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Epitactic hedenbergite whiskers on babingtonite, a second occurrence from a Triassic basalt at Lincoln Park near Paterson, New Jersey, USA

by *Thomas Armbruster*[1], *Edwin Gnos*[2], and *R. Peter Richards*[3]

Abstract

Epitactic [001] whiskers of hedenbergite $(Ca_{0.92}Na_{0.04})(Fe^{2+}_{0.76}Mn_{0.08}Mg_{0.17}Al_{0.02})Si_2O_6$ (space group $C2/c$, $a = 9.80(1)$, $b = 8.99(1)$, $c = 5.241(4)$ Å, $\beta = 105.4(1)°$, $V = 445.7$ Å^3) were analyzed on (010) plates of babingtonite $Ca_{2.00}(Fe^{2+}_{0.78}Mn_{0.12}Mg_{0.08}Na_{0.02})(Fe^{3+}_{0.95}Al_{0.05})Si_5O_{14}(OH)$ (space group $P\bar{1}$, $a = 7.4734(8)$ $b = 12.176(2)$, $c = 6.6881(5)$ Å, $\alpha = 86.134(9)$, $\beta = 93.896(8)$, $\gamma = 112.211(9)°$, $V = 561.59(11)$ Å^3. The epitactic relationships are: $(010)_{bab}$ is parallel to $(\bar{1}\bar{1}1)_{hed}$ and $(10\bar{1})_{bab}$ is parallel to $(100)_{hed}$, in agreement with a coherent continuation of silicate single-chains in both structures. The composite crystals are overgrown by prehnite $Ca_2Al_{1.38}Fe^{3+}_{0.51}Fe^{2+}_{0.06}Si_{3.06}O_{10}(OH)_2$ and minor pumpellyite-(Fe^{2+}) $Ca_{1.97}Al_{1.01}Fe^{3+}_{0.89}Fe^{2+}_{0.96}Mg_{0.12}Si_3O_{11}(OH)_2 \cdot H_2O$, calcite, and datolite $CaBSiO_4(OH)$. The sample originates from hydrothermal cavity fillings of a Triassic basalt at Lincoln Park (Watchung Mountains) near Paterson, New Jersey, USA. The epitactic relationship between babingtonite and hedenbergite is identical to the one determined for a sample from Alpine fissures in gneisses at Arvigo, Grisons, previously described. Occurrences of this relationship are known or suspected from at least seven localities.

Keywords: babingtonite, hedenbergite, prehnite, pumpellyite, epitaxy, hydrothermal alteration.

Introduction

In a recent study (ARMBRUSTER et al., 2000) we described the epitactic relationship between the two chain silicates babingtonite $Ca_2Fe^{2+}Fe^{3+}Si_5O_{14}(OH)$ (space group $P\bar{1}$, $a = 7.48$, $b = 12.16$, $c = 6.68$ Å, $\alpha = 86.1$, $\beta = 93.9$, $\gamma = 112.1°$) and hedenbergite $CaFe^{2+}Si_2O_6$ (space group $C2/c$, $a = 9.80$, $b = 8.99$, $c = 5.24$ Å, $\beta = 105.3°$). Babingtonite with *fünfer* single chains forms the host. The pyroxene hedenbergite with *zweier* single chains grows in an oriented fashion as very thin whiskers on the host. The hedenbergite [001] whiskers emerge out of the babingtonite (010) face, which contains the ends of the silicate chains in the host crystal. This hedenbergite – babingtonite assemblage, together with heulandite, clinochlore, and fluorite, was detected in low temperature Alpine fissures in gneiss at Arvigo, Val Calanca, Grisons, Switzerland. An upper temperature limit of ca. 200 °C was estimated for this occurrence from late fluid inclusions in quartz and the presence of heulandite.

In response to this previous study (ARMBRUSTER et al., 2000), R. P. Richards informed us that he found several specimens with apparently very similar epitactic whiskers on babingtonite among gratis samples offered by D. Smoley at the micromineral symposium of the Cleveland Museum of Natural History (Ohio, USA) in November 2000. These samples originate from fissures or vugs of a Triassic basalt at Lincoln Park, near Paterson, New Jersey. This paper reports on the second occurrence of epitactic hedenbergite whiskers on babingtonite. The coarser size of the whiskers allowed both hedenbergite and babingtonite to be analyzed by electron microprobe and to be compared, and permitted additional details of the whisker formation to be resolved. Furthermore, this second occurrence, together with reports of

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similar materials from the literature and from collectors, indicates that such epitactic relationships are much more common than hitherto realized.

