

**COMPOSITIONAL HETEROGENEITY  
AND ALTERATION OF POLLUCITE**

by

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B.Sc. (Queen's University)

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## ABSTRACT

The occurrence of pollucite is restricted to rare-element granitic pegmatites which have attained the highest degree of alkali fractionation. Pollucite also is a natural analogue for the phases involved in containment of  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  nuclear wastes. Poor covariance of compositional and physical parameters (particularly refractive index) prompted a re-examination of the analcime-pollucite series by the electron microprobe technique. This approach was suitable to investigate both the compositional heterogeneity, and the fine-grained minerals derived by the alteration of pollucite.

Pollucite occurs late in primary consolidation of rare-element granitic pegmatites, and crystallizes at near-solidus temperatures. This early pollucite is compositionally homogeneous and cubic and disordered with Si/Al ratios as high as 2.6. The CRK [=  $100(\text{Cs} + \text{Rb} + \text{K})/\text{cation sum}$ ] of this pollucite centers on 80 and ranges from 70 to 90. The primary, homogeneous pollucite has locally evolved to a blebby mosaic of alternating Na-enriched and Cs-enriched domains, crosscut by Cs-rich veinlets ( $\pm$  quartz) rimmed with Na-rich pollucite. These textural features result from re-equilibration along a solvus, broadening with decreasing temperature, and controlled by three principal mechanisms: (i) exsolution

into Na, Si-enriched and Cs, Al-enriched compositions, (ii) liberation of  $\text{SiO}_2$  leading to alkali cation- and Al-enriched pollucite with Si/Al ratios approaching 2.0, and (iii) cation exchange of  $\text{Na}^+$  and  $\text{Cs}^+$  in an unchanged (Al,Si) framework. The first two mechanisms dominate at high temperature (400 to 200°C) in an apparent mass balance. In contrast, at low temperature (less than 200°C) the third mechanism dominates and produces Na-enriched pollucite, to analcime with apparent loss of  $\text{Cs}^+$ .

Three stages of alteration are recognized: (i) coarse polygonal veining (2 to 10 mm wide) of variable mineralogy (albite, microcline, quartz, amblygonite, lepidolite) which is fracture-filling in pollucite bodies larger than 1m; (ii) thin braided to sub-parallel veins (0.1 to 1.0 mm wide) of fine-grained mica and spodumene. These follow grain boundaries, fractures, and (100) cleavage traces of pollucite, are a crystallization product of late fluids (possibly from the pollucite), and are common to all pollucite segregations larger than 5 cm; and (iii) late stage replacement assemblages. Spherical aggregates (1mm size) of adularia (+ Cs-rich pollucite ± mica to Rb-dominant mica ± albite ± an AlSi-bearing mineral) to Rb-dominant adularia locally replace pollucite. Later cation exchange results in analcime formation. Latest replacement is by clay minerals, some Cs-bearing, consisting mainly of montmorillonite-type smectites, illite, and kaolinite.

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## CHAPTER 1

## INTRODUCTION

Minerals within the analcime-pollucite series ( $\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O} - \text{CsAlSi}_2\text{O}_6$ ) are restricted in occurrence to the most fractionated granitic pegmatites of the rare-element class. Primary pollucite crystallizes at near-solidus temperatures in the central parts of these pegmatites (Cerny 1982). In contrast, cesian analcime occurs in low-temperature "alpine-vein" assemblages (terminology of Ginsburg 1960) of leaching cavities and fissures in pegmatites bearing the primary pollucite (Cerny 1972).

The occurrence of pollucite is central to models of rare-element granitic pegmatite petrogenesis and internal evolution. With extreme values of electronegativity and cation radius, and high volatility, cesium is concentrated by fractionation processes in granitic magmas into a residual melt. Pollucite is not stabilized in a pegmatitic melt until the final stages of consolidation. Pollucite, therefore, tends to accumulate interstitially to the commonly associated minerals quartz, spodumene, petalite, and feldspars (Dymkov 1953). Larger amounts will segregate into pods and lenses, and into larger, essentially monomineralic bodies near the central parts of pegmatites.

Analcime-pollucite minerals are of importance to

nuclear waste containment; they form on hydrated waste-glass surfaces, in low-temperature hydrothermal reactions of several waste forms with basalt and shale, and in ceramics produced by calcination of waste sludges. Pollucite has also appeared as a phase in titanite-based nuclear waste ceramics (SYNROC formulations) (Buykx et al. 1990). Pollucite is a natural analogue of phases involved in the containment of  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  nuclear wastes. The chemistry, stability, and alteration of pollucite and cesian analcime are thus of considerable interest.

The relative ease of alteration of pollucite preserves a record of late stage, low temperature hydrothermal processes within complex granitic pegmatites. The alteration products are often too fine-grained for optical identification, and alteration veins too thin for X-ray identification. Nevertheless, they are significant to nuclear waste management. They have not yet been investigated on the scale of the electron microprobe; this constituted an important part of the present study.

The main purpose of the present study is to examine in detail by electron microprobe, the extent and character of the compositional heterogeneity in pollucite.



## CHAPTER 2

## PREVIOUS WORK

## 2.1 Bulk chemistry and physical properties

Pollucite was first discovered on the island of Elba in 1846 by Breithaupt, and since then nearly 80 localities have been reported. Pollucite from Elba was the first to be analysed (Wells 1891 reviews the early determinations of the composition), and to date 71 bulk, wet chemical analyses of pollucite from at least 48 different localities have been published (Table 2.1a to 1, Appendix 1). The specific locality is not known for some of the samples.

In Figure 2.1-1a, a histogram of bulk compositions of pollucite is given, based on these 71 analyses. The composition is expressed as  $CRK = 100(Cs + Rb + K)/\text{cation sum}$  (Cerny 1974). Compositions of secondary cesian analcime are not included in the histogram. For most pollucite, the CRK ranges from 70 to 90, and the distribution centers near CRK 80. A few localities are rich in  $Na^+$ .

$Si/Al$  ranges from 2.0 to 2.6, and is scattered with respect to CRK (Fig. 2.1-1b). This scatter is reflected in a plot of  $Al (+ Fe)$  atoms p.f.u. (per formula unit) against CRK (Fig. 2.1-2). With individual  $Al (+ Fe)$  values, the sum of cation charges is also plotted. The charge imbalance (usually an alkali cation deficiency

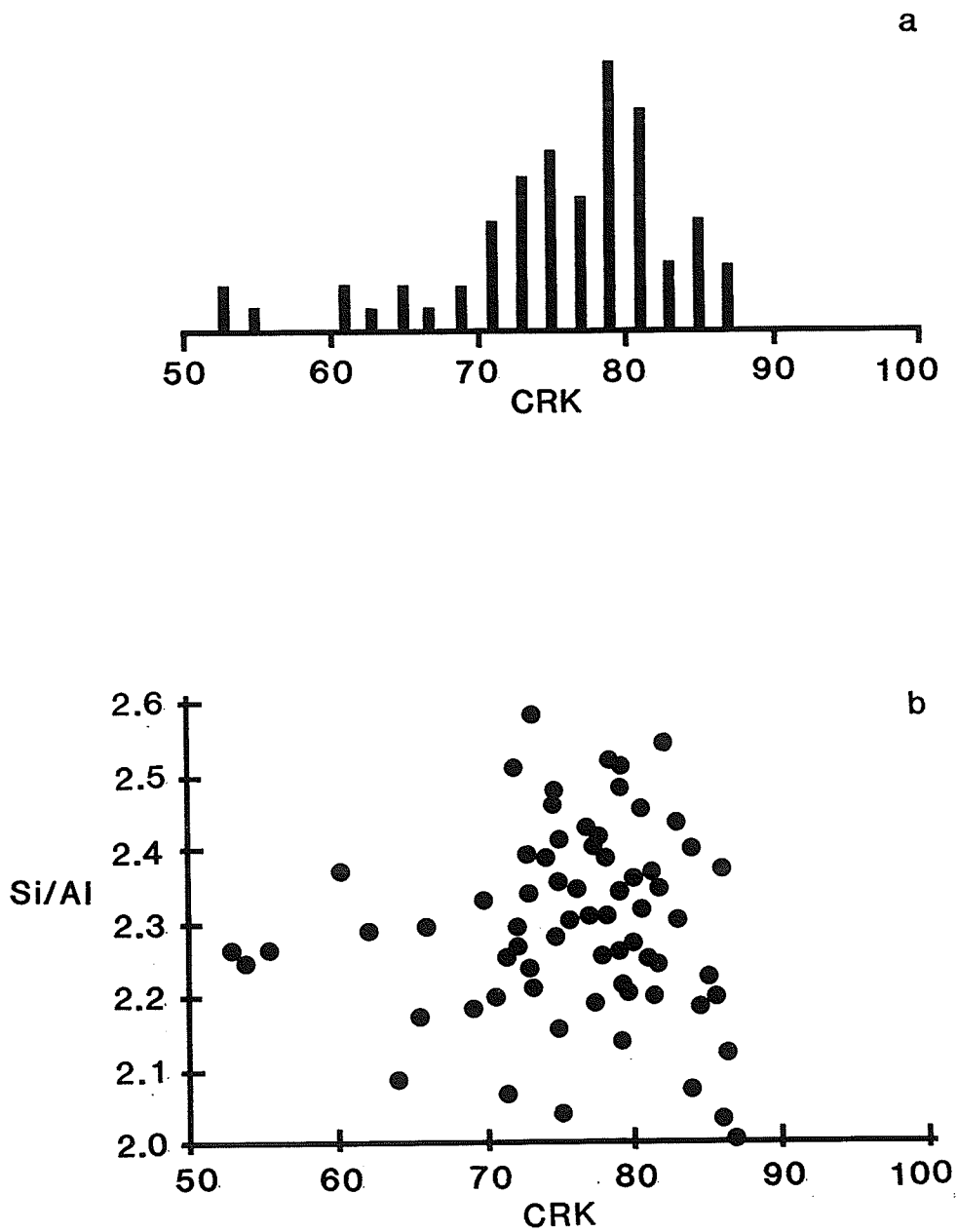


Fig. 2.1-1. a) Histogram of bulk compositions of pollucite ( $n = 71$ ), and b) Si/Al ratio vs. CRK ( $= 100(\text{Cs} + \text{Rb} + \text{K})/\text{cation sum}$ ), from the literature.

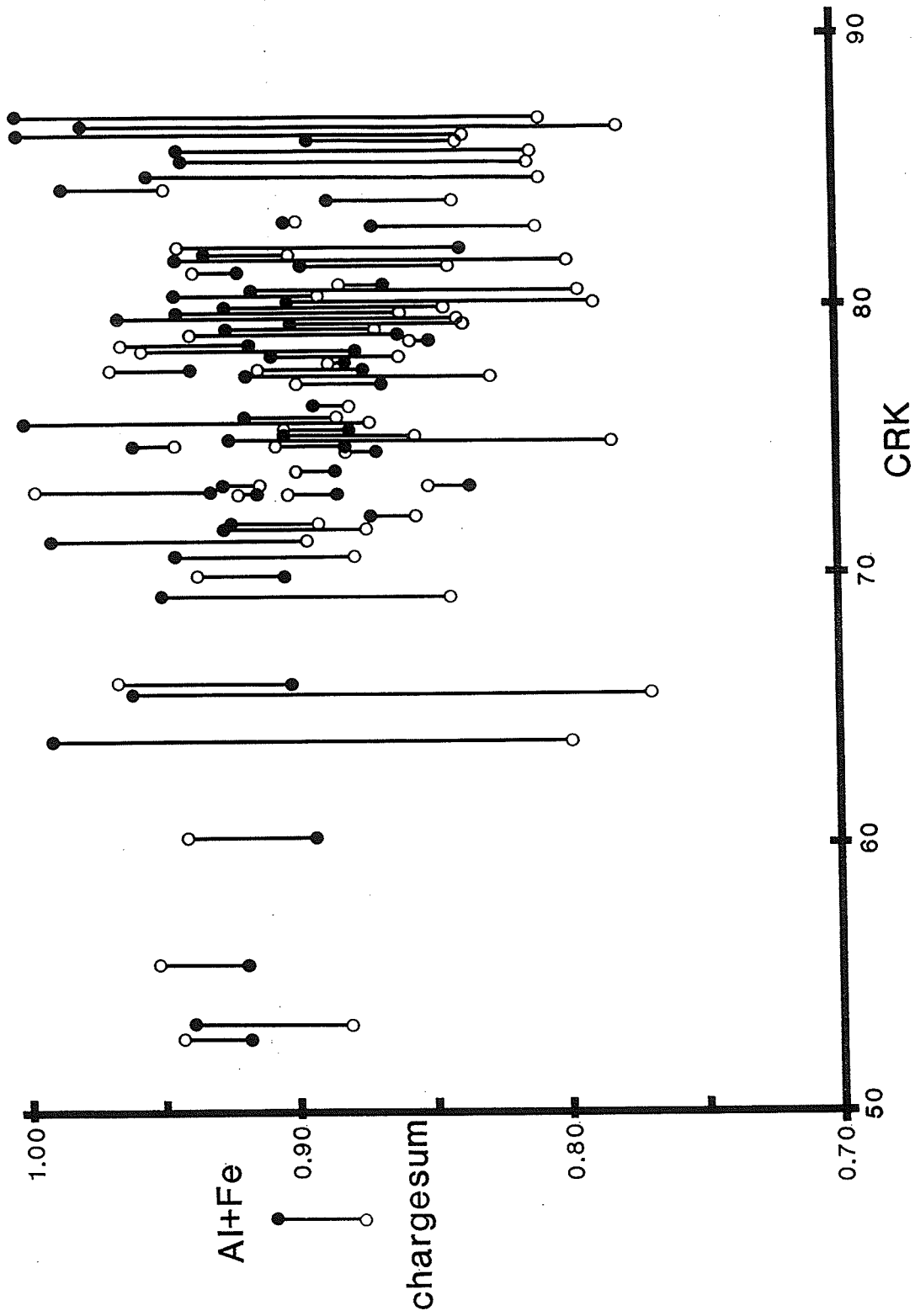


Fig. 2.1-2. Al (+ Fe), and sum of cation charges vs. CRK, from the literature.

relative to Al) appears to be largest for pollucite with CRK > 80; however, most of these analytical results were obtained in a single study (Khalili & von Knorring 1976) and may contain some systematic error.

The extent of substitution of the minor cations  $\text{Li}^+$ ,  $\text{K}^+$ ,  $\text{Rb}^+$ , and  $\text{Ca}^{2+}$  is shown in Figures 2.1-3a to d, as a function of CRK. Other cation substitutions are small (maxima reported, as wt.% : 0.63 MgO, 0.10 FeO, 0.06 MnO, 0.08 SrO, 0.07 BaO, 0.011  $\text{Tl}_2\text{O}$ , 0.32  $\text{Pb}_2\text{O}$ , 0.22 CeO).

$\text{P}^{5+}$  substitution for  $\text{Si}^{4+}$  can be substantial- up to 0.41 wt.%  $\text{P}_2\text{O}_5$ ; however, other framework substitutions are small (maxima reported, as wt.% : 0.20  $\text{Fe}_2\text{O}_3$ , 0.04  $\text{TiO}_2$ , 0.0012  $\text{Ga}_2\text{O}_3$ ). In determinations of minor constituents by Ahrens (1945, 1947) and Carobbi & Minguzzi (1948) elements below lower limits of detection are Cu, Ni, Hg, Ge, Sn, Sc, In, Ag, Y, La, Be, and W. Pollucite may also contain He (0.053  $\text{mm}^3/\text{g}$ , Cherdyntsev & Kozak 1949) and Ar (Ginsberg et al. 1975).

The plots of density, and index of refraction against CRK (Fig. 2.1-4a and b) of pollucite show considerable scatter. The lines shown are calculated using the Gladstone-Dale relationship for compositions between ideal  $\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$  and  $\text{CsAlSi}_2\text{O}_6$ , with  $\text{Si}/\text{Al} = 2.0$ , and  $\text{Si}/\text{Al} = 2.2$ , 2.4, and 2.6 assuming  $\text{Cs} + \text{Na} = \text{Al}$ , and  $\text{Na} = \text{H}_2\text{O}$ . A cell edge of 13.68 Å was used (Martin & Lagache 1975). A specific refractive energy of  $K = 0.298$  for channel  $\text{H}_2\text{O}$  in

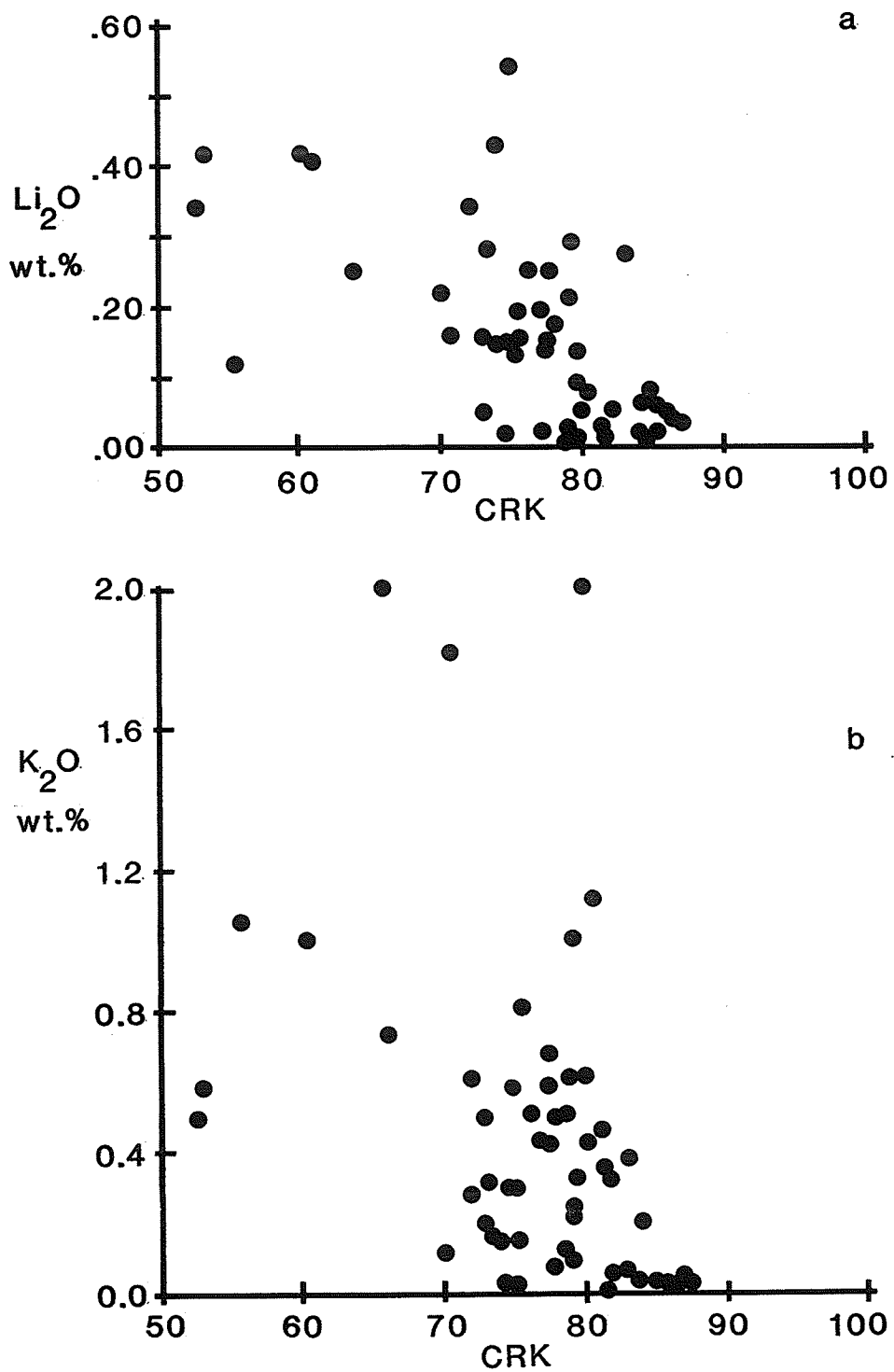


Fig. 2.1-3. a)  $\text{Li}_2\text{O}$  wt.% vs. CRK, b)  $\text{K}_2\text{O}$  wt.% vs. CRK, from bulk compositions reported in the literature.

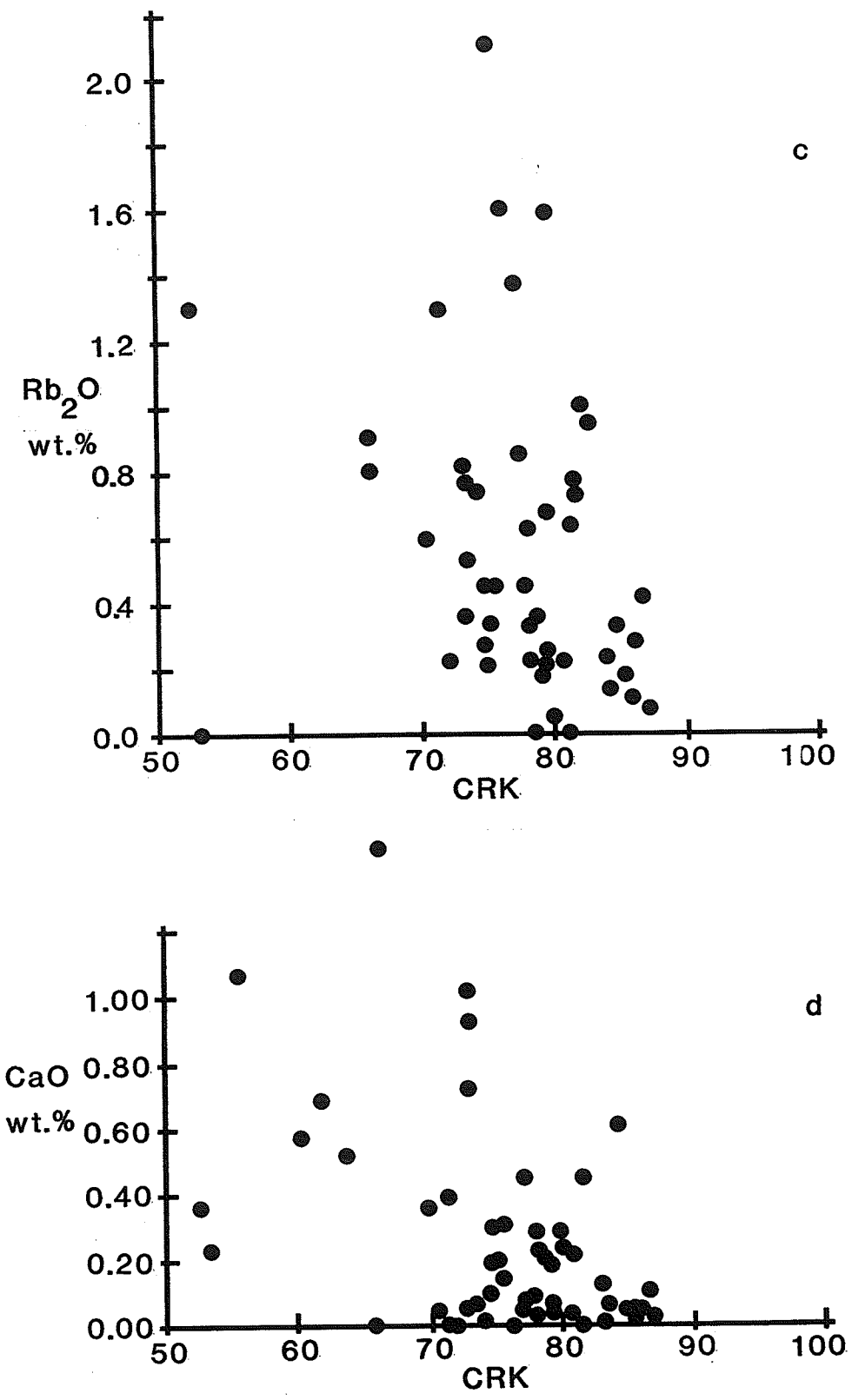


Fig. 2.1-3. c) Rb<sub>2</sub>O wt.% vs. CRK, and d) CaO wt.% vs. CRK, from bulk compositions reported in the literature.

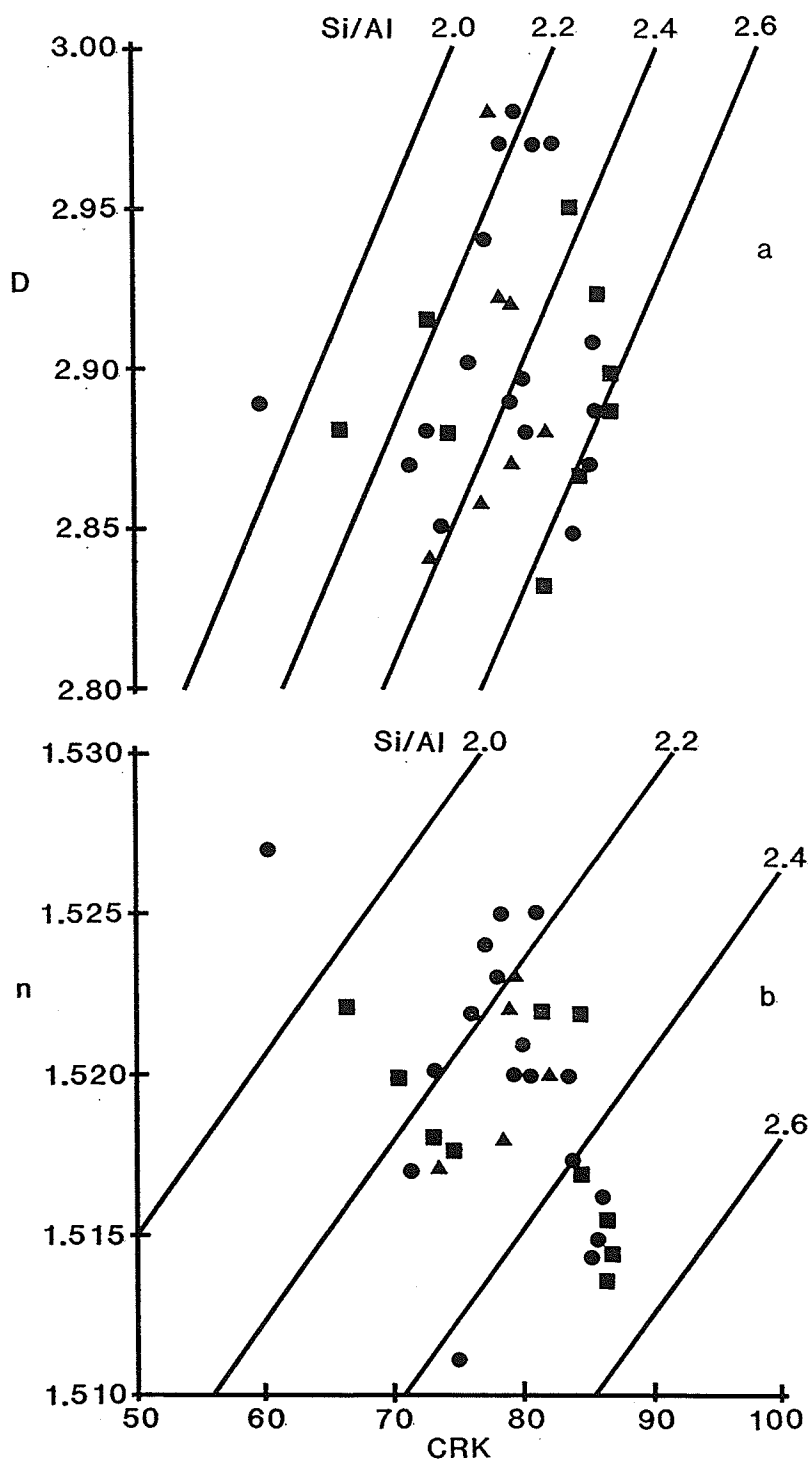


Fig. 2.1-4. a) Density (D) vs. CRK, and b) Index of refraction (n) vs. CRK, from the literature. D and n vary with Si/Al along the lines shown. Data are sorted into the ranges Si/Al less than 2.2 (squares), Si/Al 2.21 to 2.39 (circles), and Si/Al greater than 2.4 (triangles).

cordierite (Bloss & Armbruster 1982), and  $K = 0.226$  for tetrahedrally co-ordinated Al (Eggleton 1991) was used along with  $K_n$  values given in Mandarino (1976).

