

BEUSITE-TRIPHYLITE INTERGROWTHS FROM THE YELLOWKNIFE PEGMATITE FIELD, NORTHWEST TERRITORIES

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ABSTRACT

Calcium-rich beusite occurs in a pegmatite from the regionally zoned Peg swarm of the Archean Yellowknife pegmatite field, Northwest Territories. Beusite occurs as pale brown tabular lamellae, up to $5 \times 2.5 \text{ cm} \times 1 \text{ mm}$ in size, interlaminated with greenish grey triphylite lamellae. Beusite is monoclinic, space group $P2_1/c$, a 8.818(1), b 11.750(1), c 6.175(1) Å, β 99.35(1)° and V 631.3(1) Å³. Optical and single-crystal X-ray-diffraction studies confirm the presence of three crystallographic orientation relationships between beusite and triphylite, the most common being [010] of beusite parallel to [110] of triphylite. Beusite is chemically homogeneous, the Mn/(Mn + Fe) ratio varying slightly from 0.54 to 0.57. Calcium contents are higher than in other reported specimens, the maximum CaO content of the Yellowknife beusite reaching 16.1 wt.%. The elevated Ca content results in a larger b cell dimension. Beusite-triphylite intergrowths represent exsolution from a high-temperature, Li,Ca-rich graftonite-like parent phase.

Keywords: beusite, triphylite, exsolution, Yellowknife, Northwest Territories.

SOMMAIRE

Nous avons trouvé de la béusite riche en calcium dans une des pegmatites de l'essai de Peg, d'âge archéen, dans la région de Yellowknife (Territoires du Nord-Ouest). Elle forme des interlaminations tabulaires brun pâle atteignant $5 \times 2.5 \text{ cm} \times 1 \text{ mm}$ avec la triphylite gris verdâtre. La béusite est monoclinique, groupe spatial $P2_1/c$, a 8.818(1), b 11.750(1), c 6.175(1) Å, β 99.35(1)°, et V 631.3(1) Å³. Les données optiques et de diffraction X obtenues sur cristal unique confirment la présence de trois relations d'orientation entre béusite et triphylite, la plus répandue étant [010] de la béusite parallèle à [110] de la triphylite. Les cristaux sont homogènes; le rapport Mn/(Mn + Fe) ne varie que peu, entre 0.54 et 0.57. Les teneurs en calcium sont plus élevées (jusqu'à 16.1% de CaO en poids) que dans les exemples signalés auparavant, ce qui a pour effet un agrandissement de la dimension b . L'intercroissance de béusite et de triphylite résulterait d'une exsolution à partir d'un précurseur semblable à la graftonite, riche en Li et Ca, et stable à température élevée.

(Traduit par la Rédaction)

Mots-clés: béusite, triphylite, exsolution, Yellowknife, Territoires du Nord-Ouest.

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INTRODUCTION

Beusite, $(\text{Mn,Fe,Ca})_3(\text{PO}_4)_2$, is the rare manganese end-member of the graftonite-beusite series, which occurs exclusively as a primary accessory phase in granitic pegmatites. Beus (1950) was the first to report the existence of a graftonite-like mineral in which Mn exceeds Fe. A decade later, Brooks & Shipway (1960) also presented chemical data on a Mn-dominant graftonite-like phase. Beusite was given species status following the discovery of a Mn-dominant graftonite from the San Luis Province, Argentina (Hurlbut & Aristarain 1968). Since then, beusite has been found from southern Finland (Lahti 1981), the Cross Lake pegmatite field, Manitoba (T.S. Ercit, pers. comm. 1984), the Tsaobismund pegmatite, Namibia (Fransolet *et al.* 1986), and recently in the Yellowknife pegmatite field.

OCCURRENCE

Beusite occurs in a small pegmatite dyke of the regionally zoned Peg swarm of the Archean Yellowknife pegmatite field, located 75 km northeast of Yellowknife, between Upper Ross Lake and Redout Lake (Fig. 1). The pegmatite lies 3.5 km to the east of the Redout Granite, is lenticular, strikes N-S, dips 45-70° E and cuts an interlayered sequence of amphibolite and granodiorite.

