

GRAFTONITE – BEUSITE IN SWEDEN: PRIMARY PHASES, PRODUCTS OF EXSOLUTION, AND DISTRIBUTION IN ZONED POPULATIONS OF GRANITIC PEGMATITES

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ABSTRACT

Graftonite – beusite is relatively widespread in the LCT family of granitic pegmatites of the Bothnian basin and of the Utö – Mysingen field in Sweden. It commonly constitutes the matrix containing either coarse lamellae of sarcopside, which in turn hosts fine lamellae of triphylite, or abundant lamellae of triphylite with bimodal size-distribution. Deformed and recrystallized granular aggregates of graftonite with triphylite, wolfeite and sarcopside are rare. Graftonite – beusite is systematically Ca,Mn-rich and Mg-poor relative to sarcopside, which in turn is slightly Mn-rich and distinctly Mg-poor relative to the Li-concentrating triphylite. Bulk compositions of the lamellar aggregates, crystal-chemical and textural relationships among the individual phosphates, and cation distribution-coefficients indicate two patterns of exsolution that generated the lamellar intergrowths from disordered homogeneous precursors: Ca,Li-poor graftonite exsolved lithian sarcopside, which in turn broke down to sarcopside with triphylite lamellae, and Ca,Li(Mn)-rich graftonite – beusite exsolved triphylite directly. In regional zoning of pegmatite groups, Ca-bearing graftonite – beusite is either the first Fe,Mn(Li)-phosphate encountered in the beryl – columbite pegmatites, or it is found within the zone of beryl – columbite – triphylite pegmatites. Appreciable concentrations of Ca, Fe, Mn (and in part Mg), but low levels of F and Na, are required to stabilize graftonite – beusite. Good correlation of the Mn/(Mn + Fe) values of the phosphates with those of garnet suggests that the homogeneous graftonite – beusite precursor could have been generated by phosphorus-induced destabilization of garnet.

Keywords: graftonite, beusite, triphylite, sarcopside, phosphates, exsolution, granitic pegmatite, regional zoning, Sweden.

SOMMAIRE

La solution solide graftonite – béusite est relativement répandue parmi les pegmatites granitiques de type LCT dans le bassin de Bothnie et dans le champ de Utö – Mysingen, en Suède. Un de ces minéraux forme l'hôte de lamelles substantielles de sarcopside, qui à son tour contient de fine lamelles de triphylite, ou bien des lamelles abondantes de triphylite dont la taille montre une distribution bimodale. Les agrégats déformés, recristallisés et granulaires de graftonite avec triphylite, wolfféite et sarcopside sont rares. La série graftonite – béusite s'avère systématiquement enrichie en Ca et Mn, et appauvrie en Mg par rapport au sarcopside qui, à son tour, est légèrement enrichie en Mn et fortement appauvrie en Mg par rapport à la triphylite,

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qui concentre le Li. D'après les compositions globales des agrégats lamellaires et les considérations cristallochimiques et texturales parmi ces phosphates, et les coefficients de distribution des cations, deux schémas d'exsolution semblent expliquer les intercroissances lamellaires à partir d'un précurseur homogène et désordonné: graftedite à faible teneur en Ca et Li exsolve le sarcopside lithique, qui à son tour se déstabilise en sarcopside avec lamelles de triphylite; en revanche, la graftedite ou la béusite riche en Ca et Li(Mn) produit des lamelles de triphylite directement. Dans une suite de venues pegmatitiques montrant une zonation régionale, graftedite – béusite relativement calcique est le premier phosphate de Fe et Mn(Li) à cristalliser dans la zone à béryl – columbite, ou dans la zone à béryl – columbite – triphylite. Des concentrations importantes de Ca, Fe, Mn (et, en partie, Mg) et des faibles concentrations de F et de Na seraient nécessaires pour stabiliser la série graftedite – béusite. La bonne corrélation des valeurs Mn/(Mn + Fe) des phosphates et du grenat fait penser que le précurseur graftedite – béusite homogène pourrait résulter de la déstabilisation du grenat dans la magma progressivement enrichi en phosphore.

(Traduit par la Rédaction)

Mots-clés: graftedite, béusite, triphylite, sarcopside, phosphates, exsolution, pegmatite granitique, zonation régionale, Suède.

INTRODUCTION

Lamellar intergrowths of graftedite, $(\text{Fe}, \text{Mn}, \text{Ca})_3(\text{PO}_4)_2$, or beusite, $(\text{Mn}, \text{Fe}, \text{Ca})_3(\text{PO}_4)_2$, with sarcopside $(\text{Fe}, \text{Mn}, \text{Mg})_3(\text{PO}_4)_2$ or triphylite $\text{Li}(\text{Fe}, \text{Mn})\text{PO}_4$ (or both) are a relatively rare component of granitic pegmatites. Occurrences of these phosphate intergrowths are known solely from the LCT family of pegmatites (peraluminous, derived mainly from S-type granites; cf. Černý 1991); they may be locally abundant, but are virtually missing in other populations of LCT pegmatites, which may be as extensive as entire pegmatite provinces.

The first (and for 20 years the only) locality of graftedite known in Sweden was the Berg pegmatite in the vicinity of Sollefteå (Lundqvist *et al.* 1990). The other localities in the Bothnian basin were identified by the first author during a prospecting program conducted in 1981–1986. In the same period, the fifth author found the first occurrences of graftedite in the Utö – Mysingen region, in outcrops along a field-trip path visited by many geologists over the past 200 years, and additional localities have been discovered since.

Although described in the literature from several occurrences in reasonable detail, the quantitative aspects of graftedite – beusite and their lamellar intergrowths with other phosphates are poorly understood, as is their position in the paragenetic evolution of individual pegmatites and regional populations of pegmatites. Thus the main objectives of this study, complementary to the concurrent work of M.A. Wise (in prep.), are to establish the types of intergrowths of graftedite – beusite with other phosphates in the Swedish pegmatites, the composition of their component minerals and their bulk composition, to examine their origin, and to establish the position of graftedite – beusite on a regional scale of cogenetic pegmatite groups.

REGIONAL ENVIRONMENT OF THE GRAFTONITE-BEUSITE-BEARING PEGMATITE POPULATIONS

Graftedite – beusite occurs in the LCT family of rare-element pegmatites in two major regions of Sweden, occupied mainly by metasedimentary rocks. One of them is the Bothnian basin of north-central Sweden, the other the Utö region of south-central Sweden (as defined by Smeds 1990; Fig. 1, Table 1).

TABLE 1. LOCALITIES OF GRAFTONITE-BEUSITE IN SWEDEN

Locality	RAK	Phosphate intergrowth
<i>Bothnian basin</i>		
Berg	701040-157250	Gra + Sar + Tph
Brändlandet	704460-162290	Gra + Sar + Tph
Räggen, GDB	695315-149745	Gra + Tph + Sar + Wol (granular)
Sidensjö, Beusite dike	701810-162610	Beu + Tph
Tjällingsjö	703415-156130	Gra + Tph
<i>Utö-Mysingen field</i>		
Stora Persholmen	654165-164455	Beu + Tph; Gra + Sar + Tph
Ångsholmarna Norra	654120-164390	Gra + Sar + Tph
Smedshäll	654100-164327	Gra + Sar + Tph
Rånö	653590-163705	Gra + Sar + Tph; Gra + Tph
Norrö	653430-163570	Gra + Sar + Tph
Bövedden	653270-163445	Gra + Sar + Tph; Gra + Tph
Näturö	652825-163235	Gra/Beu + Tph

RAK - Coordinates of the National Topographic Grid of Sweden.

Gra = graftedite, Beu = beusite, Tph = triphylite, Sar = sarcopside, Wol = wolfeite

Bothnian basin

The Bothnian basin is a vast region of metasedimentary rocks occupying a large portion of north-central Sweden (Lundqvist 1979). Metasedimentary rocks of the Härnö Formation are dominantly turbiditic, with subordinate mafic and rare felsic to intermediate

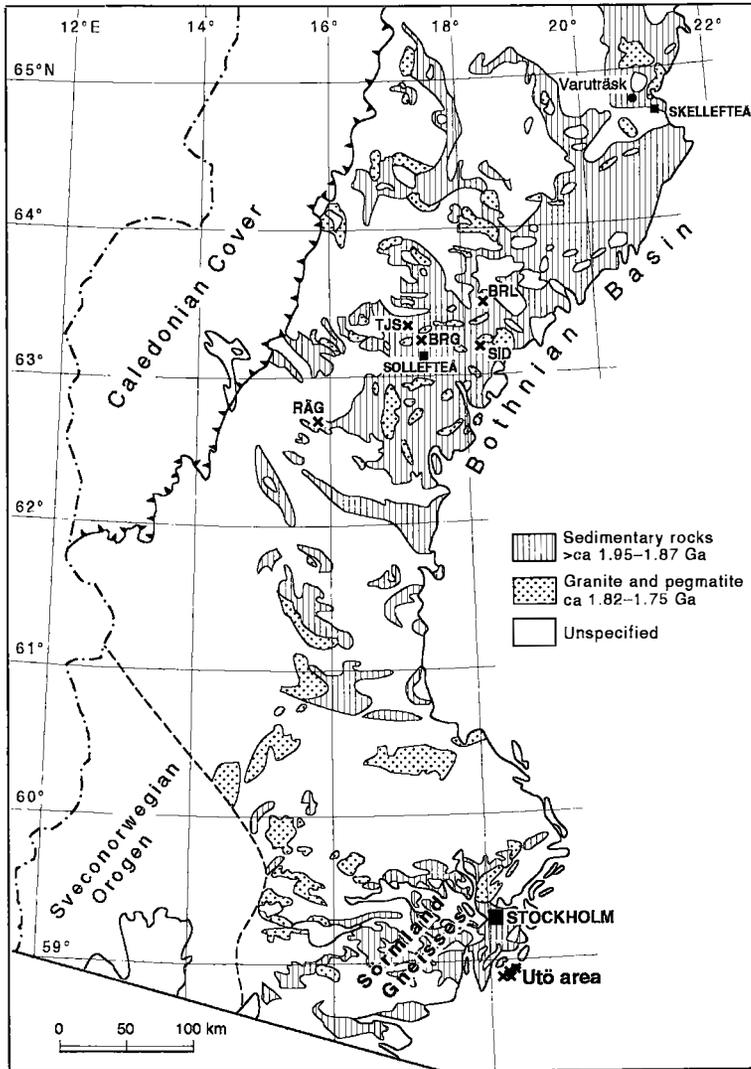
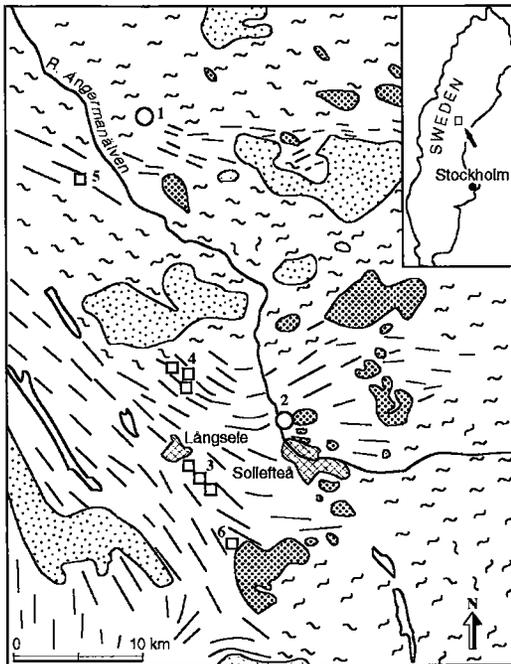


