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This study is currently published in Mineralogical Magazine as pre-publication;
Nagashima & Nishio-Hamane: TEM study of the epitaxial association of hedenbergite whiskers with babingtonite.

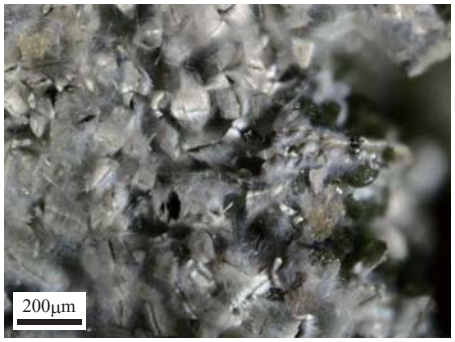
Babingtonite with epitaxial hedenbergite whiskers

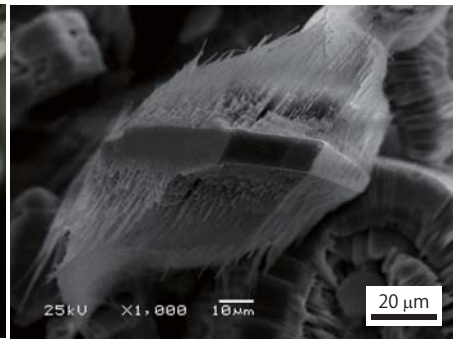
Introduction Hydrous pyroxenoid babingtonite (Bab), $Ca_2Fe^{2+}Fe^{3+}[Si_5O_{14}(OH)]$, rarely coexists with diopside-hedenbergite series clinopyroxene(Hd), $Ca(Fe^{2+},Mg)Si_2O_6$. However, several studies have reported overgrowths of Hd whiskers on the Bab as for example in a quarry at Kreimbach/Kaulbach, Germany, at Arvigo, Switzerland, and Lincoln Park, NJ, US. Based on the morphological and crystal structural features of Bab and Hd, the epitaxial growth of Hd on Bab basal was considered. However, their relation has not been directly observed.

Purpose We directly determine the orientaion between Bab and Hd with transmission electron microscopy (TEM) to provide the structural information on the interfaces between both phases, and also give the further undersanding of their formation.

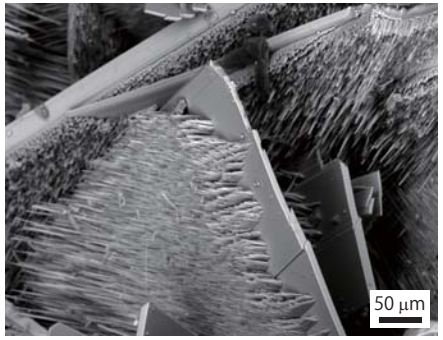
Specimens White Hd fibres are inclined at angles of 105° relative to the platy basis of the deep-green Bab at both localities.

1) Arvigo, CH (Sample No. NMBE34974 supplied by Natural History Museum of Bern)
Green central crystal of babingtonite (ca. 0.15 mm) with a dense felt of white fibres is assosicated with a few intergrown heulandite crystals covered by chlorite.
Composition: Bab $(Ca_{2.00}Na_{0.01})_{\Sigma 2.01}(Fe^{2+}_{0.50}Fe^{3+}_{0.95}Mg_{0.28}Mn^{2+}_{0.20}Al_{0.06})_{\Sigma 1.99}Si_{5.00}O_{14}(OH)$ (n = 14)
Hd fibres $CaFe_{0.5}Mg_{0.5}Si_2O_6$ ($Hd_{50}Di_{50}$)

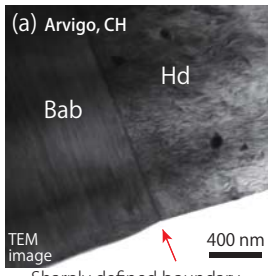




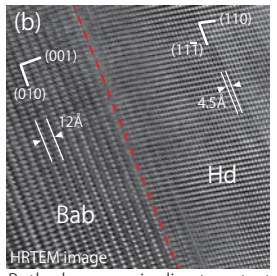
2) Kreimbach/Kaulbach, DE In the interstices of aggregates of calcite, black to greyish-green platy crystals with overgrowths of white fibres were found.
Composition : Bab $(Ca_{2.01}Na_{0.02})_{\Sigma 2.03}(Fe^{2+}_{0.72}Fe^{3+}_{0.99}Mg_{0.15}Mn^{2+}_{0.08}Al_{0.03})_{\Sigma 1.97}Si_{5.00}O_{14}(OH)$ (n = 10)
Hd fibres $(Ca_{0.88}Na_{0.11})_{\Sigma 0.99}(Fe_{0.67}Mg_{0.30}Mn^{2+}_{0.04})_{\Sigma 1.01}Si_{1.99}O_6$ ($Hd_{58}Di_{31}Ae_{11}$)



