

Hyalotekite from reedmergnerite-bearing peralkaline pegmatite, Dara-i-Pioz, Tajikistan and from Mn skarn, Långban, Värmland, Sweden: a new look at an old mineral

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Abstract

In specimens of Mn skarn from the type locality of Långban, hyalotekite, $(\text{Ba,Pb,K})_4\text{Ca}_2(\text{Si,B,Be})_{12}\text{O}_{28}\text{F}$, occurs in a matrix consisting mostly of aegirine (≤ 22 mol.% $\text{CaMnSi}_2\text{O}_6$), andradite (≤ 27 mol.% $\text{Mn}_3\text{Fe}_2\text{Si}_3\text{O}_{12}$), hematite, pectolite, quartz, calcite, baryte, barylite, and hedyphane. Melanotekite, plumbian taramellite, ferrian K-feldspar (to 6.5 wt.% Fe_2O_3), rhodonite, a talc-like mineral, apophyllite, and several Pb-As-Sb-O minerals are found in trace amounts. In a single specimen of reedmergnerite-microcline pegmatite from Dara-i-Pioz, hyalotekite occurs in close association with microcline. Other accessory minerals are albite, aegirine, pyrochlore, eudialyte, and polythionite. The optical constants for hyalotekite from Långban and Dara-i-Pioz are, respectively, $\alpha = 1.656, 1.646$, $\beta = 1.659-1.660, 1.649$, $\gamma = 1.670-1.671, 1.659$ (all ± 0.002), $2V\gamma$ (meas) = $57.2-60.5 \pm 0.5^\circ, 57.0 \pm 1.1^\circ$ ($\lambda = 589$ nm). Cell parameters of the Dara-i-Pioz hyalotekite for a body-centred triclinic cell are: $a = 11.284(2)$, $b = 10.930(1)$, $c = 10.272(8)$ Å, $\alpha = 90.35(2)^\circ$, $\beta = 90.11(3)^\circ$, $\gamma = 89.98(1)^\circ$. Electron and ion microprobe data show that Långban hyalotekite is heterogeneous even within a given sample; the most important substitutions are $\text{Pb} = \text{Ba}$, K and coupled $\text{B} = \text{Si}$ and $\text{B} = \text{Be}$. Minor constituents include Mn in the Långban hyalotekite and Na in the Dara-i-Pioz hyalotekite, which also differs in its significantly higher Ba/Pb ratio. Conditions suggested for hyalotekite formation at Långban and Dara-i-Pioz are $P \leq 4$ kbar, $T \geq 500^\circ\text{C}$, silica saturation, peralkalinity, and relatively high oxygen fugacities and low sulphur fugacities. These temperatures must have been sufficiently high to allow for miscibility of Pb with Ba (and K) despite the lone pair of electrons of Pb^{2+} .

KEYWORDS: hyalotekite, metamorphism, peralkaline pegmatite, Mn skarn, Långban, Sweden, Dara-i-Pioz, Tajikistan.

Introduction

HYALOTEKITE, approximately $(\text{Ba,Pb,K})_4\text{Ca}_2\text{-(Si,B,Be)}_{12}\text{O}_{28}\text{F}$, was described as a new mineral from Långban, Filipstad kommun, Värmland, Sweden, by Nordenskiöld (1877). Lindström (1887) published a complete chemical analysis, which showed the mineral to be a complex beryllaborosilicate of Ba, Pb and Ca, a fact that Moore *et al.* (1982) confirmed in their crystal structure refinement. Nordenskiöld (1877) and Aminoff (1918) briefly described hyalotekite as a skarn mineral occurring with hematite, 'schefferite' (manganian diopside), rhodonite, garnet, and hedyphane at Långban. Hyalotekite has also been reported together with melanotekite in veins cutting alamosite from the Artillery Peak, Arizona (Williams, 1982), and a Pb-silicate similar to hyalotekite was found in boulders of feldspathic pegmatite with cesium kupletskite and tienshanite from the alkaline complex at Dara-i-Pioz, southern Tien Shan, Tajikistan (Belakovskiy, 1991).

Despite its discovery over 100 years ago and the crystal structure solution by Moore *et al.* (1982), hyalotekite remains, in some respects, an enigmatic mineral. By studying samples not only from the classic Långban locality, but also from a new paragenesis in reedmergnerite-microcline pegmatite from Dara-i-Pioz, we have attempted to clarify some of unresolved problems with this mineral (we did not find hyalotekite in samples from Artillery Peak, the locality reported by Williams, 1982, and thus we have not considered this locality). In the present paper, we show that the optical properties of hyalotekite differ markedly from those reported by Larsen (1921; also Larsen and Berman, 1934). Hyalotekite shows both $\text{B} = \text{Si}$ and $\text{Pb} = \text{Ba,K}$ substitution over a relatively wide range; neither substitution is common in minerals. Moreover, the latter substitution could be indicative of relatively high temperatures (Moore *et al.*, 1989), and thus the parageneses of hyalotekite are relevant to the crystal chemistry of Pb^{2+} in general.

Mineralogy of the hyalotekite-bearing rocks

Dara-i-Pioz

The hyalotekite-bearing specimen, #615/368 from the Fersman Museum collection (Grew *et al.*, 1993), consists of coarse-grained reedmergnerite and light-grey (with patches of pale lavender) microcline up to 3 cm across. This sample appears veined in hand specimen: white veinlets a few mm thick separate microcline masses and the reedmergnerite is surrounded by a white margin

roughly 1–2 mm thick. Other minerals present are quartz, albite, aegirine, hyalotekite, pyrochlore, eudialyte and polyolithionite. Hyalotekite forms anhedral (rarely euhedral) grains, 0.1 to nearly 1 mm across, and typically with irregular outlines (Fig. 1). Hyalotekite is found as inclusions in the coarse microcline (in one case with polyolithionite), as a constituent in the albite-microcline veinlets, or as grains interstitial to the large microcline grains. Aegirine also occurs in microcline and the albite-microcline veinlets, whereas quartz, pyrochlore and eudialyte are found in veinlets in reedmergnerite or in the margin around reedmergnerite.