FIG. 1. Geological map of central Sweden, with graftonite – beusite localities. The map is modified after Stephens *et al.* (1994) and Teertstra *et al.* (1996). BRL: Bräntlandet, TJS: Tjälmsjön, SID: Sidensjö, Långmarken pegmatite, BRG: Berg, RÄG: Räggen GDB.

volcanic units (Lundqvist *et al.* 1990). Felsic metavolcanic rocks yield an age of 1874 ± 6 Ma (U–Pb zircon; Welin 1987). During the Svecofennian orogeny, the metasedimentary rocks were intruded by early orogenic granitic plutons with minor gabbro, and subsequently folded and to a great extent migmatized. After the major phases of folding and metamorphism, they were intruded by numerous late-orogenic granite massifs, collectively named the Härnö granites. They

constitute a peraluminous suite of biotite-, two-mica-, muscovite-, and commonly garnet-bearing bodies, formed 1822 ± 5 Ma ago (U–Pb, monazite) through partial melting of metagraywackes, with a probable contribution from early orogenic granitic lithologies (Claesson & Lundqvist 1990, 1995). The granites commonly evolve to a pegmatitic facies, and are typically accompanied by extensive schorl- and garnet-bearing pegmatitic granites and pegmatites. The



- Metagreywacke and early orogenic granitoids - gneisses and migmatites
- Metagreywacke
- Granite
- Pegmatitic granite
- Towns
- Lakes
- Graftonite localities:
1. Tjålmsjön
 2. Berg
- Triphylite localities:
3. Stockberget
 4. Gammbodberget
 5. Rotbäcken
 6. Hattkullberget

FIG. 2. Geological map of the Sollefteå region, with locations of phosphate-bearing pegmatites. The map is modified after Lundqvist *et al.* (1990).

pegmatites yield U-Pb ages from columbite between 1803 and 1794 Ma (Romer & Smeds 1994, 1997).

Approximately 100 dikes of rare-element pegmatites are hosted by andalusite-grade terranes of the basin (Smeds 1990, 1992, 1993). The dikes commonly contain minerals of Sn or Li, or both. Phosphates of the graffonite - beusite series occur in five areas: Sollefteå, Tjålmsjön - Rotbäcken, Räggen, Sidsensjö - Hinnsjön and Bräntlandet (Figs. 2 to 5). The graffonite-beusite-bearing pegmatite bodies are poorly zoned to well differentiated, with fine-grained border zones coarsening (locally through graphic units) to intermediate zones and a blocky K-feldspar + quartz core, segregated in some cases into feldspathic, phosphate-bearing core-margin and quartz core proper. The beryl-type dikes that carry the Ca-Fe-Mn-rich phosphates (Table 2) are located either closely adjacent to the Härnö peraluminous granites and associated barren pegmatites

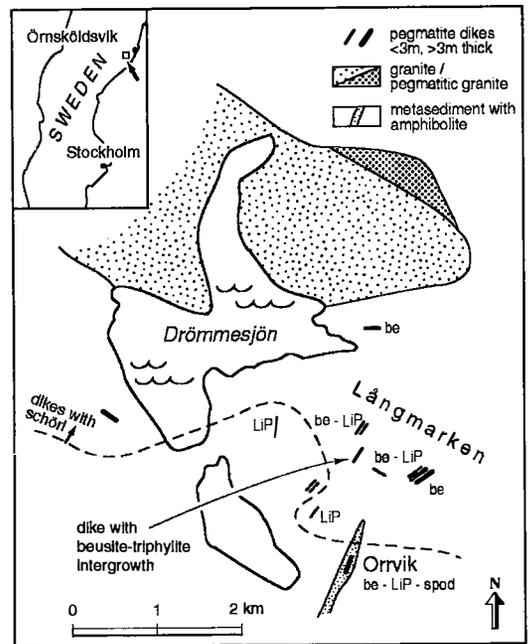


FIG. 3. Geological map of the eastern part of the Sidsensjö - Hinnsjön region. LiP: occurrence of triphylite, be: beryl.

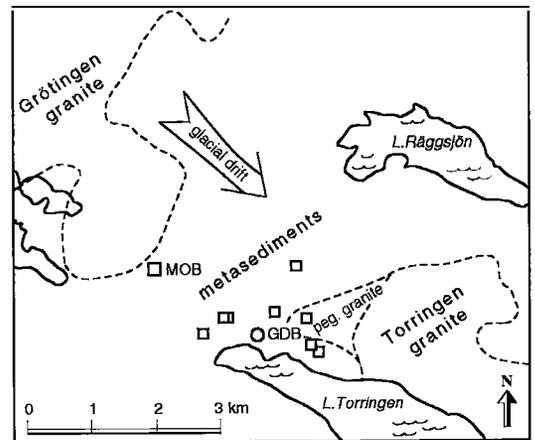


FIG. 4. Geological map of the Räggen area. MOB: the pegmatite Sn prospect, GDB: site of the graffonite-bearing pegmatite boulder.

(Sollefteå), or within the beryl + columbite + triphylite regional zone (Räggen, Sidsensjö - Hinnsjön). In other cases, the relationships between granites and different categories of pegmatites are not clearly defined at the present level of erosion (Tjålmsjön - Rotbäcken, Bräntlandet).

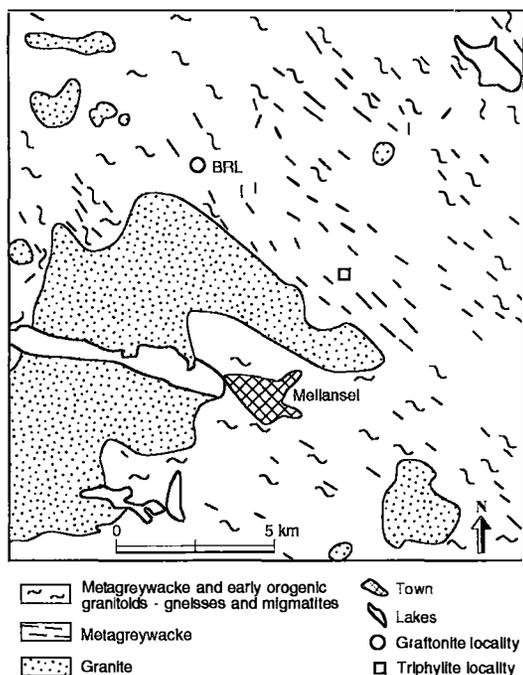


FIG. 5. Geological map of the Brantlandet area. The map is modified after Lundqvist (1987). BRL: Brantlandet pegmatite.

The Utö - Mysingen field

The bedrock of northern Utö (Fig. 6) comprises intricately folded Proterozoic metatuffitic rocks, associated with marble, skarn, oxide-facies iron formation, metasedimentary rocks and quartz porphyry (Stålhös 1982). The metasedimentary rocks are mainly well-preserved meta-argillites with muscovite, andalusite, cordierite and, locally, sillimanite. The sillimanite + cordierite + garnet-bearing metasedimentary rocks of the islets northwest of Utö Island, and of the southern part of the Utö - Nättarö chain of islands, are migmatized; they represent the southeasternmost part of the mainland sillimanite + cordierite + garnet + K-feldspar migmatites of the Sörmland gneisses to the northwest (Stålhös 1982).

A fine- to medium-grained, mildly peraluminous and garnetiferous two-mica granite, the Mysingen granite, outcrops on some islets in Mysingen Bay (Fig. 6). The Runmaren pegmatitic granite, located between the Mysingen granite and the rare-element pegmatites of northern Utö, consists of a mélange of fine-grained aplite, medium-grained two-mica granite, pegmatitic granite with graphic K-feldspar + quartz, and discrete pods of blocky potassic pegmatite.

