GRAFTONITE – BEUSITE IN GRANITIC PEGMATITES OF THE SUPERIOR PROVINCE: A STUDY IN CONTRASTS

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ABSTRACT

Graftonite – beusite occurs in only three localities of granitic pegmatites in the Superior Province of the Canadian Shield. The three occurrences illustrate the broad crystal-chemical, geochemical and genetic flexibility of this phosphate series. (i) In the granulite-facies terrane at Conifer Road in northwestern Ontario, a geochemically primitive, barren pegmatite of the Treelined Lake granitic batholith carries graftonite with inclusions of Sc-bearing, Mg-rich johnsomervilleite and exsolved sarcopside lamellae; all three phases rank with the most Fe- and Mg-rich compositions analyzed to date in terrestrial samples. (ii) In the YITT–B pegmatites, atypical members of the beryl – columbite – phosphate subtype at Bernic Lake, southeastern Manitoba, homogeneous Ca-rich beusite is Mn-dominant and associated with manganoan fluorapatite and traces of triplite and triphylite. (iii) In the #22 pegmatite, of the beryl – columbite – phosphate subtype, at Cross Lake in north-central Manitoba, homogeneous Ca-poor beusite shows the most manganoan composition known to date, and is associated with similarly Mn-rich fillowite, fluorapatite and triploidite, plus two unidentified phosphates. Low activities of Na, Li and F, combined with high concentrations of Ca, Fe, Mn (and locally Mg), are required to stabilize graftonite – beusite. These conditions cannot be expected to be widespread in the rare-element pegmatites of the Superior Province, as most of them show early enrichment in Li and F in their regional fractionation trends. Consequently, graftonite – beusite is not stable, as triphylite – lithiophilite tends to be a relatively early phase, and fluorapatite consumes all phosphate-bound Ca.

Keywords: graftonite, beusite. sarcopside, johnsomervilleite, phosphates, crystal chemistry, granitic pegmatites, fractionation, Superior Province.

SOMMAIRE

Une membre de la série graftonite – béusite a été signalé à seulement trois endroits parmi les pegmatites granitiques de la Province du Supérieur du Bouclier canadien. Ces trois exemples témoignent de la flexibilité cristallochimique, géochimique et génétique de cette série de phosphates. (i) Dans le socle granulitique de Conifer Road, dans le nord–ouest de l'Ontario, une pegmatite géochimiquement primitive et stérile du batholithe granitique de Treelined Lake contient de la graftonite avec inclusions de johnsomervilleïte riche en Mg et scandifère, et des lamelles d'exsolution de sarcopside; chacune des trois phases se classe parmi les exemples terrestres les plus riches en Fe et Mg qui soient connus. (ii) Dans les pegmatites de l'essaim YITT–B, des membres atypiques du sous-type à béryl – columbite – phosphate au Lac Bernic, au sud-est du Manitoba, contiennent de la béusite relativement calcique homogène à dominance de Mn, associée à la fluorapatite manganifère et à des traces de triplite et triphylite. (iii) Dans la pegmatite #22, du sous-type à béryl – columbite – phosphate, au Lac Cross, dans

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le centre–nord du Manitoba, la béusite homogène et à faible teneur en Ca possède la composition la plus riche en Mn qui soit connue; elle est associée à la fillowite, fluorapatite et triploïdite aussi enrichies en Mn, en plus de deux phosphates méconnus. De faibles activités de Na, Li et F, et des concentrations de Ca, Fe, Mn (et, sur le plan local, Mg), seraient nécessaires pour stabiliser la série graftonite–béusite. On ne s'attend pas que ces conditions soient répandues dans les pegmatites enrichies en éléments rares de la province du Supérieur. Dans la plupart des cas, ces pegmatites montrent un enrichissement précoce en Li et F dans leur tracé d'évolution régionale. Par conséquent, la graftonite – béusite n'est pas stable, parce que la série triphylite – lithiophilite tend à cristalliser tôt, et la fluorapatite accommode tout le calcium destiné à la fraction phosphatée.

(Traduit par la Rédaction)

Mots-clés: graftonite, béusite, sarcopside, johnsomervilleïte, phosphates, chimie cristalline, pegmatite granitique, fractionnement, Province du Supérieur.

INTRODUCTION

Minerals of the graftonite – beusite series, $(Fe,Ca)_3(PO_4)_2 - (Mn,Ca)_3(PO_4)_2$, are typical accessory phases in some populations of granitic pegmatites, but virtually absent from others. The crystal chemistry of this series was shown to be very flexible in terms of cation order–disorder, choice of cations and consequent changes in coordination of oxygen around three discrete cation sites (Steele *et al.* 1991, M.A. Wise, in prep.). The chemical composition of graftonite – beusite was shown to reflect the level of geochemical evolution of the parent pegmatites on regional as well as very local scale (Smeds *et al.* 1998), and to control, or at least strongly influence, the subsolidus exsolution of sarcopside or triphylite, or a sequence of both (Smeds *et al.* 1998, Wise, in prep., and cited earlier literature).

Graftonite – beusite minerals were not known from the numerous pegmatite fields of the Superior Province of the Canadian Shield until very recently. However, beusite was reported by Ercit *et al.* (1985, 1986) from Cross Lake, and by Anderson *et al.* (1998) from YITT–B pegmatites near Bernic Lake, both in Manitoba, and a graftonite – sarcopside intergrowth was found in 1995 by FWB in northwestern Ontario. We report here on the mineralogical and geochemical aspects of these occurrences, which represent a prime example of the paragenetic, geochemical and crystal-chemical adaptability of this phosphate series to their parent environment. Extremely Fe,Mg-rich and extremely Mn-rich compositions are encountered here in the most primitive and most fractionated parent pegmatites, respectively.

