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FLUORINE VARIATION IN HAMBERGITE FROM GRANITIC PEGMATITES

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ABSTRACT

Considerable F-for-OH substitution can occur in the structure of hambergite, $Be_2(BO_3)(OH,F)$. The substitution results in a linear reduction of the indices of refraction. This relation can be used to estimate the F content of hambergite, subject to the constraint that samples may be compositionally heterogeneous. Previous X-ray-diffraction studies of five crystals of hambergite showed that increasing amounts of F for OH lead to a decrease in the *a* dimension and an expansion of the *b* dimension. The F content of hambergite may be readily estimated if the unit-cell dimensions are known. Although originally described from an alkaline syenitic pegmatite, the majority of hambergite occurrences are in complex Li-rich granitic pegmatites of the elbaite or transitional elbaite–lepidolite subtypes. Hambergite occurs as a comparatively early phase in massive pegmatites from several localities in the Czech Republic. More commonly, hambergite is a late-crystallizing phase produced in miarolitic pockets as the highly evolved magma approaches its solidus. The F content of hambergite from pegmatites appears to reflect the F activity of the medium (melt or fluid) from which it crystallizes. Hambergite found in miarolitic pockets is typically more homogeneous in composition and commonly has a lower F content than that found in a massive pegmatite, although F-rich examples from pegmatitic pockets also are observed. It is unclear whether the lower F content of hambergite from miarolitic pockets is strictly a result of crystallization in lower-F systems, or involves precipitation in equilibrium with exsolved aqueous fluids.

Keywords: hambergite, fluorine, electron microprobe, X-ray diffraction, indices of refraction, paragenesis, granitic pegmatites.

SOMMAIRE

La structure de la hambergite, $Be_2(BO_3)(OH,F)$, fait preuve d'une étendue considérable de substitution de F pour OH. Cette substitution mène à une réduction linéaire des indices de réfraction. On peut se servir de cette relation pour prédire la teneur en F d'échantillons, toujours sous réserve que ceux-ci peuvent être hétérogènes. Les résultats d'études de cinq échantillons par diffraction X ont déjà montré qu'une augmentation de la proportion de F mène à une diminution du paramètre *a* et une augmentation du paramètre *b*. On peut facilement donc déterminer la teneur en F en connaissant les paramètres réticulaires. Quoique découvert dans une syénitique alcaline pegmatitique, la majorité des exemples connus se trouvent dans des venues de pegmatite granitique complexe montrant un enrichissement en Li, faisant partie des sous-types à elbaïte ou transitionnels à elbaïte–lépidolite. La hambergite y constitue une phase relativement précoce dans des pegmatites massives à plusieurs endroits dans la République Tchèque. Le plus souvent, la hambergite cristallise tardivement, dans des miaroles, lors de l'approche finale du solidus des magmas fortement évolués. La teneur en F de la hambergite des pegmatites semble être en relation avec l'activité du fluor dans le milieu de croissance (bain fondu ou phase fluide). La hambergite de miaroles est typiquement plus homogène et plus pauvre en fluor que celle qui cristallise précocement dans les pegmatites massives, quoique la hambergite riche en fluor se rencontre aussi dans les miaroles. Il n'est pas clair si les teneurs plus faibles en F de la hambergite des miaroles résultent uniquement d'une cristallisation dans un système magmatique à teneur plus faible en fluor, ou bien témoigne d'une précipitation en équilibre avec une phase fluide exsolvée.

(Traduit par la Rédaction)

Mots-clés: hambergite, fluor, microsonde électronique, diffraction X, indices de réfraction, paragenèse, pegmatites granitiques.