TABLE 2. PRIMARY ASSEMBLAGE OF GRAFTONITE BEARING-DIKES IN SWEDEN

	BRG	TJS	RÄG	SID	BRL	PRH	ÄHN	SMH	RAN	NRR	BTS	NTT
Biotite	s				s	s	s	s	x	x		r
Muscovite	x	x	x	x	x	x	x	x	x	x	x	x
Garnet*	s	s			s	r	s	s		s	r	r
Schorl	x				s	x	s	s		s	s	s
Beryl			r			r	r		r	r		
Zircon							r		r	r	r	
Spinel											r	
Rutile											r	
Cassiterite			x				r				r	
Columbite						r	r		r	r	r	
Monazite (Ce, Nd)	r			r	r			r		r		
Triphylite	r	r	r	r	r	r	r	r	r	r	r	r
Graftonite	s	r	r	r	r	r	r	r	r	r	r	r
Beusite				r	r							r
Sarcopside	r	r	r	r	r	r	r	r	r	r	r	r
Wolfelite	r	r	r	r	r	r	r		r	r		r
Tripliodite				r			r					
Apatite	r	r		r	r	r	r		r	r	r	r
Zwieselite							r				r	
Amblygonite											r	
Sulfides and sulfosalts of As, Sn, Fe-Zn, Pb, Cu			r				r		r	r	r	r

Berg, BRG; Tjälmsjön, TJS; Räggen GDB, RÄG; Sidensjö, SID; Brantlandet, BRL; Stora Perisholmen, PRH; Ångsholmarna Norra, ÄHN; Smedshäll, SMH; Rånö, RAN; Norrö, NRR; Båtsudden, BTS; Nättarö, NTT.
X = abundant; s = subordinate; r = rare; microcline, albite and quartz are abundant in all dikes; * almandine-spessartine.

On the islets adjacent to and on the northwestern shore of northern Utö, bodies of schorl-bearing pegmatite exhibit textural features of a pegmatitic granite similar to those of the Runmaren intrusion. The most fractionated pegmatites of the region reside inside the northern part of Utö. Thus the pegmatites of the Utö - Mysingen field, dated at 1822-1815 Ma (U-Pb columbite: Romer & Smeds 1994) can be divided into four regional zones (Smeds & Černý 1992). From northwest to southeast, the zones consist of the (1) barren pegmatites in the Mysingen granite, (2) barren pegmatites in the Runmaren pegmatitic granite, (3a) pegmatite dikes with schorl + beryl + columbite + (Fe, Mn, Ca)-phosphates + triphylite, subparallel but discordant to the NE-trending foliation of the paragneisses, (3b) dikes containing schorl - columbite - triphylite subparallel but discordant to the north-easterly trending main foliation of the metatuffitic formation, and also discordant in the east-west direction, and (4) the Li-rich pegmatites of Utö at Nyköping-sgruvan with Li-aluminosilicates and pollucite, plus some other dikes (Smeds & Černý 1989, Teertsma *et al.* 1996), all discordant in the east-west direction to the metamorphic host-rocks.

In the southern part of the Utö archipelago (Utö *sensu lato*), the pegmatites correlate with the above-mentioned zone 3a. Two distinct bodies of pegmatitic granite are known in this region, the Mälsten and Sandskär intrusions, with associated pegmatite populations that tend to be slightly zoned down the regional metamorphic gradient to the southeast.

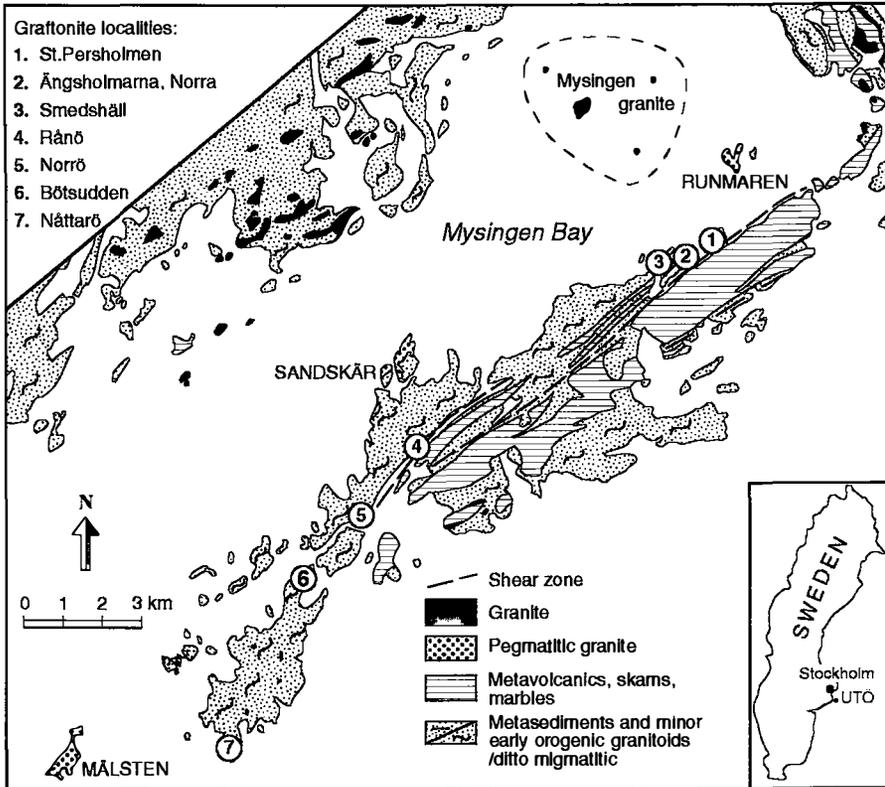


FIG. 6. Geological map of the Utö – Mysingen region, with graftonite localities. The map is modified after Stålhös (1982).

All graftonite-bearing pegmatites are confined to the zone 3a. They can roughly be described as poorly zoned assemblages of leucogranite, sodic aplite, aplite with blocky K-feldspar units, units of graphic K-feldspar + quartz megacrysts in leucogranitic matrix, and blocky quartz + K-feldspar. This last unit of potassic pegmatite carries the phosphates. The Norrö quarry (Eriksson 1946) is the only exception, containing a zone with cassiterite, amblygonite and other phases indicative of a perceptibly higher level of fractionation (Nysten & Gustafsson 1993).

General

Summarizing the above observations, we can conclude that the graftonite–beusite-based intergrowths are found as irregular nodules in blocky pods of K-feldspar + quartz (\pm book muscovite, albite to oligoclase), or locally in the adjacent intermediate unit of the parent pegmatites. Textural relationships indicate that the nodules enclose microscopic monazite-(Ce), and crystallized simultaneously with the adjacent

quartz, feldspars and muscovite. Wolfeite, zwieselite, ferroan wagnerite and fluor- (to hydroxyl-)apatite are the most commonly associated phosphates, locally with minor pyrite, arsenopyrite and (locally Cd-rich) sphalerite. The phosphates are found mainly along margins of the graftonite – beusite nodules. They apparently formed later than the graftonite – beusite, as also shown by local replacement of this phase by veinlets and dendritic aggregates of wolfeite.

Most of the phosphates were affected by secondary alteration, which generated minerals of the alluaudite group and ferrisicklerite – sicklerite series, rarely arrojadite, phosphoferrite – reddingite, and probable ludlamite. However, the secondary phosphates are beyond the scope of this study.

EXPERIMENTAL METHODS AND TREATMENT OF THE DATA

Electron-microprobe analyses of the phosphates were carried out in the wavelength-dispersion mode on a Cameca SX-50 instrument, with beam diameter of

1–2 μm and an accelerating potential of 15 kV, a sample current of 20 nA and a counting time of 20 s (40 s for F). The following standards were used: fluorapatite (for PK α , CaK α , FK α), almandine (FeK α), spessartine (MnK α), forsterite (MgK α) and gahnite (ZnK α). The following trace elements (<0.1 wt.% of oxide) were sought but not detected: Si (K α , diopside), Al (K α , pyrope), Sr (L α , SrBaNb₄O₁₂), Ba (L α , NaBa₂Nb₅O₁₂), Sc (K α , NaScSiO₄), Y (L α , YAG), La, Ce and Nd (L α , monazite), Na (K α , albite), K (K α , orthoclase) and Cl (K α , tugtupite). Data were reduced using the PAP routine of Pouchou & Pichoir (1985). Electron-microprobe analyses of the associated garnet were performed under conditions listed in Teertstra *et al.* (1996).

The conditions for the ICP (inductively coupled plasma) and INA (instrumental neutron-activation) analysis of K-feldspar from the Räggen and Sidensjö areas are quoted in Smeds (1992), those for the feldspar samples from the other occurrences in the Bothnian Basin and the Utö – Mysingen field are given in Černý *et al.* (1984).

The identities of the examined phosphates were verified by X-ray powder diffraction, using a Philips PW 1710 diffractometer.

Modal analyses were carried out with a Kontron-IBAS image-analysis system, using the differences in brightness of the back-scattered-electron images between graffonite or sarcopside and triphylite, or the differences in Ca distribution between Ca-rich graffonite and Ca-poor sarcopside. Areas of the scanned fields were in the range 16.8–88.6 mm². Some of the modal data are subject to experimental error introduced by the presence of alteration products indistinguishable from the primary phases. This is particularly noticeable in the analysis of triphylite lamellae in sarcopside, which were represented by the dark products of their oxidation and whose proportion is commonly low, thus increasing the relative error.

Densities of the phosphates, required for calculation of bulk composition of the intergrowths, were taken or interpolated from the literature. The microscopic scale of the aggregates made attempts at measurement of densities impractical. However, any error introduced in the density data is probably much smaller than the error involved in the modal analysis.

TEXTURAL RELATIONSHIPS

Three types of phosphate intergrowths involving graffonite – beusite were observed: (i) graffonite – beusite with lamellae of sarcopside, which in turn host lamellar triphylite, (ii) graffonite – beusite enclosing lamellae of triphylite, and (iii) granular aggregates of graffonite, triphylite and wolfeite.

Graffonite – beusite + sarcopside + triphylite

Graffonite – beusite is the principal host phase in these intergrowths. Sarcopside commonly forms a single set of subparallel platy lamellae, 30 to 500 μm thick. The lamellae are somewhat irregular in shape, with local bulges, constrictions and multipronged splits, but overall they are fairly straight in all sections observed (Fig. 7A). The individual lamellae, enclosed in extensive monocrystalline volumes of the graffonite – beusite host, commonly share a uniform optical orientation. A granular texture within the lamellae or the host phase is rare, and is associated with incipient deformation.