The pegmatite is of the beryl-columbite-phosphate subtype of rare-element pegmatites, as classified by Černý (1989), and exhibits well-developed, although not continuous, internal zonation. Much of the primary zonation is obscured by albitic units. The border zone consists of a fine-grained muscovite \pm quartz assemblage; it is followed by a fine-grained microcline perthite + quartz + muscovite + cleavelandite wall zone. Most of the pegmatite is composed of a coarse-grained microcline perthite (graphic) + quartz + muscovite + cleavelandite + beryl zone that hosts most of the accessory minerals. These minerals include, in addition to the beusite-triphylite intergrowths, biotite, yellow-green beryl, ferrocolumbite-ferrotantalite, ferrotapiolite, almandine and pyrite. The core is discontinuous and consists of coarse-grained quartz + microcline + accessory beryl. Replacement muscovite units are present locally.

The beusite-triphylite intergrowths occur as a

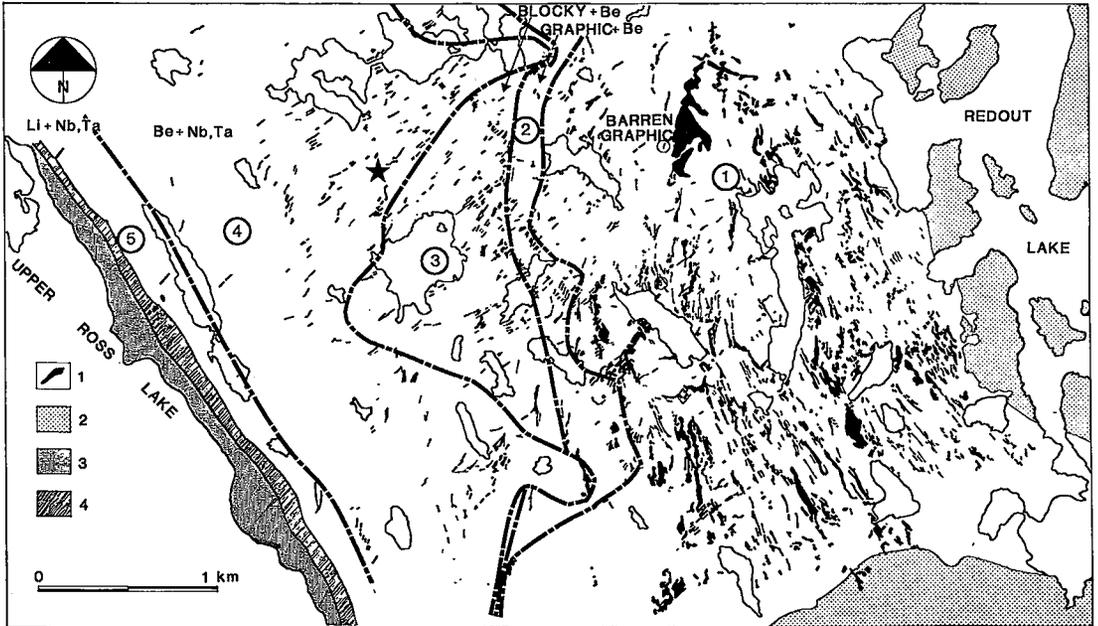


FIG. 1. Location map of the regionally zoned Peg swarm (modified after Meintzer 1987 from Hutchinson 1955), showing westward regional zoning (1–5) from the Redout Granite. 1 Pegmatites hosted by interlayered granodiorite and amphibolite, 2 Redout granite, 3 Burwash Formation and 4 Cameron River mafic volcanic suite. Star denotes location of beusite-bearing pegmatite.