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INTRODUCTION

Hambergite, $Be_2(BO_3)(OH,F)$, was first described from a nepheline syenite pegmatite on Helgeråen Island, Langesundfjord, southern Norway (Brögger 1890). It has now been reported from about 30 occurrences of pegmatite, but the original host pegmatite is of alkaline affinity in only two cases (Brögger 1890, Semenov & Bykova 1965). In all other cases, the host is commonly Li-bearing complex granitic pegmatite, mostly of the elbaite or transitional elbaite–lepidolite subtype (*cf.* Černý 1991, Novák & Povondra 1995). Despite the considerable number of localities, there remains a paucity of data about the chemistry, optics and parageneses of hambergite. Reported chemical compositions are scarce and commonly incomplete (*i.e.*, concentration of F not determined), and compositional zoning observed in many samples indicates that at least some reported bulk compositions pertain to rather heterogeneous materials. Switzer *et al.* (1965) provided the first detailed study of F variation in hambergite, and the F – OH substitution in its structure was studied by Burns *et al.* (1995). In that study, the authors examined five crystals of hambergite (from the same suite as studied here) using single-crystal X-ray-diffraction techniques, and found that there is a significant linear decrease in the *a* dimension and a significant linear expansion of the *b* dimension with increasing F.

Here, we provide new results on hambergite from several granitic pegmatites in the Czech Republic and in southern California (Table 1). Distinct parageneses of hambergite from massive pegmatite units and from miarolitic pockets are discussed with respect to their F contents. Particular emphasis is placed on the variations in F content, and its influence on the optical properties.

TABLE 1. DATA ON HAMBERGITE SAMPLES

Sample	Locality	Color	Description	Type	Source
HN-1	Himalaya mine	C	prismatic crystal, rim	P	1
HN-2	Himalaya mine	C	prismatic crystal, core	P	1
HN-3	Sušice	C	tabular grain	M	2
HN-4	Belo Horizonte	W	prismatic crystal	P	3
HN-5	Belo Horizonte	C	prismatic crystal	P	3
HN-6	Ctidružice	C	corroded grain	M	2
HN-8	Kracovice	Y	oval grain, rim	M	2
HN-9	Kracovice	Y	oval grain, core	M	2
HN-10	Kracovice	Y	oval grain, core	M	2
LL-3	Little Three	C	tabular crystal	P	4

Symbols: C colorless, W milky white, Y yellowish. P pocket hambergite, M hambergite from a massive pegmatite. Sources: 1 Mineralogical Museum, University of Manitoba, Winnipeg, 2 Moravian Museum, Brno, 3 M.C. Taylor, University of California, Riverside, 4 D. London, University of Oklahoma, Norman.

PARAGENESIS OF HAMBERGITE IN GRANITIC PEGMATITES

In most known localities associated with complex granitic pegmatites, hambergite occurs in late-stage miarolitic pockets as well-formed, isolated, commonly twinned tabular to prismatic crystals. Fragmentation or corrosion of hambergite crystals commonly occurs in association with late-stage pocket rupture or changes in fluid composition (or both). In these bodies of pegmatite, hambergite is commonly associated with multicolored tourmaline (Mn-rich elbaite), albite, quartz, K-feldspar, lepidolite, topaz, beryl, apatite, cookeite, and several other rare minerals that may be present locally (Switzer *et al.* 1965, Foord *et al.* 1986). The position of hambergite in the crystallization sequence is not clear in many cases, because physical attachment between its crystals (or their fragments) and other pocket minerals commonly has not been preserved. Where such relations are found, hambergite seems to be a relatively late pocket mineral, forming simultaneously with hydrothermal minerals such as bavenite, herderite and danburite, but prior to the formation of zeolites and clay minerals (DeVito & Ordway 1984, Kazmi *et al.* 1985, Foord *et al.* 1986, Zagorsky & Peretiazhko 1992). Switzer *et al.* (1965), however, described hambergite from the Little Three pegmatite that crystallized before most of the early pocket minerals, including quartz, microcline, and topaz.

