspars (93–265: albite, P7–1: K-feldspar).

affect the calculated *D* in any measurable way, was not included. Figure 5a shows that the calculated chondrite-normalized *REE* patterns of residual melts at high degrees of fractional crystallization do exhibit marked discontinuities at Nd and Er, which are clearly attributable to the fractional crystallization of monazite and garnet, respectively (Table 4). Another important feature is that the high degrees of fractional crystallization of monazite, garnet and other *REE*-rich minerals lead to a depletion of *REE* in the residual melts (Fig. 5a), consistent with the exceedingly low *REE* contents in many complex-type rare-element pegmatites (Černý 1990). On the other hand, melts of such low *REE* contents are obviously incapable of crystallizing the abundant *REE*-rich fluorapatite directly. Therefore, a second stage of fractional crystallization is necessary to explain the *REE* abundance in the fluorapatite of this study.

The second stage of fractional crystallization corresponds largely to that of the Separation Rapids pluton and its halo of rare-element pegmatites, involving predominantly quartz, plagioclase and K-feldspar without garnet or *REE*-rich minerals. Again, the Rayleigh fractionation model was used, and the calculated residual melt from the first stage, at 80% of fractional crystallization, was taken as the initial melt here. It should be pointed out, however, that quartz, plagioclase and K-feldspar are the predominant minerals but do not crystallize simultaneously in the Separation Rapids pegmatites. Therefore, our calculations model the combined effect of a sequential crystallization of these three minerals. We assumed that quartz, plagioclase and K-feldspar crystallized in equal proportions, although any other proportions of these minerals would not significantly alter the outcome (Fig. 5b). Also, monazite remains as a trace phase locally in the Separation Rapids pluton (but is exceedingly rare), and garnet persists even in the petalite-bearing pegmatites, but is restricted to the wall zones. However, the very low abundances of these two minerals make them insignificant in the second stage of our model calculations. Figure 5b shows that the hypothetical *REE* patterns of fluorapatite based on partitioning of *REE* between this mineral and the calculated residual melts from the second stage of fractional crystallization are in good agreement with those observed in the Separation Rapids rare-element pegmatites (Fig. 4), although Eu is excluded in our calculations for clarity. The increase in the magnitude of negative Eu anomaly in fluorapatite from the Treelined Lake granitic complex to the Separation Rapids pegmatites (Table 2) is similar to that observed between other rare-element pegmatites and their parent granites, and has been interpreted to relate to the crystallization of plagioclase (*cf.* Černý 1990).

One assumption underlying the above model calculation is that the partition coefficients of *REE* between minerals and coexisting melts are constant during the evolution of the granitic melts. Experimental studies (*e.g.*, Montel 1993), however, have shown that these coefficients vary with melt composition, temperature, and other physicochemical conditions. Therefore, this assumption is almost certainly not true but necessary, given the uncertainty of how the coefficients might change with the various physicochemical parameters.

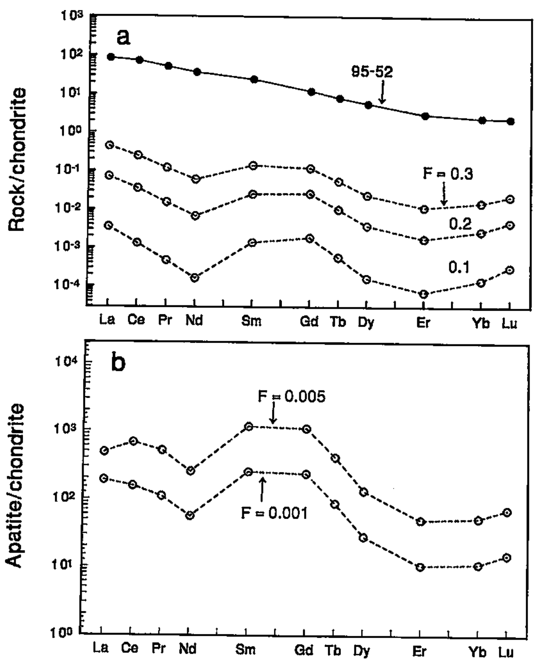


FIG. 5. Calculated *REE* patterns for a) residual melts after 70%, 80% and 90% crystallization from a granitic melt of the Treelined Lake complex (*i.e.*, 95–52), and b) apatite from a residual melt of the second-stage crystallization at 99.5% and 99.9%. The residual melt after 80% crystallization in the first-stage calculation was taken as the initial melt here.

