

THE TANCO PEGMATITE AT BERNIC LAKE, MANITOBA.

VIII. SECONDARY MINERALS FROM THE SPODUMENE-RICH ZONES

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ABSTRACT

Late hydrothermal activity produced a series of secondary minerals in the spodumene-rich zones of the Tanco pegmatite. Low-temperature alkaline solutions, operating locally without extensive migration, gave rise to cesian analcime, cesian beryl, cookeite, adularia, albite, lithiophosphate, apatite, montmorillonite-illite, calcite, and quartz. The occurrences of cesium-rich analcime and beryl are characteristically restricted to the close neighbourhood of the adjacent pollucite zone.

Cesian analcime shows strong zoning and sectorial birefringence, n_m † varying between 1.492 and 1.509. Its compositions fall into the gap in the Cs-poor range of the analcime-pollucite series. Outer parts of zoned crystals of beryl have $a = 9.213\text{Å}$, $c = 9.254\text{Å}$, $\varepsilon = 1.596$, $\omega = 1.604$, and contain among others 9.58 BeO, 1.61 Li₂O, 1.30 Na₂O, 0.80 K₂O, 0.18 Rb₂O, and 7.16 wt.% Cs₂O. Some crystals display the highest refractive indices ever known, $\varepsilon = 1.602$, $\omega = 1.610$. Rb- and Cs-poor non-perthitic adularia with ca. 1.5% Ab_{ss}* possesses high sanidine structure and optics, whereas albite (practically Ab₁₀₀) belongs to the low form. Physical properties and partial chemical analyses are given for most of the described minerals.

INTRODUCTION

The Tanco pegmatite, located on the northern shore of Bernic Lake in southeastern Manitoba, is a tantalum- and lithium-rich body containing the largest pollucite units known to date (Hutchinson 1959; Wright 1963; Crouse & Černý 1972). Besides its economical importance, it shows remarkably well many compositional and textural characteristics of lithium-rich pegmatites, a study of which contributes to the petrology of this pegmatite type.

However, late hydrothermal activity decomposing primary phases and producing new low-temperature assemblages was very restricted in those parts of the Tanco pegmatite that are presently accessible to sampling. The most widespread process seems to be the crystallization of apatite and calcite in fissures and vugs throughout the whole pegmatite body, and the ubiquitous alteration of amblygonite-montebbrasite to F-poor montebbrasite, accompanied frequently by fluorapatite and lithium micas (Černá *et al.* 1972a).

† Medium index of birefringent material.

* In solid solution.

The only other minerals that underwent considerable late alteration are petalite (Černý & Ferguson 1972) and the spodumene + quartz aggregates pseudomorphous after it. This paper deals with the alteration of the spodumene + quartz intergrowths which yielded some rare minerals associated in rather unusual assemblages: cesian analcime, cesian beryl, and lithiophosphate occur in various combinations with cookeite, adularia, albite, apatite, calcite, quartz, and clay minerals.

PARAGENESIS

Late alteration of the spodumene + quartz aggregates took place in both intermediate zones containing them in large amounts: in the lower intermediate zone (4) with the microcline-perthite + albite + quartz + spodumene (+ amblygonite-montebrazite) assemblage, and in the upper intermediate zone (5) consisting of spodumene + quartz + amblygonite-montebrazite (+ petalite) (see Crouse & Černý 1972, for location and composition of zones). Spodumene of the central intermediate zone (6), with the microcline-perthite + albite + quartz (+ beryl + wodginite) assemblage, underwent a similar alteration. However, in zones (4) and (6) the decomposition of spodumene yields only cookeite, quartz and calcite, and the study of this assemblage was not carried beyond checks of its phase composition. The present paper deals with the alteration products found within zone (5).

Typical assemblages of secondary minerals observed in this zone are listed in Fig. 1, with the crystallizational sequence indicated. Assemblages 1 to 4 are found in close neighbourhood of the pollucite zone (8) (see Figs. 2, 3, in Crouse & Černý 1972), and assemblage 5 occurs throughout the whole zone (5) without any particular relations to adjacent zones.

Besides the variations in quantitative relations of individual phases, some minerals may be frequently missing in these assemblages, and some minerals can also be found separately. Nevertheless, the assemblages shown in Fig. 1 are the most typical, and all mineral descriptions that follow below relate to minerals found in them.