CONIFER ROAD, NORTHWESTERN ONTARIO

The graftonite – sarcopside intergrowth was found in a simple barren pegmatite that cross-cuts the foliation of a peraluminous granite of the Treelined Lake granitic complex at Conifer Road, about 5 km southwest of Campfire Lake in northwestern Ontario (Fig. 1). The complex consists of a large, branching mass of S-type granites centered in the Umfreville – Conifer granulite zone of the English River Subprovince. In the area of the phosphate occurrence, the granite (with K/Rb of
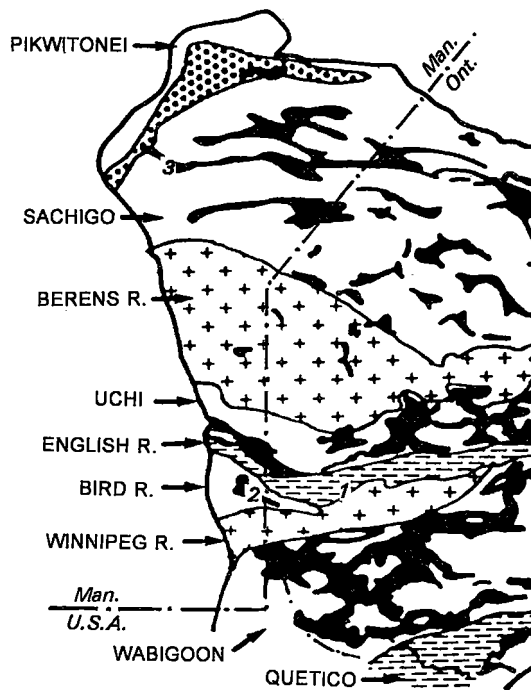

FIG. 1. Western margin of the Superior Province in Manitoba and Ontario, subdivided into the volcanoplutonic (black and white), metasedimentary (dashed), plutonic (crosses) and high-grade gneiss (dotted) subprovinces (modified from Černý 1990). The map shows the location of the graftonite and beusite occurrences at the Conifer Road (1, in the English River Subprovince), in the YITT–B swarm (2, in the Bird River Subprovince) and at Cross Lake (3, in the Sachigo Subprovince).

K-feldspar 446 to 418) penetrates the Separation Lake metavolcanic belt. The granite contains the assemblage cordierite + garnet + biotite, is coarse-grained to pegmatitic, and is laden with numerous enclaves of metagreywacke; sillimanite and orthopyroxene also
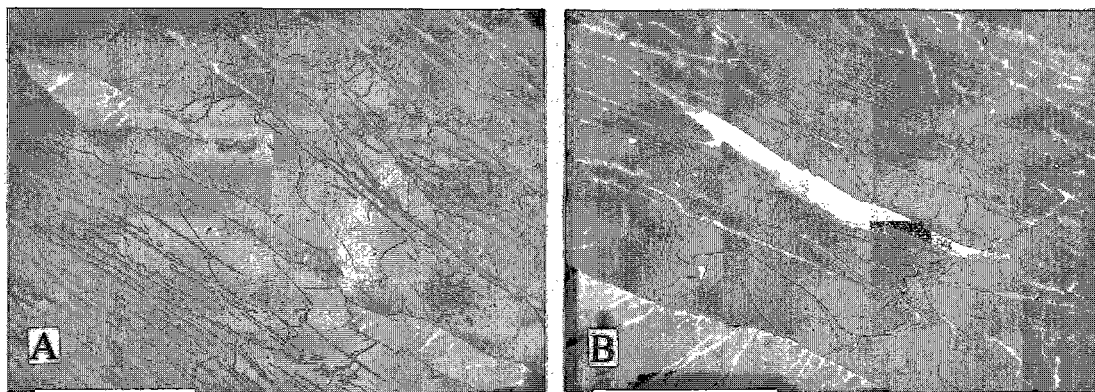
FIG. 2. A. Coarse and fine lamellae of sarcopside (dark) in graftonite matrix, Conifer Road; back-scattered electron (BSE) image. B. Lath-shaped section across a crystal of johnsomervilleite (black, near the center), surrounded by sarcopside (dark grey) exsolved from graftonite matrix, Conifer Road; BSE image; scale bars are 500 μm long.

are observed locally. The pegmatite is exposed over a 7–30 cm by 2 m area, and consists of blocky K-feldspar (with K/Rb of 209 to 187) and quartz. Subordinate to minor biotite forms randomly oriented books that enclose dark brown garnet. A dark green chlorite-group mineral may represent a pseudomorph after cordierite, which was found fresh and intergrown with quartz in a closely adjacent mass of pegmatite. Aggregates of andalusite and sillimanite are concentrated in close vicinity of the pale brown graftonite – sarcopside intergrowth, which is entirely enveloped by quartz.

The phosphate intergrowth, 1×1.5 cm in size, consists of alternating, subparallel, coarse and fine lamellae of sarcopside (0.25 to 0.50 mm and 2 to 10 μm wide, respectively) in a graftonite matrix, very similar to vein-perthitic albite in K-feldspar (Fig. 2A). Angular randomly oriented blocks of sarcopside, up to 200×400 μm in size, are extremely rare. Platy inclusions of johnsomervilleite also are scarce, randomly oriented and enveloped by sarcopside (Fig. 2B). The coarse lamellae of sarcopside are locally oxidized and hydrated along fractures.