Sample description

The sample originates from fissures or vugs in Triassic basalt exposed on Hook Mountain, part of the discontinuous Third Watchung Mountain at Lincoln Park, New Jersey (FAUST, 1975). Secondary minerals preferentially form in these pillow basalts at the intersection of multiple pillows. A geological and mineralogical review of the area is provided by PETERS and PETERS (1978). Tabular babingtonite crystals are thickly coated with brown whiskers, and overgrown by prehnite with additional minor calcite, pumpellyite, and datolite. The whiskers emerge from the babingtonite plates and are inclined 100 – 105° to them. Interestingly, PETERS and PETERS (1978) mention that thin babingtonite crystals are coated on all faces except one set of pinacoids by hairlike crystals that are very light blue or dark brown in color. These whiskers were interpreted as diopside and aegirine ('acmite'), respectively. VITALI (1978) noticed that "babingtonite from the Watchungs is usually found in some degree altered to the fibrous variety of pyroxene known as acmite". Both previous records probably describe epitactic hedenbergite whiskers similar to those investigated in this study. HENDERSON (1981) illustrated babingtonite from Lincoln Park that is apparently identical to the material we have studied, but he interpreted the overgrowths as "hairs of acmite, apparently an alteration product of the babingtonite".

Experimental

A tabular babingtonite crystal was split to produce a stalky crystal fragment elongate parallel to c. Cell dimensions were determined from 22 high angle reflections ($30° > \theta > 17°$: 0 0 8, 0 0 6, 0 10 0, 0 13 0, 9 0 0, 5 3 3 , 4 0 4, 2 9 5, 6 4 0, –5 8 8, –2 0 8, and their symmetry equivalents) on a CAD4 single crystal diffractometer using graphite monochromatized MoKα X-radiation, yielding a = 7.4734(8) b = 12.176(2), c = 6.6881(5) Å, α = 86.134(9), β = 93.896(8), γ = 112.211(9)°, V = 561.59(11) Å (in non-standard setting, e.g. ARMBRUSTER, 2000). In addition, the investigated babingtonite crystal also displayed epitactic bristles of hedenbergite for which cell dimensions (a = 9.80(1), b = 8.99(1), c = 5.241(4) Å, β = 105.4(1)°, V = 445.7 Å^3) and an orientation matrix could be determined. Babingtonite and hedenbergite single-crystal X-ray reflections could be distinguished because hedenbergite reflections are much weaker and about twice as broad compared to those of babingtonite. Miller indices of babingtonite crystal faces were determined from the orientation matrix obtained by single-crystal X-ray diffraction using a microscope attached to the single-crystal diffractometer. Subsequently the hedenbergite orientation matrix was entered and the babingtonite faces were also indexed according to hedenbergite, in order to describe the geometric relationship between the two phases.

In an independent experiment bundles of whiskers were separated from the sample, glued on the tip of a thin glass fiber, and a rotation X-ray pattern was recorded using a Siemens SMART three circle diffractometer (graphite-filtered MoKα-radiation). The resulting powder pattern was analyzed and identified as pyroxene (for detail see ARMBRUSTER et al., 2000).

A polished thin section was studied by electron microprobe (Cameca SX-50) using the same conditions as in the previous study (ARMBRUSTER et al., 2000).