× 5 × 3 cm nodule in blocky, pink microcline that appears to have undergone a moderate degree of metasomatic alteration. The microcline has been

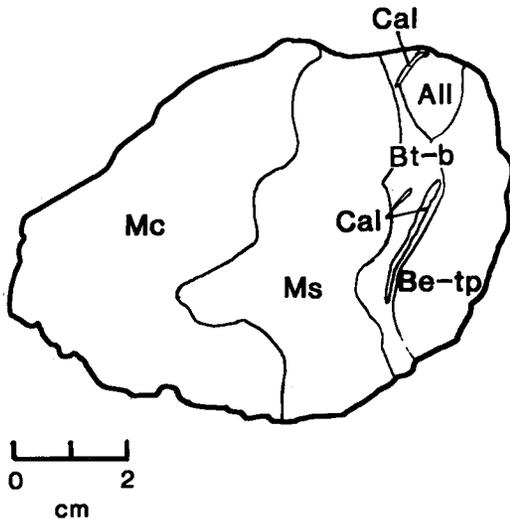


FIG. 2. Sketch of beusite-triphyllite paragenesis. Mc microcline, Ms fine-grained muscovite, Bt-b biotite-berthierine intergrowths, Be-tp beusite-triphyllite intergrowths, Cal calcite, All alluaudite-group mineral.

partly altered to a fine-grained, greenish muscovite that grades into a dark green band, 0.3–0.5 cm wide, of microscopically intermixed biotite and a berthierine-like mineral (R.E. Meintzer, pers. comm.; Fig. 2). This aggregate is in sharp contact with the beusite-triphyllite intergrowths and hosts tiny veinlets of calcite. Incipient Na-metasomatism is evident by the presence of a small patch of alluaudite-group minerals near the beusite-triphyllite intergrowth. Minor ferrisicklerite is present as a weathering product of triphylite.

PHYSICAL AND OPTICAL PROPERTIES

Beusite is pale brown, displays well-developed cleavage on {010} and {100}, vitreous luster and a pale brown streak. The density, measured on a Berman balance, is 3.600(5) g/cm³. The indices of refraction for beusite were measured by the Becke line method on crushed fragments in white light. The 2*V* angle was determined from extinction curves refined from spindle-stage data using the program EXCALIBUR (Bloss 1981). The optical properties and density of the Yellowknife beusite are compared with those of beusite from three other localities in Table 1. Using the Gladstone–Dale constants of Mandarino (1981), a compatibility index of 0.004 was obtained, indicating superior agreement between optical and chemical data and density.

TABLE 1. OPTICAL PROPERTIES FOR BEUSITE FROM YELLOWKNIFE AND OTHER LOCALITIES

	1*	2	3	4
α	1.685	1.695	1.708	1.702
β	1.688	1.696	1.711	1.703
γ	1.700	1.715	1.723	1.722
2V(°) obs	45	25	25	25
2V(°) calc	53	26	53	26
Dispersion	$\epsilon > \gamma$	$\epsilon > \gamma$	$\epsilon > \gamma$	$\epsilon > \gamma$
D(g/cm ³)	3.600	----	3.698	3.202

1 Yellowknife, this study; 2 Amanda, 3 San Salvador, 4 Los Aleros. Samples 2-4 from San Luis Province, Argentina (Hurlbut & Aristarain 1968).

*Indices of refraction measured in white light. Samples 2-4 measured in Na light.

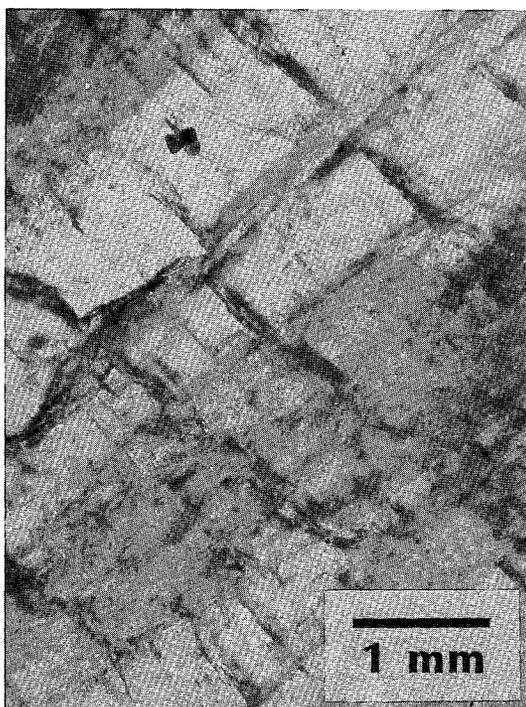


FIG. 3. Intergrowth of beusite (dark) and triphylite (light) viewed down their c axes.