The graftonite is distinctly Fe-dominant, Mg-enriched, and Ca-rich, whereas the sarcopside lamellae are even more Fe- and Mg-enriched. Zinc is minor to undetectable in both minerals (Table 1). Slight differences in composition are observed between the coarse and fine lamellae of sarcopside on one hand, and their respective matrices of graftonite on the other. The fine sarcopside shows levels of Fe and Ca higher, and of Mn and Mg lower, than its coarse counterpart, whereas the graftonite matrix of the fine sarcopside has levels of Mg and Fe lower, but of Ca and Mn higher, than the matrix of coarse lamellae of sarcopside (Table 1).

Johnsomervilleite closely matches the composition expected for a Fe-dominant counterpart of fillowite

TABLE 1. REPRESENTATIVE COMPOSITIONS OF THE CONIFER ROAD PHOSPHATES

	coarse lamellae		fine lamellae		bulk comp.	inclusion
	Gft TR-6	Sar TR-4	Gft TR-19	Sar TR-13	Gft + Sar	Jsv TR-22
P_2O_5 wt.%	42.30	43.90	42.10	42.80	43.3	44.40
Sc_2O_3	0.00	0.00	0.00	0.00	0.0	1.99
MgO	4.58	12.80	3.57	11.20	6.7	12.80
CaO	12.10	0.00	14.90	1.22	9.3	4.46
MnO	7.36	2.85	7.83	2.54	6.3	4.71
FeO	31.90	39.60	30.00	40.50	34.3	24.40
ZnO	0.00	0.00	0.07	0.00	0.0	0.40
Na_2O	0.00	0.00	0.00	0.00	0.0	5.27
K_2O	0.00	0.00	0.00	0.00	0.0	0.02
Σ	98.24	99.15	98.47	98.26	99.9	98.45
atomic contents*						
P	2.014	2.015	2.006	2.005	2.01	17.986
Sc	0.000	0.000	0.000	0.000	0.00	0.830
Mg	0.384	1.035	0.299	0.924	0.55	9.130
Ca	0.729	0.000	0.898	0.072	0.55	2.286
Mn	0.351	0.131	0.373	0.119	0.29	1.909
Fe	1.501	1.796	1.412	1.874	1.58	9.764
Zn	0.000	0.000	0.003	0.000	0.00	0.141
Na	0.000	0.000	0.000	0.000	0.00	4.889
K	0.000	0.000	0.000	0.000	0.00	0.012
Σ M	2.965	2.962	2.985	2.989	2.97	28.961

* graftonite (Gft) and sarcopside (Sar) normalized to 8 oxygen atoms and johnsomervilleite (Jsv) normalized to 72 oxygen atoms.
See Smeds et al. (1998) for conditions of electron-microprobe analysis and Kontron-IBAS modal analysis.

(Table 1). It also matches well the Mg-rich composition of the type johnsomervilleite (Livingstone 1980), but the appreciable Sc content is so far unique (cf. Fransolet et al. 1998).

The modal composition of the phosphate intergrowth was determined to be 70 vol.% graftonite and 30 vol.% sarcopside. A specific gravity of 3.49 for graftonite and 3.59 for sarcopside was calculated from average chemical compositions and unit-cell dimensions. These data gave 69 wt.% graftonite and
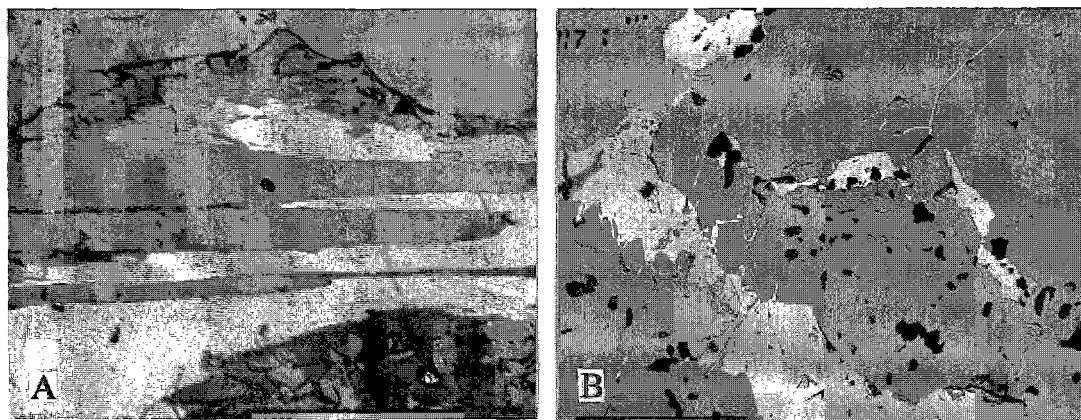
FIG. 3. A. Alteration of beusite (white) and triphylite (granular texture) to alluaudite (grey), with primary inclusions of fluorapatite (dark grey, at bottom), YITT–B; BSE image. The scale bar is 100 μm long. B. Rounded grains of fillowite (medium grey) with inclusions of triploidite (off-white) and manganoan fluorapatite (black), surrounded and possibly corroded by beusite (pale grey). Cross Lake; BSE image. The scale bar is 200 μm long.

31 wt.% sarcopside. The resulting bulk composition of the graftonite + sarcopside intergrowth is shown in Table 1.