Triphylite constitutes one set (rarely, two sets) of lamellae 10 to 50 μm thick, hosted almost exclusively by sarcopside (Figs. 7A, B). The only triphylite observed in the surrounding graffonite – beusite represents straight but short extensions of the sarcopside-hosted lamellae, restricted to only the most immediate vicinity of sarcopside. The triphylite lamellae are usually flat and slightly lenticular, gradually tapering out toward their terminations. They typically pinch out at the boundary of sarcopside with graffonite – beusite, and swell inward (Figs. 7A, B). The triphylite lamellae are extensively altered to minerals of the ferrisicklerite – sicklerite series, but the electron-microprobe data on fresh relics identify triphylite as the primary phase.

Graffonite – beusite + triphylite

In this type, graffonite – beusite hosts three types of triphylite lamellae. A single set of platy lamellae between 100 and 1000 μm thick is most common (Fig. 7C); the lamellae are straight to gently sinuous. This type is routinely accompanied by much finer lamellae of the same orientation, between 5 and 50 μm thin. Locally, the difference between the two varieties is blurred by gradual change in size, but in most cases, the difference is quite distinctive (Fig. 7C). Acicular to rod-shaped individuals of triphylite, elongate parallel to the lamellae, constitute the third variety. They vary considerably in size, from $\sim 30 \times 40$ to $\sim 200 \times 500$ μm in cross section, flattened parallel to the associated lamellae of the first and second types (Fig. 7D).

An optical examination reveals that the individual lamellae of triphylite are monocrystalline in all three textural types. The optical orientation of platy triphylite is largely uniform over large volumes of graffonite – beusite, but the acicular to rod-shaped triphylite commonly shows a distinctly different orientation. Undulose extinction of the lamellae is locally widespread, and is occasionally transitional into randomly oriented subgrains. Extensive granular aggregation, which also disturbs the otherwise smooth contacts with graffonite – beusite, seems to be restricted to phosphate nodules affected by deformation. In these

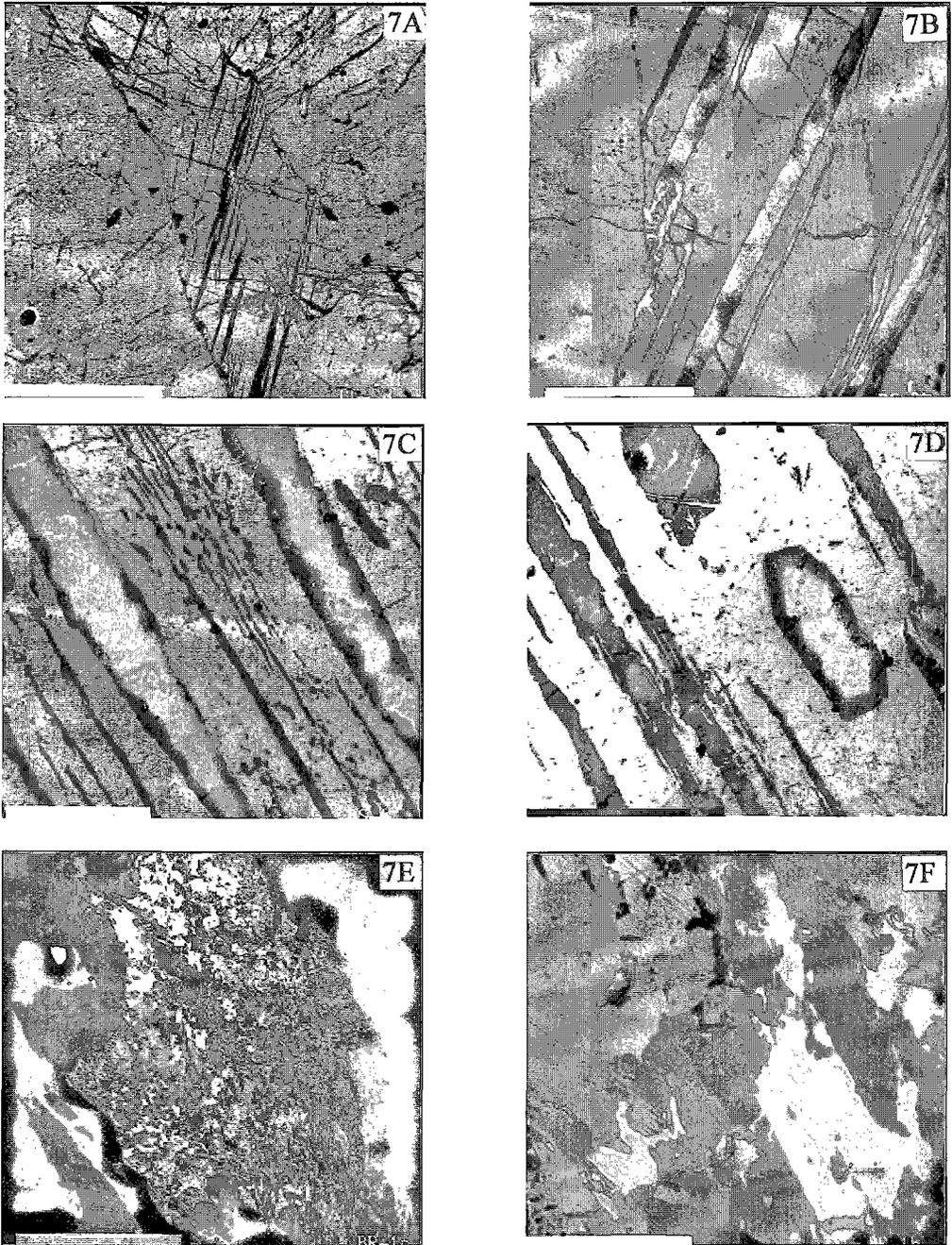


FIG. 7. Back-scattered-electron images of the graftonite–beusite-based intergrowths. A. Graftonite with coarse and bifurcating lamellae of sarcopside, which contain thin, in part oxidized lamellae of triphylite (black); Berg. B. Detail of oxidized triphylite lamellae (black) in sarcopside, tapering off toward the contact of sarcopside with graftonite (left side); Berg. C. Coarse and fine lamellae of triphylite (black) in graftonite; Böttsudden. D. Lamellae and rods of triphylite (black) in graftonite; Böttsudden. E. Granular aggregate of graftonite, triphylite and wolfeite, rimmed by apatite (dark grey); Räggen GDB. F. Detail of E, showing variable composition of the graftonite (shades of medium grey); Räggen GDB. Scale bars represent 500 μm in A, C, D and E, 100 μm in B, and 50 μm in F.

cases, the normally single-crystal graftedonite – beusite host also may show a granular texture.

Granular aggregates

As mentioned above, granular aggregates are rarely observed, and only on a microscopic scale in phosphates affected by deformation. The single distinct exception is the phosphate nodule from Räggen. It is mainly granular, and consists of an intimate mixture of graftedonite, triphylite, rare sarcopside, and wolfeite,

rinned by apatite. Graftedonite is the dominant phase, coarser-grained than the subordinate triphylite and wolfeite (≤ 150 and ≤ 70 μm , respectively). The fabric of the aggregate shows distinct preferred orientation of the constituent grains, suggestive of stress-controlled crystallization (Figs. 7E, F). A strikingly undulose extinction of the individual grains, shear zones, and kinking of associated muscovite also indicate stress-induced deformation. A lamellar intergrowth of graftedonite and triphylite was encountered only in a single small aggregate.

TABLE 3. REPRESENTATIVE CHEMICAL COMPOSITIONS OF PHOSPHATES IN THE LAMELLAR GRAFTONITE-BEUSITE+SARCOPSIDE+TRIPHYLITE INTERGROWTHS FROM SWEDISH PEGMATITES

wt. %	BRG			BRL			ÄHN			NRR		
	Gra	Sar	Tph	Gra	Sar	Tph	Gra	Sar	Tph	Gra	Sar	Tph
P ₂ O ₅	40.60	40.08	46.43	40.46	40.19	45.47	40.89	40.33	46.22	41.07	40.45	47.32
FeO	24.84	39.49	27.63	27.53	45.80	32.93	31.99	48.63	36.11	30.42	47.08	33.93
MnO	24.26	15.80	11.40	18.94	10.91	7.35	16.76	8.88	5.74	16.84	9.51	5.53
MgO	0.88	2.53	3.65	0.66	1.84	3.11	0.40	1.20	1.95	1.05	3.02	4.49
CaO	7.69	0.00	0.00	10.07	0.06	0.01	8.31	0.06	0.05	10.14	0.01	0.03
ZnO	0.09	0.00	0.00	0.24	0.07	0.01	0.31	0.09	0.19	0.25	0.06	0.00
Li ₂ O ¹	-	-	9.70	-	-	9.57	-	-	9.66	-	-	9.94
SUM	98.36	97.90	98.81	97.90	98.87	98.45	98.66	99.19	99.92	99.77	100.13	101.24
P	2.009	2.001	2.016	2.005	2.009	2.001	2.018	2.016	2.014	1.998	1.991	2.004
Fe	1.214	1.957	1.185	1.348	2.262	1.432	1.560	2.402	1.555	1.462	2.289	1.419
Mn	1.201	0.793	0.495	0.939	0.546	0.324	0.828	0.444	0.250	0.820	0.468	0.234
Mg	0.077	0.223	0.279	0.058	0.162	0.241	0.035	0.106	0.150	0.090	0.262	0.335
Ca	0.482	0.000	0.000	0.632	0.004	0.001	0.519	0.004	0.003	0.624	0.001	0.002
Zn	0.004	0.000	0.000	0.010	0.003	0.000	0.013	0.004	0.007	0.011	0.003	0.000
Li	-	-	2.000	-	-	2.000	-	-	2.000	-	-	2.000
ΣR	2.977	2.973	3.960	2.987	2.977	3.997	2.955	2.960	3.965	3.006	3.022	3.990
Mn#	0.497	0.288	0.295	0.411	0.194	0.185	0.347	0.156	0.139	0.359	0.170	0.142

Atomic contents normalized to 8 oxygen atoms. ¹calculated by charge-balancing for 8 oxygen atoms and a fixed content of 2 lithium atoms. Locality acronyms: see Table 2. Gra=graftonite-beusite, Sar = sarcopside, Tph = triphylite.