Långban

Nine hyalotekite-bearing specimens from Långban were studied in thin-section (Table 1). Five other specimens labelled as hyalotekite, # C2646, R3348,

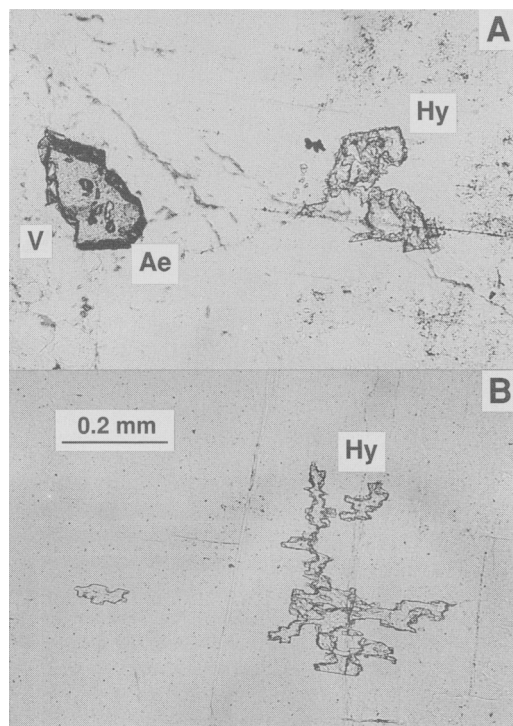


FIG. 1. Photomicrographs of hyalotekite (Hy) enclosed in microcline. (A) Analysed grain near microveinlet (V) consisting mostly of albite. Aegirine (Ae) at veinlet margin. (B) Hyalotekite with a skeletal form. Sample 615/368, Dara-i-Pioz. Plane polarized light.

TABLE 1. Mineralogy of the hyalotekite-bearing skarns from Långban.

	114716 1	114716 2	g14912	g14913	g14914*	g14915	g14917	g29826	BM- 52232	E3345
	NMNH	NMNH	SMNH	SMNH	SMNH	SMNH	SMNH	SMNH	NHM	ROM
	Pb and Ba silicates									
Hyalotekite	X	X	X	X	X	X	X	X	X	X
Melanotekite	T	T	-	X	-	T	T	X	T	T
New Pb silicate	-	-	-	T	-	T	-	-	-	-
Taramellite	-	-	-	T	-	-	-	-	-	-
Barylite	T	T	X	X	T	T	T	X	X	X
	Other silicates									
Quartz	X	X	X	X	X	T	X	X	X	X
K-feldspar	-	X	T	X	T	T	X	T	T	X
Andradite	T	X	X	X	X	T	X	X	X	X
Aegirine	X	X	X	X	X	T	X	X	X	X
Rhodonite	-	X	T	T	-	-	T	-	-	X
Pectolite	X	T	X	X	T	T	T	-	T	T
Talc-like mineral	-	-	X	T	-	-	-	-	-	-
Stilpnomelane-like min.	-	-	-	X	T	T	T	T	-	T
Apophyllite	-	-	T	-	-	T	-	-	-	-
	Non-silicates									
Hematite	T	X	T	X	T	T	X	X	T	X
Calcite	X	X	X	X	X	X	X	X	X	X
Baryte	T	X	T	T	X	T	X	T	T	X
Hedyphane	X	X	X	X	X	X	X	T	T	X
Ecdemite-like mineral	-	-	-	-	-	T	-	-	-	-
Monimolite-like mineral	-	-	-	-	-	-	-	-	-	T
Nadorite-like mineral	-	-	T	-	-	T	-	-	-	-

Note: NMNH-National Museum of Natural History (Smithsonian Institution), SMNH-Swedish Museum of Natural History, NHM-Natural History Museum, ROM-Royal Ontario Museum.

*Holotype material.

X-major constituent, T-trace constituent.

121510 and 137771 from the U. S. National Museum of Natural History and #10428 from the American Museum of Natural History, were studied in hand specimen and, except for 121510, also in thin-section, but no hyalotekite was found. The six specimens from the Swedish Museum of Natural History were representative of the 11 hyalotekite specimens in its collection.

Hyalotekite is light grey and translucent in sufficiently thin slices; the lustre is vitreous, slightly greasy; and cleavage is distinct in some fragments. It forms irregular masses up to several centimetres across in a matrix of hematite and brown aegirine. In thin section, it shows undulatory extinction and locally has a patchy extinction suggestive of incipient cross-hatch twinning reminiscent of microcline. Grains are up to 2 cm across and rarely euhedral; most are

embayed and invaded by microveinlets, commonly consisting of quartz, calcite, barylite (Fig. 2), and baryte. These minerals also form patches interstitial to the coarser hyalotekite grains.

Melanotekite is typically fine-grained, but in g14913 and g29826, it also forms grains or multigranular aggregates up to 2 mm across. It has striking brown and green pleochroism and very high relief. A colourless Pb silicate with high relief and distinct cleavage occurs as a few grains 0.2–0.4 mm across in g14913 and g14915. Chemical analyses and X-ray data indicate this mineral to be a new species, which is currently under study. Taramellite, a mineral not previously reported from Långban, was found only in g14913. It is closely associated with fine-grained aegirine, from which it is distinguished by its striking pink and yellow-brown pleochroism and

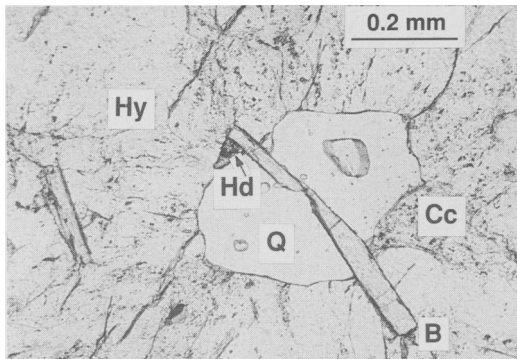


FIG. 2. Photomicrograph of coarse hyalotekite (Hy) enclosing quartz (Q), lath-shaped barylite (B), hedyphane (Hd), and calcite (Cc). Sample BM 52232, Långban. Plane light.

by being optically positive. Taramellite grains are mostly 0.05–0.15 mm in width; in places clusters up to 0.5 mm across extinguish simultaneously.

Barylite generally forms lath-shaped grains, which in places are euhedral, and are mostly 0.1–0.5 mm long in quartz-bearing veinlets and patches in hyalotekite. However, sample g14913 contains grains up to 8 mm long. Barylite can be recognized by its negative elongation, nearly parallel extinction, an optical orientation consistent with the laths being sections of {100} tablets and $\gamma = a$ (e.g. Petersen and Johnsen, 1980) The grains locally have undulatory extinction.

Medium-grained quartz (1–6 mm) commonly shows undulatory extinction. Most quartz is relatively fine-grained (0.05–0.5 mm). K-feldspar forms grains typically 0.05–0.3 mm across (up to 3 mm in g14917) commonly showing anomalous interference colours and irregular zoning. As far as these authors are aware, K-feldspar has not been previously reported from the Mn skarns (Magnusson, 1930).