YITT–B PEGMATITES, SOUTHEASTERN MANITOBA

The YITT–B pegmatite swarm is located north of the Tanco pegmatite deposit at Bernic Lake [Fig. 1; cf. Figs. 1 and 2 of Anderson et al. (1998) for details]. The swarm belongs to the Cat Lake – Winnipeg River pegmatite field, located in the Bird River Greenstone Belt, which flanks the Bird River Subprovince along its southern contact with the Winnipeg River Subprovince [cf. Černý et al. (1981) for regional geological relationships]. The YITT–B swarm generally belongs to the beryl – columbite – phosphate subtype of rare-element granitic pegmatites. It displays several anomalous features, however, which set it apart not only from other pegmatite populations encountered in the parent pegmatite field, but from most other pegmatites of this category worldwide: very low content of Li, Be, B and F, moderate enrichment of Mn over Fe, but very marked enrichment in Ta over Nb, and all primary phosphates as anhedral microscopic grains filling interstices among silicates (Anderson et al. 1998). The pegmatites are heterogeneous in terms of mineralogy, grain size and texture, but a regular concentric sequence of individual zones is rarely observed. The main constituents are K-feldspar (ranging from 53 to 24 in K/Rb for the blocky variety in central parts of the pegmatites), quartz and albite; muscovite is locally abundant but generally subordinate, and accessory garnet (Sps_{51-61}) is widespread.

TABLE 2. REPRESENTATIVE COMPOSITIONS OF THE YITT-B PHOSPHATES

		beusite		Tph	triplite	apatite
		YI-35	YI-22	YI-9	3-12	YI-14
P_2O_5	wt.%	41.50	41.20	45.30	31.56	42.00
MgO		0.00	0.00	0.06	0.17	0.00
CaO		13.30	13.30	0.17	1.25	50.70
MnO		30.20	29.00	18.70	54.54	5.18
FeO		14.10	15.00	25.20	7.32	0.59
ZnO		0.46	0.45	0.00	0.00	0.00
Na_2O		0.00	0.10	0.00	0.00	0.00
K_2O		0.00	0.00	0.00	0.01	0.00
Li_2O^*		0.00	0.00	9.46	0.00	0.00
H_2O^*		0.00	0.00	0.00	1.47	0.45
F		0.00	0.00	0.00	5.37	2.76
Cl		0.00	0.00	0.00	0.02	0.06
-O=F,Cl		0.00	0.00	0.00	-2.26	-1.17
$\sum$		99.56	99.05	98.89	99.45	100.57
		atomic contents**				
P		2.011	2.008	1.008	0.996	3.001
Mg		0.000	0.000	0.002	0.009	0.000
Ca		0.815	0.820	0.005	0.050	4.585
Mn		1.464	1.414	0.416	1.722	0.370
Fe		0.675	0.722	0.554	0.228	0.042
Zn		0.019	0.019	0.000	0.000	0.000
Na		0.000	0.011	0.000	0.000	0.000
Li		0.000	0.000	1.000	0.000	0.000
$\sum$ M		2.973	2.986	1.977	2.009	4.997
F		0.000	0.000	0.000	0.633	0.737
Cl		0.000	0.000	0.000	0.001	0.009
OH		0.000	0.000	0.000	0.366	0.255

* calculated by stoichiometry; ** beusite normalized to 8 oxygen atoms, triphylite (Tph) normalized to 4 oxygen atoms, triplite normalized to 5 anions and apatite normalized to 13 anions. See Smeds et al. (1998) for conditions of electron-microprobe analysis.

Primary phosphates, beusite, apatite and extremely rare triphylite and triplite are present in grains <1 mm in size, dispersed together with the oxide minerals of Nb, Ta and Sn in saccharoidal albite. Medium-brown beusite locally contains round inclusions of manganoan fluorapatite, but otherwise it is homogeneous. Beusite is commonly altered to extremely Ca-poor, Na□-type, Mn^{2+}- and Fe^{3+}-dominant, Fe^{2+}-"free" alluaudite, and a secondary Na,Fe,Mn,Cl-bearing carbonate-fluorapatite to carbonate-hydroxyapatite (Fig. 3A; Anderson et al. 1998).

The YITT–B beusite is Ca-rich and distinctly Mn-dominant, with subordinate content of Zn (Table 2). The associated primary phosphates also are enriched in Mn, with a difference: triplite is Mn-dominant, but triphylite is not (Table 2). Primary apatite is Mn-bearing and relatively F-poor (Table 2; cf. also Anderson et al. 1998).

CROSS LAKE, NORTH-CENTRAL MANITOBA

At Cross Lake, five series of granitic pegmatites constitute a field spread over a steep regional metamorphic gradient in, and west of, the Cross Lake Greenstone Belt, along the western margin of the Sachigo Subprovince [Fig. 1; cf. Corkery et al. (1992) for details of regional geology]. Beusite was identified in a member (#22) of the Northern Series of pegmatites classified as being of beryl – columbite subtype (Anderson 1984). The pegmatite outcrops over an area of about 40 × 7 m and shows prominent but asymmetric internal zoning. Saccharoidal to medium-grained albite and quartz are the main constituents of the outer and intermediate units. The outer unit hosts fan-shaped books of muscovite, blocks of K-feldspar, and scattered spessartine ($Sps_{34}Alm_{54}Prp_2$) and schorl; the internal unit also contains columnar beryl, apatite and Nb,Ta,Sn-bearing oxide minerals. Blocks of K-feldspar (with K/Rb of 28–21) and book muscovite mark the central parts of the pegmatite, and a quartz "core" with crystals of beryl drifts into contact with the host metagreywacke along the southeastern flank of the pegmatite (Anderson 1984).