TABLE 2. COMPARISON OF UNIT-CELL DATA FOR BEUSITE

Sample No.	a(Å)	b(Å)	c(Å)	β (°)	V(Å ³)
1	8.818(1)	11.750(1)	6.175(1)	99.35(1)	631.3(1)
2	8.797(3)	11.758(4)	6.170(2)	99.31(2)	629.8(3)
3	8.78(1)	11.52(1)	6.15(1)	99.42(35)	613.7(1)
4	8.788(3)	11.434(2)	6.255(1)	98.98(2)	620.8(4)

1 Powder data (this study), 2 single-crystal data, Yellowknife (Wise et al. 1989), 3 Los Aleros, San Luis, Argentina (Hurlbut & Aristarain 1968), 4 synthetic beusite, (Nozd & Annersten 1987).

TABLE 3. POWDER-DIFFRACTION DATA FOR BEUSITE

Beusite ¹ Yellowknife, N.W.T., Canada				Beusite ² Los Aleros, San Luis, Argentina		
hkl	d (calc)	d (obs)	I	d (calc)	d (obs)	I
111				4.293	4.29	20
121				3.607		
130				3.510	3.49	100
220				3.642		
131				3.130	3.13	40
102	3.033	3.031	100	3.021	3.01	35
131	2.995	3.000	44	2.950		
012				2.934	2.94	25
300				2.887		
040	2.938	2.942	74	2.880	2.89	30
230	2.911	2.908	69	2.873	2.863	100
221	2.875	2.876	70	2.850	2.846	20
231	2.745	2.747	12	-		
311				2.712		
202				2.701	2.708	60
212	2.642	2.642	9	-		
321				2.511		
231				2.494	2.502	5
311	2.415	2.412	34	2.402		
240				2.398	2.402	30
132				2.375	2.372	5
330	2.331	2.330	11	-		
241				2.303	2.301	5
302	2.295	2.297	18	2.286		
212	2.280	2.282	45	2.269	2.273	5
150				2.227		
132				2.224	2.222	5
222				2.147		
411	2.127	2.130	39	-		
410				2.128	2.137	5
151	2.097	2.099	14	2.063	2.064	5
340	2.064	2.064	18	-		
250				2.039		
251	2.006	2.006	13	2.034	2.038	10
242				-		
332	1.980	1.979	23	1.970		
213				1.965	1.967	5
123	1.938	1.937	24	1.932		
060				1.927	1.926	40
023	1.920	1.919	9	1.920		
412				-		
113				1.891	1.891	5
431	1.893	1.894	14	1.879		
061	1.864	1.864	23	1.876	1.874	5
152				1.831		
350				1.832	1.832	10
242				1.801		
133				1.803	1.802	5
260	1.786	1.785	14	1.805		
252	1.776	1.775	14	1.755		
431	1.746	1.747	31	1.753	1.754	5
500				1.732	1.732	5
440				1.731		
133	1.717	1.716	16	-		
510				1.713	1.712	5
351				1.680	1.680	5
043	1.671	1.671	43	1.655		
223				1.654	1.654	5
170	1.648	1.648	11	-		
402				1.640		
252				1.632	1.635	20
333				1.630		
170				1.617	1.616	10
360				1.599		
442				1.596	1.598	5
412	1.632	1.632	24			
171	1.604	1.603	13			
511	1.591	1.591	11			
270	1.566	1.566	13			
361	1.534	1.534	22			
352	1.500	1.500	36			
153	1.482	1.483	7			
304	1.449	1.449	19			
181	1.418	1.418	12			
234	1.414	1.414	15			
461	1.382	1.382	18			
281	1.372	1.371	31			
533	1.354	1.354	12			
182	1.322	1.322	23			
541	1.311	1.311	14			
115	1.228	1.228	14			

¹This study.