TABLE 4. MINERAL/MELT DISTRIBUTION COEFFICIENTS USED IN MODELING

Mineral Mode	Quartz[1] 36%	Pl[1] 21%	Kfs[1] 28%	Garnet[2] 10%	Apatite[2] 0.5%	Mon[3] 0.16%	Zircon[4] 0.2%	D	Granite (95-52)	Garnet[5]
La	0.014	0.32	0.07	0.20	27	3200	5.0	5.38	31(32.9)	0.001
Ce	0.006	0.21	0.04	0.35	34.7	3413	5.4	5.74	69(73)	0.01
Pr	0.007	0.17	0.03	0.44	45.9	3569	6.0	6.04	7.04(7.6)	
Nd	0.009	0.14	0.03	0.53	57.1	3726	6.6	6.35	26(27.9)	0.4
Sm	0.008	0.11	0.02	2.66	62.8	2859	11.0	5.21	5.15(5.43)	6.4
Gd	0.007	0.1	0.013	10.5	56.3	2144	21.7	4.82	3.73(3.78)	
Tb	0.007	0.09	0.01	17.5	53	1900	37	5.16	0.47(0.52)	
Dy	0.010	0.07	0.05	28.6	48	1429	68.3	5.54	2.18(2.41)	116
Er	0.011	0.07	0.04	42.8	37	595	101	5.65	0.77(0.88)	170
Yb	0.012	0.06	0.03	39.9	23.9	273	279	5.24	0.62(0.65)	140
Lu	0.020	0.06	0.04	29.6	20.2	174	750	4.88	0.09(0.09)	63

Sources of data on distribution coefficients: 1 Nash & Crecraft (1985), 2 Arth (1976), 3 Charoy (1986), 4 Mahood & Hildreth (1983), 5 Sisson & Bacon (1992). Mode: average of 10 samples from the Treelined Lake granitic complex; a minor amount of biotite (<5%) also is present, but is not included in the calculation. Sample 95-52 of the Treelined Lake granitic complex was analyzed by both dissolution-ICP-MS and sinter-ICP-MS (values in parentheses: Pan *et al.* 1996).

The discontinuities at Nd and Er in our model calculation are obviously not sensitive to the absolute values of the partition coefficients of individual *REE*, but rather are the result of 1) a strong preference of monazite for the light rare-earths (*LREE*), and 2) the partition coefficients of *REE* between garnet and melt peaking at Er. Monazite in all granites and granitic pegmatites is invariably strongly enriched in *LREE* relative to the heavy rare-earths (*HREE*) (*e.g.*, Charoy 1986, Demartin *et al.* 1991, Walker & Miller 1993, Bea 1996). Available data show that the partition coefficients of *REE* between garnet and siliceous melt peak at Er (Arth 1976, Sisson & Bacon 1992).

Numerous studies have emphasized the importance of accessory minerals (monazite, garnet, xenotime, zircon, among others) in influencing the *REE* characteristics of granitic pegmatites during both the partial melting and crystallization stages (*e.g.*, Yurimoto *et al.* 1990, Pichavant *et al.* 1992, Mulja *et al.* 1995, Nabelek & Glascock 1995, Wolf & London 1995). Yurimoto *et al.* (1990) were the first to relate the Nd discontinuity in whole-rock chondrite-normalized *REE* patterns of granitic pegmatites to fractionation of monazite. In the present study, we show that the marked discontinuity at Nd in fluorapatite of the Separation Rapids rare-element pegmatites also is related to fractionation of monazite. In addition, we document the presence of a marked discontinuity at Er (Fig. 4) and link it to the fractionation of garnet (Fig. 5, Table 4). Masuda *et al.* (1987) documented discontinuities at Nd and Er in a chondrite-normalized *REE* pattern of a leucogranite from Jianxi, China, and attributed these to the "tetrad" effects. It is noteworthy that a discontinuity at Er also is present (but not explained) in the whole-rock chondrite-normalized *REE* patterns of some other rare-element granitic pegmatites in the literature (*e.g.*, Yurimoto *et al.* 1990, Breaks & Moore 1992, Mulja *et al.* 1995). These granitic pegmatites are also associated with garnet-bearing granites (Breaks & Moore 1992, Mulja *et al.* 1995) Therefore, the fractionation of garnet may be responsible for the discontinuity at Er in these granitic pegmatites as well. Similarly, fractionation of garnet may be responsible for the Er discontinuity in fluorapatite from rare-element pegmatites of Black Hills, South Dakota (Walker *et al.* 1986, Jolliff *et al.* 1989), where garnet is a characteristic phase in both granites and pegmatites (*e.g.*, Norton & Redden 1990, Duke *et al.* 1992). Moreover, the very high abundance of garnet (up to 35%) in the Treelined Lake complex is clearly unusual and may not be required for the Er discontinuity. If the more recent results of Sisson & Bacon (1992) are used (Table 4), high degrees of fractional crystallization of typical granitic melts involving only 2–3% garnet will produce significant discontinuities at Er in residual melts.