Andradite that is inferred to be primary forms irregular anhedral to subhedral masses up to 7 mm across. Andradite also forms fine selvages between aegirine, and in sample 114716, it occurs as fine-grained material associated with melanotekite, barylite, quartz, hematite, and secondary aegirine in a single patch within hyalotekite; the latter paragenesis is clearly secondary. Andradite has a distinct yellow colour in thin-section.

Aegirine forms two distinct types. What appears to be primary aegirine forms aggregates that range from medium-grained (0.1–1 mm) and equigranular to coarse-grained (1–4 mm) and inequigranular. Isolated grains are commonly

euhedral, although some grain boundaries are embayed. Primary aegirine is pleochroic in brown and yellow and shows indistinct zoning, which is locally marked (g14912). Secondary aegirine is finer-grained (rarely ≥ 0.1 mm) and commonly paler in colour with a greenish or pinkish tint, but locally it is as dark brown as the primary aegirine. In places, the secondary aegirine forms spherulitic aggregates (e.g. 114716), veinlets that are continuous with fractures that have discoloured primary aegirine, or overgrowths around hematite.

Pectolite forms medium-grained aggregates and very fine-grained tufts. In g14912, it occurs with quartz that is overgrown by apophyllite, calcite, and barylite in a patch of a fine-grained, talc-like mineral. The last resembles sericite. In other samples, very-fine grained tufts appear to be mixtures of the talc-like mineral, pectolite, and other minerals.

A brown, pleochroic (locally, green) mica-like phase is tentatively identified as stilpnomelane. It has moderate birefringence. The brown stilpnomelane-like mineral in g14913 typically forms rounded aggregates 0.1–0.2 mm across of randomly oriented flakes. The brown stilpnomelane-like mineral is commonly found with K-feldspar, hematite, and quartz.

In most samples, calcite is a subordinate constituent of the matrix and hyalotekite masses, whereas in a few samples, e.g. g14915, a portion of the matrix adjacent to hyalotekite and hedyphane is composed of calcite grains in part equant and coarse (1–4 mm across) and in part flattened and finer-grained, forming a foliated aggregate.

Hedyphane forms anhedral grains up to several mm across. Most hedyphane appears weakly anisotropic; a minor amount is more noticeably birefringent. In g14912, the birefringent hedyphane occurs on the margins of the isotropic material in the vicinity of pectolite and the talc-like mineral.

Several minerals were found only in trace amounts; tentative identification is based on semi-quantitative EDS analyses. In sample g14915, an ecdemite-like mineral occurs in clusters up to 1 mm long enclosed in apophyllite and prehnite in the calcite matrix, in one case adjacent to hedyphane. It differs from hedyphane in having even higher refringence, moderate birefringence, anomalous interference colours, and lamellar twinning. A nadorite-like mineral in grains up to 0.13 mm long occurs in calcite and hyalotekite (in g14915) and in pectolite near hedyphane (in g14912). A monimolite-like mineral forms dark-brown isotropic grains (0.07 mm) with hematite in aegirine or in calcite surrounded by aegirine in sample E3345.

Optical properties

Optical properties of Långban hyalotekite and the Dara-i-Pioz hyalotekite were measured on the spindle stage (Table 2). These results support Des Cloizeaux (1878), who reported hyalotekite to be biaxial positive with a high $2V$ and to lack dispersion, but do not support Larsen (1921; see also Larsen and Berman, 1934). The higher indices of refraction for the Långban material are consistent with its higher Pb/Ba ratio (see below). The moderate indices of refraction (relief) and birefringence mean that hyalotekite is not distinctive in thin-section, and could be readily overlooked. A mineral resembling apatite or baryte and associated with other B, Be, Ba or Pb silicates (e.g. barylite, melanotekite) should be checked further.

Hyalotekite from Dara-i-Pioz fluoresces blue in short wave UV light, whereas the Långban material fluoresces yellow. Samples from both localities show blue cathodoluminescence under the electron beam of the microprobe.

X-ray diffraction

The following parameters for a body-centred triclinic cell were obtained on a single crystal of Dara-i-Pioz hyalotekite using an Enraf-Nonius CAD4 four-circle diffractometer at the University of Michigan: $a = 11.284(2)$, $b = 10.930(1)$, $c = 10.272(8)$ Å, $\alpha = 90.35(2)^\circ$, $\beta = 90.11(3)^\circ$, $\gamma = 89.98(1)^\circ$, $V = 1267(1)$ Å³ (from 25 reflections). The cell edges and volume are smaller than those reported by Moore *et al.* (1982) for #114716 hyalotekite, a difference that is consistent with the

higher Ba/Pb ratio in the Dara-i-Pioz hyalotekite (see below).

Chemical composition

Methods

Hyalotekite, taramellite, K-feldspar, aegirine, andradite, hedyphane, and the ecdemite-like mineral were analysed at the University of Maine using an extensively modified MAC 400s electron microprobe equipped with TAP, PET and LiF wavelength dispersive spectrometers (WDS) and automation developed by M.G. Yates. The minerals containing negligible Pb were analysed at 15 kV and 40 nA specimen current on quartz using silicate and oxide standards of simple composition. The data for K-feldspar, aegirine and andradite in 114716 were corrected with Bence and Albee (1968) factors, and those for andradite in g14913, with a ZAF correction scheme. The Pb-rich minerals were analysed at 20 kV and 40 nA specimen current on quartz. The standards were arsenopyrite ASP-200 (As- $L\alpha$), NBS K-456 Pb glass with 70.14 wt.% PbO and 28.60 wt.% SiO₂ (Pb- $L\alpha$ and Si- $K\alpha$), apatite (P- $K\alpha$), scapolite (Cl- $K\alpha$), scheelite (Ca- $K\alpha$ for hyalotekite and hedyphane), wollastonite (Ca- $K\alpha$ for taramellite), baryte (Ba- $L\alpha$), and hematite (Fe- $K\alpha$). We were not able to obtain satisfactory analytical totals for hedyphane and the ecdemite-like mineral. The interference between Pb- $L\alpha$ and As- $K\alpha$ forced us to use the weaker Pb- $M\alpha$ and As- $L\alpha$ peaks, and the count rates were low. The WDS data were corrected using a ZAF correction scheme (Yakowitz *et al.*, 1973). Standardization was routinely checked by running materials of known