²Hurlbut & Aristarain (1968); FeK α radiation Mn filter.

Optical studies of the Yellowknife beusite have revealed crystallographic characteristics similar to those observed by Hurlbut & Aristarain (1968). A thin-section examination shows uniform and continuous extinction for all beusite lamellae (0.1-1.5 mm thick), thus indicating a single common crystallographic orientation of this phase within its tabu-

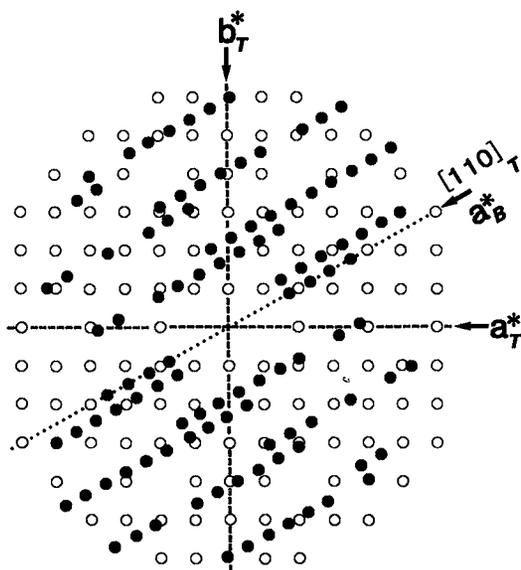


FIG. 4. Sketch of precession photograph showing crystallographic relationship of the third type of the beusite-triphylite intergrowths. Open circles: triphylite, closed circles: beusite. (Note extinction criterion in triphylite: for $hk0$, $k=2n+1$ are extinct).

TABLE 4. CHEMICAL COMPOSITION FOR BEUSITE AND TRIPHYLITE

	1*	2	3	4	5	6	7	8	9	10
P ₂ O ₅	41.40	41.6	42.1	41.8	41.0	46.2	46.1	45.7	45.6	45.6
MgO	2.64	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
CaO	4.78	16.1	15.9	14.3	15.5	0.2	0.0	0.1	0.2	0.0
MnO	36.96	23.1	23.1	25.1	23.2	18.1	19.0	18.3	17.7	18.4
FeO	14.62	19.8	19.8	19.1	19.0	27.6	26.8	26.8	27.1	26.5
Total	100.00	100.6	100.9	100.3	98.7	92.1	91.9	90.9	90.0	90.5
Beusite formula based on 8 oxygen atoms and triphylite formula based on 2(P) per formula unit.										
P ⁵⁺	2.005	1.99	2.00	2.01	2.00	2.00	2.00	2.00	2.00	2.00
Mg ²⁺	0.225	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ca ²⁺	0.293	0.98	0.96	0.87	0.96	0.01	0.00	0.01	0.01	0.00
Mn ²⁺	1.771	1.10	1.10	1.21	1.13	0.79	0.82	0.80	0.78	0.81
Fe ²⁺	0.699	0.94	0.93	0.90	0.91	1.18	1.15	1.16	1.17	1.15
Total	4.993	5.01	4.99	4.99	5.00	3.98	3.97	3.97	3.99	3.96

Beusite: 1 Los Aleros, Argentina (Hurlbut & Aristarain 1968), (2-5) Yellowknife, this study. Triphylite: (6-10) Yellowknife, this study. *Wet-chemical analysis by Dr. Jun Ito (Hurlbut & Aristarain 1968). Composition recalculated to 100% disregarding 0.14 H₂O, 1.5 SiO₂, 0.87 H₂O. All other analyses were made by the authors on the electron microprobe.

lar morphological units (Fig. 3). In contrast, the lamellae of grey to greenish grey triphylite are present in several optical orientations, two of which could be identified. Most of the triphylite lamellae (0.5-2 mm thick) are elongate along [110], which is parallel to [010] of beusite. Morphologically, the platy lamellae of triphylite are parallel to the {100} cleavage of

beusite. The contact between the two minerals is strongly undulatory. The second orientation is much less common: < 0.1- to 0.5-mm-wide lamellae of triphylite, elongate along [001], which also parallels the [010] of beusite, have straight planar contacts with beusite.