The origin of sodic wall-zones in granitic pegmatites remains unclear (London 1992). Several lines of evidence [*e.g.*, stable isotope systematics: Taylor *et al.* (1979); fluid inclusion studies: Thomas & Spooner (1988); mineralogy: London (1992)] suggest that infiltration of fluid along with other components from the host lithologies occurred during the formation of wall zones in granitic pegmatites. Therefore, the similarity in the chondrite-normalized *REE* patterns of fluorapatite in the sodic wall-zones to those of the host amphibolites (Fig. 4b) is most likely related to an introduction of *REE* (along with Ca) by fluids infiltrating from country rocks into pegmatites, providing evidence for the mobility of *REE* during the wall-rock interactions associated with the formation of pegmatites. Another important contributing factor to the *REE* characteristics of fluorapatite in the sodic wall-zones is the exceedingly low *REE* content of the rare-element pegmatites (*cf.* Černý 1990).

Miarolitic cavity of the Treelined Lake granitic complex

Fluorapatite in the miarolitic cavity of the Treelined Lake granitic complex most likely crystallized directly from a fluid phase. Again, the partition coefficients of *REE* between silicate melt and fluid (London *et al.* 1988) dictate that saturation of a fluid phase is not a major factor in determining the unusual *REE* characteristics of fluorapatite here. The high contents of Rb and Cs in K-feldspar of the miarolitic cavity (Table 3) indicate an extreme degree of fractionation (*cf.* Černý *et al.* 1985), similar to that in the Separation Rapids rare-element pegmatite group. Therefore, the unusual *REE* characteristics (*i.e.*, discontinuities at Nd and Er) of fluorapatite in the miarolitic cavity of the Treelined Lake granitic complex may also be related to fractionation of monazite and garnet, and provide geochemical evidence for a genetic link between the Separation Rapids rare-element pegmatites in the Separation Lake greenstone belt and the S-type peraluminous granites associated with granulite-facies metamorphism in the English River Subprovince.

SUMMARY

1) We have documented and interpreted the unusual *REE* characteristics of fluorapatite and coexisting minerals in the Separation Rapids rare-element pegmatite group of the Separation Lake greenstone belt and the associated S-type peraluminous granites of the English River subprovince.

2) The most striking feature of fluorapatite in both the primary and replacement units of the Separation Rapids rare-element pegmatites is the presence of marked discontinuities at Nd and Er in chondrite-normalized *REE* patterns. Model calculations show that these discontinuities are related to fractionation of monazite and garnet, respectively.

3) The *REE* characteristics of fluorapatite in the sodic wall-zones of the Separation Rapids rare-element pegmatites are similar to those of the host amphibolites, supporting fluid infiltration from country rocks during the formation of the pegmatites.

4) The presence of similar and unusual *REE* characteristics in fluorapatite from a miarolitic cavity in the Treelined Lake granitic complex provides geochemical evidence for genetic links between rare-element pegmatites in the Separation Lake greenstone belt and the S-type peraluminous granites associated with granulite-facies metamorphism in the English River Subprovince.

ACKNOWLEDGEMENTS

We thank J.-M. Montel and an anonymous referee for constructive criticisms, R.F. Martin for editorial assistance, M.E. Fleet and N.D. MacRae of the University of Western Ontario for provision of the well-characterized fluorapatite from Durango, Mexico, T. Bonli, J. Fan, J. Jain and B. Morgan of the University of Saskatchewan for analytical assistance, and NSERC for financial support. This is Lithoprobe contribution number 848.

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Received March 4, 1996, revised manuscript accepted January 2, 1997.