Pollucite tends to be disordered, with random occupancy of Al in the tetrahedral sites of the cubic (Ia3d) tectosilicate structure proposed by Beger (1969). If the framework is fully ordered, the symmetry must be less than cubic, and a Si/Al ratio of 2.0 is required.

## 2.2 Compositional heterogeneity

The first authors to note discontinuities in the analcime-pollucite series were Neuvonen & Vesasalo (1960), in a study of the Luolamäki pegmatite in Finland. They observed the co-existence of birefringent analcime and cubic pollucite, and reasoned that a clearly defined miscibility gap exists between these two minerals. In addition, the index of refraction they measured on two pollucite grains differed (1.5142, and  $1.5223 \pm 0.0005$ ). Simpson (1974) observed index of refraction to vary between 1.5195 and 1.5202 within a single hand specimen of pollucite from Tanco, Manitoba. Cerny & Simpson (1978) observed index of refraction to vary between 1.5172 and 1.5238 in Tanco pollucite and found a very poor correlation with Cs content.

In optical studies of synthetic analcime, Barrer et



al. (1953) found immiscibility between the pairs  $\text{Na}^+$  and  $\text{K}^+$ ,  $\text{Na}^+$  and  $\text{Rb}^+$ , and  $\text{Na}^+$  and  $\text{Tl}^+$ , but curiously not between  $\text{Na}^+$  and  $\text{Cs}^+$ . It is possible that the suggested unmixing of a domain structure was on a very fine scale for  $\text{Na}^+$  and  $\text{Cs}^+$ .

Cerny (1974) reviewed the chemistry and physical properties of minerals in the analcime-pollucite series and contributed new analysis of cesian analcime. Minerals in the series conform to the broad requirements of Beger's (1969) structural model; however, poor covariations of bulk chemical composition and physical parameters, particularly refractive index, were noted. Similar results are given in reviews of pollucite by Feklichev (1975) and Fel'dman & Pleskova (1978). Repeated analyses, and analyses of different samples of pollucite from the same locality (e.g. Tin Mt. in Tables 2.1a to 1) show differences in composition beyond experimental error. Dr. Scott Ercit (personal communication 1988) examined pollucite by electron microprobe, and revealed a relatively wide range of compositions within the scale of a few millimeters.

### 2.3 The alteration of pollucite

A diagnostic feature of pollucite distinguishing it in hand specimen from quartz is the widespread presence of narrow, subparallel grey or purple veinlets characteristic

of many localities. The alteration of pollucite has been noted by many authors (Landes 1925, Nel 1944, Ginsburg 1946, Neuvonen & Vesasalo 1960, Melentyev 1961, Nickel 1961, Vlasov 1966, Aramakov et al. 1975, Khalili & von Knorring 1976, Smeds & Cerny 1989), and examined as to the sequence of alteration by others (Quensel 1938, Quensel 1945, Shaub & Schenck 1954, Cerny & Simpson 1978). On the basis of these investigations, Cerny (1979, 1982) constructed a generalized alteration sequence. The consistency of this alteration sequence allowed recognition of clay pods pseudomorphous after pollucite even at localities with no relicts of the original mineral (Cerny 1978).

Shaub & Schenck (1954) observed in thin sections of pollucite from Lithia, Massachusetts, fracture filling but not replacement by any of the (spodumene and mica) vein minerals. Cooper (1964) suggested that lepidolite veining within the large pollucite body at Bikita was derived from a "residual liquor" by simultaneous crystallization of pollucite from a number of internal points. If this is the case, some of the "alteration" products of pollucite may instead coexist, perhaps in equilibrium, with pollucite.

## CHAPTER 3

## COLLECTION AND TREATMENT OF DATA

45 thin sections were examined, representing 36 specimens from 24 localities in Africa and the United States. A CAMEBAX SX-50 electron microprobe was used, and the "PAP" procedure of Pouchou & Pichoir (1985) was applied to data reduction. Back-scattered electron imaging was used to select points for analysis.

For pollucite, major element concentrations were measured using 15 kV, 20.0 nA, beam size 5  $\mu\text{m}$ , and count times of 20 s (background 10 s). Standards used were pollucite (Al  $K\alpha$ , Si  $K\alpha$ , Cs  $L\alpha$ ), and albite (Na  $K\alpha$ ). Minor elements were determined using 15 kV, 40 nA, beam size 5  $\mu\text{m}$ , and count times of 50 s (background 25 s). Standards used were rubidian microcline (Rb  $L\alpha$ ), orthoclase (Fe  $K\alpha$  and K  $K\alpha$ ), anorthite (Ca  $K\alpha$ ), hornblende (Mg  $K\alpha$ ), and vanadium diphosphate (P  $K\alpha$ ).

For analcime, major element concentrations were measured using 15 kV, 10 nA, beam size 20  $\mu\text{m}$ , and count times of 20 s (background 10 s). Standards used were albite (Na  $K\alpha$ , Al  $K\alpha$ , Si  $K\alpha$ ), and pollucite (Cs  $L\alpha$ ). Minor elements were determined using 15 kV, 10 nA, beam size 20  $\mu\text{m}$ , and count times of 50 s (background 25 s). Standards were the same as those for pollucite minor elements.

Results of the analysis were recalculated to atomic

contents on the basis of six oxygens per anhydrous subcell. The "CRK" index was calculated as  $100(\text{Cs} + \text{Rb} + \text{K})/\text{cation sum}$  (Cerny 1974), where  $\text{cation sum} = (\text{Li}) + \text{Na} + \text{Ca} + \text{Mg} + \text{Cs} + \text{Rb} + \text{K}$ . The index is based on assignment of the large cations  $\text{Cs}^+$ ,  $\text{Rb}^+$ , and  $\text{K}^+$  to the 16b site and  $\text{Li}^+$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and any other minor cations to the smaller 24c site in Beger's (1969) crystal structure model of pollucite. The CRK index is thus a crystallochemical indicator of the molar proportion of pollucite in the analcime framework, separating those cations behaving structurally as sodium, from those behaving structurally as cesium.

The assignment of minor cations to 16b or 24c was verified for the relatively abundant  $\text{Rb}^+$  and  $\text{Ca}^{2+}$  in pollucite from High Grade Dyke, Manitoba (Teertstra et al. 1992a). The validity of including  $\text{Rb}^+$  in the 16b site, and  $\text{Ca}^{2+}$  in the 24c site was tested by comparing the closeness of the alkali cation sum (in plots of Na vs Cs), to Al values, for various assignments of  $\text{Rb}^+$  and of  $\text{Ca}^{2+}$  to 16b and 24c. When  $\text{Rb}^+$  was assigned to 16b, and  $\text{Ca}^{2+}$  to 24c, cation sums fit closer to Al values than when  $\text{Rb}^+$  was assigned to 24c or  $\text{Ca}^{2+}$  to 16b.

### 3.1 The ASTM 4POLL standard

The American Society for Testing Materials (ASTM) pollucite standard consists of several grains in a polished mount located in standard block 4. Unpublished results of wet chemical analysis were supplied (4POLL in Table 2.11).

Counting parameters (for Cs, Al, and Si) were derived mainly from one grain, and 21 complete analyses were collected between September 1989 and August 1991. From the other grains, 1 analysis of homogeneous pollucite and 5 analyses of compositionally heterogeneous pollucite were collected during a single run.

#### 3.1.1 Textural relationships and alteration

Only two of the pollucite grains are entirely homogeneous. Some are cross-cut by diffuse veinlets of Cs-rich pollucite 1 to 15  $\mu\text{m}$  in width. These Cs-rich veinlets contain at their core quartz grains 1 to 5  $\mu\text{m}$  in size. The pollucite is cross-cut by veins of mica and (later?) spodumene (25  $\mu\text{m}$  wide), and spodumene (75  $\mu\text{m}$  wide). The alteration products are later than the Cs-rich veinlets. Adjacent to the alteration products, the pollucite is enriched in  $\text{Na}^+$ .

### 3.1.2 Compositional trends

A plot of Al + Fe + P vs. Si, both atoms p.f.u. (per formula unit) is shown in Figure 3.1-1. The data fit close to a line of slope -1.00, as expected for the single tetrahedral site. This suggests that all the minor elements substituting for Si have been determined. Plots of Al + Fe + P vs. Si were inspected for all localities, but not duplicated here. In the standard and all the unknowns, the atomic sum of the tetrahedral cations was always slightly above 3.00, based on 6 oxygens. Deviation of the trend from a slope of -1.00, or data points with poor fit to the theoretical line, indicates substitutions for Si of elements other than Al, Fe, or P, and this was not observed. Any data with unusually poor fit were discarded, only if there was a possibility of any analytical problems (e.g. overlap with other minerals, obvious sample burning, operator error).

A plot of Na (+ Ca + Mg) vs. Cs (+ Rb + K), both atoms p.f.u., is shown in Figure 3.1-2. The data deviate from a trend characteristic of constant Al; the sum of the alkali cations, and consequently Al, increase with enrichment in Cs<sup>+</sup>. Additional scatter in the data is due to the substitution of cation(s) which could not be analysed for (e.g. Li<sup>+</sup> or some form of H<sup>+</sup>), possible substitution of a single cation into both 24c and 16b sites, and analytical

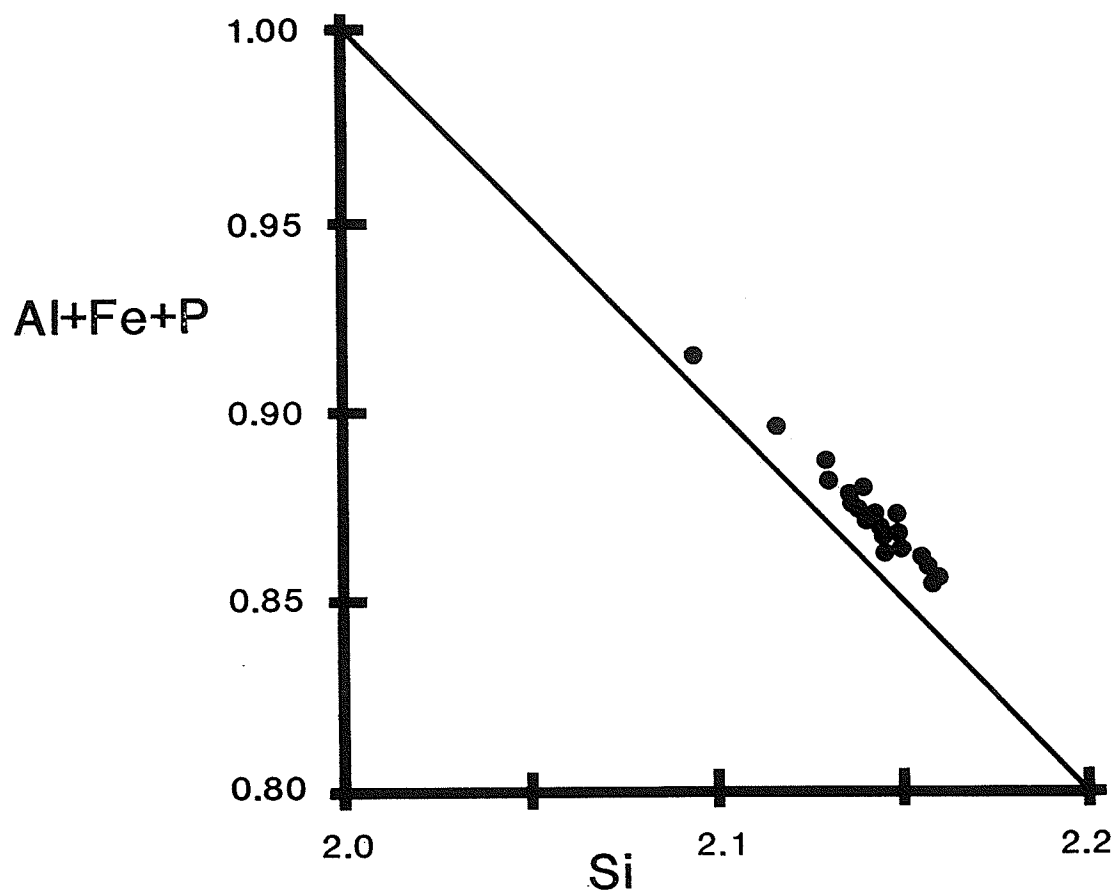


Fig. 3.1-1. Al + Fe + P vs. Si, both as atoms p.f.u. for 4POLL standard. The line shown is for the theoretical 1:1 substitution in framework silicates.

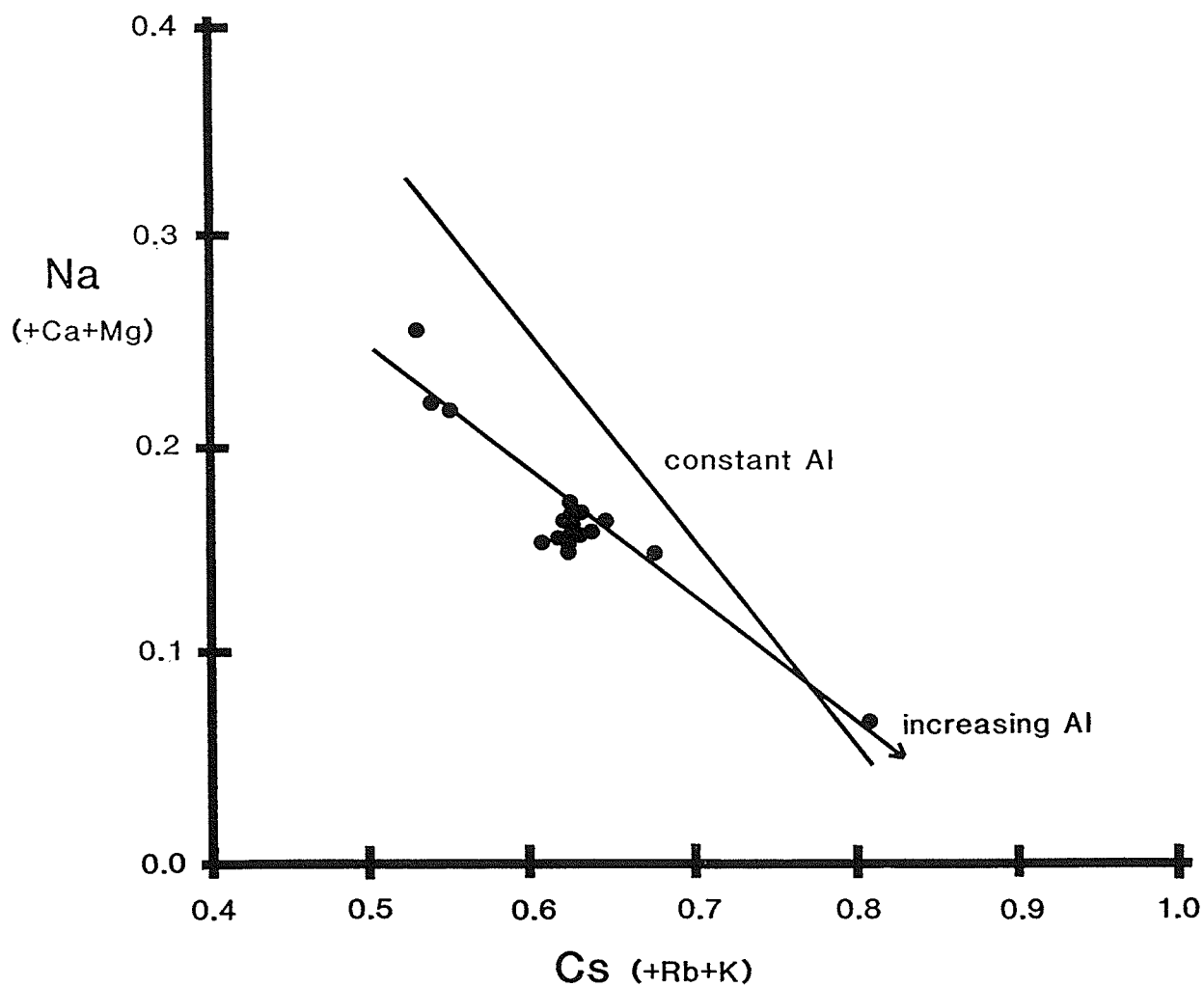


Fig. 3.1-2. Na (+ Ca + Mg) vs. Cs (+ Rb + K), both as atoms p.f.u. for 4POLL standard. The line "constant Al" is for the average Al = 0.860 of the homogeneous pollucite.



error. No further plots are given for the other localities, as information from this plot is essentially maintained in plots of sum of cation charges vs. CRK.

A plot of Si/Al vs. CRK is shown in Figure 3.1-3a. The homogeneous pollucite has Si/Al from 2.43 to 2.56 and CRK from 78.2 to 80.6; the average, and additional representative compositions are given in Table 3.1-1. The Na-enriched pollucite has CRK as low as 67.6, and Si/Al in the same range as the homogeneous pollucite. Cs-rich veinlets have Si/Al as low as 2.29 and CRK as high as 92.0.

A plot of Al (+ Fe), and sum of cation charges, both atoms p.f.u. vs. CRK is shown in Figure 3.1-3b. Two trends are apparent; one in which Al increases with CRK, and one in which Al is constant with CRK. The latter trend is defined by the Na-enriched pollucite. The sum of cation charges follows below the Al trends.

The average composition of the homogeneous pollucite (Table 3.1-1) shows excellent agreement with the bulk compositions of 4POLL in Table 2.11, and therefore was used as a reference standard. The compositions of pollucite from unknown localities, measured relative to 4POLL, varies within the range indicated by the one-sigma standard deviations given in Table 3.1-1.

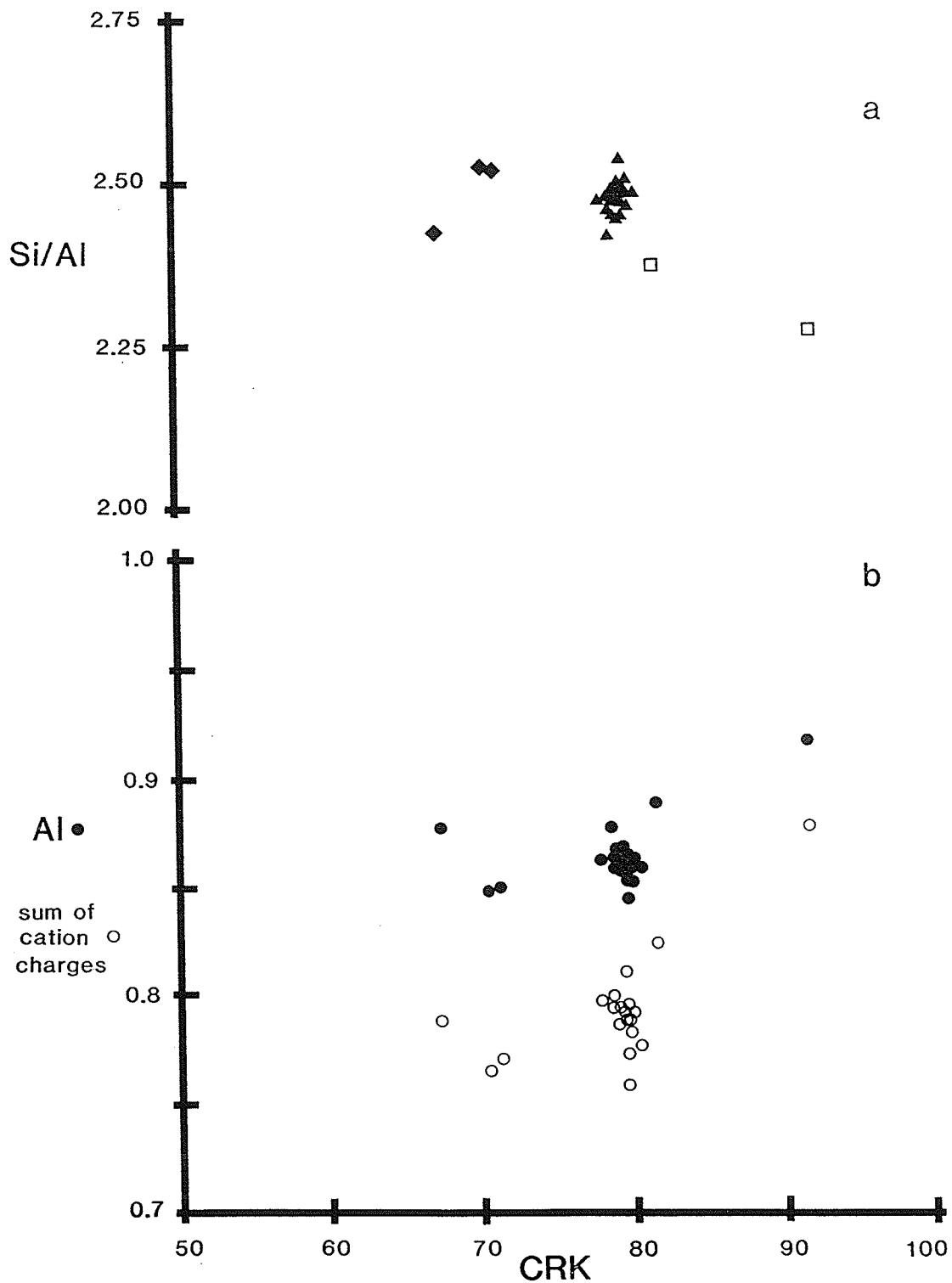


Fig. 3.1-3. a) Si/Al vs. CRK. Symbols used: homogeneous (▲), Na-enriched (◆), Na-enriched bleb (●), Cs-enriched bleb (○), Na-rich margin to Cs-rich veinlet (■), and Cs-rich veinlet (□), and b) Al (+ Fe), and sum of cation charges, both as atoms p.f.u. vs. CRK, for 4POLL standard.

Table 3.1-1: Composition of ASTM 4Poll standard.

No.	27	4	6	2	Average
SiO <sub>2</sub>	47.17	48.40	48.79	43.17	47.46 (.37)
Al <sub>2</sub> O <sub>3</sub>	16.07	16.07	16.95	16.02	16.15 (.11)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.33	0.32	0.12	0.00	0.31 (.02)
Na <sub>2</sub> O	1.82	1.80	3.00	0.74	1.84 (.06)
K <sub>2</sub> O	0.02	0.03	0.05	0.00	0.01 (.01)
Rb <sub>2</sub> O	0.58	0.56	0.00	0.58	0.56 (.05)
Cs <sub>2</sub> O	31.44	31.54	28.24	38.13	31.72 (.39)
CaO	0.00	0.01	0.00	0.01	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.43	98.71	97.15	98.66	98.06 (.60)
Si	2.142	2.158	2.140	2.094	2.144 (.010)
Al	0.861	0.845	0.876	0.916	0.860 (.006)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.013	0.012	0.005	0.000	0.012 (.003)
Na	0.161	0.156	0.255	0.069	0.162 (.006)
K	0.001	0.002	0.003	0.000	0.000
Rb	0.017	0.016	0.000	0.018	0.016 (.002)
Cs	0.609	0.600	0.529	0.789	0.611 (.007)
Ca	0.000	0.000	0.000	0.001	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.861	0.845	0.876	0.916	0.860 (.006)
Σ charges	0.787	0.773	0.787	0.878	0.789 (.010)
Si/Al	2.489	2.555	2.443	2.287	2.493 (.02)
CRK	79.6	79.9	67.6	92.0	79.6 (0.5)

27. Homogeneous pollucite

4. Homogeneous, high Si/Al.

6. Na-enriched homogeneous pollucite.

2. Cs-rich veinlet.

Average of homogeneous pollucite (N = 22).

### 3.2 Qualifying statements

The following comments examine the scope and intent of the present study, the analytical methods, and aspects of the representation of compositional range.

Preliminary results on material from other localities indicate that the data included here are typical and represent the full range of phenomena. Consequently the data from other localities are not included here. Other work, which tempered the following discussion, likewise is not included here: X-ray single crystal and powder diffraction studies, synthesis in the analcime-pollucite series, examination and analysis of secondary cesian analcime, and MAS NMR spectroscopy of natural minerals and synthetic phases in the series. Exhaustive literature reviews of studies on the natural minerals, synthesis of related phases, and relevant studies involving nuclear waste containment influenced the present study.

Two analytical programs were necessary for determining compositions across the analcime-pollucite series; one for analcime and cesian analcime ( $CRK < 50$ ) and one for pollucite ( $CRK > 50$ ). The pollucite program, used on analcime, seems to give correct atomic ratios (Si/Al and CRK), but incorrect oxide weights and a large monovalent cation deficiency relative to Al because of sample burning. Na-enriched (cation-exchanged) pollucite was found in some

samples to be susceptible to sample burning even at relatively high CRK (60 to 80). The analcime program, used on pollucite, gives oxide weights which are too high (for Al and Si), but a smaller apparent cation deficiency (relative to Al) than the pollucite program.

Many of the figures lack data for compositions in the range CRK 40 to 60. This is, in part, a result of the analytical difficulties mentioned above. As well, the most Na-enriched compositions occur closest to fractures and alteration veins, and immediately adjacent to the margins of Cs-rich veins (Fig. 4.3-4 and Fig. 4.5-1b). Because of this increasingly small exposure they are of limited accessibility even to the smallest useful beam size of 5  $\mu\text{m}$ .

The compositional diagrams do not necessarily represent the abundance of any particular composition or textural feature. These graphs may thus be used to define compositional trends but not to determine mass balance of reactions.

The levels of grey cannot be correlated between different back-scattered electron images. Within individual BSE images, the considerable difference in electron density between Na and Cs usually dominates the grey level. A secondary influence is produced by changes in Si/Al ratio, because of the variation in cation contents with Al (e.g. see Table 4.5-2 compositions with identical CRK but

different Si/Al ratios and BSE grey levels).

The images were chosen primarily to represent the textural features typical of individual localities; a secondary emphasis was placed upon documentation of unusual features. This emphasis detracts from the importance of localities in which most of the pollucite is compositionally homogeneous - an interesting feature in itself. For some localities a small sample size has limited the range of observations.

## CHAPTER 4

## POLLUCITE FROM AFRICA

## 4.1 Benson # 1, Zimbabwe

The Benson pegmatites are located in the north M'toko region of Zimbabwe. Pollucite occurs in the footwall of the core zone of Benson # 1, as rounded masses 3 or 4 ft. in diameter. The masses are surrounded by spodumene; in thin section they enclose corroded spodumene laths. Veins of lepidolite criss-cross the pollucite. Pollucite occurs also in Benson # 4, in the footwall of the core zone in association with spodumene and amblygonite (Hornung & von Knorring 1962).

Bulk compositions determined by Khalili & von Knorring (1976) are given for Benson # 1 (No. 70 in Table 2.11) and for Benson # 3 (No. 28 in Table 2.1e).

Sample numbers: PA-023 (V.T. King) and PA-079 (O.von Knorring); 2 thin sections, 34 analyses.

## 4.1.1 Properties, associations and alteration

A schematic alteration sequence is shown in Figure 4.4-1, as a guide to the following comments.

In hand specimen, the pollucite is white in colour, with some small transparent grains up to 0.5 mm in

diameter. The pollucite contains mauve lepidolite veins up to 5 mm in width, which are very irregular in thickness and continuity. Additional lepidolite grains are dispersed throughout the pollucite. One side of the hand specimen is stained rust-red and weathered to crumbly fine (1 mm) sand-sized round grains of pollucite.

In thin section, the pollucite grains contain abundant tiny acicular inclusions arranged in a three-dimensional rectangular grid, which may have crystallized at the same time as the pollucite (Fig. 4.1-2). Other scattered, abundant inclusions are spodumene and quartz grains (1 to 3 mm in size), which may have crystallized near the end of pollucite crystallization, or replaced pollucite. Lepidolite veins (+ quartz ± albite) appear to follow grain boundaries of the pollucite, outlining polyhedral areas of pollucite. The lepidolite veins may have been a product of late crystallization, as they only locally appear to replace the pollucite. Adularia is not abundant, but replaces pollucite along the surface of the lepidolite veins.