The banded intergrowth grades into a medium-grained, granular intergrowth of the same phases toward the end of the lamellae. The optical orientations within the granular intergrowth are the same as described for the lamellar intergrowths (*i.e.*, a single, uniform orientation for beusite and several orientations for triphylite). Unlike the triphylite found in the layered intergrowth, much of the granular triphylite has undergone extensive alteration to secondary phosphates.

X-RAY CRYSTALLOGRAPHY

Beusite is monoclinic with space group $P2_1/c$. Table 2 compares the unit-cell dimensions of the Yellowknife beusite with those of other samples. Unit-cell dimensions were determined for both a single crystal and powder. X-ray powder-diffraction data were obtained on a Philips PW 1710 automated powder diffractometer. Operating conditions were as follows: graphite-monochromatized CuK α_1 radiation (λ 1.54060 Å); operating voltage, 40 kV; operating current, 40 mA; scanning speed $\frac{1}{2}^\circ 2\theta/\text{min}$. Powder-diffraction data were calibrated using SiO₂ [a 4.9145(1), c 5.4061(3) Å] as an internal standard, and cell dimensions were refined by least-squares using a modified version of the CELREF program of Appleman & Evans (1973). Table 3 compares the diffraction data of the Yellowknife beusite and the type beusite from Argentina.

The cell dimensions of the single crystal were refined from the setting angles of thirteen automatically aligned intense reflections collected on a Nicolet R3m automated four-circle diffractometer. Graphite-monochromatized MoK α (λ 0.7107 Å) radiation was used at an operating voltage and current of 40 kV and 40 mA, respectively.

The unit-cell dimensions of the Yellowknife beusite, particularly the b dimension, are quite different from those of the Argentinian and synthetic samples. The anomalously large b dimension appears to be related to the elevated Ca content (Wise *et al.* 1990).

Precession photographs of a randomly selected fragment of the beusite-triphylite intergrowth, taken with the c axis of triphylite as the precession axis, show a third orientation relationship of the two phosphates, in addition to those established optically. Beusite reflections are oriented along preferred crystallographic directions of triphylite, which indicates that the [100] of beusite nearly parallels [110] of triphylite (Fig. 4).

CHEMICAL COMPOSITION

Chemical analyses were performed on a MAC 5 electron microprobe in the energy-dispersion mode. The following standards were used for analysis: apa-

tite (P,Ca), MnF_2 (Mn), and chromite (Fe,Mg). Specimen current was 0.010 mA at 15 kV. Acquisition time for each spectrum was 200 seconds. Data reduction was made using a modified version of MAGIC V (Colby 1980). Electron-microprobe data