Hambergite formed in massive units of complex granitic pegmatites is relatively rare. It has been found only in the Czech Republic to date, although at several localities. It is easily overlooked in the field, occurring as colorless to white or yellowish, equidimensional to tabular grains, up to 2 cm in diameter, with imperfectly developed crystal faces. At most localities, hambergite occurs in a blocky K-feldspar unit [Ctidružice: Čech (1957), Sušice III: Čech & Povondra (1961), Jeclov II: Němec (1988)] or locally associated with a coarse-grained graphic unit. However, minor fine- to medium-grained albite and rarely elbaite also may be present. In the latter cases, it seems likely that hambergite crystallized slightly before or almost simultaneously with the elbaite-bearing unit. Čech (1957), however, suggested that hambergite crystallized simultaneously with or slightly after elbaite. The Czech localities given in Table 1 represent typical elbaite-bearing pegmatites characterized by: (i) rare occurrence of micas (lepidolite was found only in Ctidružice, and muscovite is absent at all localities), (ii) the presence of Mn-rich elbaite, and (iii) modal dominance of K-feldspar over albite (Novák & Povondra 1995).

TABLE 2. ELECTRON-MICROPROBE DATA FOR HAMBERGITE: HOMOGENEOUS SAMPLES

sample *n*	HN-1 30	HN-2 25	HN-5 25	HN-3 15	HN-4 30	LL-3 61
BeO*	53.82	52.75	53.86	51.96	51.73	53.42
B_2O_3	37.45	36.71	37.48	36.16	36.00	37.17
H_2O*	9.61	9.40	9.44	8.43	6.51	5.98
F	0.18(.04)	0.21(.05)	0.55(.05)	1.95(.22)	5.92.(07)	7.68(.22)
O=F	−0.08	−0.09	−0.23	−0.82	−2.49	−3.23
Total	100.98	98.99	101.10	97.67	97.67	101.01
Be^{2+}	2.000	2.000	2.000	2.000	2.000	2.000
B^{3+}	1.000	1.000	1.000	1.000	1.000	1.000
OH^-	0.991	0.990	0.973	0.901	0.699	0.621
F^-	0.009	0.010	0.027	0.099	0.301	0.379

* calculated from stoichiometry: Be = 2, OH + F = 1. The data are average results of *n* analyses, expressed in wt. %. In each case, the sum of cations is 3.000, and the sum of anions (O + OH + F) is 4.

Hambergite from massive pegmatite at Kracovice exhibits a different paragenesis. Němec (1990) found hambergite in medium-grained pegmatite adjacent to the biotite-bearing granitic border unit. Relatively early crystallization of this hambergite seems to be very likely, and its indices of refraction (Němec 1990) indicate a moderate F-content. The hambergite from Kracovice examined in the current study occurs in relatively late, fine-grained saccharoidal albite with rare elbaite. On the basis of textural relationships among individual units of the pegmatite, hambergite seems to have crystallized after or in part simultaneously with the principal Li-bearing minerals (Mn-rich lepidolite to masutomilite and Mn-rich elbaite). The Kracovice pegmatite does not seem to fit the currently used subdivision of complex granitic pegmatites (*e.g.*, Černý 1991). The main host for Li is Mn-rich lepidolite to masutomilite, muscovite is absent, and the body is extremely rich in F-bearing minerals such as topaz, F-rich micas and F-rich elbaite (Němec 1990, data of the first author). This overall paragenesis indicates a much higher activity of fluorine compared to typical elbaite pegmatites, in which a high activity of boron is dominant.

CHEMICAL COMPOSITION

Mineral compositions were determined by electron-microprobe analysis, performed on a Cameca SX–50 at the University of Oklahoma. The conditions for analysis were 10 kV acceleration, a beam current of 25 nA, and a defocused spot 7 μm across; data reduction employed the PAP correction scheme (Pouchou & Pichoir 1985). The concentrations of F were established using a synthetic multilayer diffraction device with a spacing of 62.5 Å, with natural topaz used as the standard; live counting time on the peaks was 40 s, yielding a calculated 3σ minimum level of detection of <400 ppm. The samples were analyzed for B using a synthetic multilayer diffraction device with a spacing of 95.0 Å and (>99.999%) B_2O_3 glass as the standard; the counting time on the peak was 60 s. Although not calculated, the 3σ minimum level of detection for B was undoubtedly lower than analytical uncertainties arising from anisotropy of orientation of its *K*α X-ray line shape with the diffraction device used (*e.g.*, Bastin & Heijligers 1991).