Results

Babingtonite forms (010) plates a few millimeters wide and up to 0.075 mm thick forming irregular aggregates within a dense matrix of Fe^{3+}-rich prehnite $Ca_2Al_{1.38}Fe^{3+}{}_{0.51}Fe^{2+}{}_{0.06}Si_{3.06}O_{10}(OH)_2$. In thin sections, using plane-polarized light, babingtonite plates cut perpendicularly exhibit characteristic pleochroism between blue-green and brownish-pink. In addition the plates display perfect cleavage parallel to (100). Whiskers of hedenbergite, elongate parallel to $[001]_{hed}$, emerge from the (010) surface of babingtonite and form comb-like rows of platy "teeth" arranged parallel to the traces of the $(100)_{bab}$ cleavage (Fig. 1). Some of the babingtonite (010) plates display striations parallel to [001]. Viewed edgewise with the scanning electron microscope, these striations resemble step dislocations. Such steps are also preferentially decorated by the comb-like rows of hedenbergite "teeth". Free standing babingtonite plates (not overgrown by prehnite) show a well defined rim without epitactic whiskers. In contact with babingtonite, hedenbergite often forms a dense, continuous sheet (Fig. 2), allowing electron microprobe analyses (Table 1). Towards their tips the flattened hedenbergite "teeth" thin to ca. 3 µm thick cylindrical whiskers (Figs. 1 and 2). The orientation relations between babingtonite and hedenbergite are (for cell setting see Experimen-

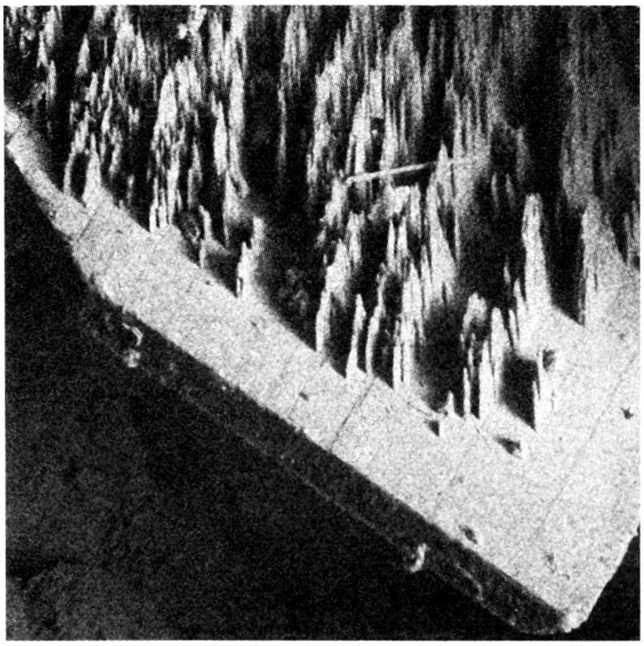

Fig. 1 Scanning electron micrograph (base of image is 0.5 mm) showing a (010) babingtonite plate with epitactic hedenbergite arranged in comb-like rows of "teeth" decorating striations (step dislocations) and cleavage traces running parallel to [001] (sample from Lincoln Park near Paterson, New Jersey, USA).

tal): $(010)_{\mathrm{bab}}$ is parallel to $(\bar{1}\bar{1}1)_{\mathrm{hed}}$ and $(\bar{1}0\bar{1})_{\mathrm{bab}}$ is parallel to $(100)_{\mathrm{hed}}$. These quantitative orientation relations are in agreement with qualitative relations given by ARMBRUSTER et al. (2000) and imply the *c*-axis of hedenbergite (the growth direction of the whiskers) to be inclined by ca. 102° to the (010) face of babingtonite. The major chemical difference between hedenbergite and babingtonite is that babingtonite contains mixed-valence iron and hydroxyl whereas hedenbergite is essentially ferrous and anhydrous (Table 1). Compared to babingtonite hedenbergite is strongly enriched in Mg and slightly enriched in Mn, leading to a diopside component of ca. 17% and a johannsenite component of ca. 8%. The increased Na content in hedenbergite is interpreted as jadeite and aegirine components of 4% each. Pumpellyite-(Fe^{2+}) (Table 1), embedded in prehnite, forms irregular clusters of flaky crystals displaying pleochroism in greenish hues. The calculated Fe^{2+}/Fe^{3+} ratio of pumpellyite and babingtonite is similar. Datolite $CaBSiO_4(OH)$ forms prismatic crystals also embedded in prehnite (Fig. 2).