TABLE 2. Optical Properties of hyalotekite

	Larsen (1921)		g14914	114716 this paper*	615/368
α	1.960(5)	1.965(3)	1.656(2)	1.656(2)	1.646(2)
β	1.960(5)	1.965(3)	1.659(2)	1.660(2)	1.649(2)
γ	1.963(5)	1.969(3)	1.670(2)	1.671(2)	1.659(2)
$2V\gamma(\text{meas})$	very small	$26^\circ \pm$ (variable)	$57.2(0.5)^\circ$	$60.5(0.5)^\circ$	$57.0(1.1)^\circ$
$2V\gamma(\text{calc})$	-	-	55.4°	62.5°	57.7°
Dispersion	-	$r < v$ (strong)	$r < v$ (weak)	$r < v$ (weak)	-

Note: Errors given in parentheses

*From measurements with the spindle stage at $\lambda = 589$ nm

TABLE 3. Analyses of hyalotekite

	g14914*	114716**	114716	114716	114716	114716	g14913	g14913	g14914	g14914	g14915	615/368
	wet chemical	crystal structure	1 A	2 A	3 A	4 B	1 B	2 B	1 B	2 B	- B	- A
SiO ₂	39.47	39.26	38.12	41.06	39.02	38.81	37.20	36.71	38.61	37.77	38.11	40.74
Al ₂ O ₃	0.18	0	≤0.01	≤0.02	≤0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	≤0.03
Fe ₂ O ₃	0.06	0	0.08	0.07	0.07	0.10	0.07	0.10	0.06	0.05	0.06	0.05
MnO	0.29	0	0.30	0.14	0.25	0.20	0.41	0.47	0.29	0.17	0.12	0
MgO	0.09	0	<0.01	<0.01	≤0.03	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
CaO	7.82	7.62	7.53	7.67	7.42	7.40	7.12	7.12	7.45	7.42	7.28	6.81
K ₂ O	0.89	0	0.65	0.79	1.09	0.59	0.70	0.65	0.80	0.67	1.13	0.74
Na ₂ O	0.17	0	≤0.02	≤0.03	≤0.10	≤0.01	≤0.02	≤0.02	≤0.02	≤0.02	≤0.02	0.77
CuO	0.09	-	-	-	-	-	-	-	-	-	-	-
PbO	25.11	17.62	29.07	24.98	28.39	26.54	26.90	27.09	27.13	28.60	25.82	17.46
Rb ₂ O	-	-	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01
SrO	-	-	<0.01	0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.07
BaO	20.08	29.56	19.63	21.87	17.87	20.54	20.61	19.78	19.96	19.40	19.77	27.30
BeO	0.75	0.65	0.28	0.97	0.33	0.81	0.14	0.13	0.22	0.32	0.57	0.57
B ₂ O ₃	3.73	4.73	5.10	3.06	4.52	3.84	5.91	5.17	6.25	5.88	5.04	4.46
F	0.99	0.98	0.77	0.93	0.83	0.22	0.21	0.18	0.25	0.22	0.25	0.77
Cl	0.06	-	-	-	-	-	-	-	-	-	-	-
-Cl,F=O	-0.43	-0.41	-0.32	-0.39	-0.35	-0.09	-0.09	-0.08	-0.11	-0.09	-0.11	-0.32
L.O.I.	0.59	-	-	-	-	-	-	-	-	-	-	-
Total	99.94	100.01	101.21	101.16	99.45	98.96	99.18	97.32	100.91	100.41	98.04	99.43
Formulae												
O	28.5	28.62	28.5	28.5	28.5	28.5	28.5	28.5	28.5	28.5	28.5	28.5
Si	9.784	9.62	9.490	10.036	9.746	9.763	9.311	9.450	9.335	9.327	9.524	9.807
Be	0.447	0.38	0.170	0.572	0.196	0.492	0.085	0.081	0.130	0.189	0.342	0.327
B	1.596	2.00	2.193	1.291	1.948	1.667	2.553	2.298	2.610	2.508	2.176	1.851
Al	0.053	0	0	0	0	0	0	0	0	0	0	0
Fe	0.011	0	0.016	0.014	0.014	0.018	0.014	0.018	0.011	0.010	0.012	0.010
Mg	0.033	0	0	0	0	0	0	0	0	0	0	0
Sum IV	11.924	12.00	11.869	11.913	11.904	11.940	11.963	11.847	12.086	12.034	12.054	11.995
Cu	0.016	-	-	-	-	-	-	-	-	-	-	-
Mn	0.061	0	0.063	0.028	0.052	0.043	0.086	0.102	0.058	0.036	0.026	0
Ca	2.077	2.00	2.008	2.008	1.984	1.995	1.909	1.964	1.931	1.962	1.950	1.757
Sr	-	-	0	0.001	0.001	0	0	0	0	0	0	0.010
Na	0.082	0	0	0	0	0	0	0	0	0	0	0.357
Sum	2.236	2.00	2.071	2.037	2.037	2.038	1.995	2.066	1.989	1.998	1.976	2.124
K	0.281	0	0.205	0.247	0.348	0.190	0.222	0.212	0.247	0.210	0.359	0.228
Rb	-	-	0	0	0	0	0	0	0	0	0	0.001
Ba	1.951	2.838	1.915	2.095	1.749	2.025	2.022	1.995	1.891	1.877	1.937	2.575
Pb	1.676	1.162	1.948	1.644	1.908	1.797	1.813	1.877	1.765	1.901	1.737	1.132
Sum	3.908	4.000	4.068	3.986	4.005	4.012	4.057	4.084	3.903	3.988	4.033	3.936
Total												
cations	18.068	18.000	18.008	17.936	17.946	17.990	18.015	17.997	17.978	18.020	18.063	18.056
F	0.776	0.76	0.605	0.720	0.658	0.172	0.165	0.143	0.193	0.176	0.198	0.584
Cl	0.025	-	-	-	-	-	-	-	-	-	-	-

Note: *From Lindström (1887). L.O.I. = loss on ignition.

**Weight % were calculated from cation occupancies determined from crystal structure refinement by Moore *et al.* (1982). Moore *et al.* reported that the F occupancy is 1.0.

#A and B refer to sessions on the ion microprobe

TABLE 4. Analysis of taramellite in g14913

	wt. %	Formula 29 O
SiO ₂	30.29	7.823
B ₂ O ₃	5.03	2.243
Al ₂ O ₃	≤0.10	-
Fe ₂ O ₃	20.19	3.923
MnO	0.17	0.038
CaO	0.15	0.043
BaO	33.93	3.434
PbO	8.50	0.591
Cl	1.08	0.477
-Cl=O	-0.24	-
Total	99.10	-
Total Cat.		18.094

Note: Ion microprobe gave B₂O₃ and Al₂O₃; Li, Be, F below detection; Na, K, Sr ≤ 0.03 wt.% oxide; and trace Mg, Ti.