ΣR=Fe²⁺+Mn²⁺+Mg²⁺+Ca²⁺+Zn²⁺+Li⁺. Mn# = Mn/(Mn+Fe) at.

TABLE 4. REPRESENTATIVE CHEMICAL COMPOSITIONS OF PHOSPHATES IN THE LAMELLAR GRAFTONITE – TRIPHYLITE INTERGROWTHS FROM SWEDISH PEGMATITES

wt. %	TJS		SID		PRH		RAN		BTS		NTT	
	Gra	Tph	Gra	Tph	Gra	Tph	Gra	Tph	Gra	Tph	Gra	Tph
P ₂ O ₅	40.93	45.73	40.13	45.90	41.12	47.91	41.46	48.38	41.81	48.23	42.70	48.37
FeO	29.44	34.43	22.70	25.92	21.38	24.21	28.64	29.51	28.70	28.24	22.11	26.93
MnO	18.17	7.60	29.87	18.47	24.18	12.31	17.14	5.57	17.65	5.95	20.30	10.18
MgO	0.44	1.83	0.10	0.34	1.13	4.79	1.95	7.73	2.18	8.16	1.22	4.85
CaO	9.69	0.06	5.45	0.11	10.86	0.02	9.92	0.04	9.60	0.01	13.03	0.03
ZnO	0.04	0.09	0.19	0.00	0.01	0.00	0.04	0.01	0.02	0.00	0.09	0.00
Li ₂ O ¹	-	9.58	-	9.60	-	9.89	-	10.19	-	10.20	-	10.00
SUM	98.71	99.32	98.44	100.34	98.68	99.13	99.15	101.43	99.96	101.24	99.45	100.36
P	2.013	2.009	2.008	2.014	2.008	2.039	2.009	2.000	2.008	1.991	2.038	2.036
Fe	1.430	1.494	1.122	1.123	1.031	1.018	1.371	1.205	1.362	1.151	1.042	1.120
Mn	0.894	0.334	1.495	0.811	1.181	0.524	0.831	0.230	0.848	0.246	0.969	0.429
Mg	0.038	0.142	0.009	0.026	0.097	0.359	0.166	0.563	0.184	0.626	0.103	0.360
Ca	0.603	0.003	0.345	0.006	0.671	0.001	0.608	0.002	0.584	0.001	0.787	0.002
Zn	0.002	0.003	0.008	0.000	0.000	0.000	0.002	0.000	0.001	0.000	0.004	0.000
Li	-	2.000	-	2.000	-	2.000	-	2.000	-	2.000	-	2.000
ΣR	2.967	3.977	2.980	3.966	2.981	3.902	2.978	4.000	2.979	4.023	2.905	3.910
Mn#	0.385	0.183	0.571	0.419	0.534	0.340	0.378	0.160	0.384	0.176	0.482	0.277

Atomic contents normalized to 8 oxygen atoms. ¹calculated by charge-balancing for 8 oxygen atoms and a fixed content of 2 lithium atoms. Locality acronyms: see Table 2; other symbols: see Table 3.

TABLE 5. REPRESENTATIVE CHEMICAL COMPOSITIONS OF PHOSPHATES IN THE GRANULAR INTERGROWTHS

wt. %	RÄG			
	Gra	Gra	Tph	Wol
P ₂ O ₅	40.85	41.60	46.52	31.94
FeO	24.01	20.78	32.32	39.27
MnO	22.21	20.37	10.74	20.14
MgO	0.21	0.09	1.11	0.96
CaO	10.47	15.33	0.21	0.16
ZnO	0.27	0.10	0.11	0.21
Li ₂ O ¹	-	-	9.70	-
H ₂ O ²	-	-	-	3.97
SUM	98.04	98.27	100.71	96.65
P ⁵⁺	2.018	2.022	2.019	1.020
Fe ²⁺	1.172	0.998	1.386	1.239
Mn ²⁺	1.098	0.991	0.466	0.644
Mg ²⁺	0.020	0.008	0.085	0.054
Ca ²⁺	0.655	0.943	0.012	0.006
Zn ²⁺	0.012	0.004	0.004	0.006
Li ⁺	-	-	2.000	-
OH ⁻	-	-	-	1.000
ΣR	2.955	2.944	3.953	1.949
Mn#	0.484	0.498	0.252	0.342

Atomic contents normalized to 8 oxygen atoms;
¹calculated by charge-balancing for 8 oxygen atoms and a fixed content of 2 lithium atoms;
²calculated by charge-balancing for 5 oxygen atoms and a fixed content of 1 H⁺.
 RÄG=Räggen, Wol=wolfeite. Other symbols: see Table 3.

CHEMICAL COMPOSITION OF THE MINERALS

Representative compositions of graftonite, beusite, sarcopsite, triphylite and wolfeite are shown in Tables 3, 4 and 5.

Compositions of individual phases

Minerals of the *grafonite – beusite series* belong in most cases to graftonite proper, but a few samples from Persholmen, Sidensjö and Berg are slightly Mn-dominant and are classified as beusite. The Ca content varies from 11 to 32 mol.% Ca₃(PO₄)₂. The Mg content is highly variable but moderate; it tends to show modestly positive correlation with Ca. Zinc is the only minor element detected in some samples in quantifiable concentrations.

Sarcopsite invariably shows Fe dominant over Mn. The Mg content is variable and generally high, attaining on average 20 mol.% of Mg₃(PO₄)₂ at Persholmen. In contrast, Ca and Zn contents are negligible.

Triphylite also is Fe-dominant in all samples examined. Calcium and Zn contents are as low as in sarcopsite, but the Mg content is in some cases very high, up to 37 mol.% of the LiMgPO₄ component at Bötsudden.

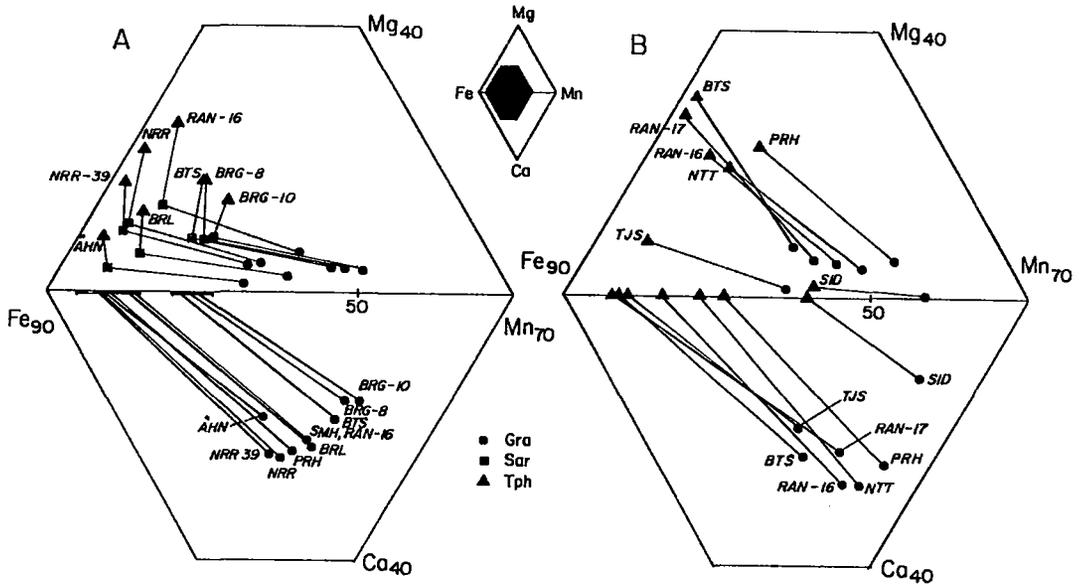


Fig. 8. Compositions of graftonite (Gra), sarcopsite (Sar) and triphylite (Tph) in the Fe – Mn – Mg and Fe – Mn – Ca (at.) diagram, averaged per locality or per specific sample (see Table 1 for locality acronyms). A. Compositions of phosphates composing the graftonite + sarcopsite + triphylite intergrowths (in the Ca diagram, the virtually Ca-free sarcopsite and triphylite plot collectively within the ranges indicated by heavy bars along the Fe–Mn sideline). B. Compositions of phosphates composing the graftonite – beusite + triphylite intergrowths.

Wolfeite from Räggen is strongly Fe-dominant, and its F content is below the detection limit under the conditions of routine electron-microprobe analysis (0.3 wt.%); thus the mineral seems to represent a virtually pure (OH) end-member. The Mg and Ca contents are minor, <3 mol.% $Mg_2(PO_4)(OH)$ and <0.25 mol.% $Ca_2(PO_4)(OH)$.

Cation partitioning among coexisting phases

In the *grafonite* – *beusite* + *sarcopside* + *triphylite* intergrowths shown in Figure 8A, *grafonite* – *beusite* shows strong preference for Mn, whereas both *sarcopside* and *triphylite* are Fe-rich. *Grafontite* also concentrates Ca, to its virtual exclusion from the other two phosphates. In contrast, Mg is strongly preferred by *triphylite*; *sarcopside* and particularly *grafonite* – *beusite* are Mg-poor.

Figure 8B shows the compositional relationships in the *grafonite* + *triphylite* intergrowths. They are very close to those observed between these two minerals in the three-membered assemblages discussed above: *grafonite* shows strong preference for Ca and Mn, whereas *triphylite* concentrates Fe and Mg.