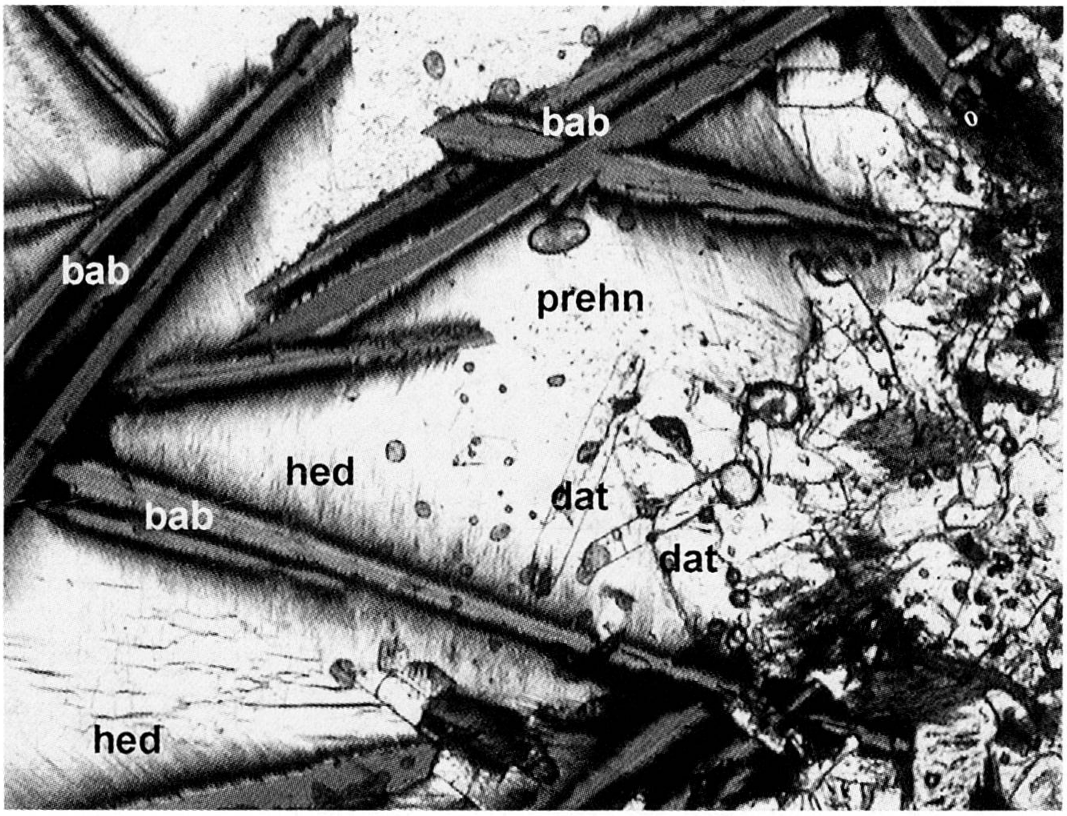

Fig. 2 Microscopic thin section (base of image is 2.15 mm) viewed with plane-polarized light showing plates of babingtonite (seen edgewise) with epitactic whiskers of hedenbergite overgrown by prehnite. The transparent columnar crystals with high relief within the prehnite matrix are datolite. Aggregates of flaky pumpellyite crystals occur in the lower and upper right corner.

Table 1 Electron microprobe analyses (wt%) of babingtonite (average of 13 point analyses), hedenbergite (average of seven point analyses), and pumpellyite (average of ten point analyses). Formulas were normalized to nine,, four, and eight cations. Fe^{2+} and Fe^{3+} were calculated to obtain 29, 12, and 24 negative charges, respectively.

	Babingtonite		Hedenbergite		Pumpellyite-(Fe^{2+})	
	average	range	average	range	average	range
SiO_2	53.14	52.64 – 53.70	50.11	49.82 – 50.44	34.54	34.15 – 35.03
TiO_2	0.01	0.00 – 0.03	0.01	0.00 – 0.04	0.02	0.00 – 0.05
Al_2O_3	0.49	0.14 – 1.04	0.35	0.28 – 0.46	9.71	8.16 – 12.16
FeO^*	9.93	20.81 – 23.58	22.64	20.88 – 24.56	12.96	21.95 – 26.93
Fe_2O_3	13.44				13.56	
MnO	1.51	0.88 – 2.21	2.33	1.57 – 3.44	0.03	0.00 – 0.09
MgO	0.55	0.14 – 0.85	2.85	2.09 – 3.49	0.89	0.78 – 1.03
CaO	19.91	19.53 – 20.33	21.58	21.26 – 22.48	20.87	20.65 – 21.25
Na_2O	0.09	0.02 – 0.14	0.54	0.28 – 0.75	0.01	0.00 – 0.03
H_2O^{**}	1.54				6.76	
Σ	100.61		100.49		99.35	
Si	5.00		2.01		3.05	
Ti^{4+}	0.00		0.00		0.00	
Al	0.05		0.02		1.01	
Fe^{3+}	0.95		0.00		0.89	
Fe^{2+}	0.78		0.76		0.96	
Mn	0.12		0.08		0.00	
Mg	0.08		0.17		0.12	
Ca	2.00		0.92		1.97	
Na	0.02		0.04		0.00	
H	1.00				4.00	