In contrast to other members of the swarm, the intermediate units of #22 carry two assemblages of phosphates. The first assemblage is rather widespread, consisting of fluorapatite and bobfergusonite (Ercit et al. 1985); the latter mineral is associated with a related but so far unidentified phosphate. The second assemblage constitutes separate nodules of medium- to coarse-grained beusite, fillowite, triplite, triploidite and Mn-rich fluorapatite (Ercit et al., in prep.). Replacement by secondary phases such as alluaudite and Mn,Fe-rich, Na,Sr-bearing chlorapatite is very limited (Ercit et al. 1985, 1986). Two unidentified phosphates also were encountered in this assemblage. The first phase forms inclusions in beusite; it has $(Na_{0.3}Ca_{0.7})$:$(Mn_{1.4}Fe_{0.6})$:P proportions very close to 1:2:2, but analytical totals

		beusite		triploidite		fillowite		apatite
		CR3-1	CR5-1	CR1-	CR4-6	CR3-5	CR2-4	CR1-7
P_2O_5	wt.%	40.40	40.90	32.10	31.40	40.80	40.60	41.00
MgO		0.10	0.02	0.54	0.21	0.42	0.12	0.00
CaO		6.05	5.52	0.35	0.07	4.31	4.19	39.50
MnO		40.40	44.20	45.80	50.90	38.20	40.80	16.90
FeO		11.20	8.51	13.60	9.58	8.63	6.42	0.32
ZnO		1.18	1.53	2.03	2.48	1.34	1.52	0.00
Na_2O		0.00	0.01	0.00	0.01	5.40	5.43	0.00
K_2O		0.00	0.00	0.00	0.00	0.05	0.04	0.00
H_2O^*		0.00	0.00	3.58	3.71	0.00	0.00	0.35
F		0.00	0.00	0.93	0.60	0.00	0.00	2.89
O=F		0.00	0.00	-0.39	-0.25	0.00	0.00	-1.22
Σ		99.33	100.69	98.54	98.71	99.15	99.12	99.74
atomic contents**								
P		2.003	2.004	1.012	0.998	18.091	18.059	3.020
Mg		0.009	0.002	0.030	0.012	0.328	0.094	0.000
Ca		0.380	0.342	0.014	0.003	2.419	2.359	3.682
Mn		2.004	2.167	1.445	1.619	16.946	18.157	1.245
Fe		0.549	0.412	0.424	0.301	3.780	2.821	0.023
Zn		0.051	0.065	0.056	0.069	0.518	0.590	0.000
Na		0.000	0.001	0.000	0.001	5.484	5.531	0.000
K		0.000	0.000	0.000	0.000	0.033	0.027	0.000
Σ M		2.993	2.989	1.969	2.005	29.508	29.579	4.950
F		0.000	0.000	0.110	0.071	0.000	0.000	0.795
OH		0.000	0.000	0.890	0.929	0.000	0.000	0.205

TABLE 3. REPRESENTATIVE COMPOSITIONS OF THE CROSS LAKE PHOSPHATES

* calculated by stoichiometry; **beusite normalized to 8 oxygen atoms, triploidite normalized to 5 anions, fillowite normalized to 72 oxygen atoms and apatite normalized to 13 anions.
See Smeds et al. (1998) for conditions of electron-microprobe analysis.

generated by electron-microprobe analysis are low by ~5 to 7 wt.%, and some light elements may be present. The second phosphate forms a rim around fillowite and beusite, with ~1 wt.% Na_2O and $(Mn_{2.3}Ca_{0.5}Fe_{0.3})$:P:O:Cl proportion close to 3:2:7.6:0.4, and electron-microprobe analytical totals close to ~99 wt.%.

Beusite forms pale flesh-colored grains 5–25 mm in size. They enclose roundish grains of fillowite and triploidite; manganoan fluorapatite forms round inclusions in the outermost parts of the fillowite grains (Fig. 3B).

Chemical compositions of all five well-defined primary phosphates of the second assemblage are extremely manganoan (Table 3, and Ercit et al. 1985, 1986). Fluorapatite attains up to 18.59 wt.% MnO. With the exception of fluorapatite, the high Zn content of the phosphates also is remarkable, particularly in triploidite; however, the Ca content of beusite is rather moderate (Table 3).