The cation distribution between *grafonite* and *triphylite* in the *granular aggregates* from Räggen is analogous to that observed in the lamellar intergrowths,

in particular to that shown in Figure 8B for the Tjålmsjön compositions. However, the Räggen *grafonite* is distinctly heterogeneous (Table 5), and the cation partitioning may be affected by the presence of *wolfeite*. Also, grains of different phases that may represent minerals coexisting at a very local scale are difficult to identify. Thus the Räggen compositions were not included in quantitative aspects of partitioning.

Table 6 shows the distribution coefficients for the major cations (except Li) in coexisting *grafonite* – *beusite*, *sarcopside* and *triphylite*. As suggested by the graphic representation of the phosphate compositions in Figure 8, the distribution coefficients are in most cases quite uniform across diverse localities, with only small deviations from average values. This applies to K_D values of Fe and Mn, in particular; the K_D values also are statistically identical for corresponding pairs of minerals in both types of intergrowths. In contrast, K_D for Mg displays a much broader spread of values.

Bulk compositions of the intergrowths

Table 7 shows the modal proportions of selected intergrowths of phosphates that were considered suitable for image analysis, and the bulk compositions calculated from the compositions, modes and densities of the individual phases. Figure 9 summarizes the relationships between the compositions of individual phosphates and the calculated bulk-compositions of the intergrowths.

The bulk composition of the *grafonite* + *sarcopside* + *triphylite* intergrowths is variable in terms of the Ca and Li contents, controlled by the proportions of *grafonite* – *beusite* and *triphylite*, respectively. The cation contents of the bulk compositions normalized to 8 atoms of oxygen total very close to 3, as the two main phases have the same stoichiometry, $R^{2+}_3P_2O_8$. The slight excess of cations is proportional to the content of

TABLE 6. CATION DISTRIBUTION COEFFICIENTS (K_D) FOR PHOSPHATES COEXISTING IN LAMELLAR INTERGROWTHS FROM SWEDISH PEGMATITES

K_D		BRG	BRG	BRL	PRH	ÅHN	SMH	RAN	NRR	NRR	BTS
		gst									
Fe	S/G	1.56	1.60	1.55	1.53	1.52	1.53	1.60	1.62	1.58	1.57
	T/S	0.74	0.70	0.74	alt.	0.75	alt.	0.68	0.69	0.73	0.70
	T/G	1.15	1.12	1.15	alt.	1.13	alt.	1.10	1.12	1.15	1.11
	G/B1	0.83	0.82			0.86					
	B2/B1	1.27	1.27			1.32					
	T/B2	0.75	0.72			0.74					
	S/G	0.66	0.66	0.59	0.50	0.55	0.58	0.58	0.56	0.56	0.66
Mn	T/S	0.67	0.75	0.66	alt.	0.67	alt.	0.61	0.60	0.61	0.68
	T/G	0.44	0.49	0.39	alt.	0.37	alt.	0.36	0.34	0.34	0.44
	G/B1	1.14	1.14			1.13					
	B2/B1	0.74	0.74			0.62					
	T/B2	0.68	0.76			0.67					
	S/G	2.74	2.87	2.92	3.23	2.88	2.84	2.85	3.07	3.02	1.61
	T/S	1.60	1.37	1.65	alt.	1.65	alt.	1.50	1.53	1.37	1.57
Mg	T/G	4.40	3.93	4.82	alt.	4.75	alt.	4.27	4.71	4.13	2.52
	G/B1	0.57	0.55			0.63					
	B2/B1	1.69	1.69			1.89					
	T/B2	1.49	1.28			1.58					
	K_D	SID	TJS	RÄG	PRH	RAN	RAN	BTS	NTT		
		gt									
	Fe	T/G	1.16	1.17	1.17	1.19	1.21	1.05	1.03	1.22	
T/B		1.09			1.09			1.03			
G/B		0.94			0.92			0.99			
Mn	T/G	0.63	0.42	0.38	0.48	0.43	0.32	0.34	0.49		
	T/B	0.70			0.59			0.44			
	G/B	1.12			1.23			1.31			
Mg	T/G	3.80	4.07	4.23	4.34	4.33	4.24	4.11	4.10		
	T/B	1.90			1.87			1.98			
	G/B	0.50			0.43			0.48			

All data in weight proportions; gst=grafonite-beusite + sarcopside + triphylite intergrowths; gt=grafonite-beusite + triphylite intergrowths; G=grafonite-beusite; S=sarcopside; T=triphylite; B1=bulk composition for gst; B2=bulk composition for sarcopside + triphylite; B=bulk composition for gt; alt.=alteration of triphylite to alluaudite-group minerals. Locality acronyms: see Table 2.

TABLE 7. BULK COMPOSITIONS OF LAMELLAR PHOSPHATE INTERGROWTHS FROM SWEDISH PEGMATITES

	BRG ₁	BRG ₂	ÅHN	BRG ₁	BRG ₂	ÅHN	SID	PRH	BTS
	B1	B1	B1	B2	B2	B2	B	B	B
Gra	62.7	62.4	72.9				66.3	60.0	63.6
Sar	34.0	31.6	25.8	91.2	83.8	95.2			
Tph	3.3	6.1	1.3	8.8	16.2	4.8	33.7	40.0	36.4
P ₂ O ₅	41.32	41.82	41.17	41.40	42.09	40.83	42.54	44.76	44.11
FeO	31.08	29.83	36.50	39.59	37.85	48.15	23.90	22.60	27.91
MnO	20.71	21.48	14.99	15.27	15.80	9.27	26.37	18.54	13.25
MgO	1.68	1.62	0.63	2.83	2.76	1.19	0.16	3.03	4.13
CaO	4.62	4.65	6.34	0.03	0.03	0.05	3.77	7.09	7.00
ZnO	0.29	0.04	0.27	0.07	0.00	0.08	0.15	0.10	0.05
Li ₂ O	0.30	0.56	0.11	0.81	1.47	0.43	3.11	3.88	3.55
ΣUM	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
P ³⁺	2.010	2.020	2.013	2.009	2.019	2.014	2.015	2.030	2.008
Fe ²⁺	1.493	1.425	1.763	1.897	1.793	2.346	1.118	1.013	1.255
Mn ²⁺	1.008	1.038	0.733	0.741	0.758	0.458	1.249	0.841	0.603
Mg ²⁺	0.144	0.138	0.054	0.242	0.233	0.103	0.013	0.242	0.331
Ca ²⁺	0.284	0.284	0.392	0.002	0.002	0.003	0.226	0.407	0.403
Zn ²⁺	0.012	0.002	0.012	0.003	0.000	0.003	0.006	0.004	0.002
Li ⁺	0.069	0.129	0.026	0.187	0.335	0.101	0.700	0.836	0.768
ΣR	3.010	3.014	2.980	3.071	3.121	3.015	3.313	3.343	3.363
Mod	0.403	0.422	0.294	0.281	0.297	0.163	0.543	0.454	0.325

Gra, Sar, Tph = modal composition of lamellar intergrowths in volume %. Oxides in weight %. Atomic contents normalized to 8 oxygen atoms. Acronyms and symbols: see Tables 2, 3 and 6.

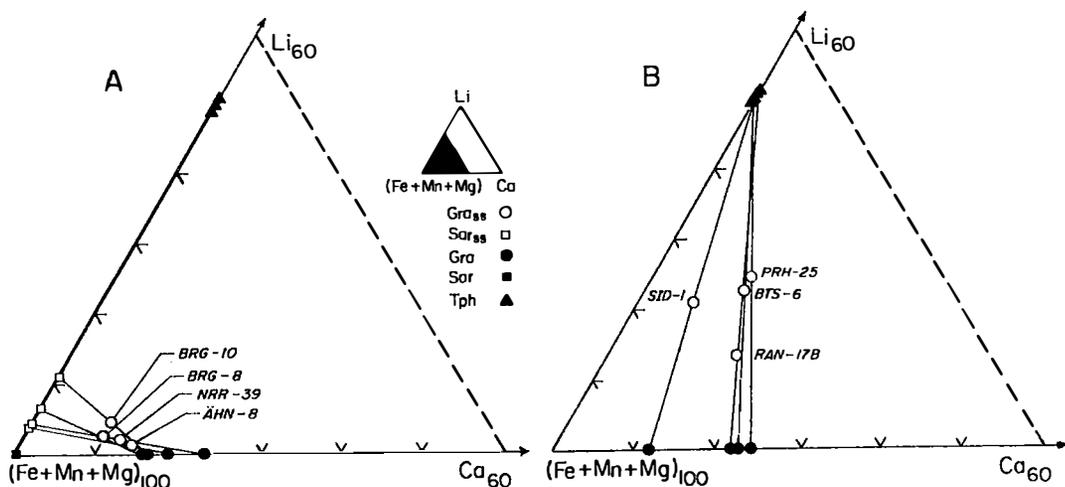


FIG. 9. Bulk compositions of the phosphate intergrowths in the (Fe + Mn + Mg)–Ca–Li (at.) diagram, averaged per individual samples (see Table 1 for locality acronyms). A. Bulk compositions of the graffonite + sarcopside + triphylite intergrowths (Gr_{ss}) and the sarcopside + triphylite intergrowths compared to the graffonite host. B. Bulk compositions of the graffonite + beusite + triphylite intergrowths (Gr_{ss}) and their component phases.

Li, hosted by triphylite $LiR^{2+}PO_4$ with a higher cation:oxygen ratio. This excess of cations, close to 50% of the total Li determined, is enhanced in the bulk compositions of the subsystem *sarcopside + triphylite*, in which the proportion of triphylite lamellae is increased relative to that of their sarcopside host.

In contrast to the preceding cases, the phase proportions in the *graffonite + triphylite* intergrowths are very similar in three out of four analyzed samples, but the ratio (Fe,Mn,Mg)/Ca is distinctly different. The above-mentioned type of cation excess, equal to 50% of the total Li present, is very prominent in these intergrowths.