MINERALOGY

Unless stated otherwise, the refractive indices were measured in immersion liquids using sodium light. X-ray powder diffraction data were collected on a Philips-Norelco diffractometer with CaF_2 annealed at 800°C ($a = 5.4620 \text{ \AA}$) used as internal standard. Unit cell dimensions were refined by aid of the self-indexing least-squares program by Evans

et al. (1963), modified by D. E. Appleman. Differential thermal analyses were performed on a DTA-13M automatic apparatus by R. L. Stone, Austin, Texas. Silica and alumina were determined by x-ray fluorescence, and other elements by atomic absorption spectrography; water was expelled by heating in dry oxygen and collected on magnesium perchlorate.

The minerals studied are marked with numbers corresponding to those of their parent assemblages as listed in Fig. 1. Additional numbers distinguish among samples separated from different specimens that belong to a single assemblage.

Cesian analcime

This mineral forms typical {211} trapezohedral crystals with subordinate to missing {100} facets, mostly waterclear, occasionally milky. The second generation of cesian analcime in assemblage 2 (sample 2-2) has a brownish-

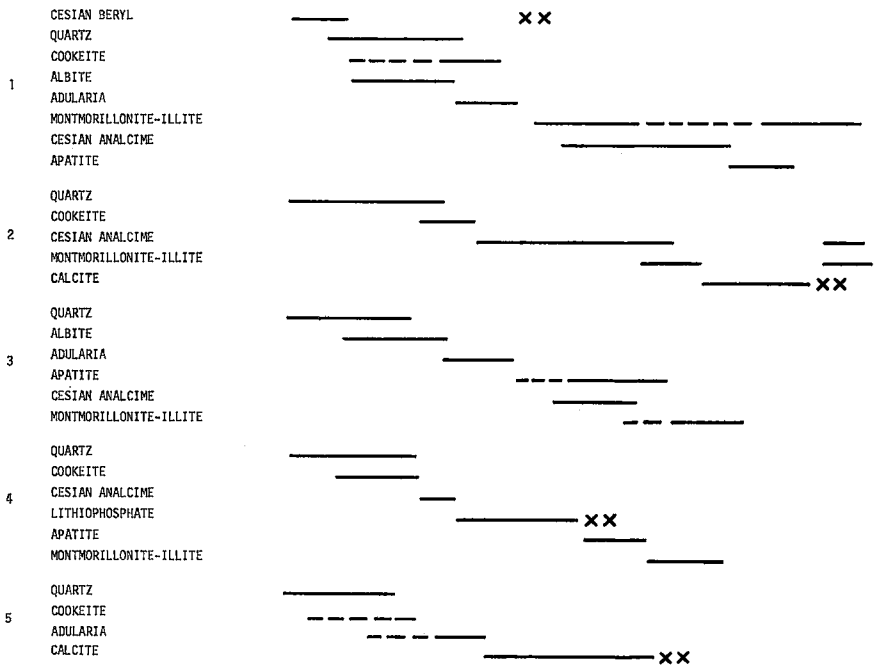
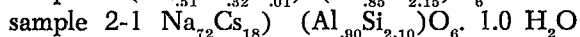
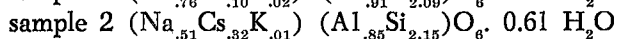
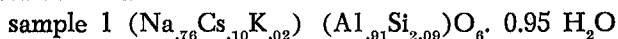


FIG. 1. Paragenetic sequences of secondary minerals in five hydrothermal assemblages from the upper intermediate zone (5) of the Tanco pegmatite; dashed line — crystallization of small amounts concurrent with precipitation of another major phase; xx — etching and corrosion.

yellow tint. Table 1 shows the chemical composition and physical properties of five samples.