#### 4.1.2 Textural relationships

The pollucite is compositionally homogeneous, with slightly Cs-enriched pollucite cementing grain boundaries; grains of pollucite form 5 to 12 sided polyhedra, 100 to





Fig. 4.1-1. Schematic alteration sequence for Benson # 1, Zimbabwe.

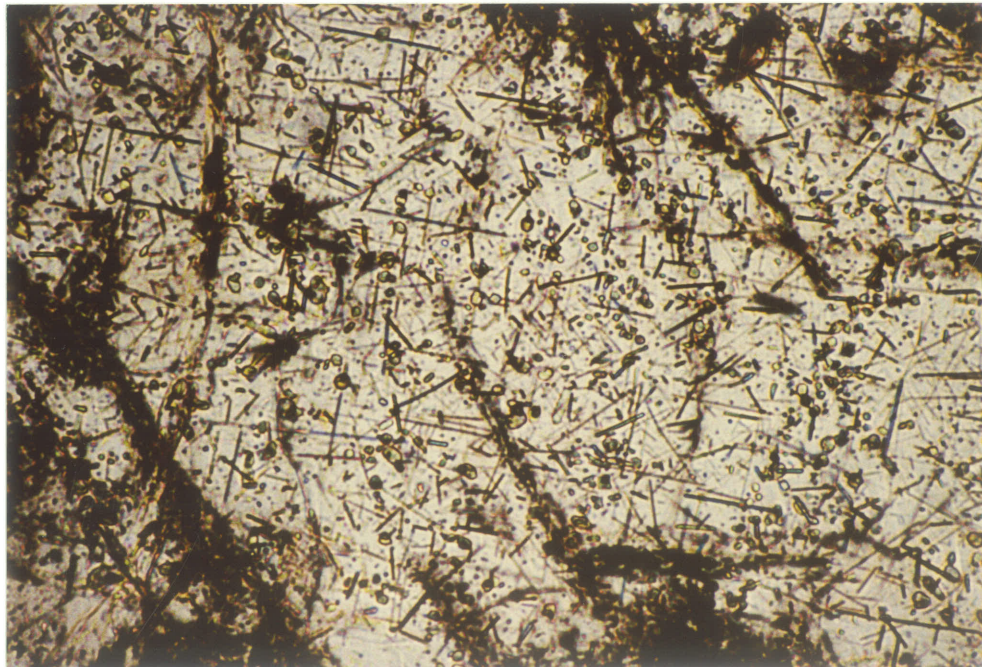


Fig. 4.1-2. Microphotograph of Benson # 1 pollucite. The pollucite contains an acicular AlP-bearing phase arranged in a three-dimensional rectangular grid. Also shown are irregular veins of fine-grained mica.

300  $\mu\text{m}$  in size (Fig. 4.1-3a). The abundant acicular inclusions (10 to 15  $\mu\text{m}$  in diameter) within grains show abundant Al and P (unknown mineral). Lepidolite, quartz, and some albite occur along the grain boundaries of pollucite. The adularia contains mica and small (1 to 2  $\mu\text{m}$ ) round grains of pollucite.

Veinlets of Cs-rich pollucite + quartz tend to follow grain boundaries, or cut across, parallel to the edges of the polyhedra. The veinlets are 5 to 10  $\mu\text{m}$  in width, and are not abundant. Rarely, a blebby structure developed, closely associated with the Cs-rich veinlets which crosscut it. A blebby structure, containing grains of quartz and the AlP mineral, is also developed adjacent to some spodumene grains (Fig. 4.1-3b).

#### 4.1.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 4.1-4a. The homogeneous pollucite has Si/Al from 2.45 to 2.53 and CRK from 82.7 to 84.4; the average and additional representative compositions are given in Table 4.1-1 and 2. Pollucite cementing the grain edges and junctions has a slightly lower Si/Al and a slightly higher CRK than the homogeneous pollucite. The Na-enriched blebs have the same Si/Al as the homogeneous pollucite, and a slightly lower CRK. Cs-enriched blebs have Si/Al as low as 2.32 and CRK as

high as 91.3. Cs-rich veinlets have Si/Al as low as 2.28 and CRK as high as 93. A plot of Al (+ Fe) and sum of cation charges vs. CRK is shown in Figure 4.1-4b. The sum of cation charges follows a trend of Al increasing with CRK.

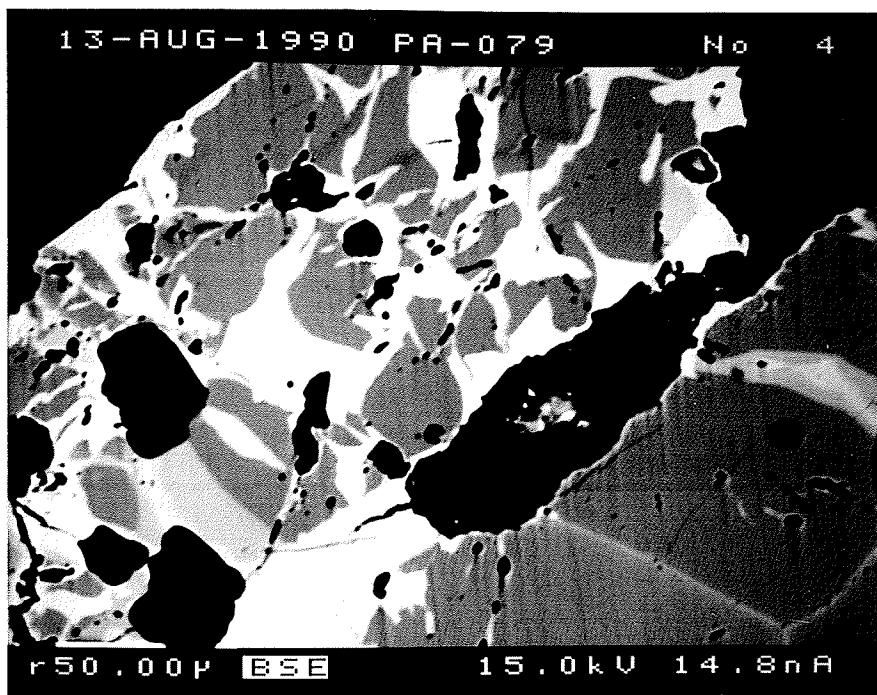
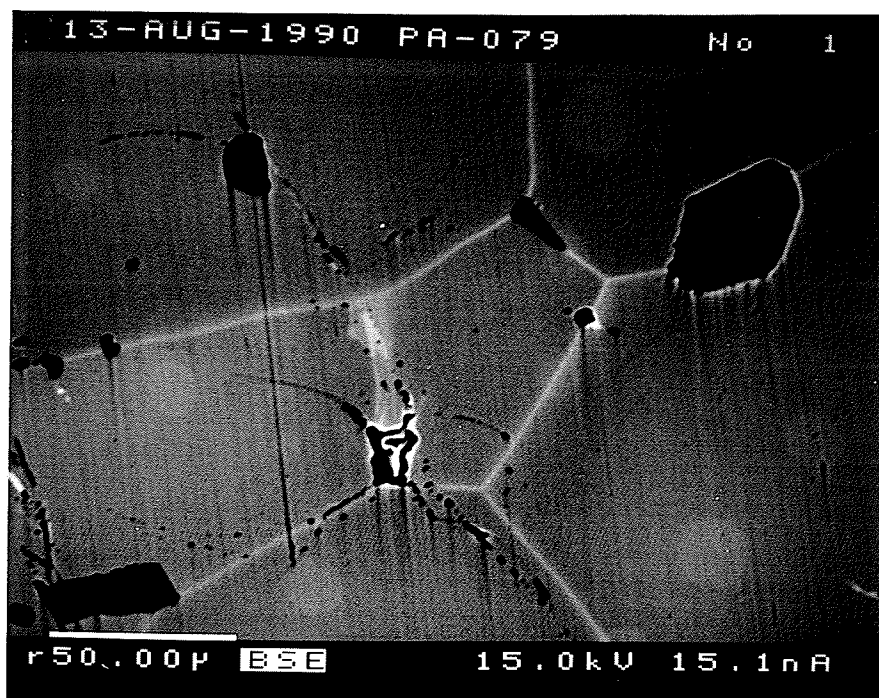


Fig. 4.1-3. Back-scattered electron (BSE) images of Benson # 1 pollucite. a) The pollucite consists of interlocking 5- to 12-sided polyhedra, "cemented" with Cs-rich pollucite. The dark grains are quartz. b) Cs-enriched blebs near lath spodumene (dark). To the top is a large area of spodumene. Within the pollucite are grains of spodumene and quartz.

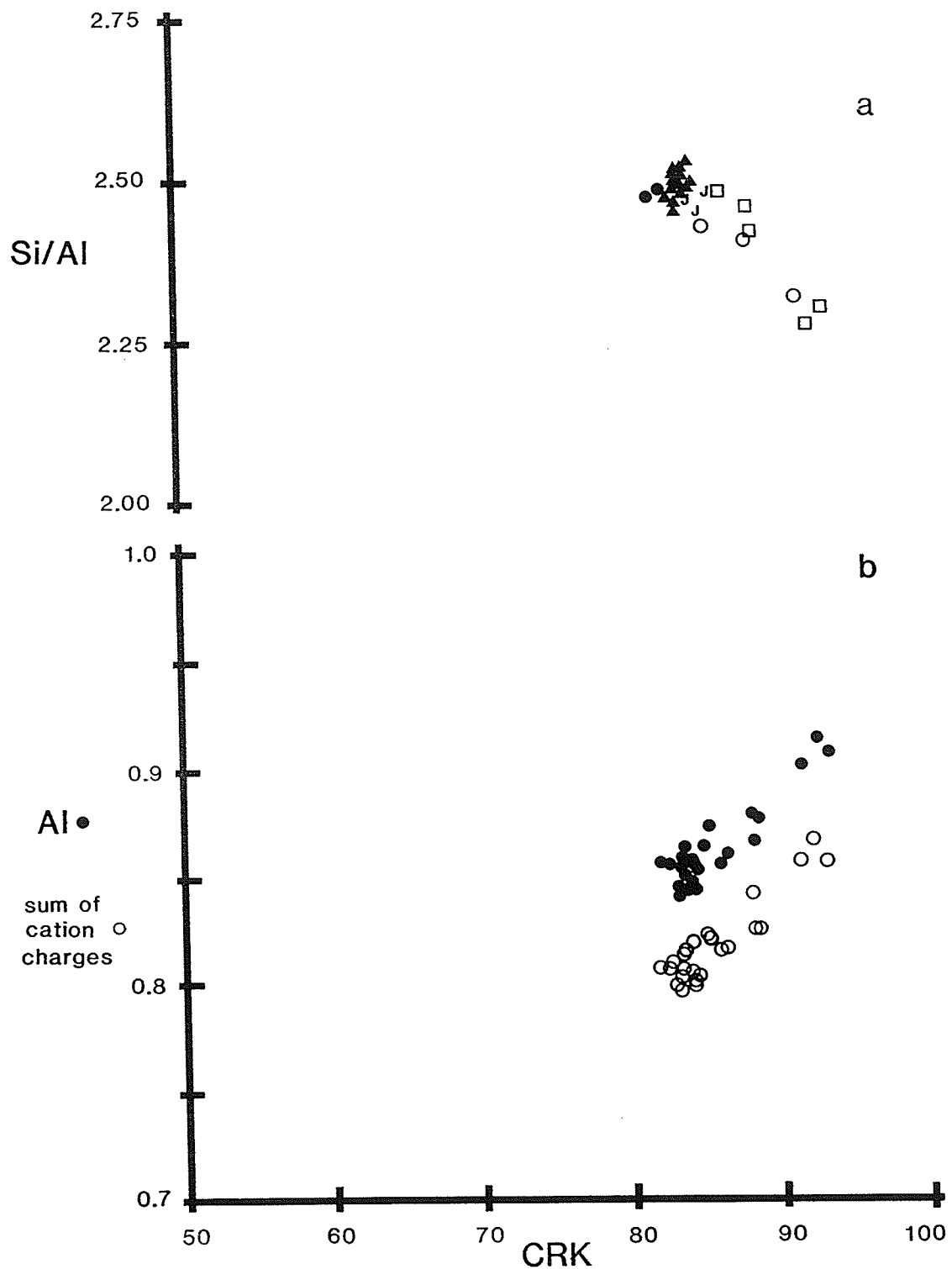


Fig. 4.1-4. Compositional trends for Benson # 1, Zimbabwe. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3; Cs-richer junctions of polyhedra (J).

Table 4.1-1: Composition of pollucite from Benson # 1, Zimbabwe.

No.	2	22	6	7	25
SiO <sub>2</sub>	47.72	46.96	46.94	46.80	47.45
Al <sub>2</sub> O <sub>3</sub>	15.85	16.11	15.82	15.89	16.10
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.02
P <sub>2</sub> O <sub>5</sub>	0.19	0.18	0.18	0.12	0.15
Na <sub>2</sub> O	1.43	1.51	1.50	1.30	1.64
K <sub>2</sub> O	0.01	0.01	0.01	0.02	0.01
Rb <sub>2</sub> O	0.95	0.85	0.88	0.97	0.69
Cs <sub>2</sub> O	33.15	33.12	32.74	34.21	32.94
CaO	0.01	0.00	0.01	0.00	0.01
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	99.31	98.75	98.07	99.31	99.02
Si	2.159	2.141	2.151	2.146	2.147
Al	0.845	0.866	0.855	0.860	0.859
Fe	0.000	0.000	0.000	0.000	0.001
P	0.007	0.007	0.007	0.005	0.006
Na	0.126	0.134	0.133	0.115	0.144
K	0.001	0.001	0.001	0.001	0.001
Rb	0.028	0.026	0.026	0.029	0.020
Cs	0.639	0.644	0.640	0.669	0.636
Ca	0.000	0.000	0.000	0.000	0.001
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.845	0.866	0.855	0.860	0.860
Σ charges	0.800	0.811	0.806	0.818	0.809
Si/Al	2.534	2.453	2.497	2.486	2.481
CRK	84.2	83.4	83.4	85.9	81.9

2. Homogeneous pollucite grain.  
 22. Homogeneous pollucite grain.  
 6. Homogeneous pollucite grain, adjacent to 7.  
 7. Cs-enriched triple junction of three pollucite grains.  
 25. Na-enriched bleb, between 24 and 26 of Table 4.1-2.

Table 4.1-2: Composition of pollucite from Benson # 1, Zimbabwe.

No.	24	26	20	21	Average
SiO <sub>2</sub>	46.28	44.39	45.97	44.03	47.35 (.23)
Al <sub>2</sub> O <sub>3</sub>	16.09	16.21	15.85	16.19	15.92 (.12)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.04	0.00	0.00	0.03	0.19 (.02)
Na <sub>2</sub> O	1.36	0.81	1.08	0.64	1.49 (.04)
K <sub>2</sub> O	0.02	0.02	0.00	0.02	0.01
Rb <sub>2</sub> O	0.80	0.72	0.83	0.78	0.87 (.05)
Cs <sub>2</sub> O	34.27	37.67	35.57	38.00	33.11 (.18)
CaO	0.00	0.00	0.00	0.01	0.01
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.85	99.81	99.31	99.69	98.95 (.32)
Si	2.136	2.105	2.140	2.101	2.152 (.004)
Al	0.875	0.906	0.870	0.910	0.853 (.005)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.002	0.000	0.000	0.001	0.000
Na	0.122	0.075	0.097	0.059	0.132 (.004)
K	0.001	0.001	0.000	0.001	0.000
Rb	0.024	0.022	0.025	0.024	0.026 (.002)
Cs	0.675	0.762	0.707	0.773	0.642 (.005)
Ca	0.000	0.000	0.000	0.001	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.875	0.906	0.870	0.910	0.853 (.005)
Σ charges	0.823	0.860	0.829	0.860	0.807 (.005)
Si/Al	2.435	2.324	2.460	2.307	2.503 (.018)
CRK	85.2	91.3	88.2	93.7	83.5 (0.4)

24. Cs-enriched bleb.

26. Cs-enriched bleb.

20. Cs-rich veinlet.

21. Cs-rich veinlet.

Average of homogeneous pollucite (N = 20).

#### 4.2 Bikita, Zimbabwe

The Bikita pegmatite is located in the Fort Victoria area, Zimbabwe. The geology of the Bikita area has been described by Tyndale-Biscoe (1952). Some aspects of the Bikita mining operations were described by Symons (1961). The pollucite bodies here are possibly the largest in the world.

In outcrop, the pollucite is cut into a coarse polygonal mosaic by veins of lepidolite 2 to 3 cm wide. The lepidolite veins are wholly contained by the pollucite bodies, and do not extend into the adjacent rock or included bodies. One large pollucite body encloses sharply bounded blocks of petalite (1.5 m in size), microcline (1.5 m), and lepidolite and quartz (6 m). The pollucite ore contains an estimated 40% quartz and other minerals (Cooper 1964). The bulk ore was found to contain albite (16%), lepidolite (13%), quartz (4%), spodumene (1%) and petalite (1%) (Dean & Nichols 1962).

Pollucite from Bikita was used in a particle desorption mass spectrometry study (Summers & Scheikert 1986).

Bulk compositions were determined by Cooper (1964) (No. 9 in Table 2.1b) and Khalili & von Knorring (1976) (No. 27 in Table 2.1e).

Sample numbers: PA-021 (D.H. Garske), PA-028 (C.



Guillemin) and PA-018 eucryptite (A.J. Anderson); 3 thin sections, 30 analyses.

#### 4.2.1 Properties, associations and alteration

A schematic alteration sequence is shown in Figure 4.2-1, as a guide to the following comments.

In hand specimen, the pollucite is white in colour and vitreous to greasy in lustre. Lepidolite veins cutting the pollucite are up to 1 cm in width. Additional lepidolite is dispersed throughout the pollucite. Subparallel very thin white veins braid around small eyelets of transparent pollucite up to 2 mm wide and 1 cm long. Embedded in the pollucite along with the thin white veins are abundant round aggregates of minerals, 1 to 2 mm in size.

In thin section, the pollucite contains rounded grains (1 to 2 mm) of spodumene (symplectitic with quartz), quartz, and lepidolite. The pollucite is filled with abundant micro-inclusions. Crosscutting the pollucite are wide lepidolite veins which are fracture-filling. Pollucite is replaced later by veins of fine-grained mica (lepidolite?) at most 1 mm in width. The mica in these thin veins is partially replaced by fine-grained spodumene. The veins are overgrown by spherical, granular aggregates (1 to 2 mm in size) of adularia and albite in

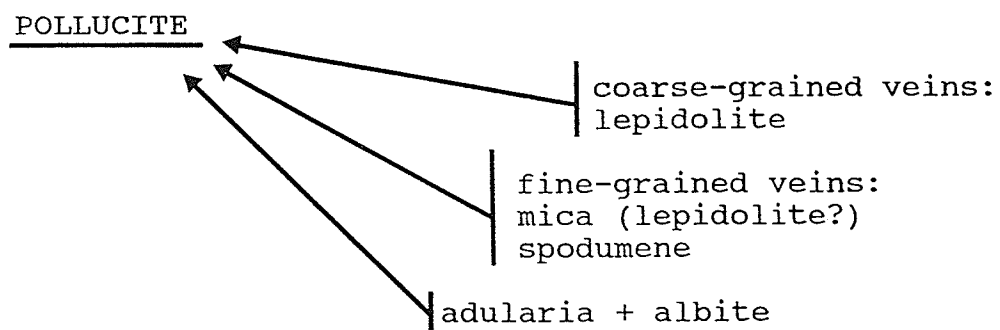


Fig. 4.2-1. Schematic alteration sequence for Bikita, Zimbabwe.

equal amounts.

#### 4.2.2 Textural relationships

The pollucite has only relatively small areas which are compositionally homogeneous. The areas are at most 5 mm in size; they have slightly different compositions and distinct boundaries.

Much of the pollucite has broken down into a blebby mosaic of Na- and Cs-enriched domains 50 to 300  $\mu\text{m}$  in size. Quartz grains ( $\pm$  feldspars) are associated with this mosaic. These domains are roughly rectangular in shape, with convex scallops along the margins. The shapes appear to have been created by replacement of successive domains, which are usually (but not always) more Cs-enriched. As a result of this complex series of replacements, the texture of the blebby pollucite ranges from brecciated to schollen or raft (Fig. 4.2-2a and b).

The blebby mosaic is crosscut by Cs-rich veinlets of two types. Early veinlets are 5 to 20  $\mu\text{m}$  in width, contain no quartz, and are fracture-filling. These are crosscut by quartz-containing veinlets 1 to 10  $\mu\text{m}$  in width, which are more Cs-rich, and braided to web-like. Locally (in areas consisting only of the quartz-containing Cs-rich veinlets in a matrix of homogeneous pollucite), the veinlets have parallel to sub-parallel orientation. Some late

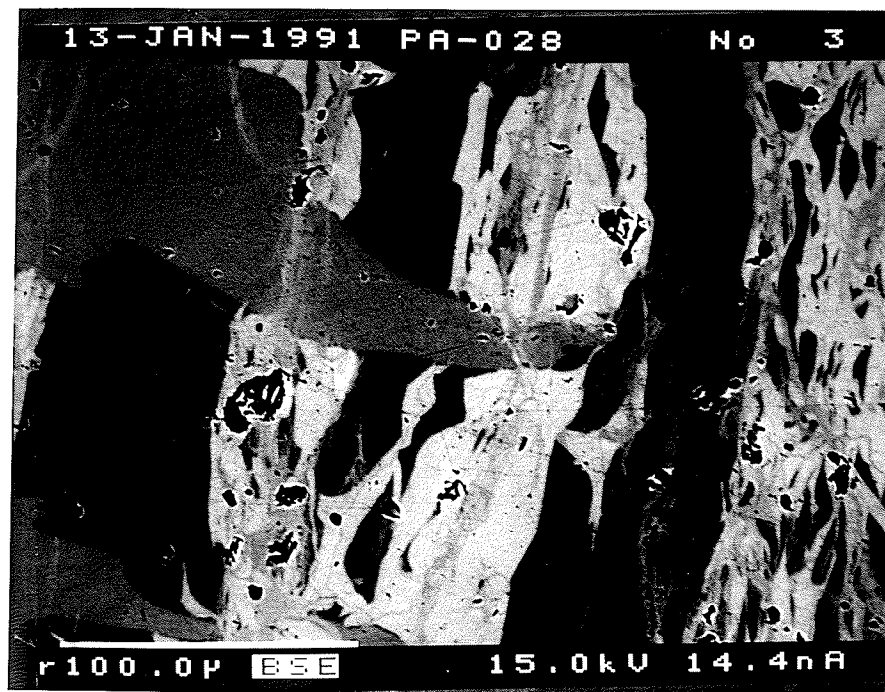
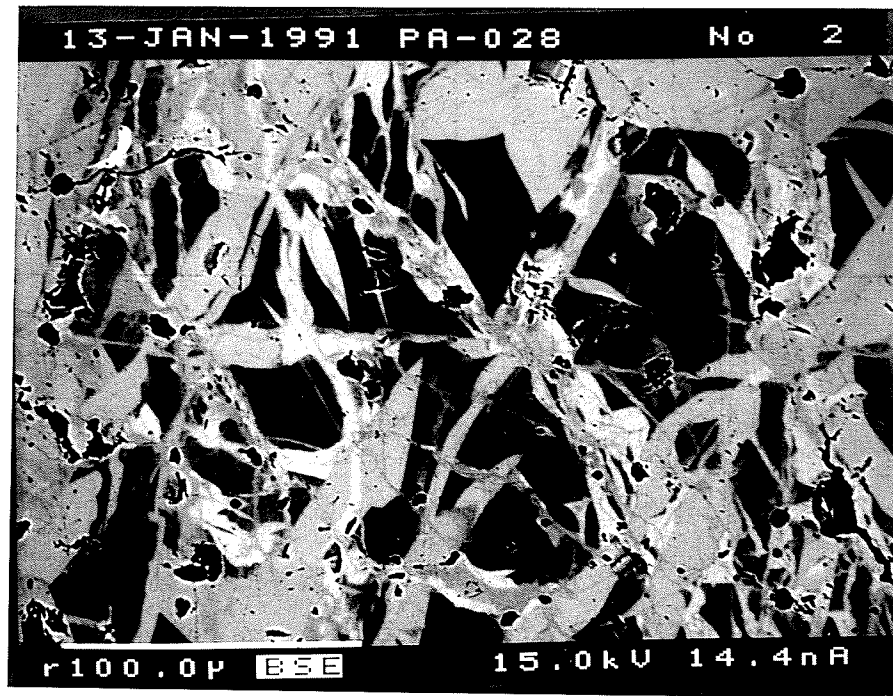


Fig. 4.2-2. BSE images of Bikita pollucite. a) and b) Multiple generations of Cs-enriched blebs in a matrix of Na-enriched blebs or remnants of homogeneous pollucite. Quartz grains are most closely associated with the Cs-enriched pollucite.

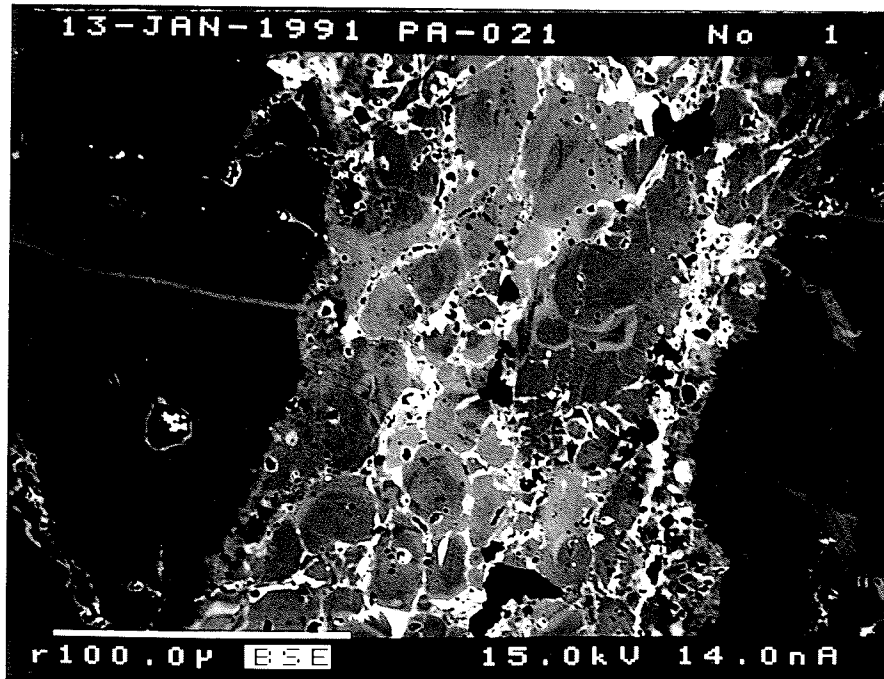
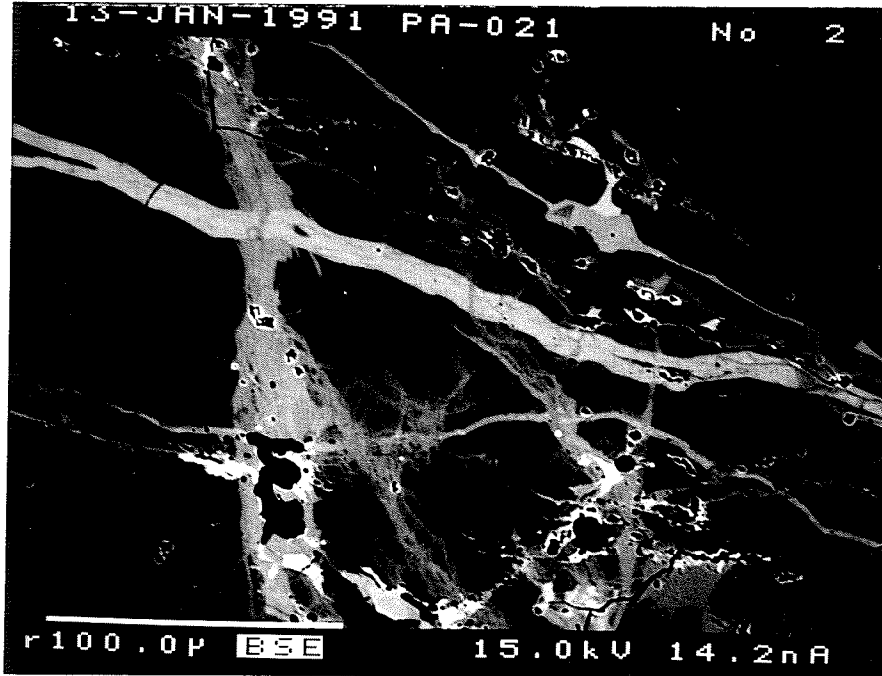


Fig. 4.2-2. c) and d) Cs-rich veinlets in areas of homogeneous pollucite. Later generations tend to be more Cs-rich and quartz-containing.

crosscutting veinlets are Na-rich (Fig. 4.2-2c and d).