Samples were analyzed for both Si and Al using a premium quality TAP device; natural plagioclase (An_{84}) was used as the standard, with counting times of 30 s on the peaks yielding a calculated 3σ minimum level of detection of <0.013 wt%. Attempts were made to analyze for oxygen directly using the multilayer diffraction device with a spacing of 62.5 Å and B_2O_3 glass as the standard. However, analytical uncertainties due to variations in thickness of the carbon coating between samples and standards made the results of limited practical use. The contents of Be (BeO) and OH (H_2O) were calculated on the basis of stoichiometry.

TABLE 3. ELECTRON-MICROPROBE DATA FOR HAMBERGITE: HETEROGENEOUS SAMPLES

sample *n*	HN-6L 4	HN-6M 9	HN-6H 1	HN-9L 10	HN-10L 3	HN-9H 15	HN-10H 22	HN-8H 20
BeO*	54.80	53.90	54.51	53.21	51.46	53.49	52.67	52.67
B_2O_3	38.13	37.51	37.93	37.03	35.81	37.22	36.65	35.81
H_2O*	9.53	8.86	7.70	8.31	5.53	4.90	4.63	4.39
F	0.71	1.79	4.47	2.69	7.88	9.97	10.24	10.75
O=F	−0.30	−0.75	−1.88	−1.13	−3.32	−4.20	−4.31	−4.53
Total	102.87	101.31	102.72	100.11	97.37	101.38	99.88	99.93
Be^{2+}	2.000	2.000	2.000	2.000	2.000	2.000	2.000	2.000
B^{3+}	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
OH^-	0.966	0.913	0.785	0.867	0.597	0.509	0.488	0.463
F^-	0.034	0.087	0.215	0.133	0.403	0.491	0.512	0.537

* calculated from stoichiometry: Be = 2, OH + F = 1. The data are average results of *n* analyses, expressed in wt. %. In each case, the sum of cations is 3.000, and the sum of anions (O + OH + F) is 4. Contents of fluorine: L low, M moderate, H high.

The analytical results are presented in Tables 2 and 3. In all cases, Al and Si contents were found to be near or below their minimum detection thresholds and are not considered further. Despite the analytical uncertainties for boron, the compositions obtained provide stoichiometries close to the ideal formula of hambergite. Most of the crystals examined, especially those formed in miarolitic pockets, are homogeneous in boron distribution (Table 2). Significant heterogeneity in F contents was observed for samples from two localities, in which hambergite occurs enclosed in massive pegmatite (Table 3). Of the samples examined, hambergite from miarolitic pockets typically shows lower F contents than those from massive pegmatite, although those found in pockets from Belo Horizonte pegmatite and Little Three pegmatite are particularly

F-rich. In general, hambergite from massive pegmatites tends to show higher F contents and greater variability in F than those from miarolitic pockets. For example, samples from the F-enriched pegmatite at Kracovice show particularly high levels of F.

OPTICAL EXAMINATION

The optical study was done with a JENA PolU polarizing microscope using crushed grains. Indices of refraction were determined using the Becke line method in a mixture of immersion liquids in sodium light (λ 589 nm). The standard errors of most determinations are estimated to be 0.001, but estimated standard errors for weathered samples and samples with abundant fluid inclusions are 0.002. The results of these determinations, and comparison to some other published data on hambergite, are presented in Table 4. As is apparent from the data, the F-for-OH substitution in the structure of hambergite produces a significant decrease in indices of refraction, which is consistent with the previous results of Switzer *et al.* (1965). The relation between F content and indices of refraction is essentially linear (Fig. 1). Indices of refraction thus serve as a useful tool for indirect estimation of F content in hambergite, within the limits of compositional heterogeneity found in some samples. The relationships are: $\alpha = -0.0764F + 1.5572$, $\beta = -0.0716F + 1.5902$ and $\gamma = -0.0743F + 1.6300$ [F given in atomic proportions at the O(4) site], and $\alpha = -0.0038F + 1.5572$, $\beta = -0.0037F + 1.5902$ and $\gamma = -0.0037F + 1.6300$ (F given in wt.%), can be used to estimate the F content if the indices of refraction are known.