Electron microprobe: P below detection.

composition as unknowns. The uncertainties in the electron microprobe analyses can be assessed from the theoretical counting statistics for four major constituents calculated for hyalotekite in sample g14914, spot 1. The 2σ errors are (in wt.%): CaO 0.10, BaO 0.24, SiO₂ 0.65, and PbO 0.76, that is, respectively, 1.4, 1.2, 1.7 and 2.8 % relative to the tabulated values.

The Link AN 10000 energy dispersive system was used for obtaining quick, semi-quantitative analyses of the minerals associated with hyalotekite.

Ion microprobe analyses were obtained from an ARL Ion microprobe mass analyser during two sessions at the Aerospace Corporation (method of Grew *et al.*, 1990). The following standards and sensitivity ratios were used: spodumene (Li/Si), surinamite (Be/Si), grandierite (B/Si), and biotite (F/Si and Ba/Si); 3.3 (Rb/Si), 3.7 (Sr/Si), and 5.5 (Cs/Si). Ion microprobe data for kornerupine (sample BM1940,39), which was analysed concurrently with hyalotekite and taramellite, are in fair to good agreement with the wet chemical data of Grew *et al.* (1990, in parentheses) for sessions A and B as follows (in wt.% oxide): Li₂O 0.17, 0.16 (0.19); BeO 0.022, 0.019 (0.022); B₂O₃ 3.81, 3.64 (4.01); and F 0.85, 0.57 (0.50). Uncertainties were estimated by Grew *et al.* (1990) to be about ± 0.5 wt.% B₂O₃ and ± 0.3 wt.% F. More significant figures than is warranted by the analytical precision in the microprobe data has been

TABLE 5. Analyses of K-feldspar

Grain Spot#	g14913	g14913	g14913
	1	2	2
	1/3	1/2	2/1
Electron Microprobe			
SiO ₂	64.26	63.90	64.43
Al ₂ O ₃	14.25	16.15	17.65
Fe ₂ O ₃	6.31	4.36	2.41
CaO	0.04	0.03	0.03
K ₂ O	15.87	15.74	16.27
Ion Microprobe*			
BeO	0.009	0.006	0.006
B ₂ O ₃	0.03	0.01	0.01
Na ₂ O	≤0.10	≤0.10	≤0.10
Rb ₂ O	0.01	0.007	0.007
BaO	0.03	0.02	0.02
Total	100.81	100.20	100.84
Formulae per 8 O			
Si	3.004	2.983	2.975
Be	0.001	0.001	0.001
B	0.002	0.001	0.001
Al	0.785	0.888	0.961
Fe	0.222	0.153	0.084
Ca	0.002	0.001	0.001
K	0.947	0.937	0.958
Ba	0.001	0	0
Total	4.964	4.964	4.981

Note: Pb near limit of detection with the electron (3σ value = 0.47 wt.% oxide) and ion probes. Ion microprobe also gave a trace of Sr and Cs below detection. Electron microprobe gave Na₂O 0.13–0.23 wt.% and BaO 0.13–0.17 wt.%.

#Analytical spot/number of analyses at this spot.

*Single ion microprobe analysis for grain 2 has been applied to both spots.

reported in Tables 3–6 in order to minimize rounding-off error in future calculations.

Results: hyalotekite

The most important constituents in hyalotekite are Si, Ca, K, Pb, Ba, Be, B and F; Mn and Na are minor constituents; and Fe barely exceeds the 3σ detection limit of 0.063 wt.% Fe₂O₃ (Table 3). Na, Mg and Al are present in amounts below the detection limits (3σ values are 0.13, 0.09 and 0.32 wt.% oxide, respectively) on the electron microprobe (except Na in 615/368), and maximum

TABLE 6. Analyses of aegirine and andradite

	Aegirine 1 114716	Aegirine 2 114716	Andradite 2 114716	Andradite 2 114716	Andradite 1 g14913
	Electron microprobe wt. %				
SiO ₂	52.27	52.70	35.85	35.86	35.63
TiO ₂	0	≤0.01	0	0	-
Al ₂ O ₃	0	≤0.03	0	0	0.04
Fe ₂ O ₃	17.66	31.16	30.07	29.72	26.87
FeO	3.35	-	-	-	-
MnO	6.57	1.79	11.14	5.14	13.01
MgO	2.39	0	≤0.01	0	0
CaO	9.06	1.79	23.33	28.72	24.53
Na ₂ O	7.90	12.17	0	≤0.02	-
Total	99.20	99.61	100.39	99.44	100.08
	Formulae				
O	6	6	12	12	12
Si	2.040	2.029	3.071	3.065	3.077
Al	0	0	0	0	0.004
Fe ³⁺	0.519	0.903	1.939	1.911	1.746
Fe ²⁺	0.109	-	-	-	-
Mn	0.217	0.058	0.808	0.372	0.951
Mg	0.139	0	0	0	0
Ca	0.379	0.074	2.141	2.630	2.270
Na	0.598	0.909	0	0	-
Total	4.0	3.973	7.959	7.978	8.048

Note: Fe presumed to be Fe₂O₃ except in column 1, where formula normalized to 4 cations and FeO/Fe₂O₃ ratio calculated accordingly.

1-Relatively coarse-grained, inferred to be primary.

2-Finer-grained, inferred to be secondary.

contents listed in Table 3 have been estimated from ion microprobe data. Ti and Cl are also below detection on the electron microprobe, and Li is below detection on the ion microprobe. Rb and Sr are present in traces (≤ 60 ppm) and Cs is close to the limit of detection in the Långban hyalotekite, whereas these constituents are more abundant in the Dara-i-Pioz sample, i.e. ≈ 30 ppm Cs.