The pollucite is rich in  $\text{Cs}^+$  adjacent to some spodumene and quartz grains, and also near the mica/spodumene veins. Pollucite inclusions within the adularia/albite aggregates are also Cs-rich.

#### 4.2.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 4.2-3a. The homogeneous pollucite has Si/Al from 2.52 to 2.64 and CRK from 79.4 to 82.1; the average and additional representative compositions are given in Table 4.2-1 and 2. The Na-enriched blebs have Si/Al slightly higher, and CRK slightly lower than the average of the homogeneous pollucite. Cs-enriched blebs have Si/Al as low as 2.39 and CRK as high as 89.2. Cs-rich veinlets have Si/Al as low as 2.27 and CRK as high as 93.5.

A plot of Al (+ Fe) and sum of cation charges vs. CRK is shown in Figure 4.2-3b. The sum of cation charges follows a trend of Al increasing with CRK.

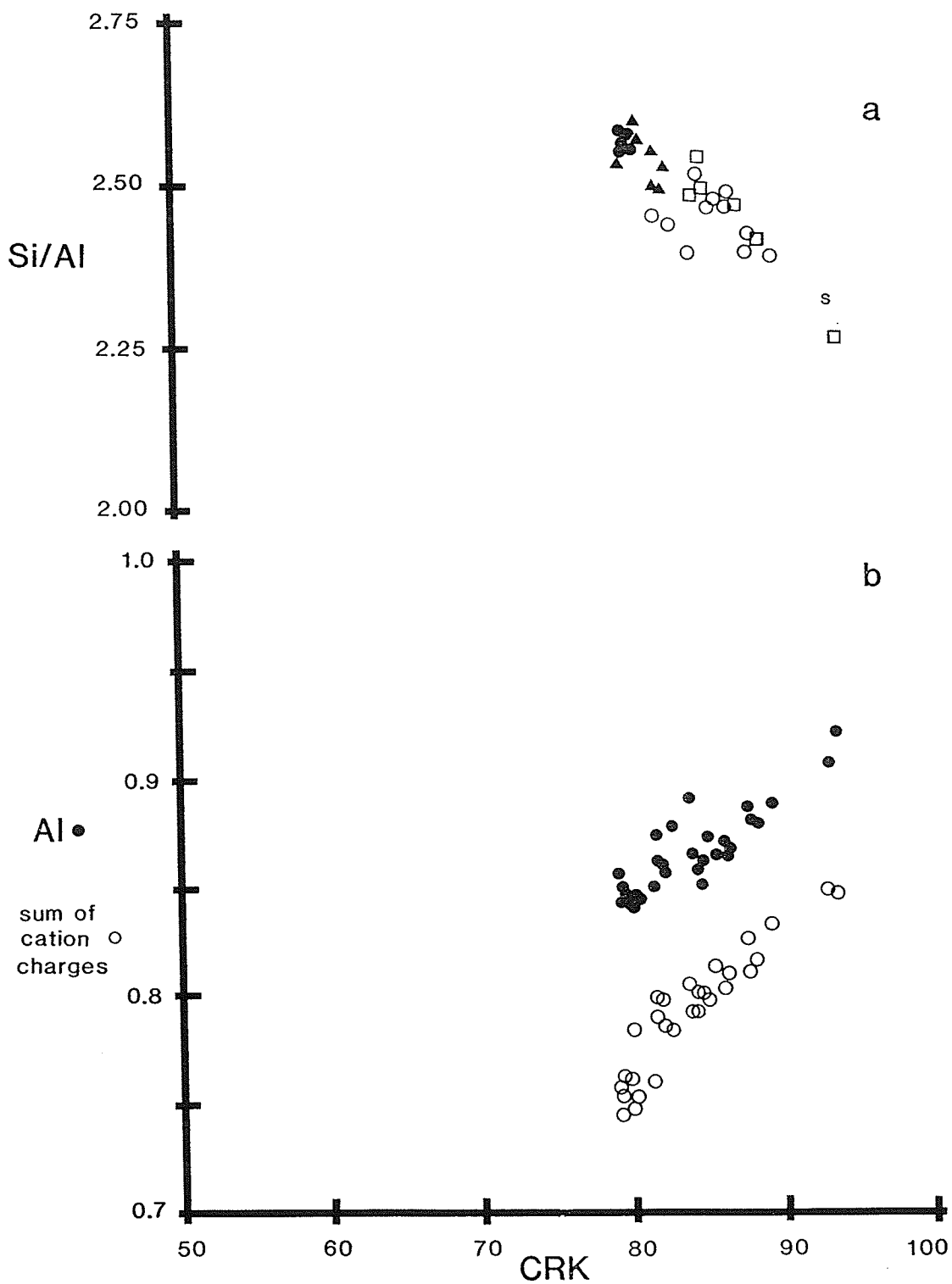


Fig. 4.2-3. Compositional trends for Bikita, Zimbabwe. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3; Cs-rich pollucite inclusion in spodumene (S).

Table 4.2-1: Composition of pollucite from Bikita, Zimbabwe.

No.	2	6	21	17	27
SiO <sub>2</sub>	48.16	50.36	49.10	47.00	45.16
Al <sub>2</sub> O <sub>3</sub>	15.89	16.21	16.19	16.39	16.02
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.03	0.37	0.37	0.38	0.00
Na <sub>2</sub> O	1.42	1.75	1.81	1.46	0.97
K <sub>2</sub> O	0.00	0.01	0.03	0.02	0.02
Rb <sub>2</sub> O	0.49	0.99	0.58	1.07	0.32
Cs <sub>2</sub> O	33.74	30.37	30.35	33.60	36.47
CaO	0.01	0.00	0.02	0.00	0.03
MgO	0.00	0.00	0.00	0.00	0.01
H <sub>2</sub> O	----	----	----	----	----
sum	98.69	100.06	98.44	98.92	98.99
Si	2.156	2.180	2.165	2.128	2.125
Al	0.857	0.826	0.842	0.875	0.888
Fe	0.000	0.000	0.000	0.000	0.000
P	0.001	0.014	0.014	0.015	0.000
Na	0.126	0.146	0.154	0.129	0.089
K	0.000	0.001	0.001	0.001	0.001
Rb	0.015	0.027	0.017	0.031	0.010
Cs	0.659	0.560	0.571	0.630	0.732
Ca	0.000	0.000	0.001	0.000	0.001
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.857	0.826	0.842	0.875	0.888
Σ charges	0.799	0.735	0.745	0.790	0.833
Si/Al	2.517	2.638	2.573	2.433	2.393
CRK	84.3	80.1	79.1	83.8	89.2

2. Homogeneous pollucite.  
 6. Homogeneous pollucite.  
 21. Na-enriched bleb.  
 17. Cs-enriched bleb.  
 27. Cs-enriched bleb.



Table 4.2-2: Composition of pollucite from Bikita, Zimbabwe.

No.	16	3	4	25	Average
SiO <sub>2</sub>	47.55	47.13	43.85	44.36	48.94 (1.1)
Al <sub>2</sub> O <sub>3</sub>	16.08	15.89	16.39	16.23	16.18 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.02	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.13	0.03	0.02	0.00	0.35 (.07)
Na <sub>2</sub> O	1.40	1.42	0.60	0.66	1.69 (.08)
K <sub>2</sub> O	0.03	0.00	0.03	0.01	0.01
Rb <sub>2</sub> O	1.01	0.49	0.48	0.54	0.95 (.06)
Cs <sub>2</sub> O	33.26	33.74	38.08	38.21	31.37 (.87)
CaO	0.01	0.01	0.01	0.00	0.00
MgO	0.00	0.00	0.01	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	99.47	98.69	99.46	100.00	99.51 (.37)
Si	2.152	2.156	2.094	2.106	2.161 (.014)
Al	0.858	0.857	0.922	0.908	0.842 (.012)
Fe	0.000	0.000	0.001	0.000	0.000
P	0.005	0.001	0.001	0.000	0.013
Na	0.123	0.126	0.055	0.060	0.145 (.005)
K	0.002	0.000	0.002	0.000	0.000
Rb	0.029	0.015	0.015	0.017	0.027
Cs	0.642	0.659	0.775	0.773	0.591 (.025)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.858	0.857	0.922	0.908	0.842 (.012)
Σ charges	0.796	0.799	0.847	0.850	0.764 (.022)
Si/Al	2.509	2.517	2.271	2.318	2.57 (.05)
CRK	84.6	84.3	93.5	92.9	81.0 (1.0)

16. Cs-enriched bleb.

3. Cs-rich veinlet.

4. Cs-rich veinlet.

25. Cs-rich pollucite adjacent to a spodumene grain.

Average of homogeneous pollucite (N = 7).

### 4.3 Helikon, Namibia

The Helikon Mine is located on Farm Okongava, Ost No. 72, near the town of Karibib, Namibia. Massive pollucite is associated with lepidolite, petalite, and amblygonite. It is traversed by lepidolite (and some quartz) veins 4 mm in thickness, as well as by numerous thin white veins up to 1 mm thick composed of an extremely fine-grained clay-like mineral (Nel 1944).

Pollucite from Helikon was used in a differential thermal analysis study (Heysteck & Schmidt 1953)

Bulk analysis were determined by Nel (1944) (No. 15 in Table 2.1c) and by Khalili & von Knorring (1976) (No. 63 in Table 2.1k). Ahrens (1945) determined (as wt.%) (0.54)  $\text{Rb}_2\text{O}$ , (0.021)  $\text{Li}_2\text{O}$ , (0.49)  $\text{K}_2\text{O}$ , (0.011)  $\text{Tl}_2\text{O}$  and (0.0012)  $\text{Ga}_2\text{O}_3$ .

Sample numbers: PA-029 (J.S. White, U.S.N.M. R 8270), PA-077 (O. von Knorring), and PA-125 (G. Friedrich); 3 thin sections, 46 analyses.

#### 4.3.1 Properties, associations and alteration

A schematic alteration sequence is shown in Figure 4.3-1, as a guide to the following comments.

In hand specimen, the pollucite is white to locally colourless, containing gemmy grains up to 5 mm in size. A

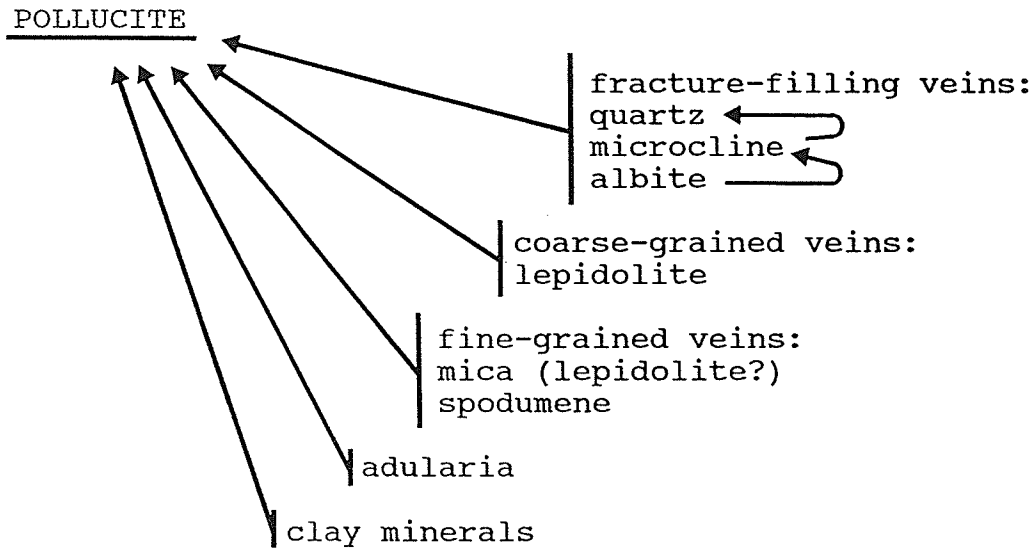


Fig. 4.3-1. Schematic alteration sequence for Helikon, Namibia.

vein of grey to slightly pink lepidolite crosscuts the pollucite; the vein is of even width (3 mm), and pinches out within the pollucite. Some small flakes of mauve lepidolite are scattered through the pollucite. Subparallel grey veins up to 1 mm wide are spaced about 5 mm apart. Fracture surfaces are coated with buff clay minerals. Associated with the clays and also scattered throughout the pollucite are thin dendritic coatings of a black (Mn-bearing?) mineral.

In thin section, pollucite is crosscut by a vein of coarsely grained quartz, about 1 cm in width. The quartz vein is cut by coarsely grained microcline ( $\pm$  quartz  $\pm$  mica), variable in width from 0.5 to 1.5 cm. The microcline, and some pollucite, are replaced by albite. Lepidolite associated with a coarse (3 mm) vein overgrows the quartz, microcline, and albite. Later fine-grained mica veins are replaced in part by spodumene, then overgrown by adularia. Adularia also occurs as rounded crystalline aggregates fully embedded in the pollucite.

#### 4.3.2 Textural relationships

The pollucite is compositionally heterogeneous to a high degree. Homogeneous areas are still present, but have mm-scale gradations of Na-enrichment, which are not visible in individual BSE images. The heterogeneous pollucite is

associated with grains of quartz ( $\pm$  an AlP-bearing mineral  $\pm$  spodumene) (Fig. 4.3-2a and b). The pollucite has a well-developed blebby structure. The blebs have an internal zonation (resembling an hourglass) parallel to the margins. The blebby structure is crosscut by Cs-rich veinlets (+ quartz) (Fig. 4.3-2c). A single very large (30 to 50  $\mu\text{m}$  wide) Cs-rich veinlet is present in apparently homogeneous pollucite (Fig. 4.3-2d).

#### 4.3.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 4.3-3a. The apparently homogeneous pollucite has a Si/Al from 2.50 to 2.57 (average  $2.539 \pm 0.018$ ) and Al (+ Fe) content of  $0.848 \pm .004$  atoms p.f.u.. The CRK varies from 64.5 to 75.1. Representative compositions are given in Table 4.3-1 and 4.3-2. Na-enriched blebs have Si/Al and CRK slightly lower than the most Na-rich homogeneous pollucite. Cs-enriched blebs have an average Si/Al near 2.40 and CRK ranging from 64 to 80. Cs-rich veinlets have a range of Si/Al from 2.35 to 2.49 and CRK from 70 to 85.

The locality is unusual in its near-constant Si/Al of the homogeneous pollucite with decreasing CRK. A traverse of Si/Al, and CRK (Fig. 4.3-4) across the large Cs-rich veinlet of Figure 4.3-2d is revealing, however: the apparently homogeneous pollucite shows a decrease in CRK

and a slight increase in Si/Al as the veinlet is approached over a relatively large distance. Pollucite in the core of the veinlet has a similar CRK (but lower Si/Al) as homogeneous pollucite 200  $\mu\text{m}$  from the veinlet. Pollucite up to 50  $\mu\text{m}$  from the vein is recrystallized; pollucite 50 to more than 200  $\mu\text{m}$  from the vein is either cation exchanged or recrystallized but maintaining a constant Si/Al in either case. From Fig. 4.3-1, it is apparent that the Cs-rich veinlet must have developed in situ by gaining  $\text{Cs}^+$  simultaneous with the enrichment in  $\text{Na}^+$  of the homogeneous pollucite. Pollucite with Si/Al 2.54 and the highest CRK (74) (No. 12 in Table 4.3-1) may be most representative of the original homogeneous pollucite.

A plot of Al (+ Fe) and sum of cation charges vs. CRK is shown in Fig. 4.3-3b. The Al (+ Fe) content is relatively constant with CRK; the sum of cation charges increases slightly with CRK.

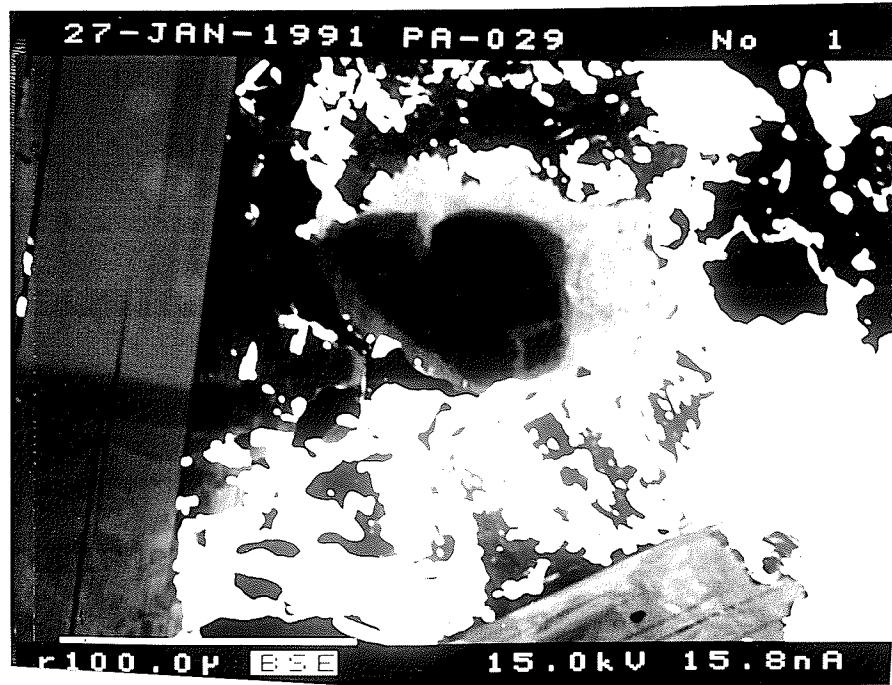


Fig. 4.3-2. BSE images of Helikon pollucite. a) Cs-rich pollucite next to a large, lightly zoned mica grain. At photo center, an adularia grain with increasing Rb from core to rim. b) Top half is an area of mica/quartz intergrowth, bottom half is a mica/pollucite intergrowth, grading to area of pollucite.

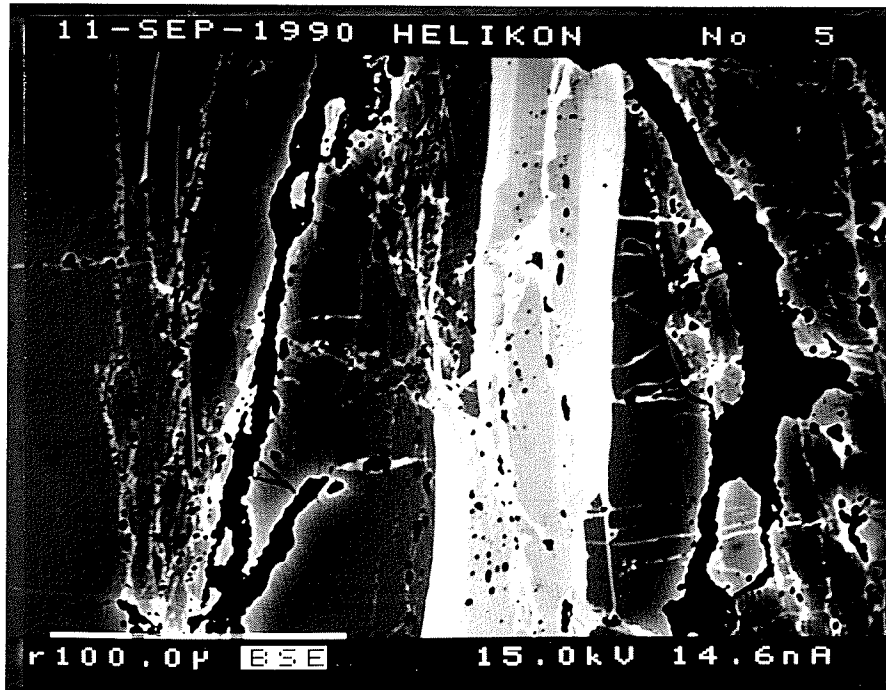
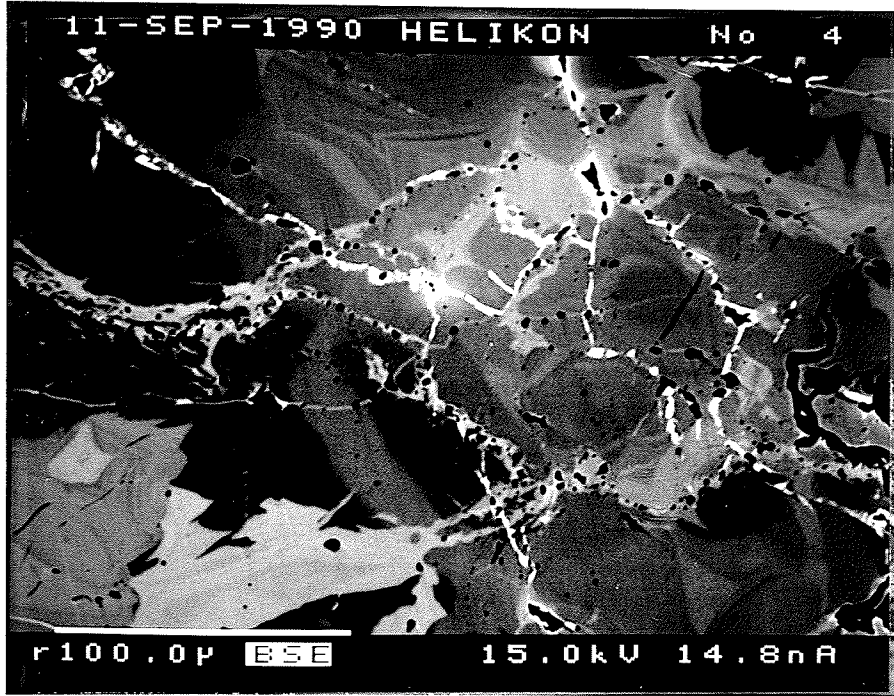


Fig. 4.3-2. c) Blebby mosaic of pollucite showing internal zonation of domains, crosscut by quartz-containing Cs-rich veinlets of typical size. d) An unusually large Cs-rich veinlet, itself containing parallel small quartz-containing Cs-rich veinlets. An E-W compositional traverse is given in Figure 4.3-4.



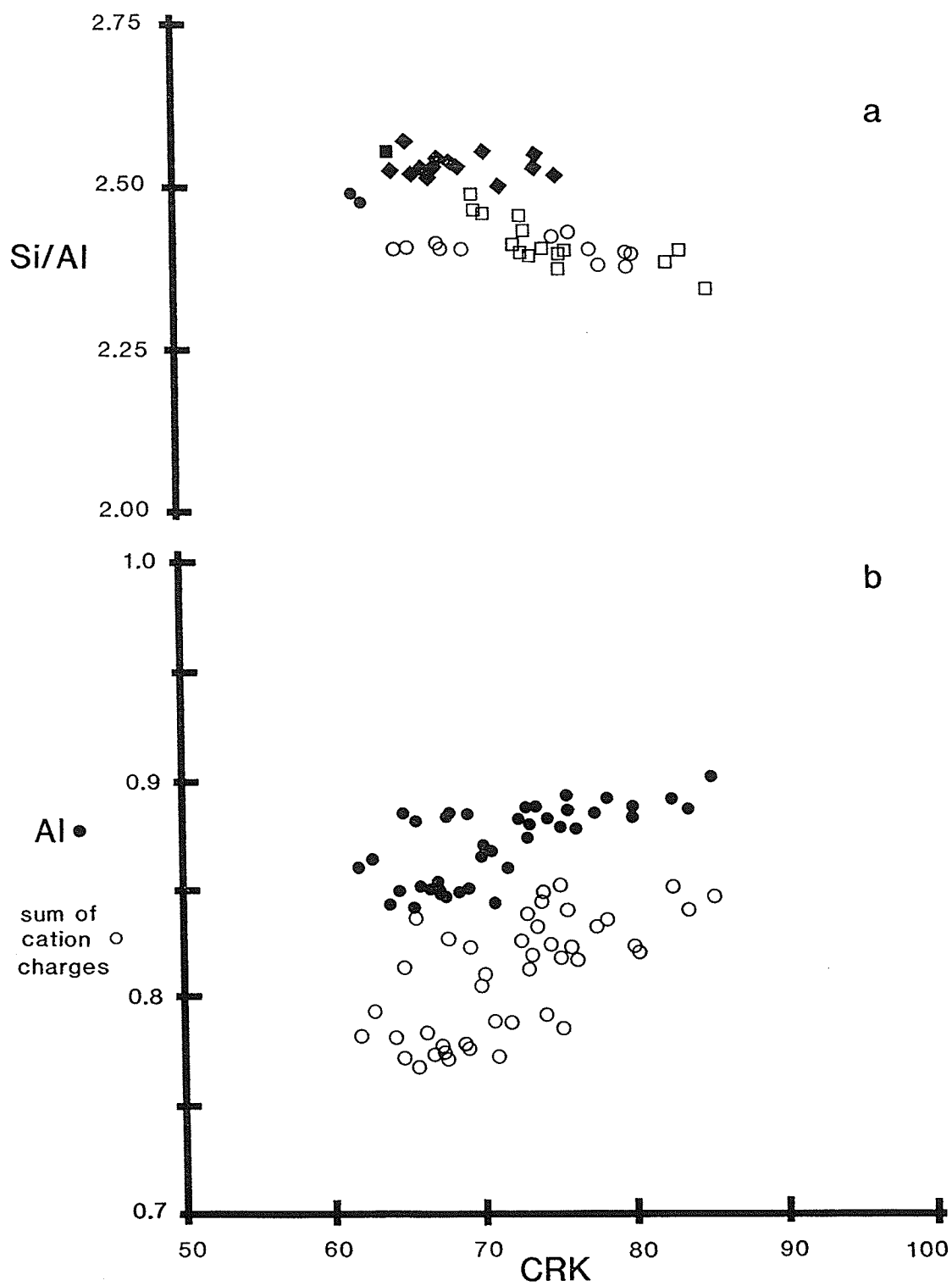


Fig. 4.3-3. Compositional trends for Helikon, Namibia. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.



Table 4.3-1: Composition of pollucite from Helikon, Namibia.

No.	12	6	34	25	30
SiO <sub>2</sub>	48.56	49.89	50.54	50.38	50.43
Al <sub>2</sub> O <sub>3</sub>	16.22	16.64	16.93	17.12	16.71
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.01	0.01	0.00	0.01
P <sub>2</sub> O <sub>5</sub>	0.33	0.33	0.33	0.32	0.37
Na <sub>2</sub> O	2.38	2.93	3.29	3.63	3.36
K <sub>2</sub> O	0.00	0.01	0.02	0.02	0.03
Rb <sub>2</sub> O	0.22	0.04	0.00	0.00	0.00
Cs <sub>2</sub> O	30.67	28.74	27.39	26.56	27.36
CaO	0.00	0.00	0.01	0.01	0.03
MgO	0.00	0.00	0.01	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.29	98.59	98.51	98.03	98.30
Si	2.150	2.156	2.154	2.145	2.156
Al	0.848	0.847	0.850	0.859	0.842
Fe	0.000	0.000	0.000	0.000	0.000
P	0.013	0.012	0.012	0.012	0.014
Na	0.205	0.245	0.272	0.299	0.279
K	0.000	0.001	0.001	0.001	0.002
Rb	0.006	0.001	0.000	0.000	0.000
Cs	0.580	0.530	0.498	0.482	0.498
Ca	0.000	0.000	0.001	0.000	0.001
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.848	0.847	0.850	0.859	0.842
Σ charges	0.791	0.777	0.773	0.782	0.781
Si/Al	2.535	2.545	2.533	2.496	2.561
CRK	74.1	68.5	64.6	61.8	64.1

12. Na-enriched.

6. Na-enriched.

34. Na-enriched.

25. Na-enriched bleb.

30. Na-enriched pollucite next to Cs-enriched veinlet.

Table 4.3-2: Composition of pollucite from Helikon, Namibia.