*Original analyses were for FeO: FeO and Fe_2O_3 given in the Table were calculated to obtain a charge-balanced formula.

**H_2O and/or OH were calculated.

babingtonite: $Ca_{2.00}(Fe^{2+}{}_{0.78}Mn_{0.12}Mg_{0.08}Na_{0.02})(Fe^{3+}{}_{0.95}Al_{0.05})Si_5O_{14}(OH)$

hedenbergite: $(Ca_{0.92}Na_{0.04})(Fe^{2+}{}_{0.76}Mn_{0.08}Mg_{0.17}Al_{0.02})Si_2O_6$

pumpellyite-(Fe^{2+}): $Ca_{1.97}Al_{1.01}Fe^{3+}{}_{0.89}Fe^{2+}{}_{0.96}Mg_{0.12}Si_{3.05}O_{11}(OH)_2 \cdot H_2O$

Discussion

FORMATION CONDITIONS

PETERS and PETERS (1978) describe a multistage formation of the secondary minerals in pillow basalts of the First Watchung Mountain; this genesis applies in general to the Lincoln Park location as well. (1) After or contemporary with solidification of the pillows and basalt flows anhydrite and glauberite, derived from lake waters, crystallize together with quartz; (2) quartz replaces basaltic glass and encrusts glauberite and anhydrite; babingtonite forms locally; (3) additional water enters the system dissolving first glauberite and later anhydrite; thus calcium is released to the aqueous system. Prehnite (± datolite) starts crystallizing and overgrows residual anhydrite. Rectangular cavities form where anhydrite dissolves after being overgrown; (4) zeolite minerals begin to form; (5) calcite crystallizes and remaining anhydrite and glauberite may be changed to gypsum and/or thaumasite.

In the sample studied (Fig. 2) it is apparent that from this entire sequence of crystallization and replacement only parts are found. Babingtonite is an early phase of the basalt-cavity fillings attributed by PETERS and PETERS (1978) to stage (2), and prehnite probably together with pumpellyite are the latest phases crystallized in the sample investigated. Similar textural relations were reported by DUGGAN (1986) for calcareous volcanic detritus in the interstices of pillow lavas from western Southland, New Zealand. Knowledge of the thermal stability of babingtonite is somewhat limited. Experimental data by SHVEDENKOV and MAZUROV (1977) indicate that babingtonite is stable over a wide range of fO_2 (defined by the Cu–CuO and Fe_2O_3–Fe_3O_4 oxygen buffers) and up to ca. 550 °C. Additional experimental data above 400 °C were summarized by GOLE (1981). There are no experimental data on the lower temperature stability of babingtonite. However, babingtonite is commonly observed in association with zeolites (BIRCH, 1983; WISE and MOLLER, 1990; NYUI-HIDEKAZU et al. 1993; GON-

ZALES DEL TANAGO and LA IGLESIA, 1998; ARMBRUSTER et al., 2000) suggesting crystallization conditions around 200 °C or even below. DUGGAN (1986) proposed an alternative interpretation, according to which babingtonite in such associations formed at higher temperatures and was preserved metastably at lower temperature during zeolite crystallization. At least for babingtonite with epitactic hedenbergite found in the Alpine fissures at Arvigo (ARMBRUSTER et al., 2000) we exclude a high temperature (>300 °C) formation of babingtonite because babingtonite grows on heulandite, and occasionally heulandite overgrowths babingtonite, suggesting a mineral assemblage. Infrequently in the Lincoln Park material, babingtonite grows on prehnite, reversing the usual sequence and indicating similar temperatures of formation for prehnite and babingtonite. In addition, WISE and MOLLER (1990) observed babingtonite as a late phase, together with quartz and prehnite, grown on chlorite in basalt cavities at Bombay, India. The estimated crystallization conditions were around 200 °C.