DISCUSSION

Crystal chemistry

The Conifer Road graftonite is Ca-rich and Mn-poor (Fig. 4). It ranks with the most ferroan and magnesian terrestrial samples analyzed to date, and so does the sarcopside [Fig. 4, 5; cf. Wise & Černý (1990) and Steele et al. (1991) for graftonite; Hurlbut (1965), Moore (1972), Fransolet (1977), Fransolet et al. (1986), Zhang (1995) and Smeds et al. (1998) for
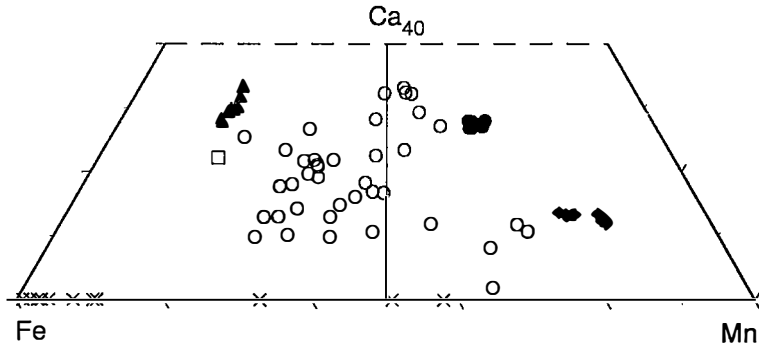
FIG. 4. Compositions of graftonite and beusite in terms of Fe – Mn – Ca (atomic proportions). Symbols: ▲ Conifer Road, ● YITT–B, ◆ Cross Lake, □ Scotland; O data from Wise & Černý (1990), Steele *et al.* (1991) and Smeds *et al.* (1998) for terrestrial phases, and × for meteorite-hosted phases.
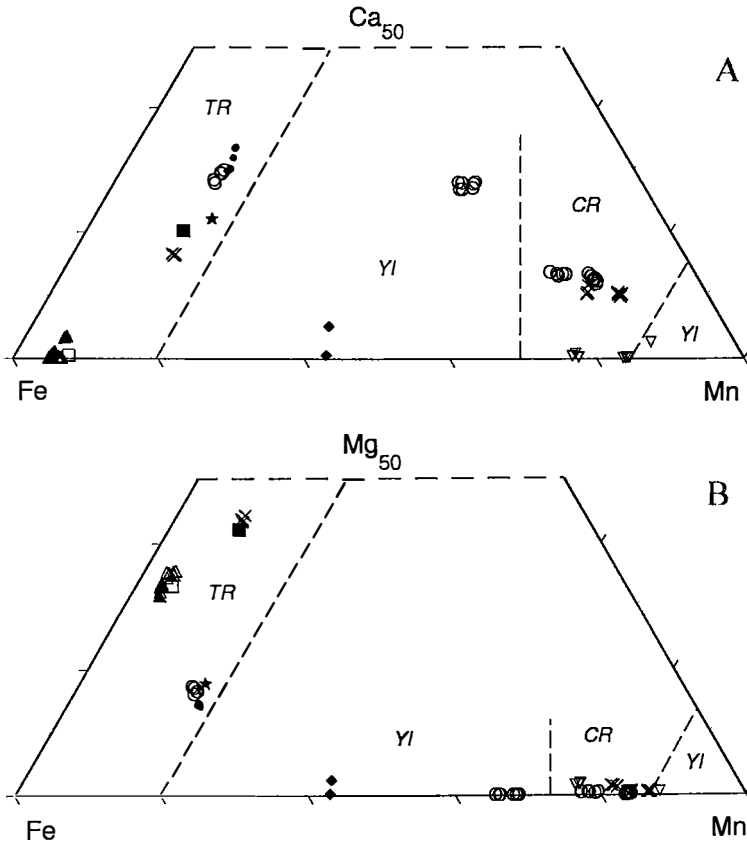

FIG. 5. Compositions of the phosphates examined from the Conifer Road (TR), YITT–B (YI) and Cross Lake (CR) pegmatites in terms of Fe–Mn–Ca (A) and Fe–Mn–Mg (B) (atomic proportions). Symbols: O: graftonite and beusite (coarse in the TR field); ●: fine-lamellar graftonite in the TR field; △: coarse sarcopside; ▲: fine-lamellar sarcopside; ×: johnsommervilleite and fillowite; ◆: triphylite; ▽: triplite and triploidite; metamorphic: ★: graftonite, □: sarcopside and ■: johnsomervilleite from Scotland.

sarcopside; and Livingstone (1980) for graftonite and "mineral A", compositionally corresponding to sarcopside]. The YITT–B beusite is strongly Mn-enriched and Mg-poor in comparison, and its Ca content is close to the highest encountered in this mineral [Figs. 4, 5; *cf.* Wise & Černý (1990), Steele *et al.* (1991)]. The Cross Lake beusite has only a modest Ca content, but its Mn/(Mn + Fe) value is the highest observed to date [Figs. 4, 5; *cf.* data collected in Wise & Černý (1990), Steele *et al.* (1991)].

In all compositions, the contents of divalent cations and of phosphorus normalized to 8 oxygen atoms are near-ideal within the limits of analytical error, and any possible Li content must be negligible. Although not directly detectable by electron microprobe, the presence of Li in the form of a triphylite – lithiophilite $Li(Fe,Mn)PO_4$ component in solid solution would be readily revealed by deviations of cations and P from the 3:2 ratio (Smeds *et al.* 1998, M.A. Wise, in prep.). The bulk composition of the Conifer Road graftonite + sarcopside intergrowths generates a simple formula, fitting the $R^{2+}_3P_2O_8$ stoichiometry common to both minerals (Table 1). Concerning a potential Li content, the same argument applies here as in the case of the individual minerals discussed above; the stoichiometry of the bulk formula is virtually integral, as are those of its individual components, leaving no space for more than traces of Li.

The composition of johnsomervilleite is analogous to that described from its type locality (Livingstone 1980). The main difference lies in the presence of appreciable Sc and in the Mg content, which is even higher than in the type material and closely approaches a 1:1 ratio with Fe^{2+} (Table 1). The contents of Na and Ca are lower than required by the condensed formula of Fransolet *et al.* (1998), $Na_5(Ca_2Na)$ $X^{2+}_{22}(PO_4)_{18}$. The vacancies apparently accommodate the excess positive charge generated by Sc^{3+} among the octahedrally coordinated X cations: $(Na_{4.89}K_{0.01}\square_{0.10})_{\Sigma5.00}$ $(Ca_{2.28}\square_{0.72})_{\Sigma3.00}(R^{2+}_{20.94}Sc_{0.83}\square_{0.23})_{\Sigma22.00}P_{17.98}O_{72}$ (Table 1). The Cross Lake fillowite also conforms to the above general formula, $(Na_{4.97}K_{0.03})_{\Sigma5.00}(Ca_{2.40}Na_{0.55}\square_{0.05})_{\Sigma3.00}$ $(R^{2+}_{21.70}\square_{0.30})_{\Sigma22.00}P_{18.07}O_{72}$; the excess Ca compensates for most of the vacancies in sites occupied by the octahedrally coordinated X cations (Table 3).