No.	11	26	16	20	32
SiO <sub>2</sub>	46.41	48.88	47.53	45.26	49.04
Al <sub>2</sub> O <sub>3</sub>	16.53	17.23	16.56	16.36	16.70
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.03	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.33	0.15	0.12	0.02	0.00
Na <sub>2</sub> O	1.88	3.39	2.24	1.37	2.84
K <sub>2</sub> O	0.02	0.01	0.01	0.01	0.02
Rb <sub>2</sub> O	0.24	0.00	0.07	0.10	0.00
Cs <sub>2</sub> O	33.35	28.31	32.38	35.90	29.91
CaO	0.02	0.02	0.01	0.01	0.03
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.77	98.00	98.94	99.03	98.53
Si	2.113	2.126	2.133	2.113	2.152
Al	0.887	0.883	0.876	0.900	0.863
Fe	0.000	0.000	0.001	0.000	0.000
P	0.013	0.006	0.004	0.001	0.000
Na	0.165	0.286	0.195	0.124	0.242
K	0.001	0.001	0.000	0.001	0.001
Rb	0.007	0.000	0.002	0.003	0.000
Cs	0.647	0.525	0.620	0.714	0.560
Ca	0.001	0.001	0.000	0.001	0.001
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.887	0.883	0.877	0.900	0.863
Σ charges	0.822	0.814	0.816	0.845	0.804
Si/Al	2.383	2.407	2.434	2.348	2.493
CRK	79.8	64.7	76.1	85.2	69.8

11. Cs-enriched bleb.  
 26. Cs-enriched bleb.  
 16. Cs-enriched bleb.  
 20. Cs-rich veinlet.  
 32. Cs-rich veinlet.

#### 4.4 Morrue Mine, Mozambique

The Morrue Mine is located in the Alto Ligonha area, Mozambique. Bulk compositions were determined by Khalili & von Knorring (1976) (No. 69 in Table 2.11).

Sample number: PA-032 (O. von Knorring); 1 thin section, 26 analyses.

##### 4.4.1 Properties, associations and alteration.

A schematic alteration sequence is shown in Figure 4.4-1, as a guide to the following comments.

In hand specimen, the pollucite is white but contains some transparent gemmy eyelets up to 5 mm in size. The pollucite is penetrated by a 2 to 3 mm wide vein of mauve lepidolite. Veins of fine-grained white mica are mostly 1 mm thick, but locally up to 5mm. Fractures in the pollucite contain a buff to white clay mineral, associated with a black dendritic (Mn-bearing?) mineral and red (hematite?) staining.

In thin section, the wide lepidolite vein contains analcime ( $\pm$  quartz, apatite, and spodumene). Lepidolite in the core of this vein is relatively coarse-grained; towards the vein margins finer grains are randomly oriented in the analcime. The smaller veins of fine-grained mica are later; they also contain analcime, and are partially replaced by

spodumene. Adularia occurs along both surfaces of the fine-grained mica veins, and as spherical aggregates completely embedded in the pollucite.

The pollucite contains a three-dimensional rectangular grid of hollow (leaching?) channels, about 10  $\mu\text{m}$  wide, later than the adularia (Fig. 4.4-2). The pollucite immediately adjacent to these channels is enriched in  $\text{Na}^+$ .

#### 4.4.2 Textural relationships

The bulk of the pollucite shows a slight breakdown to a blebby structure. The blebs are rectangular in shape, variable in size from 10 to 300  $\mu\text{m}$ , and internally heterogeneous (Fig. 4.4-3a). The margins of the Na-enriched domains have the highest  $\text{Na}^+$  content; the margins of the Cs-enriched domains have the highest  $\text{Cs}^+$  content (Fig. 4.4-3b). Pollucite inclusions in spodumene are Cs-rich.

Along contacts with most of the alteration products, and also the hollow leaching channels, the pollucite grades quickly towards analcime compositions over a distance of less than 10  $\mu\text{m}$ .

#### 4.4.3 Compositional trends

A plot of Si/Al vs. CRK is given in Figure 4.4-4a. The homogeneous pollucite has Si/Al from 2.42 to 2.53 and CRK

from 86.0 to 88.4; the average and additional representative compositions are given in Table 4.4-1. The slight breakdown of this pollucite is seen in Figure 4.4-4b, Al (+ Fe), and sum of cation charges vs. CRK, as an Al-enrichment trend. An analysis of the Na-enriched pollucite near one of the hollow channels shows a direct loss of  $\text{Cs}^+$  for  $\text{Na}^+$  increase, with no change in Si/Al ratio.

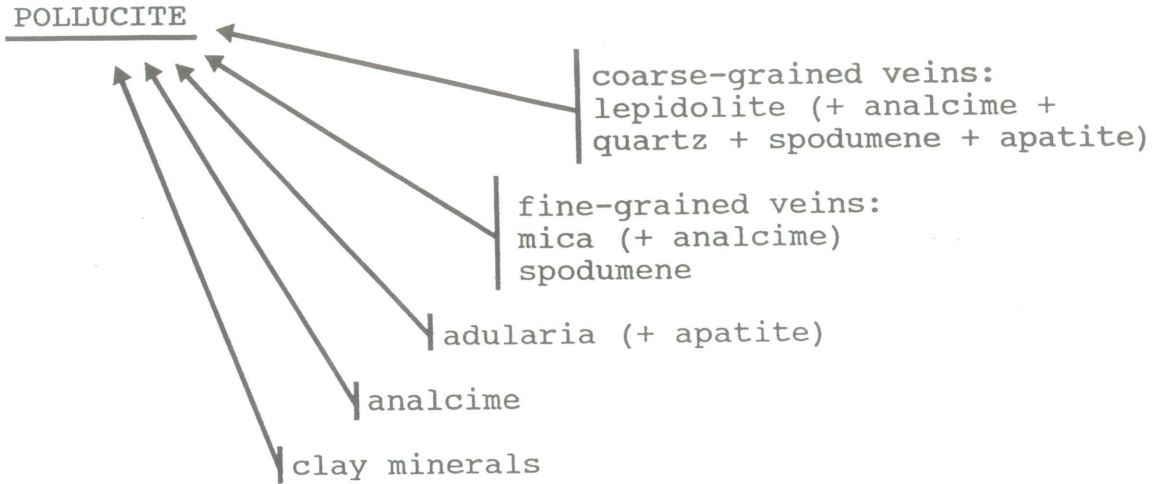


Fig. 4.4-1. Schematic alteration sequence for Morrua Mine, Mozambique.

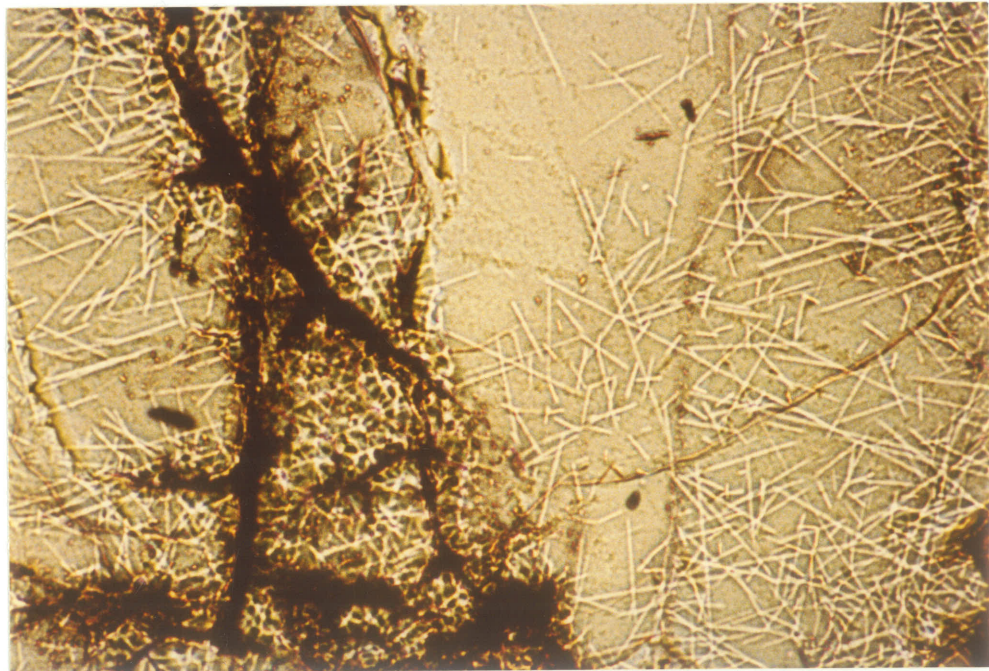


Fig. 4.4-2. Microphotograph of Morrua pollucite. Three-dimensional rectangular grid of thin, hollow channels. They are more abundant near the veins of fine-grained micas.





Fig. 4.4-3. BSE images of Morrua pollucite. a) and b) A slight heterogeneity of "homogeneous" pollucite: incipient development of Na-enriched and Cs-enriched domains.

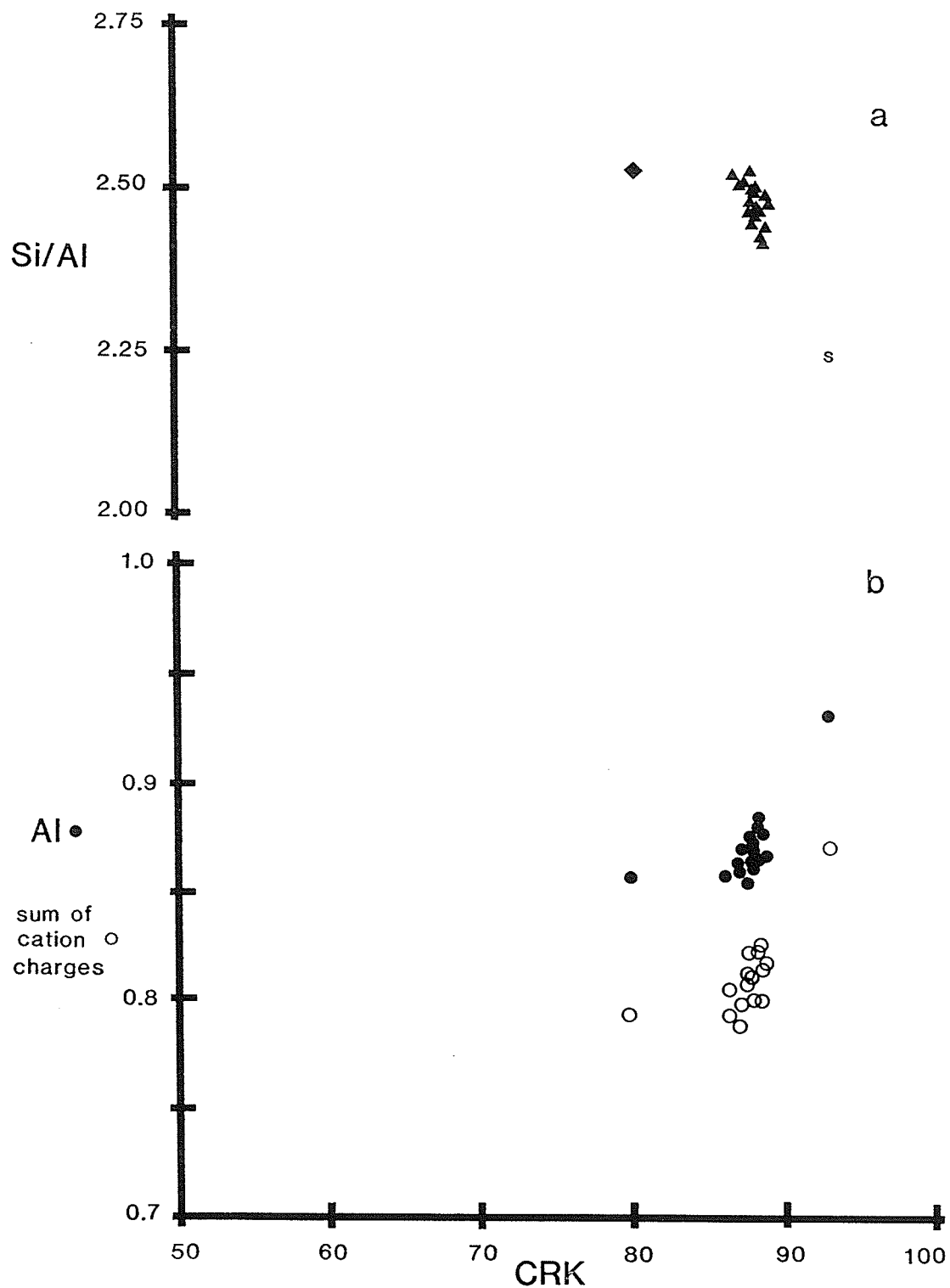


Fig. 4.4-4. Compositional trends for Morruea Mine, Mozambique. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3; inclusion of Cs-rich pollucite in spodumene (S).

Table 4.4-1: Composition of pollucite from Morrua Mine, Mozambique.

No.	20	16	22	21	Average	
SiO <sub>2</sub>	45.37	46.49	48.02	42.90	45.92	(.42)
Al <sub>2</sub> O <sub>3</sub>	15.91	15.66	16.16	16.25	15.78	(.12)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00	
P <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.12	0.00	0.00	
Na <sub>2</sub> O	1.04	1.10	1.81	0.64	1.06	(.05)
K <sub>2</sub> O	0.06	0.06	0.02	0.07	0.05	(.02)
Rb <sub>2</sub> O	0.34	0.36	0.22	0.14	0.34	(.02)
Cs <sub>2</sub> O	35.28	34.59	32.46	38.49	34.78	(.18)
CaO	0.06	0.05	0.06	0.03	0.05	(.01)
MgO	0.00	0.01	0.00	0.00	0.00	
H <sub>2</sub> O	----	----	----	----	----	
sum	98.06	98.31	98.75	98.53	97.97	(.35)
Si	2.133	2.157	2.160	2.085	2.145	(.007)
Al	0.881	0.856	0.856	0.930	0.869	(.008)
Fe	0.000	0.000	0.000	0.000	0.000	
P	0.000	0.000	0.000	0.000	0.000	
Na	0.094	0.099	0.158	0.060	0.097	(.004)
K	0.004	0.004	0.001	0.004	0.003	(.001)
Rb	0.010	0.011	0.006	0.004	0.010	(.001)
Cs	0.707	0.684	0.623	0.798	0.693	(.011)
Ca	0.003	0.002	0.003	0.002	0.003	(.001)
Mg	0.000	0.000	0.000	0.000	0.000	
Al + Fe	0.881	0.856	0.856	0.930	0.869	(.008)
Σ charges	0.821	0.804	0.793	0.870	0.809	(.009)
Si/Al	2.421	2.521	2.522	2.241	2.468	(.030)
CRK	88.1	87.3	79.7	92.8	87.7	(0.6)

20. Homogeneous pollucite grain, low Si/Al.

16. Homogeneous pollucite grain, high Si/Al.

22. Na-enriched pollucite near leaching channel.

21. Cs-rich pollucite inclusion in spodumene vein.

Average of homogeneous pollucite (N = 22).

#### 4.5 Muiane Mine, Mozambique

The Muiane mine is located in the Alto Ligonha area, Mozambique. Bulk compositions were determined by Khalili & von Knorring (1976) (No. 67 in Table 2.11).

Sample number: PA-035 (O. von Knorring); 1 thin section, 28 analyses.

##### 4.5.1 Properties, associations and alteration

In hand specimen, the pollucite is white in colour and resinous in lustre, and contains some transparent vitreous grains 2 to 3 mm in size, and a crystal of spodumene 1 cm in size. The pollucite is cut by a pink to grey veins of lepidolite 2 to 3 mm wide. Fracture surfaces of the pollucite are lightly coated with white to buff clays associated with a slight red staining (hematite?) and small amounts of a black dendritic (Mn-bearing?) mineral.

In thin section, the crystal of spodumene is fresh, contains a small amount of quartz, and does not appear to replace the pollucite. The pollucite contains scattered quartz grains a maximum of 1 mm in size. The pink veins contain lepidolite + quartz ( $\pm$  apatite); grain size decreases from core to margin where apatite is most abundant. The width of the vein is constant, it appears to be fracture-filling, and it terminates by pinching out

within the pollucite.

#### 4.5.2 Textural relationships

Local areas of pollucite up to 5 mm in size and free from inclusion consist of compositionally and texturally homogeneous pollucite. One area of homogeneous pollucite has a distinct composition. Much of the pollucite, however, has broken down to a blebby structure; some areas of the pollucite consist entirely of Cs-rich blebby domains. The domains are 25 to 100  $\mu\text{m}$  in size, and rounded to hourglass-like and elongate in shape. They are internally heterogeneous, containing zones parallel to the margins. Complex replacement of some zones by others, usually but not necessarily more Cs-rich, has produced a raft texture in what seems to be remnants of the homogeneous pollucite (Fig. 4.5-1a). The Cs-rich veinlets (+ quartz) are most abundant near the homogeneous pollucite, but also cross-cut the blebby structure. They locally show Na-enrichment of the adjacent pollucite at their margins (Fig. 4.5-1b).

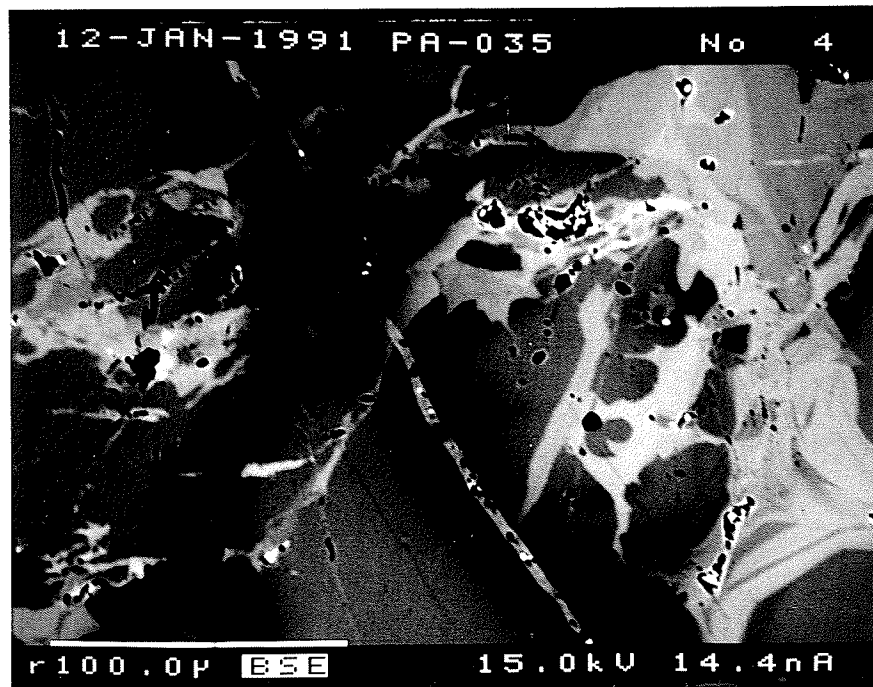


Fig. 4.5-1. BSE images of Muiane pollucite. a) Blebby mosaic showing internal zonation of blebs and association with quartz grains (darkest). b) Na-rich rims of quartz-containing Cs-rich veinlet, cutting blebs and homogeneous pollucite, but all in turn are cut by Na-enriched pollucite along a fracture.

#### 4.5.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Fig. 4.5-2a. The homogeneous pollucite has Si/Al from 2.53 to 2.58 and CRK from 78.2 to 80.0; the average and additional representative compositions are given in Tables 4.5-1 and 2. The Cs-enriched blebs have Si/Al as low as 2.41 and CRK as high as 85.5. Two adjacent blebs with the same CRK but different Si/Al were found to have noticeably different brightness in a back-scattered electron (BSE) image (see Table). Similarly, the internal zonation of the Cs-enriched blebs seems to consist of slight variations in Si/Al with constant CRK. Analysis of Na-rich pollucite adjacent to a Cs-rich veinlet gave Si/Al 2.52 and CRK 76.0. Cs-rich veinlets have Si/Al as low as 2.30 and CRK as high as 90.3.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 4.5-2b. The sum of cation charges follows a trend of Al increasing with CRK.

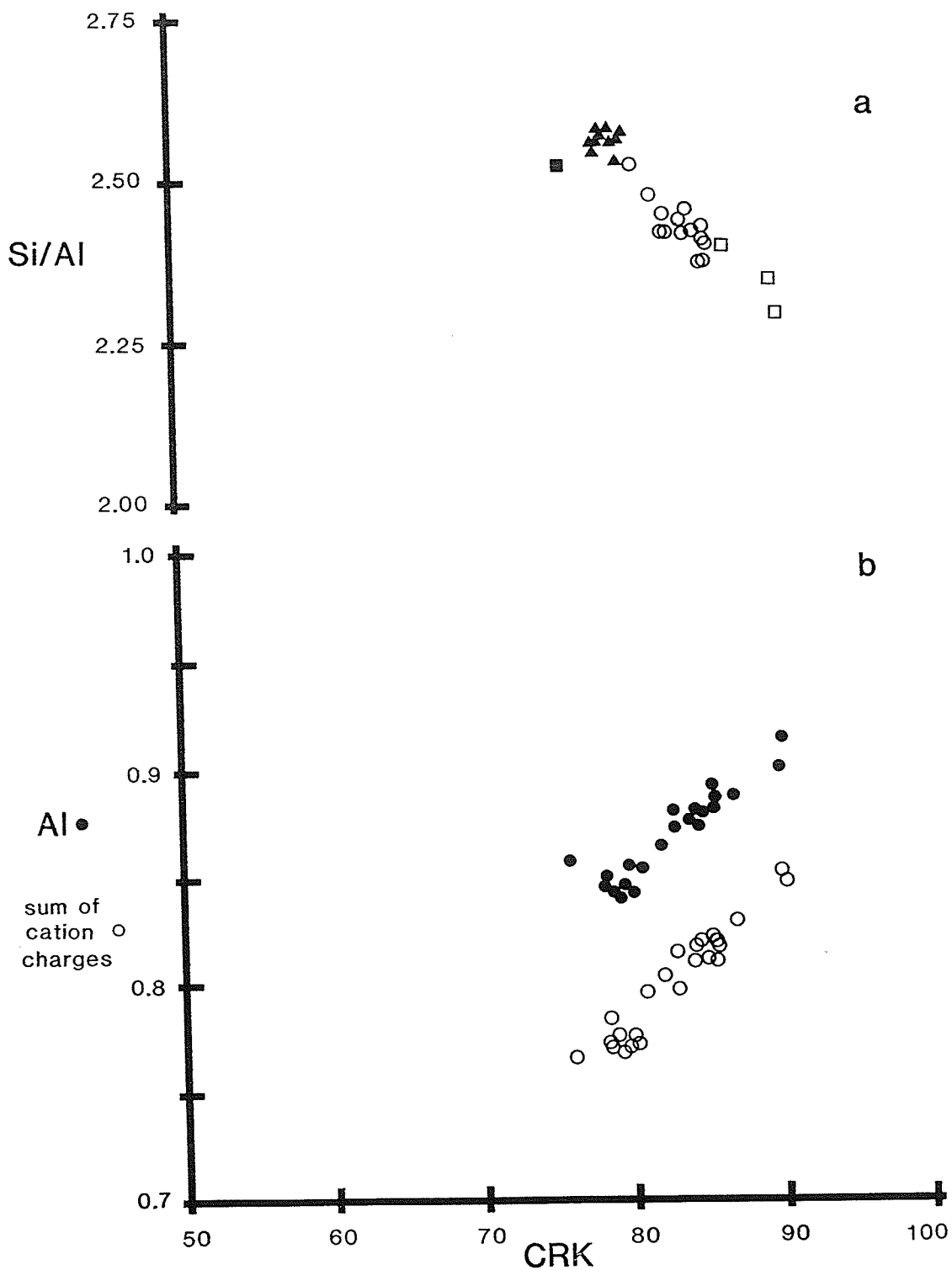


Fig. 4.5-2 Compositional trends for Muiane Mine, Mozambique. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.



Table 4.5-1: Composition of pollucite from Muiane Mine, Mozambique.

No.	23	27	5	1	28
SiO <sub>2</sub>	47.09	47.53	48.56	48.81	49.09
Al <sub>2</sub> O <sub>3</sub>	16.32	16.25	16.06	16.06	16.54
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.00	0.00	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.24	0.26	0.25	0.21	0.23
Na <sub>2</sub> O	1.56	1.66	1.79	1.86	2.15
K <sub>2</sub> O	0.01	0.01	0.01	0.01	0.01
Rb <sub>2</sub> O	0.17	0.18	0.24	0.16	0.04
Cs <sub>2</sub> O	33.91	34.01	32.09	31.78	30.95
CaO	0.00	0.00	0.00	0.00	0.01
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	99.30	99.90	99.00	98.91	99.02
Si	2.135	2.140	2.163	2.168	2.155
Al	0.873	0.863	0.843	0.841	0.856
Fe	0.000	0.000	0.000	0.000	0.000
P	0.009	0.010	0.010	0.008	0.009
Na	0.137	0.144	0.155	0.161	0.183
K	0.000	0.001	0.000	0.000	0.001
Rb	0.005	0.005	0.007	0.005	0.001
Cs	0.656	0.653	0.610	0.602	0.579
Ca	0.000	0.000	0.000	0.000	0.001
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.873	0.863	0.843	0.841	0.856
Σ charges	0.797	0.803	0.771	0.767	0.766
Si/Al	2.447	2.480	2.567	2.579	2.518
CRK	82.9	82.0	80.0	79.1	76.0

23. Homogeneous area of pollucite with high CRK.

27. Homogeneous area of pollucite with high CRK.

5. Homogeneous pollucite grain.

1. Homogeneous pollucite grain.

28. Na-rich pollucite adjacent to Cs-rich veinlet.

Table 4.5-2: Composition of pollucite from Muiane Mine, Mozambique.

No.	7	14	15	8	Average
SiO <sub>2</sub>	45.95	45.79	46.18	44.24	48.65 (.21)
Al <sub>2</sub> O <sub>3</sub>	16.19	16.35	16.13	16.36	16.16 (.06)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.02	0.00	0.02	0.00
P <sub>2</sub> O <sub>5</sub>	0.24	0.23	0.00	0.00	0.24 (.03)
Na <sub>2</sub> O	1.31	1.33	1.33	0.89	1.89 (.07)
K <sub>2</sub> O	0.00	0.00	0.00	0.01	0.00
Rb <sub>2</sub> O	0.25	0.16	0.14	0.22	0.16 (.07)
Cs <sub>2</sub> O	34.72	35.22	35.33	37.48	32.04 (.20)
CaO	0.00	0.01	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.68	99.09	99.12	99.20	99.15 (.29)
Si	2.123	2.116	2.135	2.101	2.161 (.004)
Al	0.882	0.891	0.879	0.915	0.846 (.004)
Fe	0.000	0.001	0.000	0.001	0.000
P	0.010	0.009	0.000	0.000	0.009 (.001)
Na	0.118	0.119	0.119	0.082	0.162 (.006)
K	0.000	0.000	0.000	0.000	0.000
Rb	0.008	0.005	0.004	0.007	0.005 (.002)
Cs	0.685	0.694	0.696	0.759	0.607 (.005)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.882	0.892	0.879	0.916	0.846 (.004)
Σ charges	0.810	0.818	0.819	0.847	0.773 (.005)
Si/Al	2.408	2.375	2.428	2.295	2.555 (.016)
CRK	85.5	85.5	85.5	90.3	79.0 (0.6)

7. Cs-enriched bleb.

14. Cs-enriched bleb, low Si/Al but same CRK as 15.

15. Cs-enriched bleb, high Si/Al, but higher BSE level.

8. Cs-rich veinlet.

Average of homogeneous pollucite (N = 10).