The chemical composition of samples 1, 2, and 2-1 shows a good correlation with the general pollucite formulae proposed by Nel (1944) and Neuvonen & Vesasalo (1960), when recalculated to 6 oxygens per anhydrous formula:



Partial analyses of two small samples contaminated by quartz and an insoluble phase are also shown in Table 1. Sample 63568 has been studied independently by Drs. R. V. Gaines and Jun Ito. The alkali contents

TABLE 1. CHEMICAL COMPOSITION AND PHYSICAL PROPERTIES OF CESIAN ANALCIME

	1	2	2-1	2-2	3	63568
SiO ₂	54.85	52.10	52.93	n.d.	—	n.d.
Al ₂ O ₃	20.36	17.60	19.27	n.d.	—	n.d.
CaO	0.05	0.13	0.06	0.27	—	n.d.
Na ₂ O	10.30	6.46	9.29	4.91	—	7.5
K ₂ O	0.40	0.19	0.19	1.84	—	0.2
Rb ₂ O	0.01	0.06	n.d.	0.11	—	0.01
Cs ₂ O	6.00	18.00	10.50	14.90	—	7.9
H ₂ O ⁺	7.51	4.45	7.76	n.d.	—	n.d.
H ₂ O ⁻	0.41	0.14	—	n.d.	—	n.d.
	99.89	99.13	100.00	—	—	—
<i>n</i> m	1.492	1.500- —1.509	1.496- —1.497	1.505	1.505	1.493- —1.496
<i>a</i> (Å)	13.694 ±.001	13.693 ±.003	—	13.684 ±.001	13.685 ±.003	—
sp. gr.	2.35	2.63	—	2.60	—	—

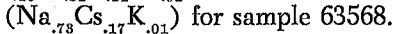
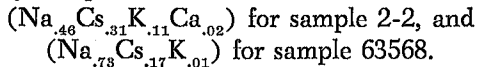
Analysts: sample 63568 — Dr. Jun Ito (Harvard University); other specimens — K. Ramlal and R. M. Hill (Dept. of Earth Sciences, University of Manitoba).

Sample 2-1 calculated from the mixture with eucryptite 3a quoted by Černý (1972a), assuming the Al_{0.9}Si_{2.1} tetrahedral content as found in samples 1 and 2.

Sample 2-2 slightly contaminated by quartz.

Sample 63568 slightly contaminated by quartz and by a phase insoluble in hot HNO₃ + HF.

adjusted to the $Al_{.9}Si_{2.1}$ tetrahedral composition found in the completely analyzed specimens are :



Sample 2-2 is close to the composition of sample 2 except for the partial substitution of K for Na, and sample 63568 is very similar to sample 2-1.

Refractive indices and densities are intermediate between those of analcime and pollucite, and show a positive correlation with Cs-content. All crystals show concentric zoning with respect to the refractive indices that decrease from the centres to the margins. The maximum difference observed is 0.003, with the exception of sample 2 (see below). All crystals are anisotropic, consisting of several radical sectors (octants?) with uniform extinction. The birefringence varies slightly among the concentric zones displaying different refractive indices.

Cesian analcime 2 shows only two zones, but the difference in their n values is the largest observed, 0.008 — 0.009. In individual anisotropic sectors, the outer zone is optically homogeneous but the core consists of fine lamellae showing two or three optical orientations and different refractive indices (Fig. 2). In the sequences of parallel lamellae, the indices



FIG. 2. A fragment of cesian analcime 2 in oil immersion ($n = 1.512$), plane polarized light. The homogeneous zone at left has $n_m = 1.500-1.501$, the "cross-hatched" part at right has an average $n_m = 1.509$. Note the perthite-like character of the right part, corresponding to the core of the crystal.

alternate rapidly suggesting a "perthitic" breakdown of an intermediate cesian analcime into components approaching the sodic and cesian end-members.

Unit cell dimensions do not show any simple changes that could be correlated with chemistry; the cell edges of analcime and pollucite are almost identical and vary, as proved for analcime by Saha (1961), mainly with the Si/Al ratio. In accordance with the observed birefringence, the 422, 521, 640, and 800 peaks are slightly broadened but never actually split, suggesting a monoclinic distortion of the cubic structure comparable to that found in wairakite (e.g., Seki 1966, and Liou 1970). The intensities of x-ray powder reflections show regular changes with changing chemical composition: the intensities of 321, 440, 532, 631, and 721 increase, and those of 211, 220, 422, 431, 521, 640, and 800 decrease with increasing Cs-content.

Only pure analcime, and pollucites with up to 45 mol. % analcime have been known to date in the analcime-pollucite series. The cesian analcimes reported here fall into the analcime-rich gap, and deserve further study to check their homogeneity and structure.