The microprobe analyses gave reasonable totals (average = 99.61 wt.%) and stoichiometries in good agreement with the crystal structure deduced by Moore *et al.* (1982), that is average Si + B + Be + Fe = 11.961, Ca + Mn + Na + Sr = 2.033, Pb + Ba + K = 4.007, and average total cations, 18.001. The Pb/Ba ratio of sample 114716 exceeds that reported by Moore *et al.* (1982) for the same sample, but agrees with electron microprobe data on 114716 obtained by P. Dunn

(pers. comm., 1993): PbO 27.5, BaO 19.2 wt.%. The Pb/Ba ratio given by Moore *et al.* (1982) is lower than that in any of the Långban hyalotekites, including Lindström's, a situation for which Moore *et al.* (1982) had no explanation. Microprobe values for major constituents in the Långban hyalotekite, including g14914, the holotype specimen analysed by Lindström, are in good agreement with Lindström's (1887) values obtained by wet chemical methods if allowance is made for the compositional variations discussed below. However, F values are lower than those reported by Lindström and Moore *et al.*, particularly those obtained during the second of two sessions (B, Table 3) on the ion microprobe (F ≤ 0.25 wt.%). F values are less reproducible than those for B (Grew *et al.*, 1990), and thus the ion microprobe data are not conclusive evidence for a deficiency in F relative to the one F p.f.u. reported

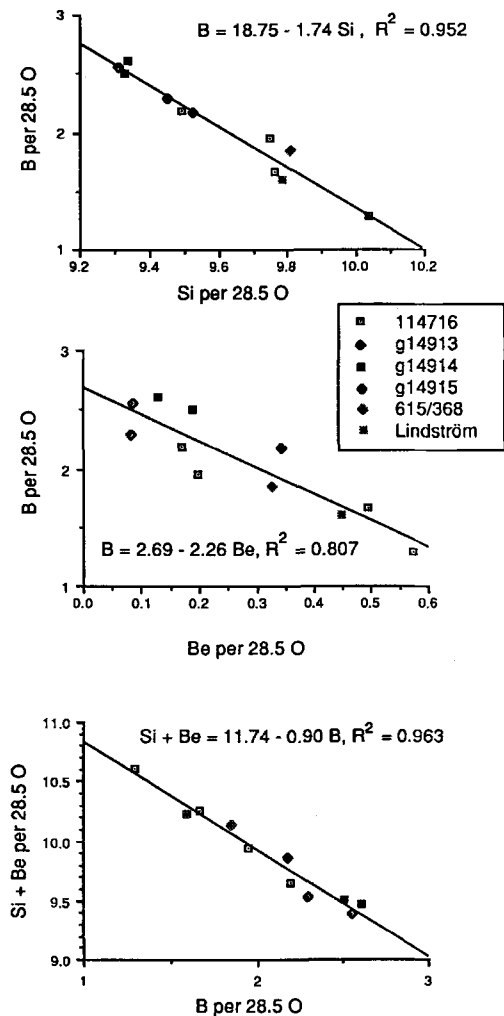


FIG. 3. Plots of Be, B and Si contents of hyalotekite. Lindström refers to the wet chemical analysis reported by Lindström (1887), which was included in the least-squares regression.

by Moore *et al.* (1982). Charge balance calculated from the occupancies reported by Moore *et al.* (1982) requires $0.76 F + 0.24 O$ on the F site, in good agreement with Lindström's F content (Table 3). Alternatively, a $K + F = (Ba, Pb) + O$ substitution could increase the F content in the formula of Moore *et al.* until $F = 1$. Lindström obtained 0.59 wt.% loss on ignition, but did not mention the possibility that this could be essential H_2O . Further study is needed to resolve this ambiguity regarding the anion composition of hyalotekite.

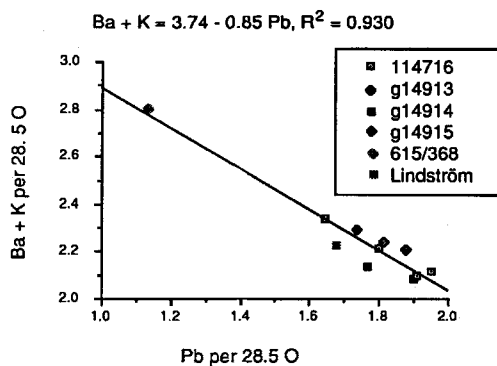


FIG. 4. Plot of Pb, Ba and K contents of hyalotekite. Lindström refers to the wet chemical analysis reported by Lindström (1887), which was included in the least-squares regression.

Långban hyalotekite is heterogeneous, not only from sample to sample, but also from grain to grain within a given sample. The major difference between the Långban and Dara-i-Pioz hyalotekites is Pb/Ba ratio; the Dara-i-Pioz hyalotekite is also sodic and contains no Mn. The compositional variations can be related by the substitutions $Be + Si = 2B$ and $Pb = Ba, K$ (Figs. 3,4). Lindström's analysis plots close to the trends determined by the microprobe data, which is consistent with his analysis being an average of heterogeneous material having compositions similar to those analysed by microprobe in the present study.

The substitution $Be + Si = 2B$ probably results from coupling of $Si = B$ and $Be = B$ substitutions on the B and Si(1) sites of Moore *et al.* (1982). These tetrahedral sites share corners as part of a 4-membered ring. This substitution is consistent with the T-O bond lengths reported by Moore *et al.* (1982): 1.496 Å for the B site, somewhat higher than $[^{41}B-O = 1.46$ Å, and 1.597 Å for the Si(1) site, somewhat lower than for either $[^{41}Be-O = 1.62$ and $[^{41}Si-O = 1.61$ Å (radii from Shannon, 1976, assuming $[^{21}O]$). The chemical data in Table 5 indicate that there is too little B to fill the B site in some hyalotekite, an excess of B in others. The tetrahedral composition $Si_8(Si_{1.5}Be_{0.5})B_2O_{28}$ proposed by Moore *et al.* (1982) should be generalized to $Si_8(B_{2+2x}, Si_{1.5-x}, Be_{0.5-x})_{\Sigma 4}O_{28}$. Hyalotekite thus joins kornrupine, serendibite, synthetic $KBSi_3O_8$, and possibly sapphirine as one of the few silicates with substantial $B = Si$ substitution (see Van Derveer *et al.*, 1993; Kimata, 1993), and the gadolinite group, as a silicate with $Be = B$ substitution (Semenov *et al.*, 1963). However, hyalotekite is unique, as far as we

are aware, in that B = Si substitution is coupled with B = Be substitution. In summary, a generalized formula for hyalotekite consistent with the available analytical and crystallographic data is (given that hyalotekite is associated with hematite, Fe is assumed to be Fe^{3+} , which would prefer a tetrahedral site to the other, much larger sites): $(\text{Ba}, \text{Pb}, \text{K})_4(\text{Ca}, \text{Na}, \text{Mn}, \text{Sr})_2[(\text{Si}, \text{B}, \text{Be}, \text{Fe})_{12}\text{O}_{28}]\text{F}$.

The clear-cut evidence for Pb = Ba, K substitution (Fig. 4) appears to be inconsistent with the distinctive stereochemical behaviour of Pb^{2+} resulting from its lone pair of s^2 electrons. Moore *et al.* (1989) listed hyalotekite as a mineral that might have crystallized above a temperature where the lone-pair electrons no longer have an effect and Pb miscibility with K and Ba would be possible. Hyalotekite could be a relatively high-temperature mineral, as discussed below.