#### 4.6 Neinein, Namibia

The Neinein Mine is located about 30 km south of Uis, in the Omaruru district, Namibia.

Sample number: PA-124 (G. Friedrich); 1 thin section, 26 analyses.

##### 4.6.1 Properties, associations and alteration

In hand specimen, the pollucite is white in colour, and dull in lustre. It contains some scattered white to slightly green flakes of mica (lepidolite?) 1 mm in size. Fracture surfaces are coated with buff clays.

In thin section, the pollucite contains some scattered quartz grains up to 0.5 mm in size. Fine trains of microinclusions are present, oriented perpendicular to cross-cutting veins of fine-grained mica. The mica veins, up to 1 cm long and less than 0.5 mm wide, are partially replaced by spodumene.

##### 4.6.2 Textural relationships

About half of the pollucite is compositionally homogeneous. Elongate Cs-enriched blebs associated with quartz are abundant (Fig. 4.6-1). The blebs are up to 50  $\mu\text{m}$  wide and 300  $\mu\text{m}$  in length. Some Cs-rich veinlets + quartz

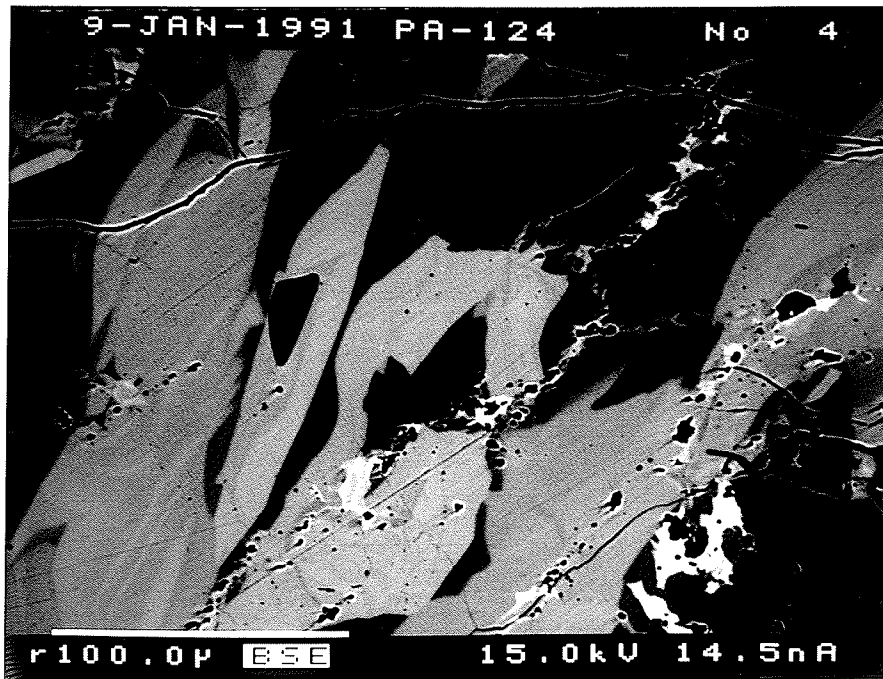


Fig. 4.6-1. BSE image of Neinein pollucite. Elongated Cs-enriched blebs in an area of homogeneous pollucite, cross-cut by poorly developed quartz-containing Cs-rich veinlets.

crosscut the blebby structure. Mica veins crosscut the heterogeneous pollucite.

#### 4.6.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 4.6-2a. The homogeneous pollucite has Si/Al from 2.51 to 2.58 and CRK from 73.0 to 74.2; the average and additional representative compositions are given in Table 4.6-1. The Cs-enriched blebs have Si/Al as low as 2.35 and CRK as high as 78.3. The Cs-rich veinlets have Si/Al as low as 2.33 and CRK as high as 79.5.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 4.6-2b. The sum of cation charges follows a trend of Al increasing with CRK.

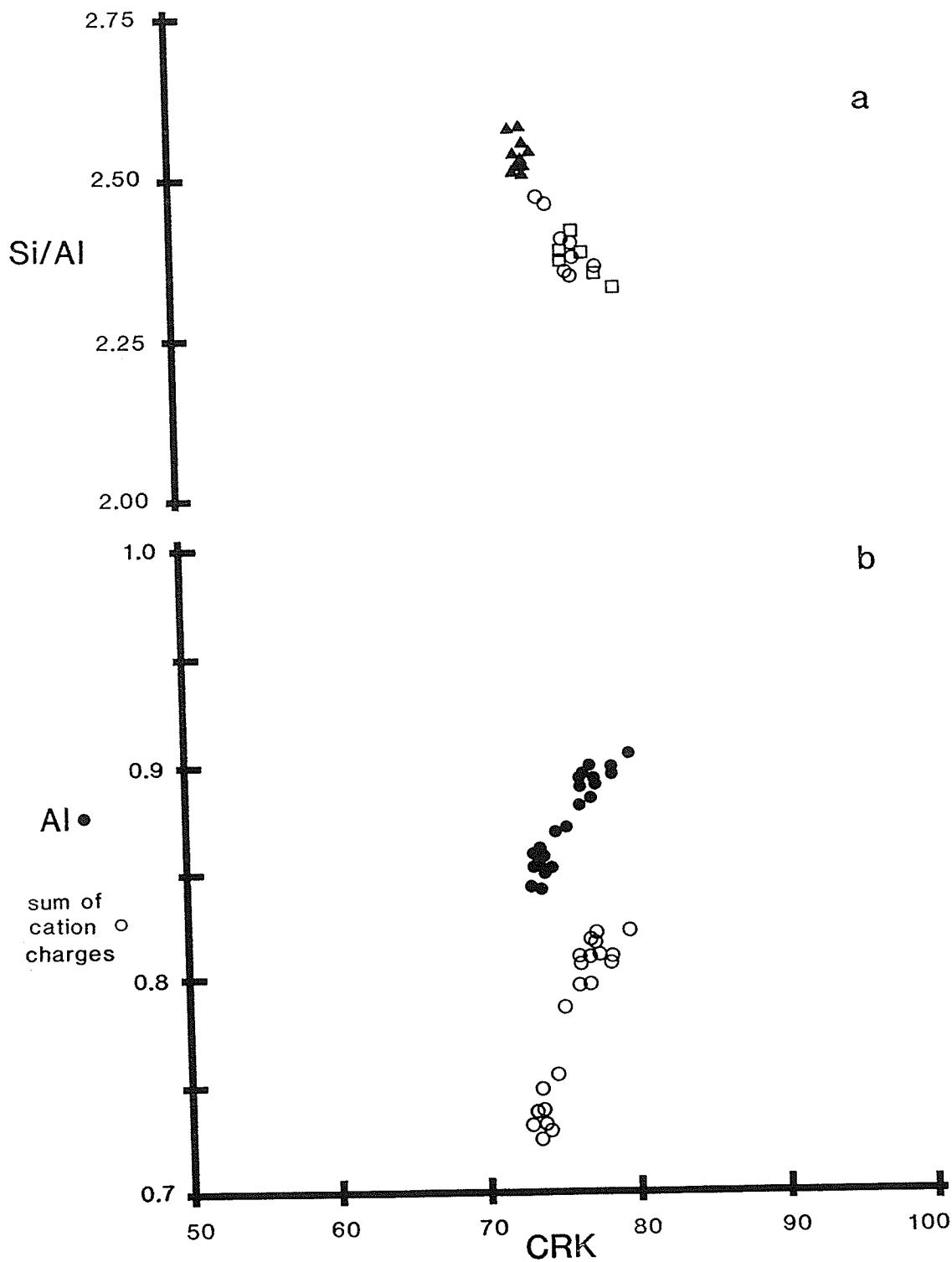


Fig. 4.6-2. Compositional trends for Neinein Mine, Namibia.  
 a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 4.6-1: Composition of pollucite from Neinein Mine, Namibia.

No.	18	23	8	26	Average
SiO <sub>2</sub>	50.24	46.61	46.41	46.00	50.38 (.47)
Al <sub>2</sub> O <sub>3</sub>	16.91	16.84	16.71	16.77	16.86 (.07)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.01	0.00	0.00	0.02 (.01)
P <sub>2</sub> O <sub>5</sub>	0.33	0.27	0.30	0.14	0.31 (.01)
Na <sub>2</sub> O	2.28	2.11	1.98	1.88	2.32 (.04)
K <sub>2</sub> O	0.00	0.01	0.02	0.00	0.01
Rb <sub>2</sub> O	0.00	0.00	0.00	0.00	0.00
Cs <sub>2</sub> O	29.35	32.22	32.62	33.39	29.43 (.15)
CaO	0.01	0.01	0.01	0.01	0.01
MgO	0.01	0.00	0.00	0.01	0.01
H <sub>2</sub> O	----	----	----	----	----
sum	99.15	98.08	98.06	98.19	99.33 (.49)
Si	2.160	2.110	2.111	2.109	2.162 (.007)
Al	0.857	0.899	0.895	0.906	0.853 (.005)
Fe	0.000	0.000	0.000	0.000	0.001
P	0.012	0.011	0.012	0.005	0.011 (.002)
Na	0.190	0.186	0.175	0.167	0.193 (.004)
K	0.000	0.000	0.001	0.000	0.001
Rb	0.000	0.000	0.000	0.000	0.000
Cs	0.538	0.622	0.633	0.652	0.539 (.005)
Ca	0.001	0.001	0.001	0.001	0.001
Mg	0.001	0.000	0.000	0.000	0.001
Al + Fe	0.857	0.899	0.895	0.906	0.853 (.005)
Σ charges	0.732	0.810	0.811	0.822	0.735 (.006)
Si/Al	2.521	2.346	2.358	2.327	2.535 (.027)
CRK	73.7	76.9	78.3	79.5	73.5 (0.4)

18. Homogeneous pollucite grain.

23. Cs-enriched bleb.

8. Cs-enriched bleb.

26. Cs-rich veinlet.

Average of homogeneous pollucite (N = 10).

## CHAPTER 5

## POLLUCITE FROM THE UNITED STATES

## 5.1 Brown Derby # 1, Colorado

Sample number: PA-080 (P. Cerny); 4 thin sections, 1 grain mount, 15 analyses.

## 5.1.1 Properties, associations and alteration

A schematic alteration sequence is shown in Figure. 5.1-1, as a guide to the following comments. In hand specimen, the pollucite is white to colourless. It is crosscut by veins (1 to 2 mm wide) of coarse-grained pink to grey mica (lepidolite?). Sets of smaller (at most 1 mm wide) subparallel veins containing finer-grained mica are spaced 1 to 3 mm apart and braided around eyelets of pollucite. White to buff clays have replaced the pollucite, locally entirely, leaving the early mica veins intact.

In thin section, the pollucite contains scattered grains (up to 1 mm) of mica. An irregular vein of microcline seems to be earlier than the coarse-grained mica veins. All the mica veins contain analcime ( $\pm$  quartz). Surfaces of the veins are overgrown with adularia ( $\pm$  albite  $\pm$  quartz). Clay minerals replace pollucite along fractures and mineral contacts.



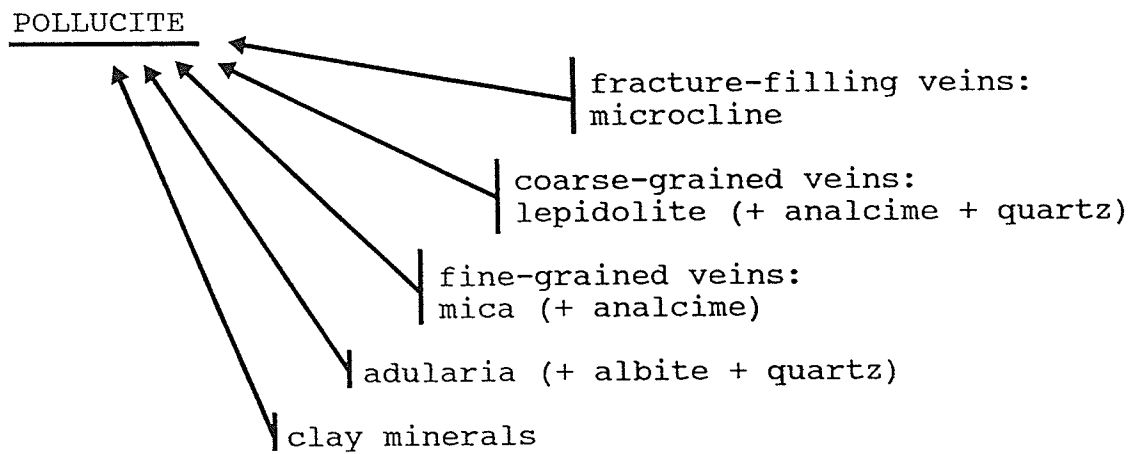


Fig. 5.1-1. Schematic alteration sequence for Brown Derby # 1, Colorado.

### 5.1.2 Textural relationships

The pollucite is mostly homogeneous. Within 100 to 300  $\mu\text{m}$  from the alteration minerals, however, there are steep gradations towards Na-enriched compositions. Some Cs-enriched blebs are present, as are Cs-rich veinlets (+ quartz).

### 5.1.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.1-2a. The homogeneous pollucite has Si/Al from 2.52 to 2.59 and CRK from 77.4 to 79.0; the average and additional representative compositions are given in Table 5.1-1. Qualitative analysis of the Na-enriched areas indicates CRK as low as 43 and Si/Al within the same range as the homogeneous pollucite. The Cs-enriched blebs have Si/Al as low as 2.40 and CRK as high as 83.2. The Cs-rich veinlets have Si/Al as low as 2.32 and CRK as high as 87.8.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.1-2b. For the Na-enriched pollucite (CRK less than 70) the sum of cation charges decreases with constant Al, due to sample burning. For CRK greater than 70, the sum of cation charges follows a trend of Al increasing with CRK.

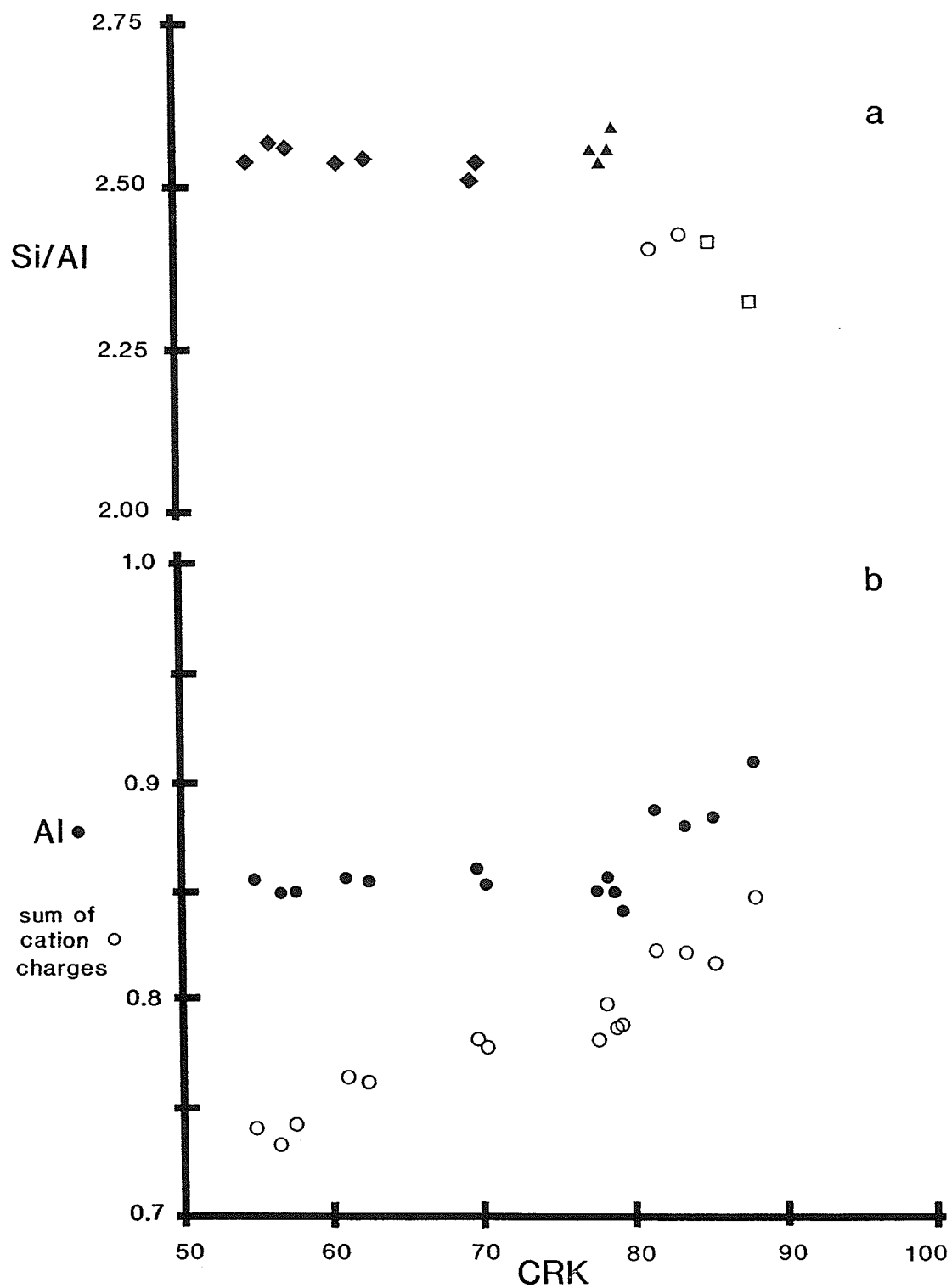


Fig. 5.1-2. Compositional trends for Brown Derby # 1, Colorado. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 5.1-1: Composition of pollucite from Brown Derby, Colorado.

No.	2	9	13	14	Average
SiO <sub>2</sub>	47.94	50.74	46.01	44.35	47.73 (.24)
Al <sub>2</sub> O <sub>3</sub>	15.74	16.99	16.29	16.24	15.88 (.11)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.01	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.10	0.10	0.10	0.07	0.08 (.02)
Na <sub>2</sub> O	1.89	3.58	1.72	1.12	1.95 (.06)
K <sub>2</sub> O	0.00	0.03	0.00	0.00	0.00
Rb <sub>2</sub> O	0.27	0.14	0.14	0.23	0.24 (.04)
Cs <sub>2</sub> O	31.84	25.14	33.64	36.41	31.58 (.32)
CaO	0.01	0.03	0.00	0.01	0.00
MgO	0.00	0.01	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.79	96.75	97.90	98.43	97.47 (.25)
Si	2.169	2.164	2.125	2.103	2.163 (.005)
Al	0.839	0.854	0.886	0.908	0.848 (.007)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.004	0.004	0.004	0.003	0.003
Na	0.166	0.296	0.154	0.104	0.171 (.005)
K	0.000	0.002	0.000	0.000	0.000
Rb	0.008	0.004	0.004	0.007	0.007 (.001)
Cs	0.615	0.457	0.663	0.737	0.610 (.007)
Ca	0.000	0.002	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.839	0.854	0.886	0.908	0.848 (.007)
Σ charges	0.788	0.763	0.821	0.847	0.789 (.007)
Si/Al	2.585	2.535	2.397	2.316	2.551 (.025)
CRK	79.0	60.8	81.3	87.8	78.3 (0.7)

2. Homogeneous.

9. Na-enriched.

13. Cs-enriched bleb.

14. Cs-rich veinlet.

Average of homogeneous pollucite (N = 4).

## 5.2 Tin Mountain, South Dakota

The Tin Mountain pegmatite is located in the central part of Sec. 35, Twp. 3 S, Rge. 3 E, Custer County, South Dakota. It is one of the pegmatites around the Harney peak granite, described by Schwartz (1930) and more recently by Walker (1986). Pollucite is found in the core zone with quartz, spodumene and lepidolite, and in the adjacent albite, quartz and spodumene zone. At least four separate masses have been found, and about 45000 kg mined. A large amount of this had 32 wt.%  $\text{Cs}_2\text{O}$ , but some was so altered as to contain only 1 wt.%  $\text{Cs}_2\text{O}$  (Kennedy 1938).

Bulk compositions were determined by Connolly & O'Hara (1929) (quoted in Schwartz 1930; No. 4 in Table 2.1a), Wells & Stevens (1937) (No. 12 in Table 2.1b) and Roberts & Rapp (1965) (No. 46 in Table 2.1h). Ahrens (1947) determined minor elements (as wt.%): (0.006)  $\text{Li}_2\text{O}$ , (0.19)  $\text{K}_2\text{O}$ , (0.25)  $\text{Rb}_2\text{O}$ , (0.0019)  $\text{Tl}_2\text{O}$  and (0.0005)  $\text{Ga}_2\text{O}_3$ .

Sample numbers: PA-008, PA-022, PA-025 and PA-034 (D. Garske), PA-127 (P. Cerny), and PA-136 and PA-137 (M. Wise, U.S.N.M. 97634 and 95890); 6 thin sections, 40 analyses.

### 5.2.1 Properties, associations and alteration

A schematic alteration sequence is shown in Figure 5.2-1, as a guide to the following comments. In hand

specimen, the pollucite is white in colour, and has a greasy to earthy lustre. It contains some spodumene grains up to 3 cm in size. Veins of coarse-grained lepidolite up to 5 mm wide cross-cut the pollucite, as well as a few grey, fine-grained sub-parallel veins up to 1 mm in width. Most of the pollucite is altered to buff clays, along fractures and penetrating throughout. A black dendritic (Mn-bearing?) mineral occurs in small amounts along fracture surfaces. Locally the pollucite is completely replaced by clay minerals (distinct in terms of color: white to buff, green, and orange-red) leaving the early veining intact.

In thin section, the pollucite contains scattered grains of muscovite, quartz and some albite, up to 1 mm in size. Abundant micro-inclusions are present throughout the pollucite. The wide lepidolite veins contain quartz. The fine veins are mostly mica, locally replaced by spodumene. Adularia is present along surfaces of the veins and as aggregates embedded in the pollucite. Clay minerals replace the pollucite completely in some areas.

#### 5.2.2 Textural relationships

Only locally are areas of the pollucite homogeneous. Most of the pollucite has considerable enrichment in  $\text{Na}^+$ , with compositions grading well towards analcime (Fig. 5.2-

2a). Much has broken down to a blebby aggregate consisting mostly of Cs-enriched domains, and some Na-enriched domains or remnants of the homogeneous pollucite (Fig. 5.2-2b, c and d). Cs-rich veinlets containing quartz (+ spodumene in some cases) are present but not particularly abundant (Fig. 5.2-2e and f) . A single Na- and Si-rich veinlet crosscuts a Cs-rich veinlet (Fig. 5.2-2g). Quartz within the coarse lepidolite vein contains round inclusions of pollucite.

### 5.2.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.2-3a. The homogeneous pollucite has Si/Al from 2.31 to 2.48 and CRK from 74.8 to 81.4; the average and additional representative compositions are given in Tables 5.2-1 and 2. Na-enriched compositions have CRK extending to near end-member analcime, with the same Si/Al as the homogeneous pollucite. Na-enriched blebs have Si/Al and CRK within the same range as the homogeneous pollucite. Cs-enriched blebs have Si/Al as low as 2.27 and CRK as high as 86.4. Cs-rich veinlets have Si/Al as low as 2.24 and CRK as high as 90.8. The unique Na, Si-rich veinlet has Si/Al as high as 2.68 and CRK as low as 76.3. A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.2-3b. The sum of cation charges follows a trend of increasing Al with CRK for CRK greater than 70, but constant with CRK less than 70.

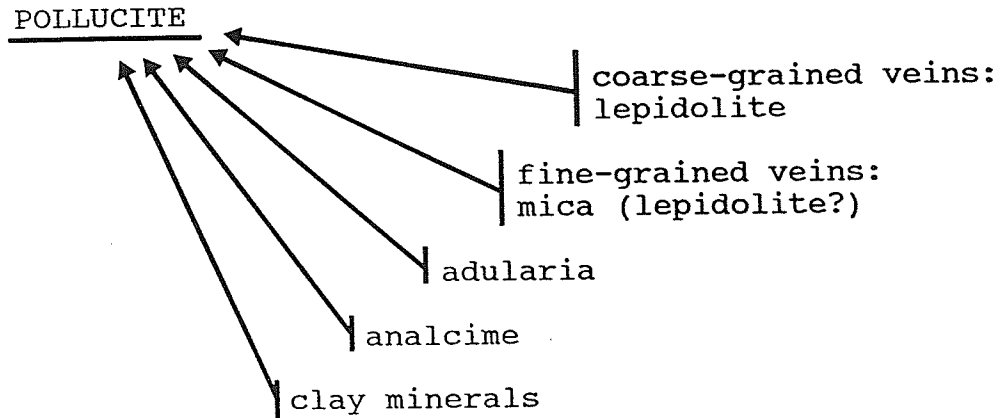


Fig. 5.2-1. Schematic alteration sequence for Tin Mountain, South Dakota.

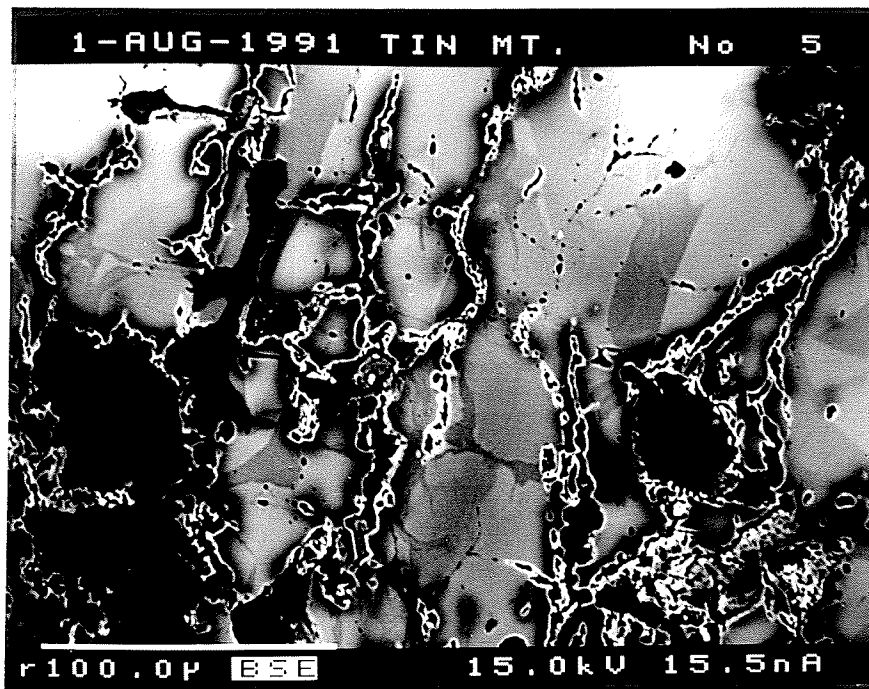


Fig. 5.2-2. BSE images of Tin Mt. pollucite. a) An area of strong "analcimization" associated with clay minerals. The highest Na contents are near veinlets of relatively Cs-rich compositions which contain trains of quartz grains.