The textural relationships of graftonite and sarcopside in the Conifer Road specimen, and the partitioning of Ca, Fe, Mn and Mg between the two phases, match those observed in graftonite + sarcopside intergrowths from other localities, at which they are documented and interpreted as products of exsolution [Fig. 5; *cf.* Smeds *et al.* (1998) and M.A. Wise (in prep.) for review of the literature and new data]. The bulk composition of the intergrowth conforms to that of a homogeneous precursor with the same overall stoichiometry.

The compositional differences in the coarse and fine intergrowths suggest slightly different patterns and

TABLE 4. UNIT-CELL DIMENSIONS OF
SELECTED PHOSPHATES

		TR - Gft	TR - Sar	CR - Beu
a	Å	8.900(1)	10.386(6)	8.80(1)
b		11.580(2)	4.781(2)	11.63(2)
c		6.187(1)	6.019(3)	6.174(7)
β	°	97.49(2)	90.82(7)	99.4(1)
v	Å³	632.2(1)	298.9(2)	623.1(9)

TR = Conifer Road, CR = Cross Lake
Refined from X-ray powder diffraction data determined on the
Siemens D5000 apparatus in transmitted mode, with silicon internal
standard (*a* = 5.4309 Å); FIX peak-positioning program of Ercit
(1986), modified CELREF program of Appleman & Evans (1973).

degrees of unmixing. The graftonite matrix of the fine-lamellar sarcopside, which is Mg,Fe-poor and Ca,Mn-rich relative to the matrix of the coarse lamellae of sarcopside, shows a higher degree of exsolution of the sarcopside component. However, the compositional changes in the corresponding lamellae of sarcopside match those in graftonite only in Fe and Mn; Ca and Mg suggest an opposite trend, but the shifts are negligible. These relationships suggest a two-stage exsolution, observed at other localities in analogous textural patterns, but not in terms of different chemical compositions (Smeds *et al.* 1998, M.A. Wise, in prep.).

Table 4 presents the unit-cell dimensions of the graftonite, beusite and sarcopside (except the YITT–B beusite, which was not suitable for refinement). The data are generally in good agreement with those published for phases of similar composition (Moore 1972). In particular, the *b* parameter of graftonite – beusite increases with the Ca content (Wise *et al.* 1990, Steele *et al.* 1991). However, detailed relationships between unit-cell dimensions and chemical composition are far from fully understood in the graftonite – beusite series; thus our data cannot be interpreted in more detail.

Geochemical and genetic considerations

The three broadly different occurrences of graftonite – beusite examined here closely reflect the petrological and geochemical characteristics of their host pegmatites. The phase assemblages and mineral compositions at the individual localities match local conditions and document a remarkable adaptability of the graftonite – beusite minerals to extensive differences among their parent environments. However, the scarcity of their occurrences across the pegmatite populations of the Superior Province also indicates that some parameters critical to their stabilization are only rarely encountered.

The graftonite + sarcopside intergrowth from Conifer Road is geochemically the most primitive ever found in granitic pegmatites, and it comes from the

TABLE 5. GEOCHEMICAL CHARACTERISTICS OF
THE THREE PHOSPHATE ASSEMBLAGES

	Conifer Road	Yitt-B	Cross Lake
K-feldspar			
K/Rb	415	24-53	21-28
Sarcopside			
Fe/(Fe+Mg)	0.81(1)	-	-
Mn/(Mn+Fe+Mg)	0.05(1)	-	-
Zn/(Zn+Fe+Mn+Mg)	0.00(0)	-	-
Graftonite-Beusite			
Fe/(Fe+Mg)	0.91(1)	1.00(0)	1.00(0)
Mn/(Mn+Fe+Mg)	0.19(1)	0.67(1)	0.82(3)
Zn/(Zn+Fe+Mn+Mg)	0.00(0)	0.01(0)	0.03(0)
Ca/(Ca+Fe+Mn+Mg+Zn)	0.23(2)	0.21(0)	0.09(0)
Johnsomervilleite-Fillowite			
Fe/(Fe+Mg)	0.71(0)	-	0.98(1)
Mn/(Mn+Fe+Mg)	0.12(0)	-	0.84(3)
Zn/(Zn+Fe+Mn+Mg)	0.01(0)	-	0.03(0)
Ca/(Ca+Fe+Mn+Mg+Zn)	0.09(0)	-	0.07(0)

all ratios calculated on wt.% basis; (standard deviation).

petrogenetically least evolved magmatic environment to ever yield graftonite (Table 5). The Treelined Lake granitic complex is, in the broadest of terms, parental to the Separation Lake pluton and derived rare-element Li,Be,Ta-bearing pegmatites (F.W. Breaks *et al.,* in prep.), but the graftonite + sarcopside-bearing pegmatite resides in an anchi-autochthonous granite bordering its granulite-facies source region. The low Mn content of the Conifer Road phosphates, their high Mg content and Fe/Mn ratio and the absence of Li match the simple mineralogy and very low degree of alkali fractionation attained by the parent pegmatite. The exsolution of sarcopside from a parent graftonite-type phase at Conifer Road supports the contention of Smeds *et al.* (1998) and Wise (in prep.) that the relatively high-temperature (~550 to 600°C), Ca-poor precursor becomes Ca-enriched and stabilized as graftonite by exsolution of Ca-free sarcopside, which is stable only at low temperatures (≤500°C), as discussed by Smeds *et al.* (1998).