TABLE 4. INDICES OF REFRACTION OF HAMBERGITE

Sample	Locality	α	β	γ	Δ
HN-1	Himalaya mine	1.557(1)	1.589(1)	1.629(1)	0.072
HN-2	Himalaya mine	1.557(1)	1.589(1)	1.630(1)	0.073
	Himalaya mine*	1.552	1.588	1.628	0.076
HN-3	Sušice	1.548(1)	1.583(1)	1.621(1)	0.073
	Sušice**	1.556	1.588	1.630	0.074
HN-5	Belo Horizonte	1.556(1)	1.590(1)	1.630(1)	0.074
LL-3	Little Three	1.526(1)	1.561(1)	1.598(1)	0.072
	Little Three*	1.543	1.580	1.617	0.074
HN-6	Ctidružice	1.528(2)	1.566(2)	1.599(2)	0.071
HN-8	Kracovice rim	1.537(2)	n.d.	1.608(2)	0.071
HN-10	Kracovice core	1.520(2)	1.555(2)	1.595(2)	0.075

* Data of Switzer *et al.* (1965). ** Data of Čech & Povondra (1961).

DISCUSSION

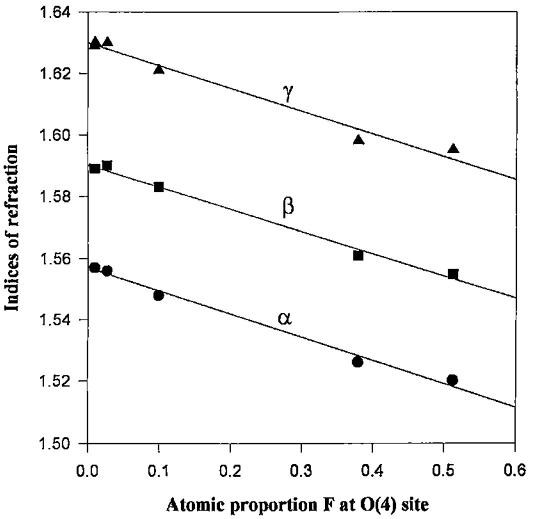

FIG. 1. Indices of refraction *versus* atomic proportions F at O(4) site in hambergite. Data for the samples HN–1, HN–2, HN–3, HN–5, LL–3 and HN–10 are presented in the diagram.

In addition to hambergite, hambergite-bearing complex pegmatites commonly also contain additional Be-minerals, including beryl, bertrandite, bavenite and herderite. Beryl typically occurs in a different textural and paragenetic unit (Čech 1957, Čech & Povondra 1961, Němec 1988, 1990). Its crystallization is believed to be favored by neutral environments (Černý 1968), whereas most borates typically form in alkaline systems (*e.g.*, London 1986, 1987). The presence of these two phases in the same locality seems to indicate changes in relative acidity–alkalinity throughout pegmatite evolution. The presence of both hambergite and beryl from a single pocket of the Little Three pegmatite is noteworthy (Switzer *et al.* 1965), and may be a direct consequence of high F activity during fractionation. Although mutual stability relationships for these phases are not well known, variations in relative acidity throughout pocket crystallization in time and space appear to be both possible and reasonable. For example, London (1986) suggested that increasing contents of B and F in residual melts are a natural consequence of progressive fractionation in evolved deposits such as the miarolitic pegmatites of the San Diego District (including the Little Three). Increased contents of B or F, or both, in turn, promote migration of late residual melts toward peralkaline bulk compositions (*e.g.*, London 1986, 1987). Bavenite and bertrandite typically represent late hydrothermal replacements of beryl, and apparently form after hambergite. At Ctidružice, for example, bertrandite replaces both beryl and hambergite (Čech 1957). Calcic phases such as bavenite and herderite are almost always texturally late with respect to most pocket minerals;

they commonly are associated with danburite and manganoan epidote or zoisite, and are typically succeeded only by clay minerals, including Ca,Mg-bearing smectite-group minerals. The liquids giving rise to complex lithian pegmatites are extremely evolved, and are usually exceedingly poor in Ca and particularly Mg. Thus, it is likely that the calcic phases form in response to the influx of wallrock-derived fluid components when pegmatite consolidation is nearly complete, perhaps just prior to or accompanying pocket rupture (*e.g.*, Foord *et al.* 1986, London 1986).