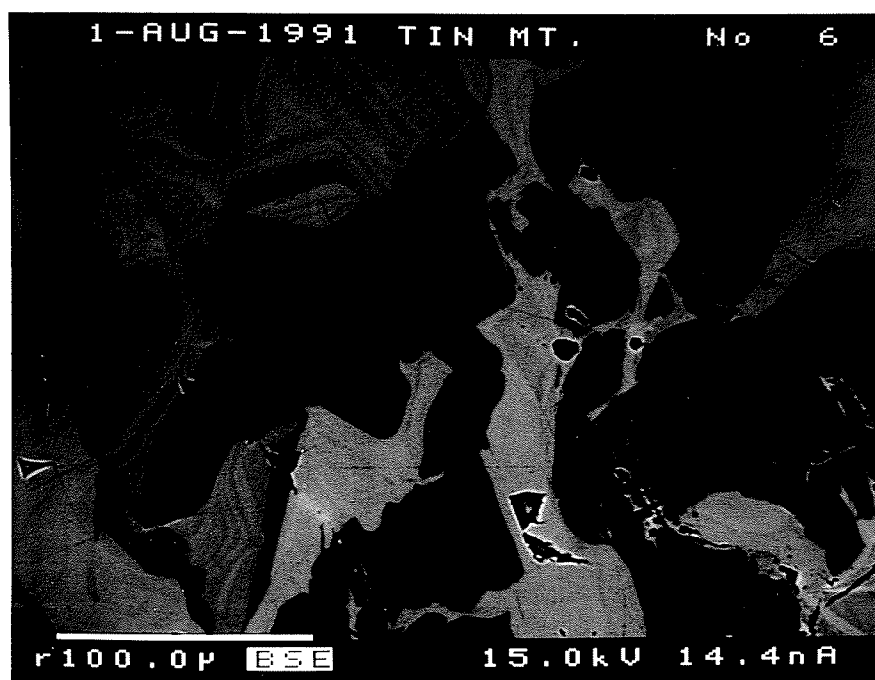
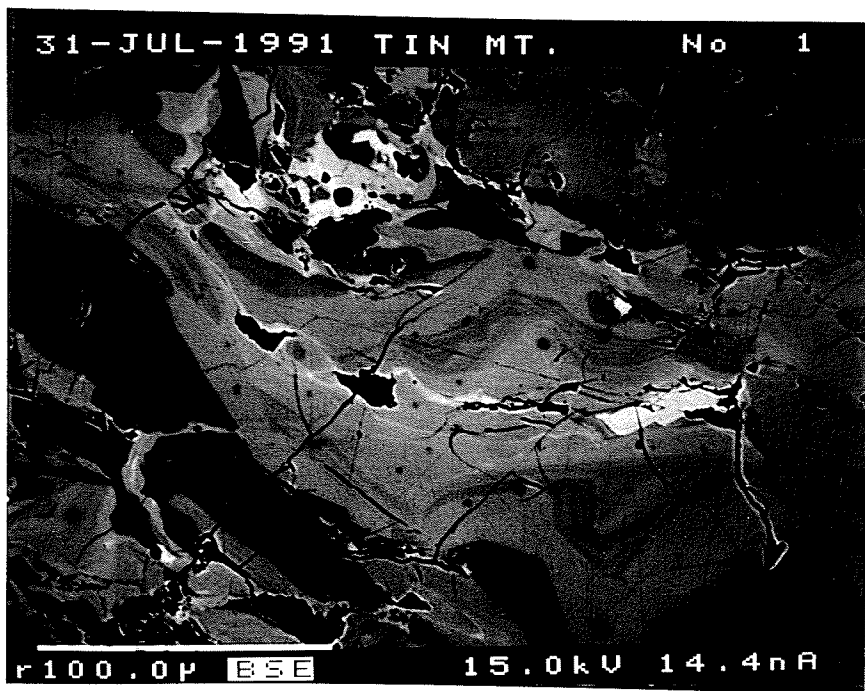


Fig. 5.2-2. b) and c) Internally zoned, blebby mosaics.  
Dark grains are quartz.

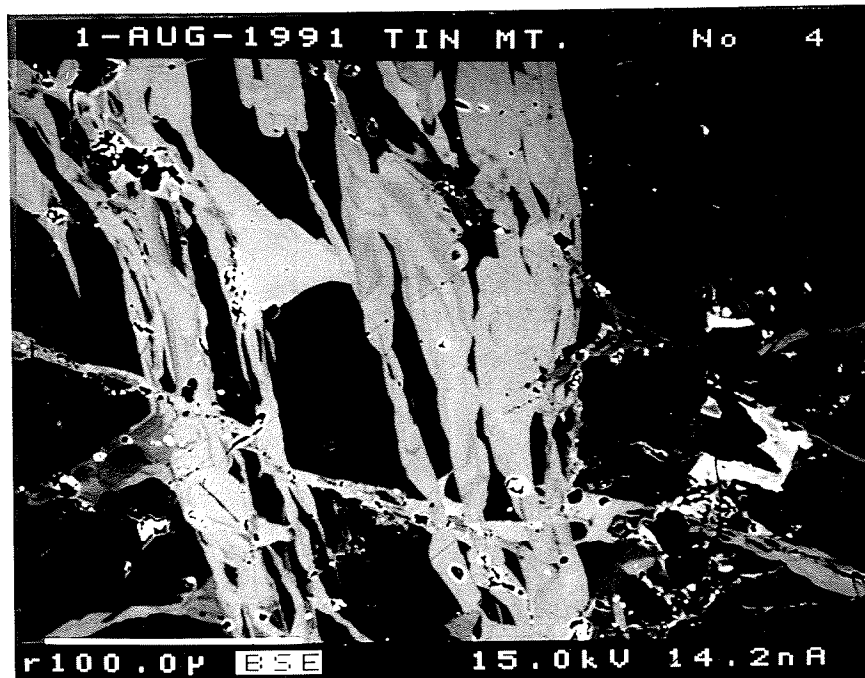
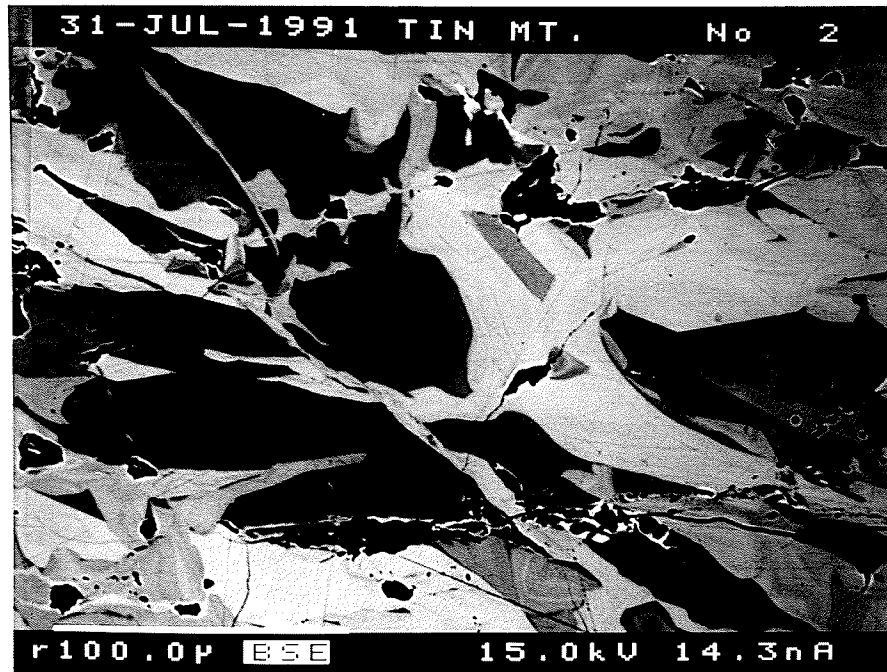


Fig. 5.2-2. d) Remnants (dark) of homogeneous pollucite within domains of Cs-enriched blebs. e) Multiple generations of Cs-rich veinlets crosscut a homogeneous area.

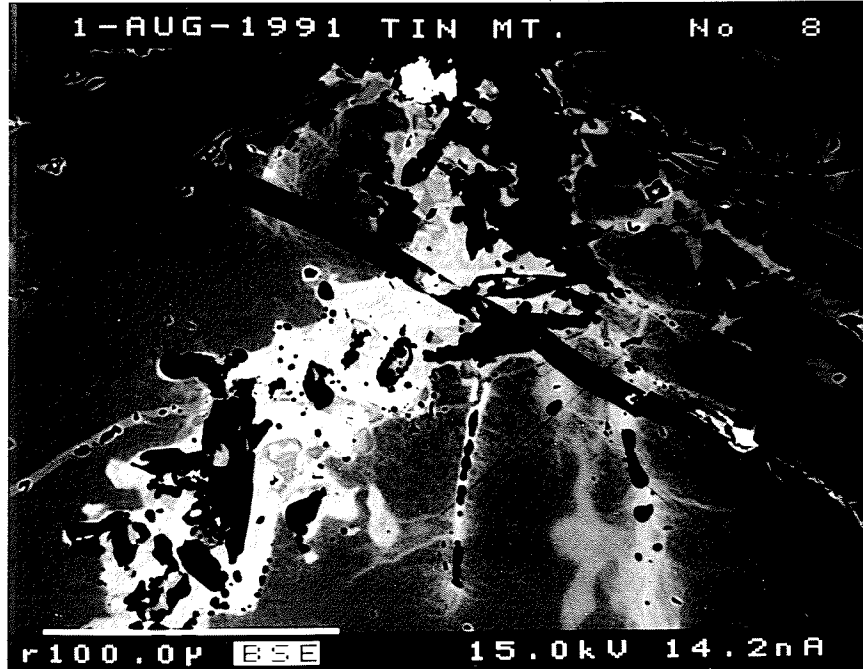
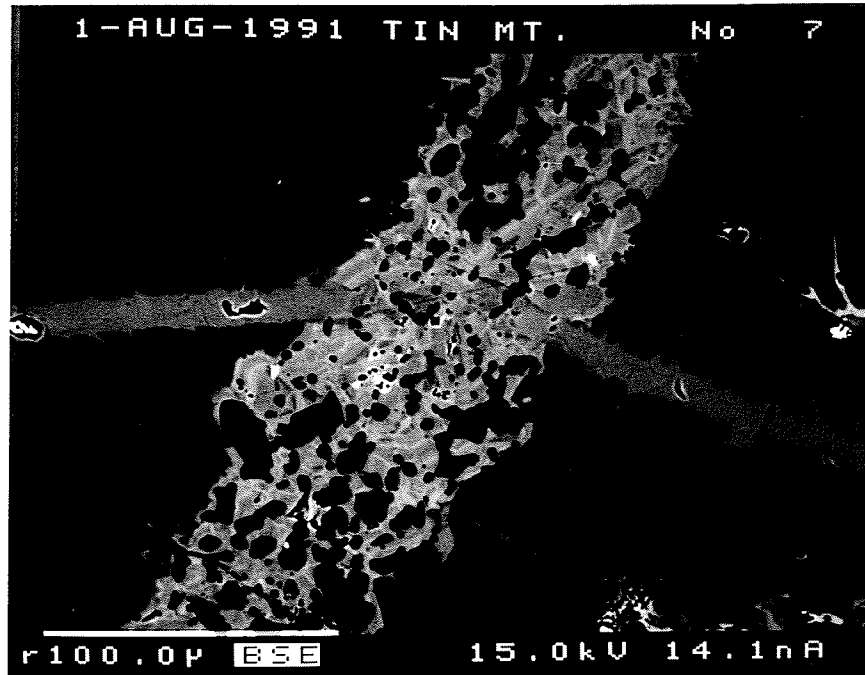


Fig. 5.2-2. A veinlet of Cs-rich pollucite containing grains of spodumene and quartz in an area of homogeneous pollucite. f) A later(?) veinlet of Cs-rich pollucite, fracture-filling. g) Further along this veinlet, a later veinlet of Na- and Si-rich pollucite.

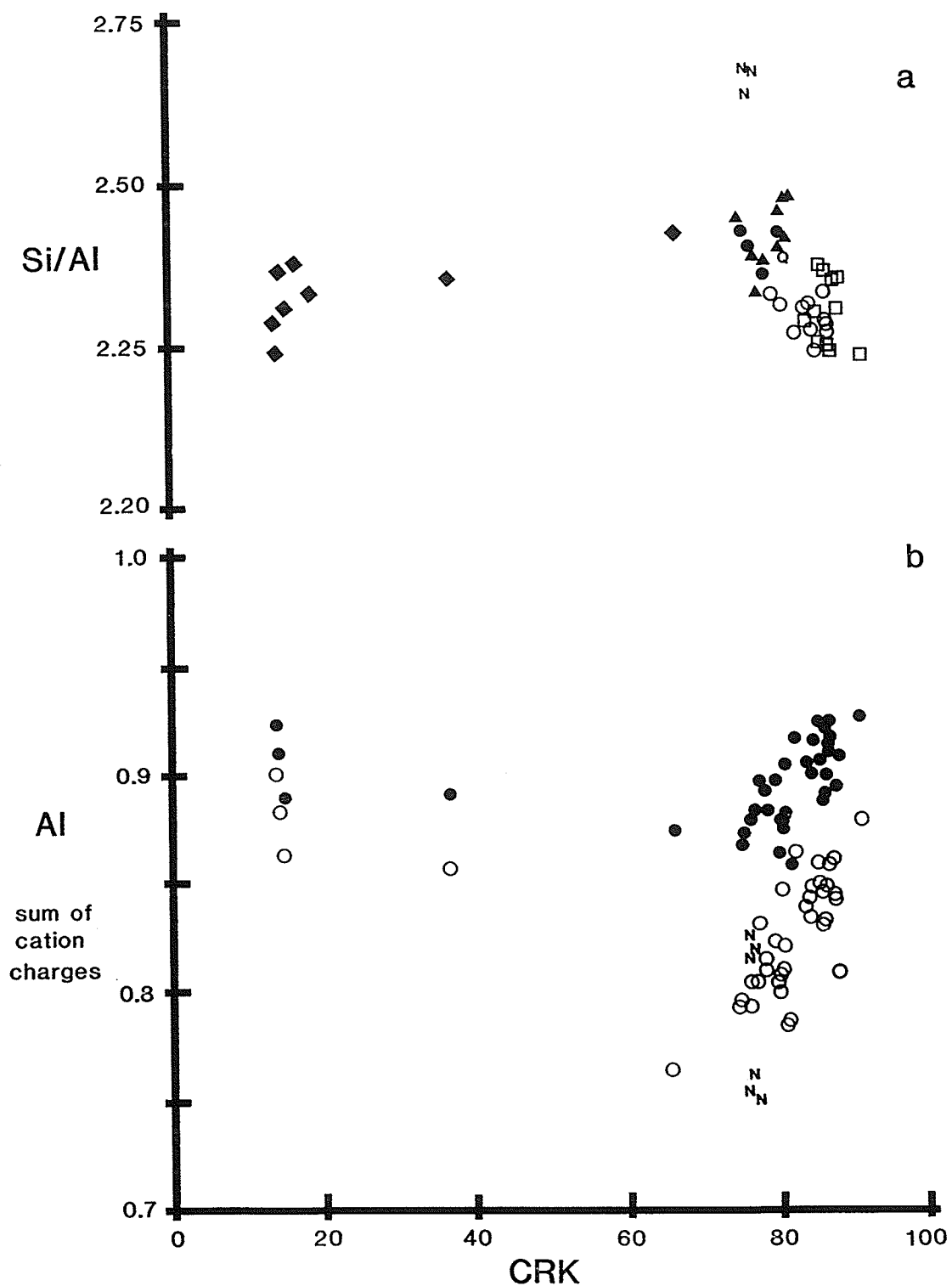


Fig. 5.2-3. Compositional trends for Tin Mountain, South Dakota. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Pollucite inclusion in quartz (Q). Na- and Si-rich veinlet (N).

Table 5.2-1: Composition of pollucite from Tin Mt., South Dakota.

No.	46	16	31	33	26
SiO <sub>2</sub>	54.25	47.43	46.63	49.21	45.75
Al <sub>2</sub> O <sub>3</sub>	19.35	16.44	15.95	15.60	16.45
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.04	0.30	0.36	0.00	0.14
Na <sub>2</sub> O	9.70	2.31	1.66	2.01	2.01
K <sub>2</sub> O	0.06	0.02	0.01	0.01	0.01
Rb <sub>2</sub> O	0.00	0.00	0.72	0.67	0.00
Cs <sub>2</sub> O	1.27	31.00	31.76	29.50	32.39
CaO	0.01	0.00	0.00	0.00	0.01
MgO	----	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	90.96	97.49	97.09	97.02	96.76
Si	2.115	2.134	2.139	2.196	2.116
Al	0.889	0.871	0.862	0.821	0.896
Fe	0.001	0.000	0.000	0.000	0.000
P	0.001	0.011	0.014	0.000	0.006
Na	0.733	0.201	0.147	0.174	0.180
K	0.003	0.001	0.001	0.000	0.000
Rb	0.000	0.000	0.022	0.020	0.000
Cs	0.125	0.595	0.621	0.561	0.638
Ca	0.001	0.000	0.000	0.000	0.000
Mg	-----	0.000	0.000	0.000	0.000
Al + Fe	0.890	0.871	0.862	0.821	0.896
Σ charges	0.864	0.797	0.790	0.754	0.818
Si/Al	2.379	2.448	2.481	2.675	2.361
CRK	14.9	74.8	81.4	76.9	78.0

46. Na-enriched.  
 16. Homogeneous  
 31. Homogeneous.  
 33. Na, Si-rich veinlet.  
 26. Na-enriched bleb.

Table 5.2-2: Composition of pollucite from Tin Mt., South Dakota.

No.	42	3	5	11	Average
SiO <sub>2</sub>	49.41	43.97	42.88	45.22	46.25 (.63)
Al <sub>2</sub> O <sub>3</sub>	17.20	16.31	16.24	16.06	16.21 (.22)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.00	0.01	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.37	0.02	0.00	0.26	0.30 (.04)
Na <sub>2</sub> O	3.10	1.25	0.87	1.74	1.92 (.23)
K <sub>2</sub> O	0.02	0.00	0.00	0.01	0.00
Rb <sub>2</sub> O	0.00	0.05	0.05	0.00	0.25
Cs <sub>2</sub> O	27.25	36.12	38.69	33.28	32.04 (.65)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.36	97.73	98.73	96.58	97.03 (.36)
Si	2.135	2.098	2.082	2.117	2.125 (.012)
Al	0.876	0.918	0.929	0.886	0.878 (.013)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.014	0.001	0.000	0.010	0.012 (.001)
Na	0.259	0.115	0.081	0.158	0.166 (.027)
K	0.001	0.000	0.000	0.001	0.000
Rb	0.000	0.001	0.001	0.000	0.007
Cs	0.502	0.735	0.802	0.664	0.628 (.055)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.876	0.918	0.929	0.886	0.878 (.013)
Σ charges	0.763	0.852	0.884	0.824	0.806 (.044)
Si/Al	2.437	2.286	2.241	2.391	2.422 (.048)
CRK	66.0	86.4	90.8	80.8	78.8 (2.3)

2. Na-enriched.

9. Cs-enriched bleb.

13. Cs-rich veinlet.

14. Pollucite grain in quartz.

Average of homogeneous pollucite (N = 9).

### 5.3 Strickland Quarry, Connecticut

The Strickland Quarry is located near Portland, Connecticut among the spodumene pegmatites of the Middletown district.

Sample number: PA-138 (M. Wise, U.S.N.M. 112525); 1 thin section, 8 analyses.

#### 5.3.1 Properties, associations and alteration

In hand specimen, the pollucite is white to colourless, and vitreous in lustre. Very thin veins crosscut the pollucite; these have a slight red (hematite?) staining.

In thin section, abundant micro-inclusions follow around small areas (up to 1 mm) of pollucite. Thin veins (3 mm long, 30  $\mu\text{m}$  wide) of fine-grained mica ( $\pm$  quartz) contain small amounts of Cs-rich pollucite. Later veins (2 cm long, 1 mm wide) of medium-grained mica locally contain analcime.

#### 5.3.2 Textural relationships

Most of the pollucite is homogeneous. Locally the pollucite is Na-enriched particularly near fractures. Rare Cs-enriched blebs are present, as are Cs-rich pollucite

veinlets containing small grains of quartz.

### 5.3.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.3-1a. The homogeneous pollucite has Si/Al from 2.54 to 2.59 and CRK from 81.1 to 81.7; the average and additional representative compositions are given in Table 5.3-1. Qualitative analyses indicate the Na-enriched compositions have CRK as low as 58 and Si/Al within the same range as that of the homogeneous pollucite. The Cs-enriched blebs have Si/Al as low as 2.48 and CRK as high as 84.1. The Cs-rich veinlets have Si/Al as low as 2.39 and CRK as high as 86.4.

A plot of Al (+ Fe), and the sum of cation charges vs. CRK is shown in Figure 5.3-1b. The sum of cation charges follows a strong trend of increasing Al with CRK.





Table 5.3-1: Composition of pollucite from Strickland Quarry, Connecticut.

No.	1	9	4	2	Average
SiO <sub>2</sub>	47.38	47.12	46.00	44.96	47.21 (.29)
Al <sub>2</sub> O <sub>3</sub>	15.58	15.77	15.73	15.94	15.64 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.00	0.00	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.21	0.20	0.20	0.01	0.19 (.02)
Na <sub>2</sub> O	1.64	1.64	1.47	1.26	1.67 (.03)
K <sub>2</sub> O	0.01	0.02	0.03	0.01	0.02
Rb <sub>2</sub> O	0.09	0.17	0.17	0.15	0.15 (.04)
Cs <sub>2</sub> O	33.00	32.90	34.16	36.15	32.79 (.16)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.93	97.86	97.77	98.48	97.67 (.28)
Si	2.163	2.156	2.139	2.123	2.160 (.002)
Al	0.838	0.849	0.863	0.887	0.843 (.006)
Fe	0.001	0.000	0.000	0.000	0.000
P	0.008	0.008	0.008	0.000	0.007
Na	0.145	0.145	0.133	0.115	0.148 (.003)
K	0.001	0.001	0.002	0.000	0.001
Rb	0.003	0.005	0.005	0.005	0.004 (.001)
Cs	0.642	0.641	0.677	0.728	0.640 (.004)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.839	0.849	0.863	0.887	0.843 (.006)
Σ charges	0.791	0.792	0.816	0.848	0.793 (.004)
Si/Al	2.581	2.539	2.480	2.392	2.561 (.023)
CRK	81.7	81.7	83.8	86.4	81.4 (.03)

1. Homogeneous.

9. Homogeneous.

4. Cs-enriched bleb.

2. Cs-rich veinlet.

Average of homogeneous pollucite (N = 5).

#### 5.4 Walden Gem Mine, Connecticut

The Walden Gem Mine (a.k.a. Gotta-Walden) is located near Portland, Connecticut about 1500 m north of the Strickland pegmatite.

Sample number: PA-139 (M. Wise, U.S.N.M. R 16708); 1 thin section, 7 analyses.

##### 5.4.1 Properties, associations and alteration

In hand specimen the pollucite is clear and colourless with a vitreous lustre. It is cut by a lens-shaped vein of a grey to pink fine-grained mineral. Very small amounts of buff clays occur along fracture surfaces.

In thin section, the vein contains mica and quartz  $\pm$  analcime. The tip of the lens is extended to a thin vein. Small round aggregates of adularia extend from the vein and are also embedded in the pollucite.

##### 5.4.2 Textural relationships

The pollucite is almost entirely homogeneous; near some fractures it is slightly enriched in  $\text{Na}^+$ .

### 5.4.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.4-1a. The homogeneous pollucite has Si/Al from 2.54 to 2.60 and CRK from 77.9 to 82.9; the average and additional representative compositions are given in Table 5.4-1. The Na-enriched compositions have CRK as low as 70.7 and the same Si/Al as the homogeneous pollucite.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is given in Figure 5.4-1b. Al is constant with CRK. The sum of cation charges decreases with decreasing CRK, due to sample burning of the Na-enriched pollucite.

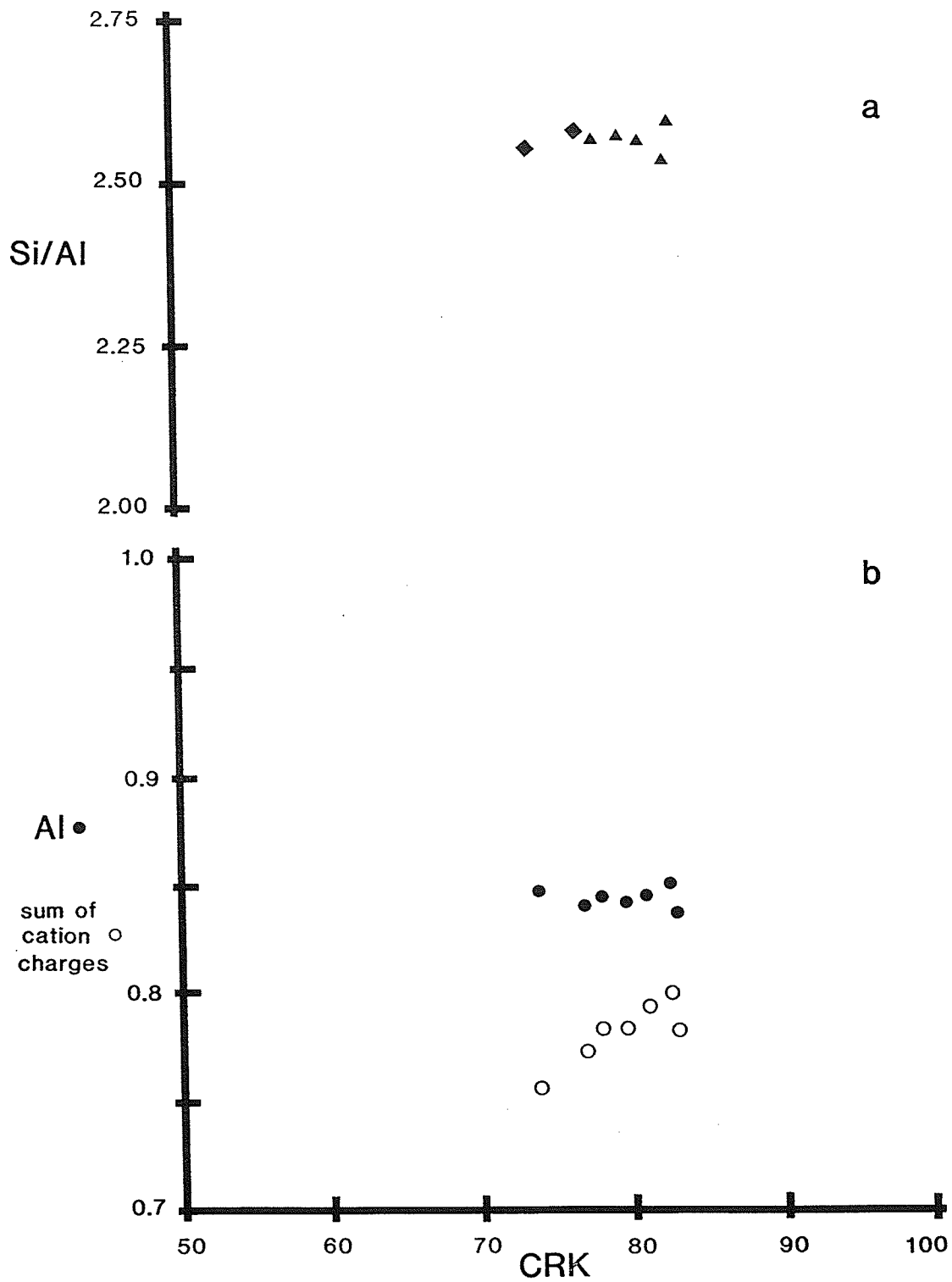


Fig. 5.4-1. Compositional trends for Walden Gem Mine, Connecticut. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 5.4-1: Composition of pollucite from Walden Gem Mine, Connecticut.

No.	1	8	4	5	Average
SiO <sub>2</sub>	47.38	47.12	46.00	44.96	47.21 (.29)
Al <sub>2</sub> O <sub>3</sub>	15.58	15.77	15.73	15.94	15.64 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.00	0.00	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.21	0.20	0.20	0.01	0.19 (.02)
Na <sub>2</sub> O	1.64	1.64	1.47	1.26	1.67 (.03)
K <sub>2</sub> O	0.01	0.02	0.03	0.01	0.02
Rb <sub>2</sub> O	0.09	0.17	0.17	0.15	0.15 (.04)
Cs <sub>2</sub> O	33.00	32.90	34.16	36.15	32.79 (.16)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.93	97.86	97.77	98.48	97.67 (.28)
Si	2.163	2.156	2.139	2.123	2.160 (.002)
Al	0.838	0.849	0.863	0.887	0.843 (.006)
Fe	0.001	0.000	0.000	0.000	0.000
P	0.008	0.008	0.008	0.000	0.007
Na	0.145	0.145	0.133	0.115	0.148 (.003)
K	0.001	0.001	0.002	0.000	0.001
Rb	0.003	0.005	0.005	0.005	0.004 (.001)
Cs	0.642	0.641	0.677	0.728	0.640 (.004)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.839	0.849	0.863	0.887	0.843 (.006)
Σ charges	0.791	0.792	0.816	0.848	0.793 (.004)
Si/Al	2.581	2.539	2.480	2.392	2.561 (.023)
CRK	81.7	81.7	83.8	86.4	81.4 (.03)

1. Homogeneous.

9. Homogeneous.

4. Na-enriched.

2. Na-enriched.

Average of homogeneous pollucite (N = 5).