Cesian beryl

This mineral has been found only rarely, in assemblage 1. It forms either larger crystals (up to 4 x 12 mm) with colourless core overgrown by a pale pink zone, or small colourless prisms (up to 1.5 x 6 mm) that seem to have crystallized later than the larger crystals. Both types of beryl are deeply corroded, and the etching grooves and crosscutting fractures are usually filled with clay minerals.

The pink outer zone of the larger crystals shows remarkably higher refractive indices than the colourless core (Table 2). The clear prisms of the second generation have even higher indices, equal to the highest values ever found in beryl (Schaller *et al.* 1962), and higher than any recorded to date for beryl with negligible Fe, Mg, and Mn (Beus 1960; Feklitchev 1964; Evans & Mrose 1966).

TABLE 2. PHYSICAL PROPERTIES OF CESIAN BERYL

			ϵ	ω	$\epsilon - \omega$	$a(\text{\AA})$	$c(\text{\AA})$
1-1.	core	colourless	1.589	1.596	0.007	9.2176 \pm .0009	9.2469 \pm .0020
	rim	pink	1.596	1.604	0.008	9.2132 \pm .0007	9.2541 \pm .0013
1-2.		colourless	1.602	1.610	0.008	—	—

Unfortunately, partial chemical analysis could be performed only on the pink outer zone of the larger crystals. This material was found to contain 9.58 BeO, 0.32 MgO, 0.01 CaO, 0.12 FeO, 0.02 MnO, 1.61 Li₂O, 1.30 Na₂O, 0.80 K₂O, 0.18 Rb₂O, and 7.16 wt. % Cs₂O. These contents agree well with the unit cell dimensions of the pink zone (Table 3), when compared with the data for the Marropimo morganite (Vorma *et al.* 1965), the Antsirabe cesium beryl (Evans & Mrose 1966) and for other Cs-rich beryls quoted by Bakakin *et al.* (1969, 1970). The latter authors confirmed in a quantitative way the increase of *c* with alkali enrichment, and the increase of *a* with Fe, Mg substitutions for Al in beryl.

The non-analyzed clear cores of the larger beryl crystals have lower *c* and refractive indices, corresponding roughly to the beryls containing *ca.* 4.5 Cs₂O, 1.3 Li₂O, 2 % Na₂O, *etc.* (see Sosedko 1957; Bakakin *et al.* 1970; and others). The high indices of the colourless crystals of the second generation are probably not caused by Fe, Mg and/or Mn and the composition of this beryl could be very close to that described by Evans & Mrose (1966).

Cookeite

This mineral is a rather widespread constituent of the secondary assemblages. Unfortunately, it occurs disseminated as isolated fine flakes in other minerals and cannot be separated in amounts sufficient for chemical analysis. Thus, only refractive indices and unit cell dimensions are given for four samples that are considered characteristic for their respective assemblages (Table 3). These properties show considerable differences in some cases, but they may be caused by so many compositional changes that it is impossible to derive reasonable estimates of the composition (Černý 1970).

Albite

Albite is rather rare, and is found only where the matrix surrounding the secondary assemblage contains cleavelandite. Thus, it seems to be

TABLE 3. PHYSICAL PROPERTIES OF COOKEITE

colour	α	γ	$\gamma - \alpha$	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)	β
1. pink	1.573	1.595	0.022	5.14 ₁	8.89 ₄	14.16 ₂	96.8°
1-1. white	1.575	1.598	0.023	5.15 ₀	8.88 ₂	14.17 ₅	96.8°
2. white	1.576	1.600	0.024	5.14 ₄	8.89 ₈	14.16 ₈	96.9°
5. greenish white	1.574	1.596	0.022	5.15 ₁	8.90 ₈	14.17 ₈	97.1°

formed only by recrystallization, more or less *in situ*. Albite forms platy crystals flattened on {010}, with {001} and {10 $\bar{1}$ } predominant over prismatic faces. Simple albite twins are most common, occasionally lamellar twinning // to {010} is also observed, and the extinction angles of these polysynthetic twins on {001} suggest the possible presence of other twinning laws. The crystals are usually etched and exhibit a silky dull luster. Oriented overgrowths of adularia on {001} and {10 $\bar{1}$ } are common.