The homogeneity of F contents in hambergite from pegmatite pockets is likely a result of the process of pocket formation. Miarolitic pockets are generally produced in the late stages of pegmatite consolidation, as the magmas approach their solidus (*e.g.*, London 1986). The magmas (or potential aqueous fluids derived from them) contained within pockets represent near end-products of the differentiation process. They are thus unlikely to be affected by the large changes in F activity of the melt that could accompany fractionation of the bulk magma to form earlier textural and paragenetic units within the pegmatite. Moreover, the pockets themselves are spatially discrete entities that are typically small in comparison to the entire mass of a pegmatite body. Nucleation densities (and growth rates?) of minerals in pockets should be reduced by high dissolved volatile contents of the late-stage melts. It is thus easy to envision why the phases produced in pockets should be more homogeneous than those produced in larger, perhaps more rapidly crystallizing massive pegmatite.

One point of curiosity left unresolved by the current work is whether the observed F contents of hambergite from pegmatitic pockets is strictly an indication of crystallization from F-poor magmatic systems, or in some cases could be due to growth from an exsolved aqueous fluid. Because fluorine in melt–vapor systems preferentially partitions into the melt phase (*e.g.*, Kogarko *et al.* 1968, Manning 1981, London *et al.* 1989), it seems reasonable that hambergite crystallized from a given melt would have a higher F content than that crystallized from its exsolved aqueous fluid. In this respect, it is interesting to note that hambergite from the Little Three pegmatite is texturally early and has high F contents, perhaps suggesting crystallization directly from a F-enriched hydrous residual melt (London 1986), whereas F-poor hambergite from the Himalaya dike is among the latest of the pocket minerals, forming after pocket rupture (Foord *et al.* 1986). We are not aware of any hambergite–melt or hambergite–fluid partitioning data for F. Such information, together with other data from mineral chemistries, which may provide an indication of the apparent F activity of a pegmatite-forming melt, could conceivably be used to discern whether hambergite had precipitated from a melt or fluid. Precipitation from a melt and fluid, however, could help to explain variations in the F contents of hambergite from distinct pockets of the same locality (*e.g.*, compare HN–4 and HN–5 from the Belo Horizonte pegmatite in Table 4), although compositional variability between individual pockets is possible as well.

SUMMARY

The reduction in indices of refraction accompanying the substitution of F for OH in the structure of hambergite is linear, and the relationship can be used to estimate F contents, subject to the constraint that hambergite may be compositionally heterogeneous. Contrary to the results of Switzer *et al.* (1965), the examination of five crystals using X-ray diffraction (Burns *et al.* 1995) showed that increasing F for OH substitution causes a decrease in the *a* dimension and an expansion of the *b* dimension.

Hambergite has been observed as a comparatively early phase in massive complex Li-rich pegmatites from several localities in Czech Republic, which are undoubtedly Be-, B-, and F-rich and probably neutral to alkaline in character. More commonly, hambergite is a late-crystallizing phase in miarolitic pockets produced as the highly evolved B-rich magmas approach their solidi. The F contents of hambergite crystals from pegmatites seem to reflect the F activity of the medium of growth. Hambergite produced in miarolitic pockets is more homogeneous in composition and commonly has a lower F content than those produced in massive pegmatite, although high-F examples from pegmatitic pockets are observed. In the absence of F partitioning data, it is as yet unclear whether the F contents of hambergite provides a direct (*e.g.*, quantifiable) monitor of F activities in the melt, or whether the lower average F content of hambergite from miarolitic pockets may indicate precipitation from an exsolved aqueous fluid.

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