LEVEL OF FRACTIONATION OF THE GRAFFONITE-BEUSITE-BEARING PEGMATITES

Two geochemical aspects of the environments generating graffonite + beusite are of interest: (i) the K/Rb value of K-feldspar indicative of the general grade of geochemical evolution of pegmatites, which decreases from barren to lithium-enriched dikes, and (ii) the Mn/(Mn + Fe) value of garnet as the main ferromanganous phase consistently present in the phosphate-bearing pegmatites.

The K/Rb value of blocky K-feldspar is somewhat variable among the various graffonite + beusite-bearing pegmatites. The overall variation extends from 78 to 234, but the ranges are smaller in individual dikes (Table 8). The data are intermediate relative to the ranges shown by the total zoned populations of

the parent pegmatite groups, which is consistent with the intermediate location of the phosphate-bearing pegmatites in the regional zoning patterns.

Geochemical data from a single locality provide the only information about the relationship between the graffonite + beusite + sarcopside + triphylite and graffonite + beusite + triphylite intergrowths. At St. Persholmen, the first type [with Mn/(Mn + Fe) of 0.40 to 0.38 for graffonite and 0.18 to 0.17 for sarcopside] was found in a blocky pod with K/Rb of the K-feldspar varying from 234 to 196, whereas the second type [with Mn/(Mn + Fe) of 0.55 to 0.51 for beusite and 0.40 to 0.28 for triphylite] is hosted by another pod in which the K-feldspar has a K/Rb value of 114 to 102. The higher degree of alkali fractionation in the K-feldspar correlates with the Mn- and Li-enriched, Fe-depleted phosphates.

Correlation of Mn/(Mn + Fe) in garnet with that of the graffonite + beusite-based intergrowths is somewhat complicated. In some of the pegmatite bodies examined, garnet is found in the blocky core-margin assemblages, but usually separate from the phosphate nodules; it was found in contact with the phosphates only in one occurrence, at Nättarö. In other dikes, garnet occurs only in the outer zones, characterized by a granitic to aplitic texture. As garnet commonly undergoes enrichment in Mn relative to Fe during the course of solidification of individual pegmatite bodies, compositional correlation with the phosphates located in separate units is rather tenuous. Also, the systematic differences in Mn/(Mn + Fe) between the individual phases of the phosphate

TABLE 8. FRACTIONATION INDICATORS IN PHOSPHATES, GARNET, AND BLOCKY K-FELDSPAR FROM THE GRAFTONITE-BEUSITE BEARING PEGMATITES OF SWEDEN

	Phosphates			Garnet		K-feldspar	
	graftonite	sarcopside	triphylite	aplitic, granite	pegmatitic units	range in pegmatite	range in field
	Mn/(Mn+Fe)					K/Rb	
Berg	0.41-0.51	0.26-0.32	0.26-0.32	0.40-0.50	0.36-0.38	121-177	
Bråntlandet	0.39-0.42	0.20-0.23	0.18-0.19				
Räggen, GDB	0.33-0.49	0.20-0.21	0.16-0.30			78	34-110
Sidensjö beusite peg.	0.57-0.60		0.40-0.44			69	17-310
Tjålmsjön	0.38-0.42		0.17-0.21	0.37-0.39		88-129	
Stora Persholmen	0.38-0.56	0.17-0.19	0.28-0.39	0.24-0.26		102-234	6-376
Ångsholmarna Norra	0.36-0.49	0.16-0.19	0.13-0.25		0.37-0.38	91-164	6-376
Smedshäll	0.40-0.43	0.20-0.22	0.15-0.16	0.24-0.25		96	6-376
Rånö	0.41-0.47	0.20-0.22	0.17-0.23				
Norrö	0.36-0.39	0.17-0.22	0.14-0.21		0.33-0.35	30-160	
Bötsudden	0.38-0.46	0.26-0.28	0.15-0.26		0.36-0.38		
Nättarö	0.47-0.52		0.27-0.31		0.31-0.34*		

All data in wt.% ratios; * garnet coexisting with graftonite.

intergrowths make simple correlations difficult, as bulk compositions of the phosphate nodules are only available for a limited number of occurrences. Nevertheless, Table 8 shows that in most cases, Mn/(Mn + Fe) of the garnet falls within the range of values of the main phases of the phosphate nodules, and very close to the values of the bulk compositions of the phosphate intergrowths.

DISCUSSION

Crystal-chemical relationships of the phosphates

The composition of the monoclinic *graftonite* – *beusite* (Ca,Fe,Mn)₃(PO₄)₂ is very variable in terms of Mn/(Mn + Fe), Ca content (0 to 0.98 Ca atoms per formula unit, *apfu*), and Mg content. The largest of the three cation sites, *M1*, accommodates preferentially Ca in the sequence Ca > Mn > Fe; coordination decreases with diminishing Ca content from 8- to 6-fold, tending to 5-fold (Calvo 1968, Wise *et al.* 1990, Steele *et al.* 1991). The *M2* site is 5-fold-coordinated and prefers Fe (Nord & Ericsson 1982), whereas *M3* is 6- to 5-fold coordinated and is the second choice for Mn (after *M1*), at least in Ca-free phases. The structure contains open interstices that could accommodate additional cations of small to medium radius (less than or equal to ~0.70 Å).

The composition of *triphylite* (– *lithiophilite*) Li(Fe,Mn)PO₄ is less variable relative to *graftonite* – *beusite*, complicated only by a largely subordinate Mg

substitution for (Fe,Mn). The orthorhombic structure corresponds to that of an olivine or, more specifically, monticellite (Finger & Rapp 1970), based on octahedrally and tetrahedrally coordinated sites within a hexagonally closest-packed array of oxygen atoms.

Natural *sarcopside* (Fe,Mn,Mg)₃(PO₄)₂ is Mn-poor, and its Mg content varies considerably but is minor. The stoichiometry of *sarcopside* is identical with that of *graftonite* – *beusite*, but the mineral is isotypic with *triphylite* – *lithiophilite* (Mrose & Appleman 1961, Moore 1972). The only structural difference is a slight monoclinic distortion, and ordered vacancies in 50% of the *M1* octahedral positions; thus the derivation of *triphylite* from *sarcopside* is achieved by the substitution Li₂(□R²⁺)₋₁. *Sarcopside* can be considered a polymorph of Ca-free *graftonite*, which is so far known only from meteorites (Steele *et al.* 1991).

In contrast to the above relationships, *graftonite* – *beusite* and *triphylite* differ in both structure and stoichiometry. However, Li can be accommodated in the *M2* sites of *graftonite* – *beusite* and in adjacent interstices by the substitution Li₂(□R²⁺)₋₁, as suggested by Moore (1982) and confirmed by M.A. Wise (in prep.).

Origin of the *graftonite*–*beusite*-based intergrowths

The phosphate intergrowths have been interpreted in three ways: (i) exsolution was advocated by Peacor & Garske (1964), Hurlbut (1965) and Hurlbut & Aristarain (1968) for *graftonite* + *sarcopside*, Peacor

(1969) for sarcopside + triphylite, Fransolet (1977) for sarcopside + triphylite, Wise & Černý (1990) for (grafonite-) beusite + triphylite, and by Moore (1972) for all of these cases; (ii) metasomatic replacement was favored by Fransolet (1977), except for the case of sarcopside in triphylite, and Malló *et al.* (1995) for triphylite + sarcopside; (iii) simultaneous crystallization was considered a possibility for quasi-graphic or "pegmatitic" textures by Peacor (1969), Huvelin *et al.* (1971) and Fransolet (1977). However, the work of Wise (in prep.) and our results confirm exsolution as the exclusive process, as far as lamellar intergrowths are concerned.

In the *grafonite – beusite + sarcopside + triphylite* aggregates, exsolution proceeds in two stages: a homogeneous precursor unmixed to grafonite – beusite + lithian sarcopside, and triphylite exsolves from the lithian sarcopside. Textural relationships and stoichiometry, subparallel orientation of tie-lines connecting the compositions of coexisting phases (Fig. 8A) and uniform distribution-coefficients (Table 6) in various bodies of pegmatites, indicative of local equilibrium among the intergrown phosphates, all clearly point to exsolution of lithian sarcopside from grafonite – beusite. This interpretation also is supported by crystallographic control of orientation in grafonite + sarcopside intergrowths (Hurlbut 1965). Experimental work strongly suggests that sarcopside may be stable only at low subsolidus temperatures, as it is routinely generated under hydrothermal conditions at 500 to 300°C from a grafonite – beusite phase synthesized at ~800°C (Ericsson *et al.* 1986, Charalampides *et al.* 1988). A high-temperature, homogeneous, and relatively Ca,Mn-poor, Fe,Mg-enriched, Li-bearing grafonite phase evidently crystallizes in natural assemblages, probably with a highly disordered structure. This primary precursor exsolves into a Ca,Mn-enriched, Li-free grafonite host with virtually Ca-free, Fe, Mg-enriched and Li-bearing lamellae of disordered sarcopside (Wise, in prep.). The subsequent exsolution of triphylite from Li-bearing sarcopside concludes the overall trend toward stoichiometric and structurally ordered low-temperature phases.

For the *grafonite – beusite + triphylite* intergrowths, single-stage exsolution is supported by uniform distribution-coefficients of cations (Table 6), subparallel arrays of tie-lines connecting coexisting phases (Fig. 8B), inverse correlation between size and density of distribution of triphylite lamellae (Figs. 7C, D), and a structurally controlled orientation of the intergrown phosphates (Hurlbut & Aristarain 1968, Wise & Černý 1990), rarely violated by either incoherent nucleation of the precipitate or recrystallization. The homogeneous precursor must have been again a grafonite – beusite-like phase, but in most cases richer in Ca and generally much more enriched in Li relative to the lithian-sarcopside-exsolving phase above (Fig. 9). The Li incorporation attained up to 42% of the

M2 site population in the homogeneous parent (Table 7, PRH), which subsequently exsolved triphylite.