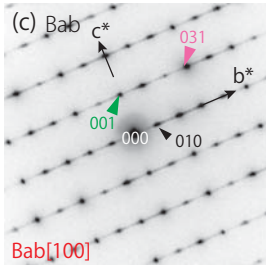
TEM observation : Bab and Hd at their junctions

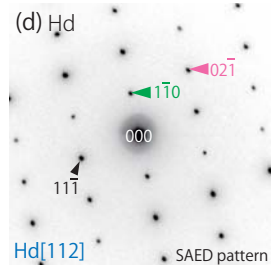


Sharply defined boundary



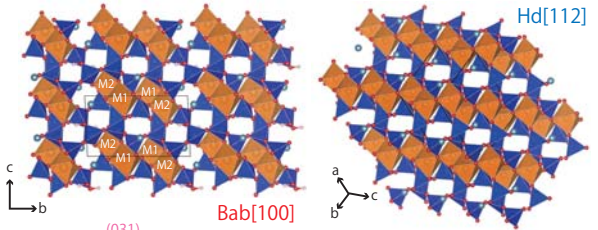
Both phases are in direct contact.

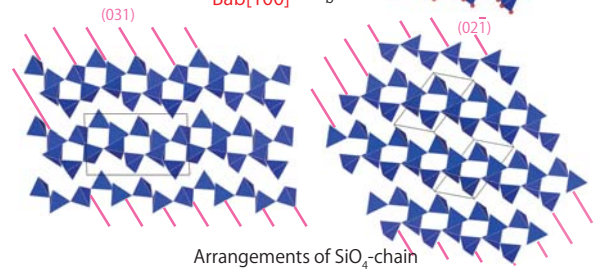




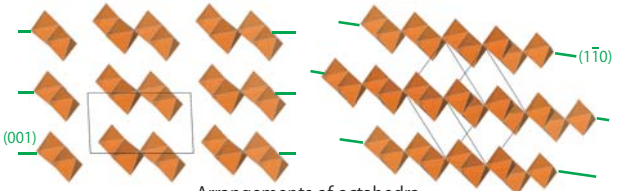
Bab(031) & Hd(02$\bar{1}$), and Bab(001) & Hd(1$\bar{1}$0) are observed at identical positions.

Structural orientation determined by TEM





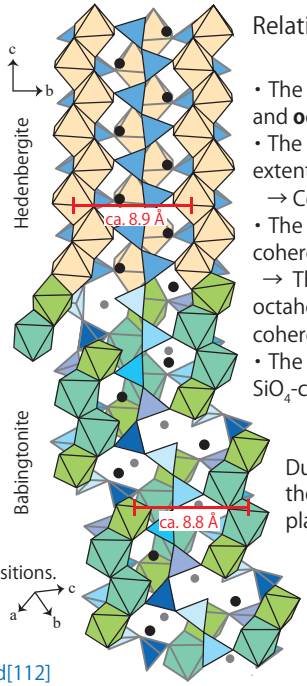
Arrangements of SiO_4-chain



Arrangements of octahedra

The topological relationship of Bab and Hd is explained by the arrangements of the SiO_4-chain units and the octahedra.

Factors controlling coherent growth between pyroxenoid and clinopyroxene



Relationship between babingtonite and hedenbergite

- The arrangements of both **the SiO_4-chain** and **octahedra** are similar.
- The calculated angle between the chain extention direction of cpx and Bab is 16.1° .
  → Consistent with the observation
- The octahedral cluster of Bab transforms coherently to the octahedral ribbon in Hd.
  → The different topologies and individual octahedral sizes do **NOT** disturb their coherent growth.
- The distance between ribbons across a SiO_4-chain in Bab and Hd is largely consistent.

Due to the close orientation relationship, the Hd fibres grew on a base of {010} plate of Bab as epitaxial phase.

- For epitaxial pairs a close chemical relationship is not a requirement, but it may be favourable for the coherent growth.

Epitaxy vs Topotaxy

- Cpx and Px are coexisted as intergrowth phase (e.g. johannsenite and rhodonite). ← Topotaxial intergrowth

<Common principle> Topological relation is governed by the orientaion of SiO_4-chain.

Epitaxial growth		Topotaxial intergrowth
Sharply difined. No addtional phase	Phase boundary	Continuous change. Metastable mixture showing intermediate composition is present.
Epitaxial guest formed under different condition after the host mineral primary formed (Non-equilibrium)	Formation process	Diffusion-controlled solid state reaction (Equilibrium)

Epixial pairs are formed by supercritical hydrothermal fluids, supersaturated solutions or a vapour medium.

Key to epitaxial coherence

Strong surface relationship between the contact planes of the host and guest phases.

In the case of chain silicates, the spots where the silicate chains emerge should preferably be "identical" .

Almost nucleation-energy-free!?

Crystallisation of the guest phase without supplying the complete nucleation energy.

Drawn with VESTA3 (Momma & Izumi 2011)