For the Arvigo occurrence of epitactic hedenbergite on babingtonite we speculated (ARMBRUSTER et al., 2000) that hedenbergite nucleated on preexisting babingtonite when the oxygen fugacity in the fissures decreased. Notice that the major chemical difference between babingtonite and hedenbergite is the valence of Fe. For an idealized composition, babingtonite is hydrous and has $Fe^{2+}/Fe^{3+} = 1$ whereas hedenbergite is anhydrous and has only Fe^{2+} (Table 1). However, hedenbergite is stable at high fO_2 as will be shown below. The hypothesis of fluctuations in fO_2 and water activity, not unusual for multi-stage hydrothermal crystallization processes, may be applied also to the second occurrence of epitactic hedenbergite on babingtonite in the Watchung Mountains. At a later stage both fO_2 and water activity could have increased again (e.g., during cooling) because prehnite overgrowing the composite crystals is very rich in ferric iron. Such prehnite compositions seem to be characteristic of low-temperature basalt-cavity fillings (ZOLOTUKHIN et al., 1965; DUGGAN, 1986; WISE and MOLLER, 1990).

As thermodynamic data for babingtonite are not well known we used hedenbergite as a proxy to estimate babingtonite formation conditions because both minerals occur in close association and are chemically similar. Hedenbergite associated with babingtonite has near end-member composition. The approach followed was to test if in the given chemical system, as marked in Fig. 3A (estimated from the mineral chemistry and association), hedenbergite is a stable phase under low-grade metamorphic conditions. The calculations were performed using the computer-code THERIAK-DOMINO (DE CAPITANI and BROWN, 1987) in combination with the updated thermodynamic database JUN92 of BERMAN et al. (1988) in the system Si–Al–Fe–O–H ± C without excluding phases. For the estimated bulk composition hedenbergite is stable with prehnite at low metamorphic grade and below the hematite-magnetite buffering reaction, and at higher temperatures with prehnite and magnetite (Fig. 3B). At lower temperature, as suggested by the observed paragenesis, the stability of hedenbergite depends mainly on the bulk and fluid composition; with decreasing a_{H_2O} the hedenbergite stability field tends to widen. SHVEDENKOV and MAZUROV (1977) suggest that babingtonite is stable over a wide range under oxygen fugacity conditions of magnetite.

Hedenbergite and prehnite are shown to react to wollastonite, chlorite, and quartz. Experimental work and natural observations (GUSTAFSON, 1974) suggest that xonotlite $Ca_6Si_6O_{17}(OH)_2$ is probably stable instead of wollastonite below ~ 200 °C. If CO_2 is present calcite and quartz may represent the stable assemblage instead. Moreover, ilvaite $CaFe^{2+}{}_2Fe^{3+}Si_2O_8(OH)$ may also be stable in such a system (e.g., WISE and MOLLER, 1990). Whether ilvaite and babingtonite, ilvaite and hedenbergite, or babingtonite and hedenbergite are stable may depend mainly on the prevailing fO_2 and the bulk Si/Ca ratio.

The obtained stability field for hedenbergite (± prehnite) under low-grade metamorphic conditions agrees well with the natural babingtonite-hedenbergite occurrences associated with zeolites (e.g., ARMBRUSTER et al., 2000) or prehnite–pumpellyite or julgoldite (e.g., BUNGERT et al., 1992). The association babingtonite–hedenbergite seems thus characteristic of the boundary region between the zeolite and lower prehnite-pumpellyite metamorphic fields and not only of greenschist-facies skarn conditions. At slightly higher metamorphic conditions amphiboles such as ferroactinolite replace hedenbergite.