Graftonite – beusite in the regional zoning of pegmatite populations

The grafonite – beusite series and the exsolved sarcopside are among the geochemically most primitive phosphates of the rare-element pegmatites, exhibiting moderate to high contents of Ca and Mg. Consequently, in regional zoning of cogenetic pegmatite groups of the LCT family (Fig. 10), grafonite – beusite should signal the onset of the beryl – columbite – phosphate subtype, developing outward from the beryl or beryl – columbite category (Trueman & Černý 1982, Černý 1991). The exsolution lamellae of triphylite would then represent the first Li-bearing phase, preceding discrete grains of triphylite, which in turn precede the appearance of lithium silicates (Moore 1982). Unfortunately, there is virtually no information available about the regional setting of the reported grafonite – beusite occurrences in the literature. Meintzer (1987) and Wise & Černý (1990) reported the beusite + triphylite intergrowth from the inner boundary of triphylite occurrences within the beryl – columbite (– phosphate) zone of the PEG group, Yellowknife field, with most of the triphylite localities spread outward, including the outermost zone of spodumene-bearing pegmatites. The occurrences of grafonite in the pegmatite field of the southern Black Hills, in South Dakota, reside in the beryl – columbite – phosphate pegmatites, exterior to the parent granite and to the immediately adjacent barren pegmatites.

In the Swedish pegmatite populations studied here, two patterns of the grafonite – beusite position in zoned pegmatite populations can be distinguished. In some cases, the grafonite – beusite-based intergrowths are the first Fe,Mn-phosphates encountered in the beryl-type pegmatites, before the appearance of discrete triphylite. In other terranes, grafonite – beusite is found within the regional zone of beryl – columbite – triphylite pegmatites.

The first case is best exemplified by the Utö – Mysingen field. It is well documented in the Runmaren group, and also suggested in the southern Mälsten – Sandskär group (Smeds *et al.* 1996). A narrow zone of beryl – columbite – grafonite – beusite pegmatites follows after barren dikes, whereas pegmatites with discrete triphylite are disposed still farther to the southeast (Fig. 6). The same pattern is shown at Sollefteå; the Berg pegmatite is closely adjacent to (or within) a pegmatitic granite, but the triphylite-bearing dikes are ~8–10 km to the west (Fig. 2).

In contrast, the Räggen grafonite-bearing dike is well within the beryl – columbite – triphylite regional zone of a well-zoned pegmatite group, despite the uncertain provenance of the drift boulder that represents the GDB locality (Fig. 4). The erosional surface exposes here a section through the regional zoning

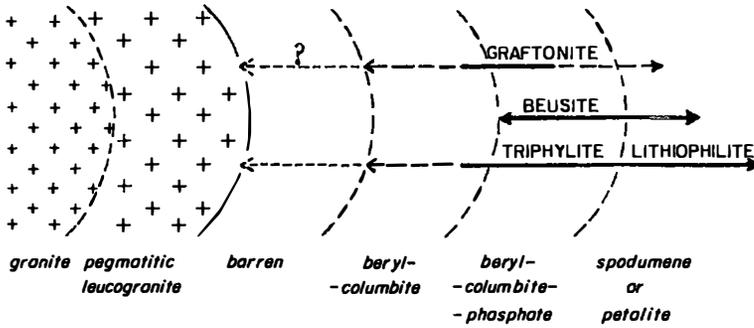


FIG. 10. Schematic distribution of grafftonite – beusite occurrences in regional zoned populations of granitic pegmatites. Heavy arrow shafts: main distribution, heavy dashes: local extensions, light dashes: relatively rare extensions of the main ranges.

populated by beryl – columbite – triphylite dikes, including an occurrence of triphylite inside the parent body of pegmatitic granite (Fig. 4). The same is true for the regional setting of the Långmark beusite-bearing dike in the Sidensjö – Hinnsjön area (Fig. 3).

In the Sollefteå area, two topographically distinct pegmatite populations are distinguished: the Tjålsjön – Rotbäcken dikes are separated from the others (Fig. 2). However, the distribution of the phosphate-bearing pegmatites, and the spatial relationship between the granites and the pegmatites, are such that no definite conclusions can be drawn about their genetic links and regional zoning. However, the grafftonite-bearing dikes invariably reside in, or closer to, migmatitic terranes that are associated with the Härnö-type granites, whereas the triphylite-bearing pegmatites are located further down the metamorphic gradient, in andalusite-bearing metasedimentary units. The metamorphic environments of the grafftonite- and triphylite-bearing pegmatites in the Bräntlandet area are closely analogous; the two pegmatites encountered here are equidistant from the closest outcrops of Härnö-type granite (Fig. 5).

The regional distribution of the grafftonite – beusite + triphylite intergrowths relative to that of the grafftonite – beusite + sarcopside + triphylite aggregates is largely ambiguous. At five localities, both types occur in a single body of pegmatite, and all other occurrences are isolated and randomly distributed.

Geochemical factors

The stabilization of the homogeneous precursors of the intergrowths requires appreciable enrichment of Ca and Mg, and low concentrations of F and Na, to prevent crystallization of apatite, wyllieite, fellowite, griphite, wagnerite or triplite – zwieselite. Such condi-

tions are encountered in relatively poorly evolved pegmatite-forming melts, generating oligoclase-bearing beryl – columbite pegmatites. However, these conditions are inconsistent with more fractionated pegmatites, particularly those hosting the Mn-dominant beusite. Very localized depletion in F may be responsible for these cases, although coexistence of beusite with F-bearing triplite and Na-bearing fellowite is locally observed (Černý *et al.* 1998). At the Swedish localities, a late increase in F and Na generates rims of fluorapatite, zwieselite and ferroan wagnerite around the grafftonite-beusite-based intergrowths.

In view of the general character of the grafftonite-beusite-bearing pegmatites, their overall level of fractionation should be moderate. This is well borne out by the K–Rb data on blocky K-feldspar (Table 8). Note that the feldspar of the beusite-bearing pegmatites usually shows somewhat lower K/Rb values, and that the regional zoning of the pegmatite groups with beusite commonly extends into spodumene- or petalite-bearing dikes. This applies not only to the Swedish pegmatite populations examined here (Sidensjö – Hinnsjön, Runmaren), but also to other terranes [Los Aleros in Argentina: Hurlbut & Aristarain (1968); Kitee – Tohmajärvi, southeastern Finland: Kallio & Alviola (1975); Eräjärvi, southern Finland: Lahti (1981); PEG group in the Yellowknife field: Wise (1987, Wise & Černý 1990); Turkestan Range: Beus (1950)]. However, exceptions are documented from several terranes (Anderson *et al.* 1998, Černý *et al.* 1998); differential depth of erosion affecting individual pegmatite groups may play a significant role here.

London *et al.* (1995) examined silicate – phosphate equilibria in peraluminous pegmatite-forming melts and established a reaction relationship involving biotite, which converts with increasing P content to equivalent Fe–Mn–Mg phosphates (such as triplite, grafftonite –

sarcopside, triphylite – lithiophilite) at ~0.8 wt.% P_2O_5 in the melt. They also documented a reaction of spessartine (– almandine) to manganoan apatite, beusite or the above phosphates at ~0.4 to ≤ 2.0 wt.% P_2O_5 in the melt, with the higher values associated with the Fe-bearing phosphates. Our compositional data indicate that the precursors of the graffonite–beusite-based intergrowths were generated by local phosphorus-induced destabilization of garnet. Except for a single case in the Nättarö pegmatite, the phosphate nodules occur separated from garnet, and the Mn/(Mn + Fe) value of the broadly associated garnet is close to or overlaps that of the phosphate intergrowths.

CONCLUSIONS

(1) Phosphates of the graffonite – beusite series are found in twelve bodies of the LCT-family pegmatites of the Bothnian basin and of the Utö – Mysingen field, in north-central and south-central Sweden, respectively. They constitute a matrix hosting either coarse lamellae of sarcopside, which themselves contain finely lamellar triphylite, or abundant lamellae of triphylite with a bimodal size-distribution. Deformed and recrystallized granular aggregates of graffonite with triphylite, wolfeite and rare sarcopside were encountered at a single locality.

(2) The graffonite – beusite matrix is invariably Ca- and Mn-rich, whereas sarcopside is virtually Ca-free and Fe(Mg)-enriched; triphylite also is Ca-free and shows an even stronger preference for Fe and particularly Mg.

(3) Crystal-chemical relationships among the above phosphates show a diversity of structural and stoichiometric relationships that indicate solid-solution potential for triphylite in sarcopside, lithian sarcopside in graffonite – beusite, and triphylite in graffonite – beusite. Voids in the graffonite – beusite structure can accommodate half of the Li required for the $Li_2(\square R^{2+})_{-1}$ substitution.

(4) Textural relationships, bulk compositions, subparallel orientation of tie-lines connecting compositions of coexisting phases, and rather uniform distribution-coefficients for the cations indicate an exsolution origin of both types of lamellar intergrowths. A two-stage process initially generates lamellae of Li-bearing sarcopside from a relatively Ca, Li-poor disordered graffonite-type precursor; the Li-bearing sarcopside subsequently exsolves triphylite. A single-stage exsolution yields triphylite lamellae from a Ca, Li(Mn)-rich graffonite – beusite precursor.

(5) In regionally zoned groups of cogenetic pegmatites, the Ca-bearing graffonite – beusite is the first Fe, Mn(Li)-phosphate encountered in the beryl – columbite subtype of pegmatites, before the appearance of discrete grains of triphylite, or within the zone of beryl – columbite – triphylite pegmatites. This is in

accord with appreciable concentrations of Ca, Fe, Mn (and in part Mg), but low levels of F and Na, which are required to stabilize graffonite – beusite, and which are not routinely attained in bodies of more fractionated pegmatite.

(6) The generally good correlation of Mn/(Mn + Fe) of the phosphate aggregates examined with those of broadly associated garnet supports the thesis of London *et al.* (1995) that these phosphates are generated by phosphorus-induced destabilization of garnet.

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