Results: other minerals

Långban taramellite differs from previously analysed material (Alfors and Pabst, 1984) in its negligible Ti content and substantial Pb = Ba substitution, which gives a formula close to $(\text{Ba}, \text{Pb})_4\text{Fe}_4\text{B}_2\text{Si}_8\text{O}_{29}\text{Cl}_{0.48}$ (Table 4). However, there is an excess of anionic charge in this formulation. In other taramellites, Cl is presumed to balance the extra charge of tetravalent cations substituting for octahedral Fe^{3+} (Mazzi and Rossi, 1980). In the absence of detectable P, there is no evidence for extra charge on tetrahedral sites. Mazzi and Rossi (1980) calculated that no oxygen atom is seriously deficient in charge even if only Fe^{3+} occupied the octahedral sites, and concluded that OH was absent in taramellite. On the other hand, Matsubara (1980) did a similar calculation for the isostructural mineral nagashimalite, in which V^{3+} substitutes for Fe^{3+} , and reported that one oxygen atom was deficient. Possibly the Pb = Ba substitution in the Långban taramellite results in a local charge deficiency that is balanced by OH = O substitution.

The Mn_2O_3 contents of melanotekite $[\text{Pb}_2(\text{Fe}^{3+}, \text{Mn}^{3+})_2\text{Si}_2\text{O}_5]$ range from less than 0.5 (below detection) to 1.6 wt.%, equivalent to about 6 mol.% kentrolite (EDS analyses).

The only significant compositional variation in K-feldspar is the presence of Fe_2O_3 , with a range of 0.7–6.5 wt.% (EDS analyses and Table 5). Fe^{3+} = Al substitution could account for the optical zoning. These are the most ferrian K-feldspars reported from a metamorphic rock; other ferrian K-feldspars occur in alkaline volcanic rocks (e.g. ≤ 14.5 wt.% Fe_2O_3 , Linthout and Lustenhouwer, 1993).

Andradite is manganoan (calderitic) with negligible Al and Mg (Table 6). In g14913, the deficiency in Fe could be made up by Mn^{3+} ; $\text{Mn} + \text{Ca} = 3.221$ p.f.u., significantly above the ideal stoichiometry. Fine-grained, secondary andradite in 114716 is heterogeneous, ranging from 12–27 mol.% calderite and nearly ideal Fe contents ($\text{Fe} \geq 1.891$ p.f.u., e.g. Table 6).

Aegirine, inferred to be primary, including coarse- and medium-grained varieties, contains substantial $\text{CaMnSi}_2\text{O}_6$ and $\text{CaMgSi}_2\text{O}_6$ (Table 6 and EDS analyses). Finer-grained and not strongly coloured aegirine, which is presumably secondary, generally contains at least 90 mol.% $\text{NaFeSi}_2\text{O}_6$, whereas the dark-brown variety in E3345, also presumed to be secondary, contains 20 mole.% $\text{CaMnSi}_2\text{O}_6$ and < 5 mol.% $\text{CaMgSi}_2\text{O}_6$ (EDS analyses). Rhodonite in samples g14913 and 114716 is close to $(\text{Mn}, \text{Ca})\text{SiO}_3$ with 16–18 mol.% Wo; minor MgO (≤ 0.6 wt.%) and Na_2O (≤ 1 wt.%) are present in 114716 (EDS analyses). Pectolite contains from 0.8–14 wt.% MnO (samples g14913, g14915, 114716), corresponding to atomic $\text{Mn}/(\text{Mn} + \text{Ca}) = 0.02$ –0.3 (EDS analysis).

EDS analysis of the sericitic talc-like mineral in sample g14912 gives a talc stoichiometry in which 4% of the Mg has been substituted by Fe, and 7% by Mn. Analyses of the talc-like tufts in sample g14913 yield larger amounts of Mn, as well as variable amounts of Na, K, Ca, Fe and Pb; these tufts are undoubtedly mixtures.

Hematite contains ≤ 0.6 wt.% Mn_2O_3 and negligible TiO_2 (≤ 0.2 wt.%, WDS and EDS analyses). Calcite contains ≤ 5 mol.% each of MnCO_3 and BaCO_3 ; Mg, Fe and Pb contents are just above the limit of detection or below it (EDS analyses).

Weakly anisotropic hedyphane grains analysed semi-quantitatively by WDS are barian and phosphatian: $\text{Pb}_{3.04}\text{Ca}_{1.34}\text{Ba}_{0.66}\text{As}_{2.57}\text{P}_{0.41}\text{O}_{11.93}\text{Cl}_{1.12}$ (g14913) and $\text{Pb}_{3.68}\text{Ca}_{1.21}\text{Ba}_{0.43}\text{As}_{2.63}\text{P}_{0.24}\text{O}_{11.95}\text{Cl}_{1.10}$ (g14915), similar to the compositions reported in other samples from Långban by Rouse *et al.* (1984). Birefringent hedyphane differs from associated nearly isotropic hedyphane in sample g14912 mainly in its negligible Ba content (EDS analyses). Constituents other than Pb, As and Cl were not detected in EDS scans of the ecdemite-like mineral. Semi-quantitative WDS analyses gave PbO, As (as As_2O_3), and Cl in roughly the same proportions as Nordenskiöld (1877) reported for ecdemite.

The EDS analyses of the monimolite-like mineral gave an approximate formula $(\text{Pb}_{1.85}\text{Ca}_{0.74}\text{Mn}_{0.05})_{\Sigma 2.64}(\text{Sb}_{1.84}\text{Fe}_{0.51})_{\Sigma 2.35}\text{O}_8$,

which was written assuming that Fe is trivalent and substitutes for Sb^{5+} . (Moore, 1968, who reported that Sb^{5+} and Fe^{3+} are disordered over one site in melanotibite.) The mineral resembling nadorite [$\text{PbSb}^{3+}\text{O}_2\text{Cl}$] contains major Pb, Sb, Cl and minor Fe, Ca (<1 wt.% oxide, EDS analyses).

Paragenesis

Dara-i-Pioz. Textures suggest two periods of crystallization of the reedmergnerite-bearing pegmatites overall, including the hyalotekite-bearing specimen (Grew *et al.*, 1993): (1) coarse-grained reedmergnerite, quartz, polyolithionite and orthoclase; and (2) fine-granular quartz and fine-grained albite, orthoclase, aegirine, pyrochlore, hyalotekite, eudialyte, leucospheinite, leucophanite and zektzerite. This suite of rare minerals formed by reaction of reedmergnerite and orthoclase with fluids containing Li, Be, F, Zr, Nb and U during the second period, after which orthoclase inverted to microcline. It is possible that the Pb and Ba in hyalotekite originated from orthoclase. Temperatures and pressures are inferred not to have exceeded 450–500°C and a few kilobars, respectively, which are the conditions estimated for reedmergnerite crystallization during the first period (Grew *et al.*, 1993).