OTHER LOCALITIES WITH EPITACTIC WHISKERS ON BABINGTONITE

W.A. HENDERSON (personal communication to Richards, May 2001) indicates that he has samples of overgrown babingtonite, visually identical to the Lincoln Park material, from Prospect Park, New Jersey; Peabody, Massachusetts; and Bull Run, Loudoun County, Virginia, all from quarries in the Triassic basalt belt. In this study we have

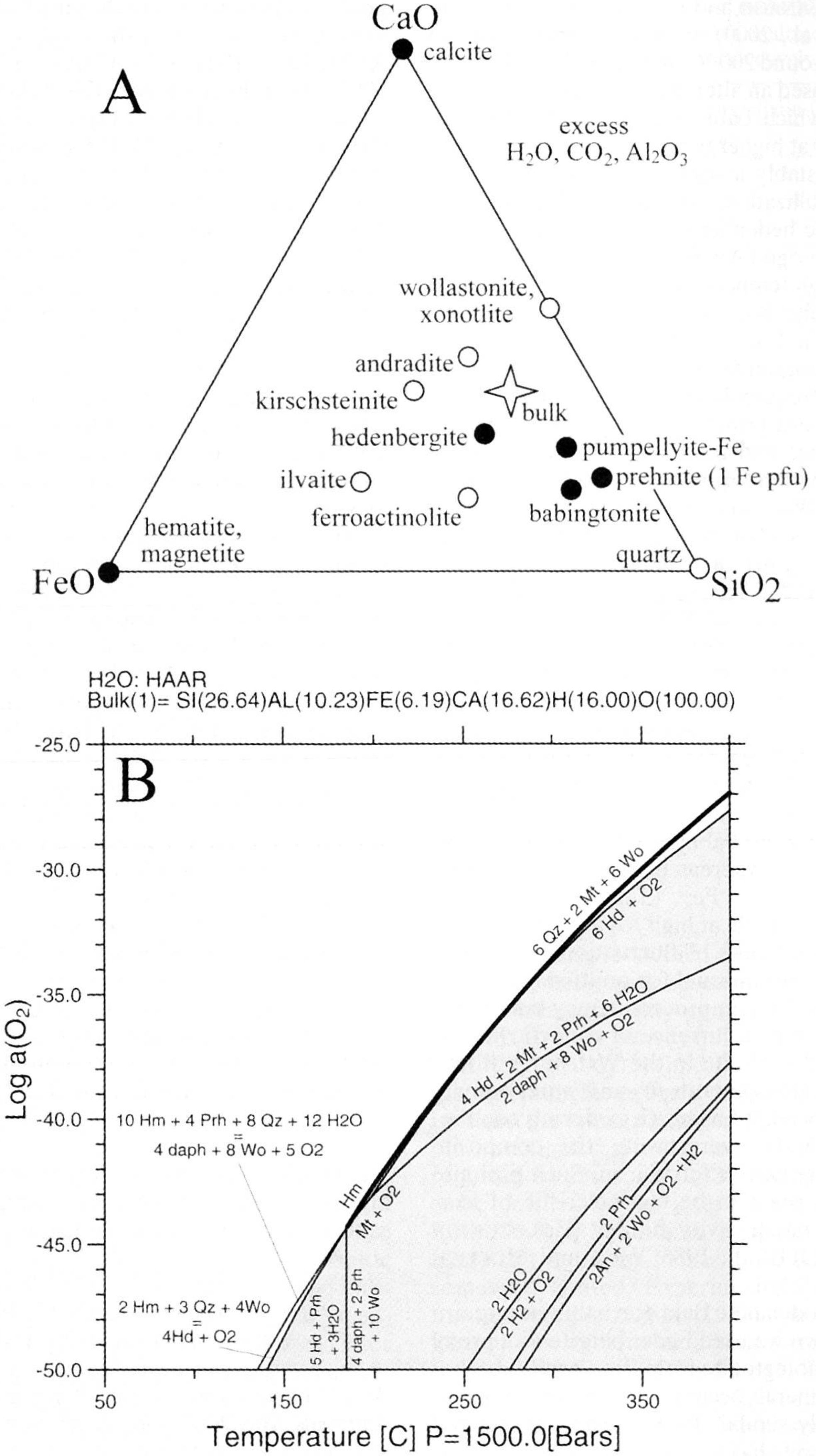

Fig. 3 (A) Mineral phases in the system Fe–Si–Ca–O–H (± Al) characteristic for low-grade metamorphic rocks. Filled circles indicate phases present in the New Jersey sample. The star marks the estimated bulk composition used for calculations. (B) Oxygen fugacity versus temperature diagram illustrating that hedenbergite with babingtonite is likely to occur near the magnetite-hematite buffer reaction (bold). Note: daph (Fe-chlorite) = $Fe_5Al_2Si_3O_{10}(OH)_8$.