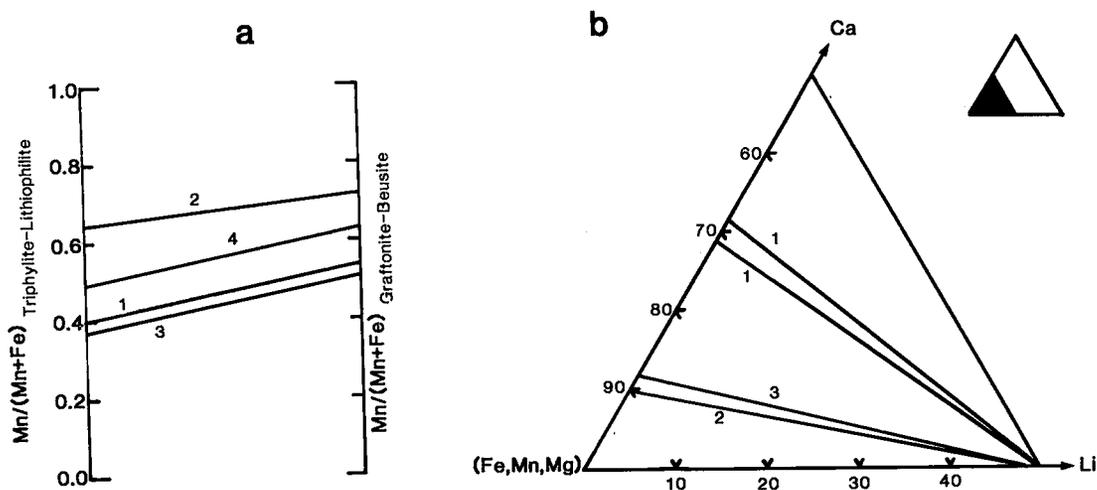


FIG. 5. Compositions of coexisting beusite and triphylite pairs from 1 the Yellowknife pegmatite field, 2 the Los Aleros pegmatite (Hurlbut & Aristarain 1968), 3 the Tsaobismund pegmatite (Fransolet *et al.* 1986) and 4 the Jussinuovi pegmatite (Lahti 1981). a) Comparison of Mn/(Mn + Fe) values, b) comparison of compositions in terms of Ca, (Fe + Mn + Mg) and Li (at. %).

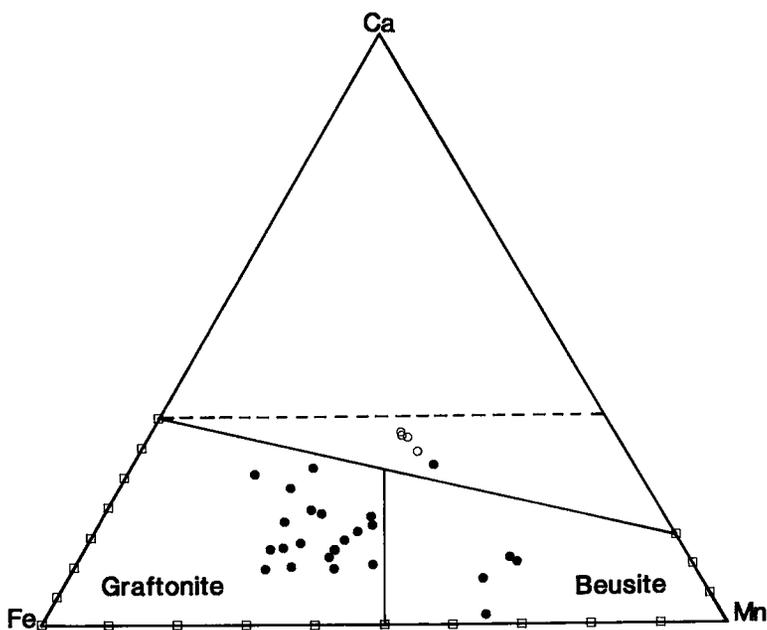


FIG. 6. Graftonite-beusite compositions in terms of Ca, Fe and Mn. Dashed line denotes hypothetical limit of Ca solubility based on the Yellowknife data. Dots: data of Hurlbut & Aristarain (1968) and Fransolet (1977), open circles: this study. Open squares: synthetic data of Nord (1982) and Nord & Ericsson (1982).

of Yellowknife beusite and triphylite are presented in Table 4. Because lithium is not detectable on the microprobe, the results listed for triphylite reflect partial analyses only.

The Yellowknife examples of beusite are homogeneous in composition. The range of Mn/(Mn + Fe) values is very narrow, from 0.54 to 0.57, and somewhat lower than for the Argentinian examples (0.60–0.72; Hurlbut & Aristarain 1968). By contrast, the Ca content found in beusite from Yellowknife is high. In all cases of natural beusite and terrestrial graffonite, some Ca is present, but the beusite from Yellowknife contains the highest amount of Ca recorded to date (16.1 wt. % CaO). Unlike all other known examples of beusite, no Mg was detected in the Yellowknife sample.