Långban

Magnusson (1930) proposed a 4-period sequence of mineral parageneses for the Långban skarns and associated rocks. Primary minerals and the earliest skarn minerals, e.g. braunite, hematite, and quartz, formed at relatively low temperatures during period A. Hyalotekite, hedyphane, aegirine, barylite, melanotekite (in part), and andradite (in part) crystallized late in period B when most of the high-temperature skarn minerals formed, and continued into period C, when the vug minerals formed. Magnusson (1930) reported a second thermal high late in period C, which was followed by the fissure mineral period D, when minerals such as pectolite and apophyllite formed. Overall, the Långban area has been affected by regional metamorphism in the amphibolite-facies succeeded by contact metamorphism associated with the post-orogenic Hyttsjö and Filipstad granite massifs (Björk, 1986; Baker *et al.*, 1988; Rickard, 1988).

Textures in the Långban hyalotekite-bearing skarns are broadly consistent with the interpretation of Magnusson (1930, 1970). Crystallization of medium- to coarse-grained, nearly isotropic hedyphane, hyalotekite, primary aegirine, rhodonite, andradite, barylite and melanotekite appear

to represent an early, relatively high-temperature event corresponding to period B (Åberg and Charalampides, 1986). The fine-grained aggregates dominated by secondary aegirine, quartz and calcite with accessory barylite, baryte, melanotekite, andradite, K-feldspar, rhodonite and taramellite are inferred to represent recrystallization during a later event at temperatures approaching those of the preceding event, that is, period C. Pectolite, apophyllite and minerals resembling stilpnomelane, talc, and ecdemite, as well as the new Pb silicate, are inferred to have formed during a third event at substantially lower temperatures, which could be equated with Magnusson's (1930) period D and the retrogressive phase mentioned by Baker *et al.* (1988). In the hyalotekite-bearing skarns, the late minerals are not restricted to discrete fissures or druses, but appear in patches and aggregates dispersed throughout.

Temperatures and pressures for the two earlier events, which could have resulted from the contact effects of nearby intrusives belonging to the Hyttsjö granite and Filipstad granite (see the map in Björk, 1986), can only be estimated from circumstantial evidence. Cordierite, sillimanite, and andalusite in rocks associated with the Långban skarns, including a sample containing andalusite and microcline, are inferred to have crystallized during period C, somewhat later than the main skarn crystallization (Magnusson, 1930). We examined an andalusite–microcline–biotite–quartz–cordierite rock (Swedish Museum of Natural History #700208). Cordierite, andalusite, and microcline appear to have formed coevally. Subsequently, cordierite was largely replaced by sericite, and biotite was partially altered to chlorite. Conditions for the transition of andalusite to sillimanite and for the andalusite + K-feldspar assemblage are estimated from experimental data to have been $T \geq 600^\circ\text{C}$, $P < 2$ kbar (assuming $X_{\text{H}_2\text{O}} = 1$ in the andalusite–microcline rock) or $T \geq 540^\circ\text{C}$, $P < 3$ kbar (at $X_{\text{H}_2\text{O}} = 0.3$, e.g. Holdaway, 1971; Kerrick, 1972). Assemblages with aegirine solid solutions, calderitic andradite, and rhodonite are found in Labrador in Fe–Mn rich rocks metamorphosed under amphibolite-facies conditions corresponding to the kyanite + staurolite zone in pelitic rocks (Klein, 1966). Temperatures estimated for this zone are roughly 500–600°C at 4 kbar and increase with pressure (e.g. Thomson and Guidotti, 1989, Fig. 7). Other indications of relatively high temperatures are the presence of ferrian K-feldspar, a mineral of alkalic volcanics (Smith and Brown, 1988) and taramellite, a mineral most typical of, though not restricted to, contact deposits (Alfors and Pabst,

1984). In summary, temperatures of 500–600°C and *P* as low as 2 kbar for hyalotekite formation are consistent with available information.

Comparison of the two hyalotekite parageneses

The parageneses at Dara-i-Pioz and Långban at first glance appear to be very different. Nonetheless, they have several important features in common, features that may be critical in stabilizing hyalotekite. In addition to the availability of Pb, Ba, B and Be, the common features are silica-saturation, peralkalinity, high oxygen and halogen fugacities, low contents of S and Mg, low pressures and moderate temperatures (possibly $P \leq 4$ kbar, $T \geq 500^\circ\text{C}$).

Quartz is found in the hyalotekite-bearing rocks from both Dara-i-Pioz and Långban, but only in the Långban skarns is it directly associated with quartz. In general, at Långban silica-saturated Mn skarns are less common than silica-undersaturated Mn skarns, in which olivine minerals are reported to be relatively common (Magnusson, 1930). Peralkalinity in Långban and Dara-i-Pioz parageneses results from the excess of Na + K over Al. Although the Långban hyalotekite-bearing skarns appear to contain little alkali, their Al content is even less, so that the small amount of Na present is incorporated in aegirine. In both parageneses, K is incorporated in K-feldspar, which is abundant at Dara-i-Pioz, but sparse at Långban. The low Al₂O₃ contents, together with the high oxygen fugacities, favoured the substitution of Fe³⁺ for Al in the Långban feldspar, as suggested by Hall (1982) for ferrian sanidine in a minette.

Iron in both parageneses is largely ferric; indeed firm evidence for Fe²⁺ in the Långban skarns is lacking. On the other hand, measurable Mn in hematite nearly free of Ti and in melanotekite suggests that minor Mn³⁺ is present in the Långban hyalotekite-bearing skarns. The presence of rhodonite instead of braunite + quartz implies oxygen fugacities above those buffered by hematite-magnetite and below those buffered by Cu₂O–CuO if the effect of Ca incorporation in rhodonite is ignored (Abs-Wurmbach *et al.*, 1983).

Sulphur contents of the Långban and Dara-i-Pioz parageneses are low and (Pb + Ba) ≫ S, so that Ba and Pb are incorporated in silicate phases in addition to baryte. The activities of SO₃ and CO₂ in the reedmergerite- and hyalotekite-bearing rocks were only sufficient to stabilize baryte (Långban) and calcite with quartz, but not anglesite, cerussite, Ba carbonate, or rhodochrosite. Oxygen fugacities were too high for

sulphides to form. As a result, Pb formed silicate, arsenate, and antimonate minerals instead of galena and the Pb sulphosalts typical of hydrothermal ore deposits.

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