The unit cell dimensions and optical properties given in Table 4 indicate a pure low albite, when compared with the data published by Wright & Stewart (1968), Bambauer *et al.* (1967), Morse (1969), and Martin (1969b). The physical parameters suggest an absence of any appreciable Ca and K, and the structural state as indicated by the unit cell dimensions and Δ corresponds to the lowest known.

Adularia

Potassium feldspar forms mostly cauliflower-like, radial aggregates of hatchet-shaped crystals typical of the adularia habit. It occurs also as oriented overgrowths on either early cleavelandite protruding from the walls of fissures, or on the late albite described above.

The unit cell dimensions of this feldspar given in Table 4 coincide with those of high sanidine (Orville 1967, Table 3A, and Wright & Stewart 1968, Fig. 2b and Table 2). The refractive indices in the {010} plane compare well with the true α and γ of a pure potassium feldspar; they indicate high sanidine optics with optic axial plane // to {010}.

A partial chemical analysis of material contaminated by cleavelandite from the fissure walls and by quartz gave 11.70 K₂O, 2.20 Na₂O, 0.12 Rb₂O, and 0.18 wt. % Cs₂O. X-ray estimates of solid solution by the method of Orville (1967) indicate 1.5 % Ab_{ss} in adularia and 0 – 0.5 % Or_{ss} in cleavelandite. Since albite does not incorporate more than a few tens of ppm of Cs and Rb (*e.g.* Heier 1962), no Rb and Cs were subtracted when calculating the corrections for the admixed phases. The alkali content of the adularia thus appears to be 16.50 K₂O, 0.18 Na₂O, 0.14 Rb₂O, and 0.26 wt. % Cs₂O, corresponding to 97.3 Or, 1.5 Ab, 0.5 Rb-f, and 0.7 wt. % Cs-f.

Lithiophosphate

This species has been known to date only from the Kola Peninsula (Matias & Bondareva 1957; Matias 1961) and from the Kings Mountain spodumene pegmatites (White 1969). At both localities, lithiophosphate originated in late stages of the parent pegmatite. It replaces montebraşite

in Kola, and occurs in fissure with albite, quartz, and rhodochrosite at Kings Mountain.

In the Tanco pegmatite, lithiophosphate forms monocrystalline fillings of cavities lined with quartz, cookeite, and cesian analcime. Individuals of lithiophosphate reach up to 5 cm in these fillings. The pale-beige mineral is always slightly etched, showing silky luster and very fine vertical striation parallel to the cleavages.

The refractive indices compare well with those found in earlier studies (Table 5). Optical orientation, related to cleavages as re-oriented by White (1969; pers. comm., 1970), is $X//a$ (normal to the best $\{100\}$ cleavage), $Y//b$, $Z//c$ ($//$ to both $\{100\}$ and $\{110\}$ cleavages). Unit cell dimensions are close to those given by Swanson *et al.* (1965) and to those of the Kings Mountain material (Table 5).

Apatite

Apatite shows a wide variety of aggregations, colours, and refractive indices. This variability is characteristic not only of material from different assemblages but also of different specimens within a single assemblage. The very small size of both crystals and aggregates and the lack of larger accumulations exclude any chance of getting sufficient chemical data. The physical properties of the most frequent types are given in Table 6.

TABLE 4. PHYSICAL PROPERTIES OF THE FELDSPARS

	adularia 1	albite 3
$a(\text{\AA})$	$8.5967 \pm .0025$	$8.1383 \pm .0025$
$b(\text{\AA})$	$13.0360 \pm .0031$	$12.7858 \pm .0044$
$c(\text{\AA})$	$7.1705 \pm .0024$	$7.1579 \pm .0010$
α	90°	$94^\circ 14.3' \pm 1.2'$
β	$115^\circ 59.3' \pm 1.1'$	$116^\circ 37.3' \pm 0.8'$
γ	90°	$87^\circ 44.6' \pm 1.2'$
$V(\text{\AA}^3)$	$722.36 \pm .24$	$664.01 \pm .18$
α' on (010)	1.517	1.528
γ' on (010)	1.523	1.538
$\alpha\Lambda\alpha$ on (010)	3°	20°
$\alpha\Lambda\alpha$ on (001)	0°	4.5°
$\Delta 2\theta$ ($\bar{2}01$ fsp — 113 KBrO_3)	0.75_8°	1.84_2°
$\Delta 2\theta$ (131 — $\bar{1}\bar{3}1$)	0.00°	1.08°