DISCUSSION

In nature, graffonite-beusite commonly occurs intergrown with a member of the triphylite-lithiophilite series or with sarcopside. Crystallization of graffonite-beusite is regarded as primary, generally occurring during the stages of inner intermediate zone and core consolidation. The interlamination has been examined by previous workers and is considered to represent exsolution from a high-temperature, (Li,Ca)-rich graffonite-like parent phase. Subsequent decrease in temperature resulted in the migration and concentration of Li into triphylite and Ca into beusite. Hurlbut (1965) suggested that the role of Li in the parent phase determines the type of intergrowth that will form. If Li is absent, sarcopside-graffonite intergrowths will occur; where Li is present, graffonite will occur with triphylite-lithiophilite. Hurlbut & Aristarain (1968) interpreted the granular intergrowth of beusite-lithiophilite as being caused by rapid cooling from crystal boundaries, with reduced ionic mobility.

According to published compositions of graffonite-beusite, all terrestrial examples of the series contain some Ca, typically between 3 and 13 wt. % CaO. Only graffonite found in fine octahedrite meteorites is known to be Ca-deficient (Olsen & Fredriksson 1966). The most prominent feature of the Yellowknife beusite is its high Ca content. A metasomatic, cation-exchange introduction of Ca into beusite might be envisaged at hydrothermal stages. However, late fluids that have locally deposited calcite with apatite inclusions in the parent pegmatite would have affected triphylite first; triphylite-lithiophilite minerals are sensitive to fluoride-, carbonate- and calcium-bearing fluids, yielding (manganous) apatite and calcite as alteration products (unpublished observations of P. Černý and A.-M. Franolet). The triphylite lamellae in the Yellowknife beusite have not suffered this alteration, which suggests that the levels of Ca observed in the

beusite are a direct reflection of the composition of the parent mineral before exsolution.

A comparison of the Mn/(Mn + Fe) values in triphylite and beusite from four different localities shows that the graffonite-structured mineral tends to incorporate Mn more readily than triphylite (Fig. 5a). Similarly, the graffonite structure is better suited to incorporating Ca than that of triphylite (Fig. 5b).

The compositions of the Yellowknife beusite extend the previously experimentally determined boundaries of $\text{Fe}_3(\text{PO}_4)_2$ - $\text{Ca}_3(\text{PO}_4)_2$ and $\text{Mn}_3(\text{PO}_4)_2$ - $\text{Ca}_3(\text{PO}_4)_2$ solid solutions. Nord & Ericsson (1982) and Nord (1982) determined the maximum solubility of Ca to be about 35 atom % in $\text{Fe}_3(\text{PO}_4)_2$ and 15 atom % in $\text{Mn}_3(\text{PO}_4)_2$. The low solubility of Ca in $\text{Mn}_3(\text{PO}_4)_2$ is surprising in light of the flexible nature of the graffonite structure. Ideally, one third of the cation sites are six- or seven-coordinated, depending on the presence of large cations such as Mn or Ca. Calvo (1968) showed that Ca^{2+} occupies and prefers the unique seven-coordinated $M(1)$ site over the $M(2)$ and $M(3)$ sites. However, Nord & Ericsson (1982) showed that in spite of the larger size of the $M(1)$ site, the substitution of Ca for Fe in the graffonite structure takes place at both the $M(1)$ and $M(3)$ sites, with virtually no preference. Since one third of the cation sites are sufficiently large to accommodate the available Ca, there is no obvious reason why the maximum solubility of Ca should be less in beusite than in graffonite. In most graffonite-beusite specimens examined to date, the amount of Ca present is not sufficient to completely fill the $M(1)$ site; the difference is made up by the incorporation of Mn or Fe. Based on cation abundances, the $M(1)$ site of the Yellowknife beusite may be nearly filled with Ca, as Ca closely approaches 33% of the total cations. Based on our present understanding of the crystal chemistry of the graffonite-beusite series and on the Yellowknife data, we suggest that Ca could constitute one third of all cations in both graffonite and beusite (Fig. 6). The substitution may, however, be different in Ca-disordered phases and in those with Ca restricted to the $M(1)$ site.

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