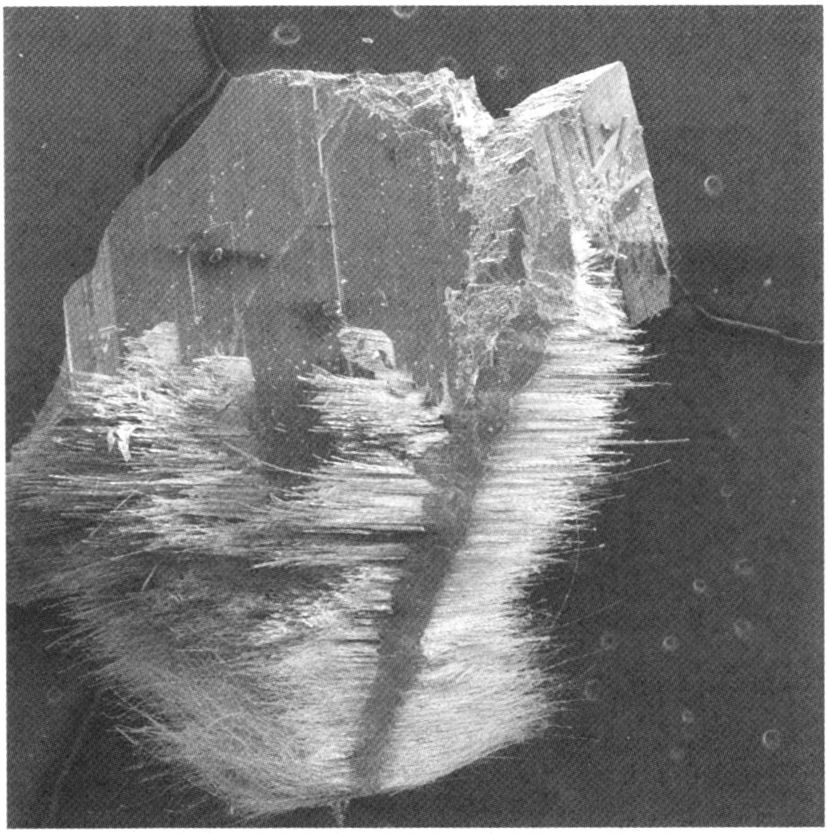

Fig. 4 Scanning electron micrograph (base of image is 1.5 mm) showing a (010) babingtonite plate with epitactic hedenbergite (sample from Manassas Quarry, Prince William County, Virginia, USA).

also analyzed (scanning electron microscopy with energy dispersive system) samples from New Goose Creek Quarry (Loudoun County, Virginia) and from Manassas Quarry (Prince William County, Virginia) and confirmed epitactic hedenbergite on babingtonite (Fig. 4).

BUNGERT et al. (1992) described babingtonite from hydrothermal alteration zones of volcanic rocks in the Saar-Nahe area (Germany). These authors also display epitactic whiskers on babingtonite, very similar to our Fig. 1. However, these whiskers were interpreted as pectolite without stating the identification criteria. A sample from this locality, kindly received from Prof. W. Hofmeister (University of Mainz, Germany), was reanalyzed by us using the same single-crystal X-ray technique as used for the Lincoln Park sample. The results revealed hedenbergite whiskers with $a = 9.810(3)$, $b = 8.978(2)$, $c = 5.247(1)$ Å, $\beta = 105.30(4)°$, $V = 445.2$ Å^3 grown on (010) babingtonite plates following the same epitactic relationships as found for the Lincoln Park sample. Similar epitactic associations were also described (without final identification) by HEIDTKE (1986) from Kreimbach (Saar-Nahe area, Germany). If the unverified observations also represent hedenbergite epitactic on babingtonite, the number of descriptions stands at ten, and the number of localities is at least seven. This association appears to be much more common than previously recognized.

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