Globular radial aggregates in assemblage 1 belong largely to a podolite- or francolite-type carbonate apatite, because of the low refractive indices. A thin outermost layer, however, must have a quite different composition. Brown apatite 3-1 shows a remarkably high birefringence, close to that of dehrnite and/or lewistonite (alkali-rich apatites described by Larsen & Shannon 1930a, 1930b), but a partial chemical analysis gave only 0.44 Na₂O and 0.07 K₂O. The other apatites listed in Table 6 seem to belong to mixed (F,OH) apatites.

Montmorillonite-illite

The two clay minerals compose dense aggregates with fibrous structure, white in assemblages 2 and 3, and pale apple-green in assemblages 1 and 4. They replace quartz and to a lesser extent spodumene, and form globular aggregates with a radial inner structure in the vugs.

TABLE 5. PHYSICAL PROPERTIES OF LITHIOPHOSPHATE

	<i>a</i> (Å)	<i>b</i> (Å)	<i>c</i> (Å)		
Synthetic, Swanson <i>et al.</i> (1965)	5.2340	6.1155	4.8452		
Tanco pegmatite, 4	5.2301 ± .0007	6.1130 ± .0009	4.8402 ± .0010		
Kings Mountain, this study	5.2296 ± .0007	6.1123 ± .0011	4.8415 ± .0006		
	α	β	γ	$\gamma - \alpha$	+2 <i>V</i>
Synthetic, Swanson <i>et al.</i> (1965)	1.550	—	1.538(?)		large
Synthetic, Tien & Hummel (1961)	1.550	1.555	1.562	0.012	~ 80°
Kola, Matias & Bondareva (1957)	1.550	1.557	1.567	0.017	69°
Kola, Matias (1961)	1.550	1.557	1.567	0.017	88°
Kings Mountain, White (1969)	1.553	1.558	1.566	0.013	~ 80°
Tanco pegmatite, 4	1.550	1.556-7	1.564	0.014	large

TABLE 6. PHYSICAL PROPERTIES OF APATITE

	ϵ	ω	$\epsilon - \omega$	<i>a</i> (Å)	<i>c</i> (Å)
1. inner	1.624	1.629	0.005	9.430 ± .001	6.876 ± .001
outer	1.639-40	1.645	0.005-6	—	—
3-1. brown	1.631	1.641	0.010	9.396 ± .001	6.900 ± .001
3-2. pink core	1.638	1.644	0.006	—	—
surface	1.641	1.647	0.006	—	—
4. beige	1.638	1.643	0.005	—	—

As indicated by the relative intensities of characteristic peaks on x-ray powder diffractometer records, and by the intensities of reactions in the DTA patterns, the proportions of the two minerals vary in different assemblages: illite prevails in assemblages 1 and 4, montmorillonite predominates in assemblage 3, and both phases are present in about equal amounts in assemblage 2. Besides these two minerals, the clay in assemblage 1 seems to contain also small admixture of dickite. This material was analyzed for alkalis, and was found to contain 0.50 Na₂O, 2.61 K₂O, 0.67 Li₂O, 0.16 Rb₂O, 0.39 Cs₂O, and 0.09 CaO.

Calcite

Rhodochrosite is found frequently in the Tanco pegmatite, but calcite is the only carbonate associated with the secondary silicates and phosphates described here. In assemblage 2 it was only rarely followed by a second generation of cesian analcime and montmorillonite-illite. In most cases it fills completely the remaining space among crystals of the earlier minerals, or it leaves only tiny vugs. Refractive indices of the colourless calcite from the assemblage 2 correspond to pure calcite, whereas those of calcite from assemblage 5 are slightly higher. The latter carbonate is frequently pinkish and develops a dark brown stain upon weathering. This suggests a low content of manganese.

Quartz

Greyish, milky or waterclear, quartz is one of the earliest minerals in all assemblages. It frequently forms well-shaped columnar crystals bearing only striated prismatic faces and the rhombohedra in terminations; no trapezohedra typical of low quartz were observed. The crystals are occasionally aligned in subparallel coatings that cover sizeable areas on fissure walls. A slight etching of the crystals is quite common.