The YITT–B beusite crystallized at ~450 to 350°C, in saccharoidal albitic assemblages of beryl – columbite – phosphate pegmatites that are geochemically much more evolved than the barren pegmatite at Conifer Road (Table 5). The beusite precipitated as a homogeneous phase and did not exsolve sarcopside, possibly because of the high primary Ca and Mn contents. The Ca and Mn contents are substantially higher, and the Mg and Fe/Mn values are much lower than in the homogeneous precursor of the Conifer Road intergrowth.

At Cross Lake, the homogeneous beusite is hosted by about the same category of pegmatite as in the YITT–B series, since the parent Northern Series corresponds to the beryl – columbite subtype. However, it displays somewhat elevated overall F content, and the beusite-bearing dike #22 shows extremely advanced fractionation of Mn over Fe, plus a distinct enrichment in P (Table 5). All primary phosphates represent the most Mn-enriched compositions ever encountered (Ercit *et al.* 1986), and ferromanganoan silicates and oxide minerals also show considerable enrichment in Mn (Anderson 1984). This high Mn content (and low Fe content) of the Cross Lake beusite, in conjunction with the otherwise moderate Ca content, served to stabilize the original precipitate at a low temperature (~450 to 350°C), and to prevent exsolution of sarcopside.

The compositional changes in the graftonite – beusite series in the three localities examined are matched by the shifts in composition of the associated phosphates (Tables 1, 2, 3 and 5). These are best illustrated by the remarkable differences between johnsomervilleite at Conifer Road and fillowite from Cross Lake, both present as inclusions in the graftonite – beusite host. The johnsomervilleite is very Fe,Mg-rich and Sc-bearing, whereas the fillowite has the most manganoan composition encountered to date, contains appreciable Zn, and its Mg content is negligible. It is noteworthy that the geochemically primitive compositional features of the johnsomervilleite + graftonite + sarcopside assemblage from Conifer Road are almost identical to those of the same assemblage from Loch Quoich, Scotland, which occurs in "metamorphic-segregation phosphate-rich pods" in the kyanite – sillimanite-grade Moine gneiss (Livingstone 1980).

Finally, the scarcity of the graftonite – beusite occurrences in the Superior Province should be addressed. As stated by Smeds *et al.* (1998), appreciable activities of Ca, Fe and Mn are required, and Mg also is a significant component in the case of the graftonite + sarcopside intergrowth from Conifer Road. Low activities of F and Na also are necessary to restrict or prevent stabilization of fluorapatite, wagnerite, triplite – zwieselite, wyllieite, fillowite, alluaudite or griphite. Low activity of Li is required to prevent crystallization of primary, discrete triphylite – lithiophilite (as opposed to exsolution of these minerals from Li-bearing graftonite – beusite: Smeds *et al.* 1998, M.A. Wise, in prep.). The principal conditions of high activity of Ca and low activities of Li and F are relatively commonly achieved in poorly fractionated, geochemically primitive pegmatites; the Conifer Road occurrence of Fe,Mg-rich phosphates is a prime example. In pegmatites that show an advanced level of geochemical evolution, such as the YITT–B and #22 bodies, Ca is generally very low, and it must be primarily the low activity of F in these pegmatites that stabilizes graftonite – beusite. Low activity of F and very low concentration of Li were documented in the YITT–B swarm (Anderson *et al.* 1998), and confirmed in the present study, which identified mere traces of

triphylite and triplite. At Cross Lake, the #22 dike does not carry any lithium-rich minerals, and no F-bearing silicates are present except for tourmaline.

It is significant that limited quantities of Na,Li, F-bearing phosphates (johnsomervilleite, fillowite, triphylite, triplite, triploidite, apatite) *preceded* the crystallization of graftonite and beusite at their respective localities. Precipitation of these phases evidently reduced the chemical potential of Na, Li and F, and thus facilitated the subsequent crystallization of the Na,Li,F-free graftonite – beusite.

In general, the above conditions favorable to graftonite – beusite stabilization should not be expected to be widespread in the rare-element pegmatites of the Superior Province. As shown by Černý (1990), the LCT-family pegmatite populations of the Superior Province generally show early and significant enrichment in Li and F in their trends of regional zonation. Consequently, triphylite is commonly the first ferromanganoan phosphate to crystallize in cogenetic pegmatite sequences (occasionally even in parent pegmatitic leucogranites; Černý & Brisbin 1982, Smeds *et al.* 1998), and fluorapatite commonly accounts for all phosphate-bound Ca.

CONCLUSIONS

The rare occurrences of graftonite – beusite in granitic pegmatites of the Superior Province demonstrate the broad compositional flexibility of this mineral series and the variety of environments in which its members can be generated. The Conifer Road occurrence of the Ca-rich (Fe > Mg ≅ Mn) graftonite + (Fe > Mg ≅ Mn) sarcopside exsolution-induced intergrowth, with inclusions of johnsomervilleite, is the first one recorded from a deep-seated primitive magmatic environment of an anchi-autochthonous association of peraluminous granite with barren pegmatite, bordering their granulite-facies source region. The beusite localities at Bernic and Cross lakes belong to the beryl – columbite – phosphate subtype of pegmatites. The Ca-rich, Mg-free, Zn-bearing (Mn>Fe) beusite from the YITT–B pegmatites is associated with manganoan fluorapatite plus minor triplite and triphylite. The Ca-poor, Zn-enriched, Mg-free beusite from Cross Lake, with inclusions of fillowite, triploidite and fluorapatite, is the closest to the manganoan end-member found to date; it was generated in an extremely Mn-rich environment, unique in granitic pegmatites. At all three localities, stabilization of the graftonite – beusite minerals was facilitated by relatively to extremely low activities of F, Na and Li. Such conditions are expected to be rare in pegmatite populations of the Superior Province, which explains the scarcity of graftonite – beusite in this region.

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