DISCUSSION

The described group of secondary minerals is a typical example of a low-temperature association produced in late stages of pegmatite evolution, under conditions of "alpine vein" crystallization as characterized by Ginzburg (1955, 1960). There is plentiful evidence that these minerals were formed at low temperature (and possibly low pressure) in an alkaline hydrothermal environment, at the expense of the surrounding early minerals, and that only local migration of chemical constituents was involved in this process.

A low-temperature hydrothermal environment is indicated by the presence of some characteristic minerals like the lithium chlorite cookeite, lithiophosphate, and clay minerals. The remarkable scarcity of albite in the examined assemblage, and the quite widespread occurrence of cesian analcime, also suggest very low temperatures below the albite stability range (Senderov & Chitarov 1966 — 190°C; Martin 1969b — 330-220°C decreasing with increasing alkalinity). Albite seems to have crystallized only when seeded by cleavelandite in walls of the fissures. The presence of considerable Cs and Li in the examined natural assemblage may have shifted the equilibria from the p-T conditions found for simple sodic systems, nevertheless, the general relationships and the order of temperature are probably the same.

The varieties of apatite, beryl, and K-feldspar also suggest late hydrothermal crystallization. Carbonate-apatite is known from low-temperature environment generally, and from late alterations in pegmatites in particular (e.g. Landes 1925; Fraser 1930). Cs-rich beryl crystallizes usually in pegmatite cavities with other low-temperature phases (e.g. Hurlbut & Wenden 1951). The adularia variety of K-feldspar is characteristic of low-temperature hydrothermal environment, and its structural state of high sanidine suggests temperatures insufficient for any ordering detectable by x-ray diffraction. This is the more significant because alkaline environment is known to catalyze ordering in feldspars (Martin 1969a,b), and the mineral assemblage under examination strongly suggests an alkaline character of the parent solutions.

Chlorites, montmorillonite, illite, calcite, analcime, and K-feldspars all require alkaline environment for their hydrothermal crystallization (e.g. Stringham 1952; Hemley & Jones 1964; Senderov & Chitarov 1966; Höller 1970). Albite, apatite (particularly carbonate-apatite, as shown by Simpson 1967), and quartz may originate from alkaline solutions. Alkali-rich varieties of beryl seem to crystallize characteristically in alkaline environment, contrary to the common alkali-low types formed in earlier stages of pegmatite formation.

The wide variability in mineral composition of the secondary assemblages, and in chemical composition of some single species in different assemblages as well as within a single assemblage, indicate a local character of the alteration. Residual solutions, dissolving earlier phases and depositing new minerals more or less *in situ*, are most probably responsible for the origin of the described minerals. Solutions pervading large volumes of the pegmatite would probably produce more uniform associations, and would tend to equalize the composition of such chemically variable phases as cookeite, apatite, and cesian analcime. The large-scale distribution of Cs

is also characteristic in this respect : cesian analcime and beryl are absent in the major part of the spodumene-rich zone (5) but relatively abundant in the spodumene + quartz aggregates closely adjacent to the pollucite zone (8).

Despite the great variability, all secondary assemblages follow the same general sequence of crystallization (Fig. 1). Feldspars, beryl, quartz and cookeite are the earliest minerals, followed by cesian analcime and montmorillonite-illite; phosphates and carbonates are the latest phases, frequently contemporaneous with, or overgrown by, clay minerals. This sequence suggests a temperature decrease during the crystallization of these assemblages. The Cs-content typically increases during crystallization of the early beryl, but invariably decreases during precipitation of late analcime. Ca enters apatite and calcite in late stages, whereas most of the alkalis precipitate in the early and intermediate-stage minerals.

In conclusion it is interesting to note that some of the secondary assemblages described here resemble that reported by Neuvonen & Vesasalo (1960) from the petalite- and pollucite-rich Luolamäki pegmatite in Finland. Despite the similarity in mineral composition, the chemistry of individual phases seems to be remarkably different. Adularia-like potassium feldspar is a Rb-rich highly triclinic microcline in the Finnish pegmatite, and the refractive indices of analcime indicate a very pure Na-phase without appreciable Cs.