## 5.5 Leominster, Massachusetts

Pollucite was discovered in 1937 in one of a series of ledges of spodumene pegmatites extending into the town of Leominster. Irregular masses up to 3 cm in diameter are associated with spodumene, quartz, feldspars, amblygonite, green tourmaline, and topaz. Bulk compositions were determined by Richmond & Gonyer (1938) (No. 44 in Table 2.1h). In Massachusetts, pollucite is also known from the Barrus pegmatite in the town of Goshen (Shaub & Schenck 1954). Sample number: PA-020 (J.S. White, U.S.N.M. 103386); 2 thin sections, 20 analyses.

### 5.5.1 Properties, associations and alteration

In hand specimen, the pollucite is white to colourless and granular in texture. It is associated with quartz, spodumene, white mica, microlite(?), apatite, and green tourmaline.

In thin section, the pollucite occurs adjacent to and interstitially with lath spodumene (symplectitic with quartz), quartz, and feldspar. The pollucite is cut by thin (0.5 mm wide) subparallel veins of fine-grained mica which pinch out within the pollucite. Scattered small grains of quartz are present in the pollucite. Minor adularia is present along surfaces of the mica veins.

### 5.5.2 Textural relationships

The pollucite is mostly homogeneous but is enriched in  $\text{Na}^+$  adjacent to the veins of mica. Locally the pollucite is cut by Cs-rich veinlets (+ quartz). A 100  $\mu\text{m}$  wide band of Cs-rich pollucite contains a 25  $\mu\text{m}$  wide band with even higher Cs contents immediately adjacent to some of the lath spodumene. Quartz adjacent to the pollucite contains round pollucite inclusions up to 50  $\mu\text{m}$  in size. The pollucite is Cs-rich at contacts with adularia grains.

### 5.5.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.5-1a. The homogeneous pollucite has Si/Al from 2.47 to 2.53 and CRK from 79.3 to 82.0. Na-enriched pollucite has CRK as low as 66 and the same Si/Al as the homogeneous pollucite. The Cs-rich veinlets have Si/Al as low as 2.34 and CRK as high as 88.9. The Cs-rich pollucite adjacent to spodumene laths has Si/Al as low as 2.24 and CRK as high as 92.2.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.5-1b. The sum of cation charges follows a trend of Al increasing with CRK, except for the Na-enriched pollucite, where Al is constant with CRK.



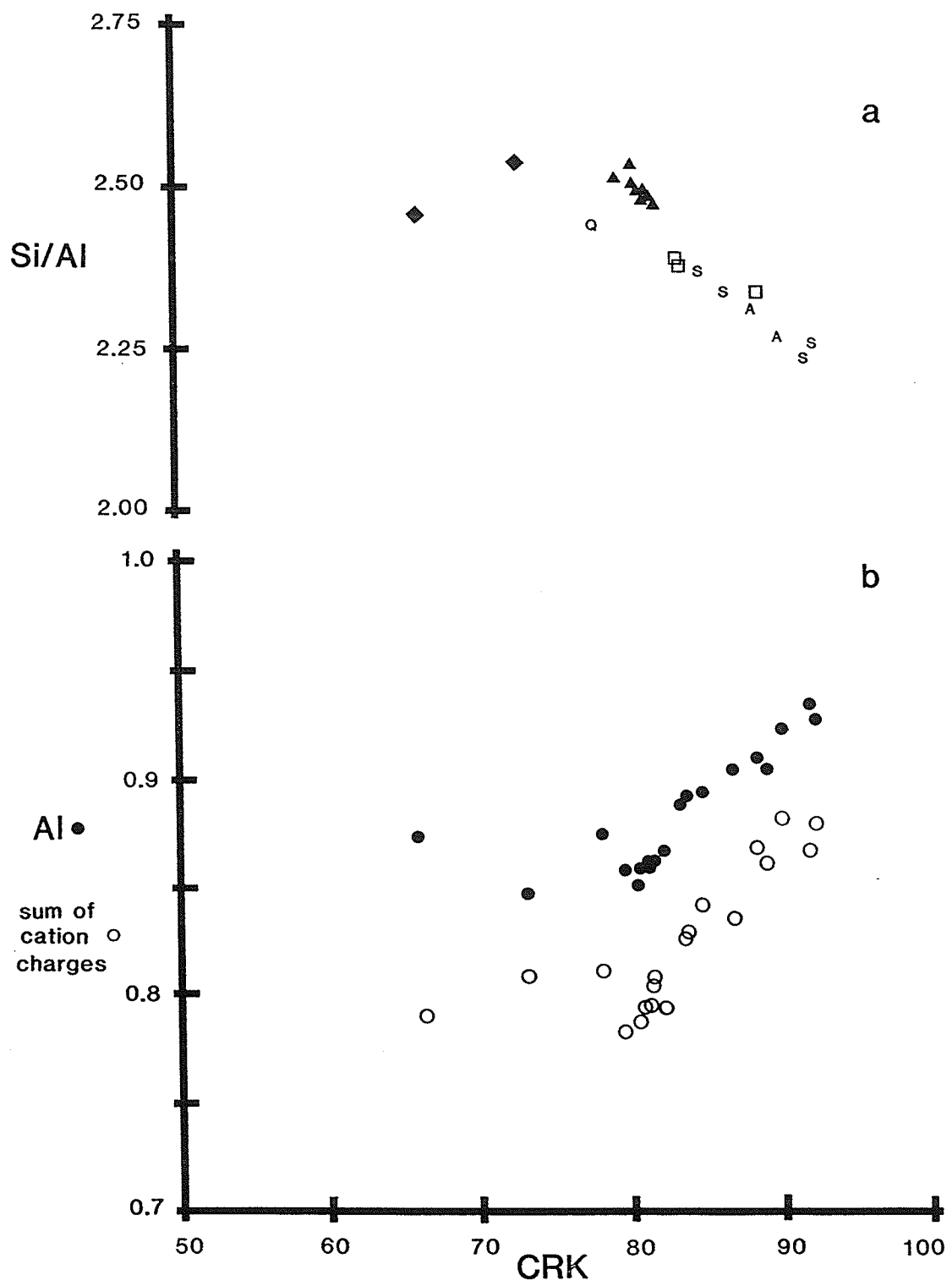


Fig. 5.5-1. Compositional trends for Leominster, Massachusetts. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Inclusion in quartz (Q) and in adularia (A). Bands of Cs-rich pollucite adjacent to lath spodumene (S).

Table 5.5-1: Composition of pollucite from Leominster, Massachusetts.

No.	1	10	18	8	4
SiO <sub>2</sub>	46.53	47.23	47.17	48.81	43.61
Al <sub>2</sub> O <sub>3</sub>	15.96	15.95	15.80	16.86	15.83
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.01	0.00	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.22	0.21	0.25	0.21	0.00
Na <sub>2</sub> O	1.60	1.82	1.75	3.13	1.02
K <sub>2</sub> O	0.01	0.01	0.00	0.04	0.00
Rb <sub>2</sub> O	0.19	0.08	0.14	0.00	0.15
Cs <sub>2</sub> O	32.78	31.74	32.29	27.82	36.74
CaO	0.00	0.03	0.01	0.03	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.30	97.08	97.41	96.90	97.36
Si	2.141	2.152	2.154	2.139	2.109
Al	0.866	0.857	0.850	0.871	0.902
Fe	0.000	0.000	0.000	0.000	0.000
P	0.009	0.008	0.010	0.008	0.000
Na	0.143	0.161	0.155	0.266	0.096
K	0.001	0.001	0.000	0.002	0.000
Rb	0.006	0.002	0.004	0.000	0.005
Cs	0.643	0.617	0.628	0.520	0.758
Ca	0.000	0.001	0.000	0.001	0.000
Mg	-----	0.000	0.000	0.000	0.000
Al + Fe	0.866	0.857	0.850	0.871	0.902
Σ charges	0.793	0.782	0.787	0.790	0.859
Si/Al	2.473	2.513	2.534	2.456	2.338
CRK	82.0	79.3	80.3	66.2	88.9

1. Homogeneous.
10. Homogeneous.
18. Homogeneous.
8. Na-enriched.
4. Cs-enriched veinlet.

Table 5.5-2: Composition of pollucite from Leominster, Massachusetts.

No.	17	5	12	13	Average
SiO <sub>2</sub>	46.26	42.63	44.81	42.53	46.64 (.38)
Al <sub>2</sub> O <sub>3</sub>	16.07	15.96	16.01	15.98	15.84 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.02	0.00	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.20	0.00	0.06	0.01	0.23 (.05)
Na <sub>2</sub> O	1.99	0.93	1.40	0.71	1.71 (.07)
K <sub>2</sub> O	0.00	0.03	0.01	0.01	0.01
Rb <sub>2</sub> O	0.19	0.14	0.09	0.08	0.16 (.08)
Cs <sub>2</sub> O	31.85	37.61	34.99	38.54	32.49 (.39)
CaO	0.01	0.01	0.02	0.02	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	96.58	97.34	97.39	97.87	97.09 (.27)
Si	2.132	2.088	2.118	2.087	2.146 (.005)
Al	0.873	0.921	0.893	0.925	0.859 (.005)
Fe	0.000	0.001	0.000	0.000	0.000
P	0.008	0.000	0.002	0.000	0.009 (.001)
Na	0.178	0.088	0.129	0.067	0.152 (.005)
K	0.000	0.002	0.001	0.000	0.001
Rb	0.006	0.004	0.003	0.003	0.005 (.001)
Cs	0.626	0.785	0.705	0.806	0.637 (.011)
Ca	0.001	0.000	0.001	0.001	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.873	0.922	0.893	0.925	0.859 (.005)
Σ charges	0.811	0.881	0.840	0.878	0.795 (.008)
Si/Al	2.442	2.268	2.373	2.257	2.499 (.019)
CRK	77.9	89.9	84.5	92.2	80.9 (0.8)

17. Inclusion in quartz.

5. Inclusion in adularia.

12. Wide Cs-rich band next to spodumene.

13. Thin Cs-rich band next to spodumene.

Average of homogeneous pollucite (N = 8).

## 5.6 Mt. Rubellite, Hebron, Maine

The locality of the historic second find of pollucite in the world was Mt. Rubellite (C. Francis, personal communication 1991), and not Cusham Farm or the Owl's Head property (both mined by W.D. Neville, both reportedly pollucite localities). Pollucite was found in two cavities, one with etched quartz, the other within a pile of clay.

Wells (1891) determined bulk compositions (No. 41 in Table 2.1g and No. 55 in Table 2.1j). More than 500 g of fresh material was available to Wells, associated with quartz, cesian beryl, and psilomelane. The pollucite is clear, colorless, vitreous, and contains only a single embedded crystal of quartz. Perpendicular to fracture surfaces is a series of parallel holes of rectangular cross-section. Fleischer & Ksanda (1940) determined dehydration curves of this pollucite (1.46 wt.% H<sub>2</sub>O).

Sample number: PA-074 (J.S. White. U.S.N.M. 96627); 1 thin section, 11 analyses.

### 5.6.1 Properties and composition

The pollucite is clear and inclusion-free, colourless, and vitreous in lustre. It is compositionally homogeneous, with Si/Al from 2.37 to 2.40 and CRK from 85.2 to 87.3; the average and typical compositions are given in Table 5.6-1.

Table 5.6-1: Composition of pollucite from Mt. Rubellite, Hebron, Maine.

No.	1	2	3	4	Average
SiO <sub>2</sub>	45.02	45.40	45.07	44.90	45.09 (.20)
Al <sub>2</sub> O <sub>3</sub>	16.06	16.19	15.98	16.07	16.13 (.24)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.00	0.00	0.00
Na <sub>2</sub> O	1.26	1.38	1.28	1.22	1.24 (.06)
K <sub>2</sub> O	0.02	0.03	0.02	0.04	0.04 (.01)
Rb <sub>2</sub> O	0.04	0.04	0.02	0.02	0.02 (.01)
Cs <sub>2</sub> O	35.79	35.85	36.04	35.79	37.78 (.22)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.19	98.90	98.41	98.04	98.26 (.32)
Si	2.122	2.121	2.123	2.121	2.123 (.003)
Al	0.892	0.891	0.888	0.894	0.891 (.003)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.000	0.000	0.000	0.000	0.000
Na	0.115	0.125	0.117	0.111	0.114 (.005)
K	0.001	0.002	0.001	0.002	0.002 (.001)
Rb	0.001	0.001	0.001	0.001	0.009
Cs	0.719	0.715	0.724	0.721	0.718 (.006)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.892	0.891	0.888	0.894	0.891 (.003)
Σ charges	0.837	0.843	0.843	0.835	0.835 (.007)
Si/Al	2.380	2.381	2.392	2.371	2.382 (.010)
CRK	86.2	85.2	86.1	86.7	86.4 (0.6)

1. Homogeneous.

2. Homogeneous.

3. Homogeneous.

4. Homogeneous.

Average of homogeneous pollucite (N = 11).

## 5.7 Mt. Marie, Paris, Maine

Sample number: PA-076 (D. Garske); 1 thin section, 15 analyses.

### 5.7.1 Properties, associations and alteration

In hand specimen the pollucite is white in colour with a greasy lustre. A few thin (1 mm) veins of mica crosscut the pollucite as well as a set of very thin subparallel veins spaced 1 to 3 mm apart. A light coating of buff clays occurs along fracture surfaces and near veins.

In thin section, the mica veins are irregular in width and diffuse. The pollucite has abundant micro-inclusions, and a few grains of spodumene. Apatite is associated with the clays.

### 5.7.2 Textural relationships

The pollucite is mostly homogeneous. It is Na-enriched near fractures. Cs-enriched blebs are rare. Diffuse Cs-rich veinlets (Fig. 5.7-1a) are associated with small grains of quartz ( $\pm$  spodumene  $\pm$  mica). The pollucite contains some elongate lens-shaped grains (up to 150  $\mu\text{m}$ ) of Cs-rich pollucite, surrounded by Na-enriched pollucite (Fig. 5.7-1b).

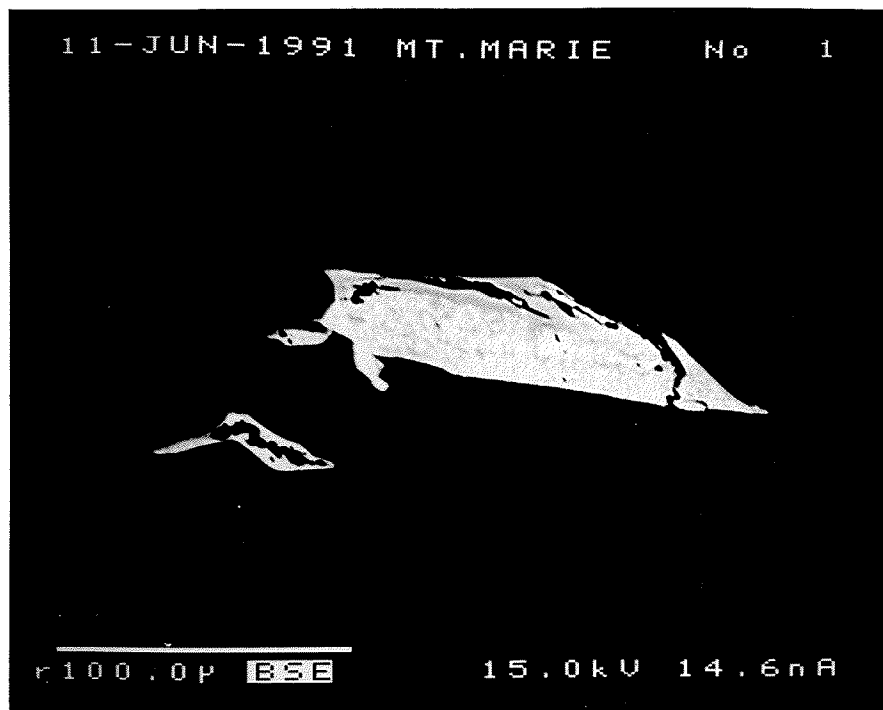
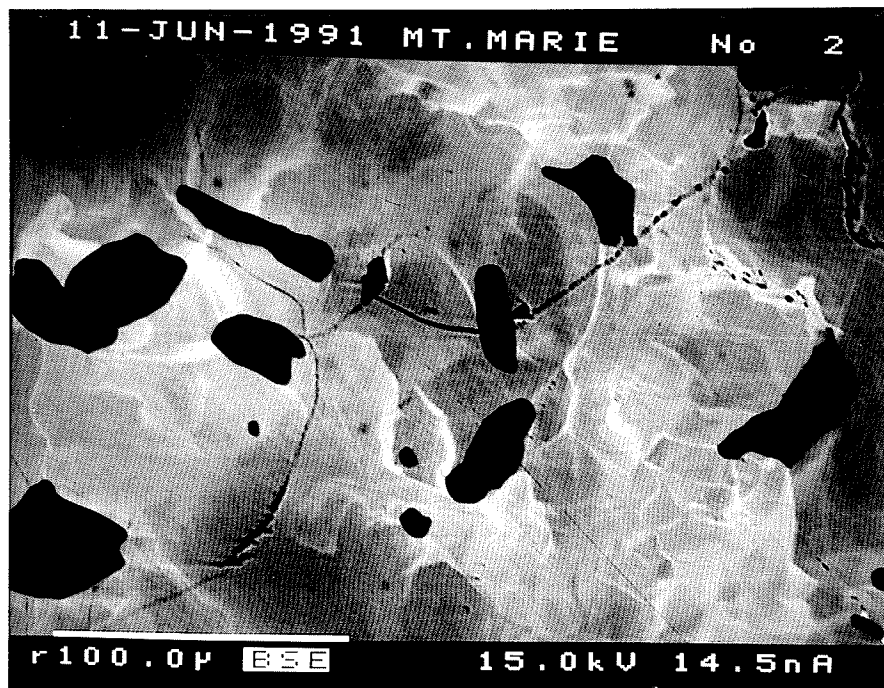


Fig. 5.7-1. BSE images of Mt. Marie pollucite. a) Diffuse Cs-rich veinlets and quartz grains in a large area of homogeneous pollucite. b) Cs-rich grain with strongly Na-enriched rims in a large area of homogeneous pollucite.

### 5.7.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.7-2a. The homogeneous pollucite has Si/Al from 2.52 to 2.58 and CRK from 77.7 to 79.5; the average and additional representative compositions are given in Table 5.7-1. Qualitative analyses of the Na-enriched pollucite indicate CRK as low as 71.6 and Si/Al within the same range as the homogeneous pollucite. Cs-enriched blebs have Si/Al as low as 2.34 and CRK as high as 82.5. Cs-rich veinlets have Si/Al as low as 2.27 and CRK as high as 86.8.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.7-2b. The sum of cation charges follows a trend of Al increasing with CRK.



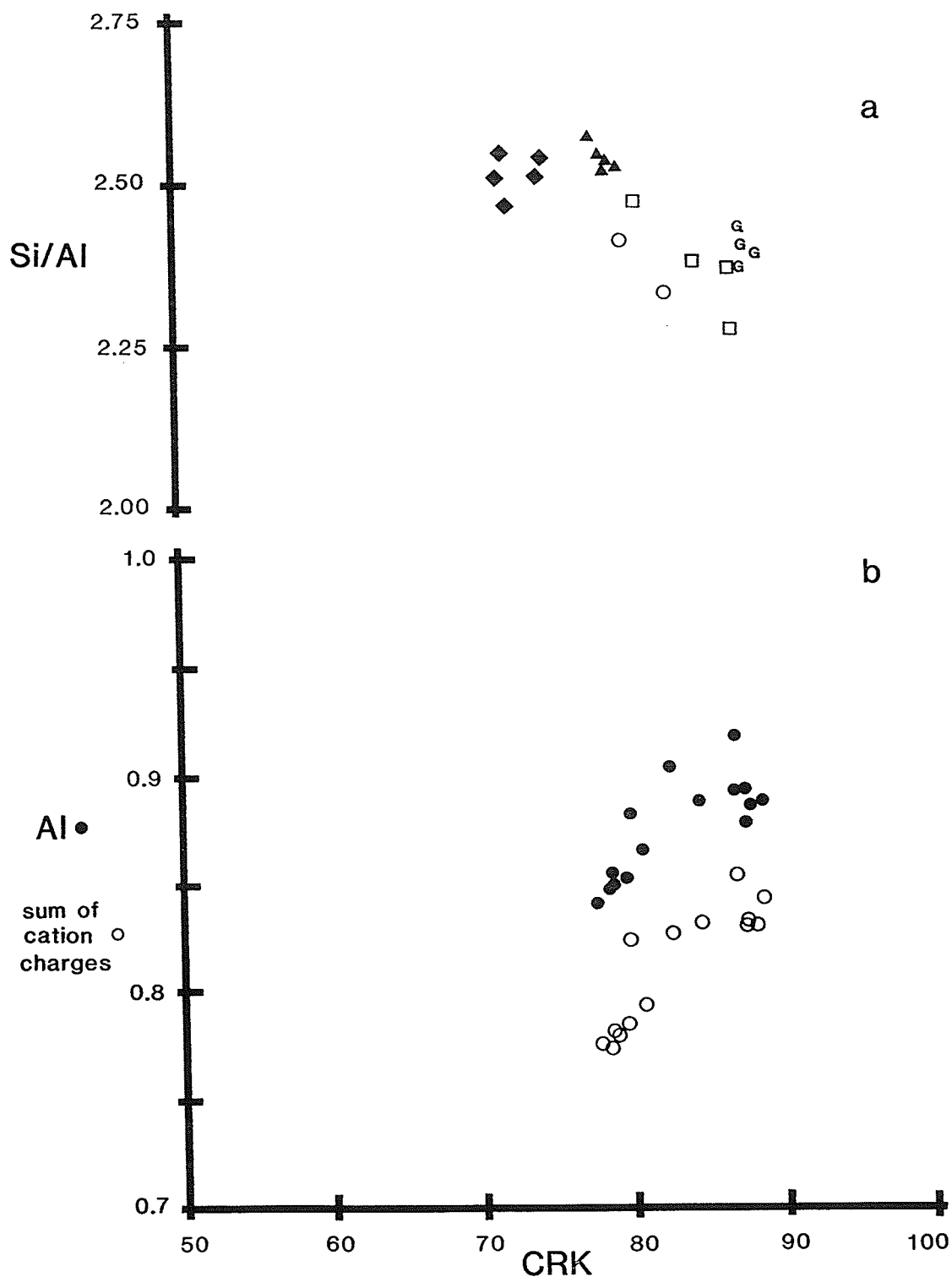


Fig. 5.7-2. Compositional trends for Mt. Marie, Paris, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Grains of Cs-rich pollucite (G).

Table 5.7-1: Composition of pollucite from Mt. Marie, Paris, Maine.

No.	18	7	19	13	Average
SiO <sub>2</sub>	47.82	44.89	45.37	43.75	47.98 (.40)
Al <sub>2</sub> O <sub>3</sub>	16.00	15.91	16.49	16.34	16.01 (.06)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.01	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.22	0.04	0.18	0.26	0.27 (.03)
Na <sub>2</sub> O	1.89	1.06	1.61	1.21	1.91 (.06)
K <sub>2</sub> O	0.01	0.00	0.02	0.00	0.02 (.01)
Rb <sub>2</sub> O	0.00	0.00	0.00	0.05	0.00
Cs <sub>2</sub> O	31.89	37.00	34.33	36.37	31.86 (.22)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.84	98.91	98.00	97.99	98.06 (.35)
Si	2.156	2.122	2.108	2.086	2.156 (.004)
Al	0.850	0.887	0.903	0.918	0.848 (.005)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.009	0.002	0.007	0.010	0.010 (.001)
Na	0.166	0.097	0.145	0.113	0.167 (.005)
K	0.001	0.000	0.001	0.000	0.001
Rb	0.000	0.000	0.000	0.001	0.000
Cs	0.613	0.746	0.680	0.739	0.611 (.005)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.850	0.887	0.903	0.918	0.848 (.005)
Σ charges	0.780	0.842	0.826	0.853	0.779 (.004)
Si/Al	2.537	2.393	2.335	2.272	2.543 (.021)
CRK	78.8	88.5	82.5	86.8	78.6 (0.7)

18. Homogeneous.

7. Cs-rich grain.

19. Cs-enriched bleb.

13. Cs-rich veinlet.

Average of homogeneous pollucite (N = 5).

## 5.8 Merrill Mine, Paris, Maine

The sample was mis-labelled as Merritt mine, but is probably from the mine of Loren B. Merrill, at Mt. Mica. The composition is the same as pollucite from Mt. Marie, however, the associated minerals are different.

Sample number: PA-033 (I. Cerná, M48); 1 thin section, 16 analyses.

### 5.8.1 Properties, associations and alteration

In hand specimen, the pollucite is white, granular, and crumbly. It is associated with light blue cleavelandite, is crosscut by very thin white veins, and is associated with small amounts of buff clays.

In thin section, the pollucite occurs between grains and as fracture-fillings of the albite. It is cut by veins of coarse-grained mica. Later fine-grained mica veins are partially replaced by spodumene and pinch out within the pollucite segregation.

### 5.8.2 Textural relationships

About half of the pollucite is compositionally homogeneous. This half contains later Cs-enriched blebs, crosscut by quartz-containing Cs-rich veinlets. The

pollucite between albite laths is Cs-rich and shows a symplectitic intergrowth with quartz (Fig. 5.8-1a and b).

### 5.8.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.8-2a. The homogeneous pollucite has Si/Al from 2.53 to 2.59 and CRK from 75.7 to 78.8; the average and additional representative compositions are given in Table 5.8-1. The Cs-enriched blebs have Si/Al as low as 2.43 and CRK as high as 82.6. The Cs-rich veinlets have Si/Al as low as 2.33 and CRK as high as 90.1. The Cs-rich pollucite symplectitic with quartz has Si/Al as low as 2.28 and CRK as high as 89.1.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.8-2b. The sum of cation charges follows a trend of Al increasing with CRK.

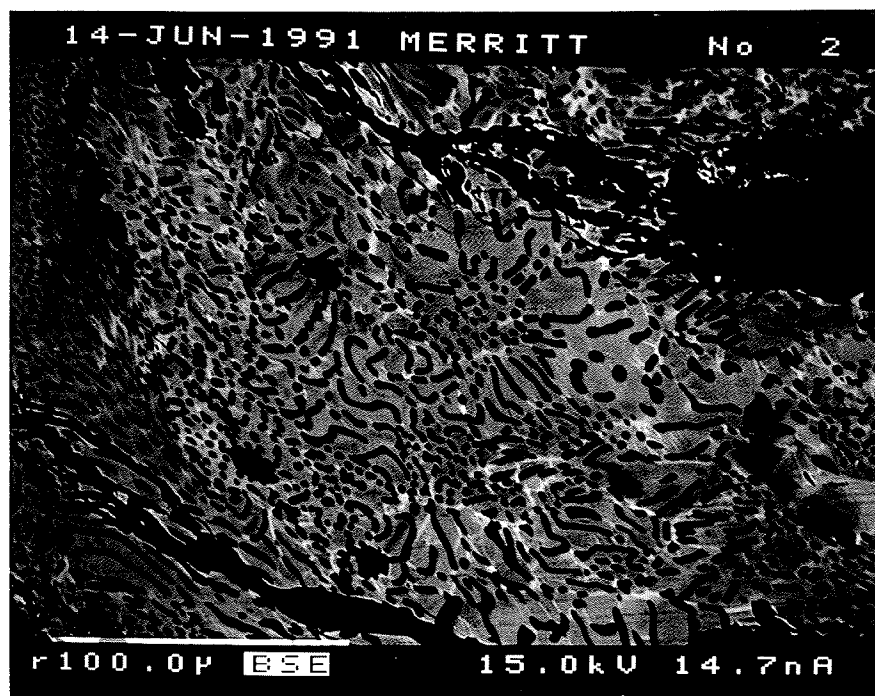
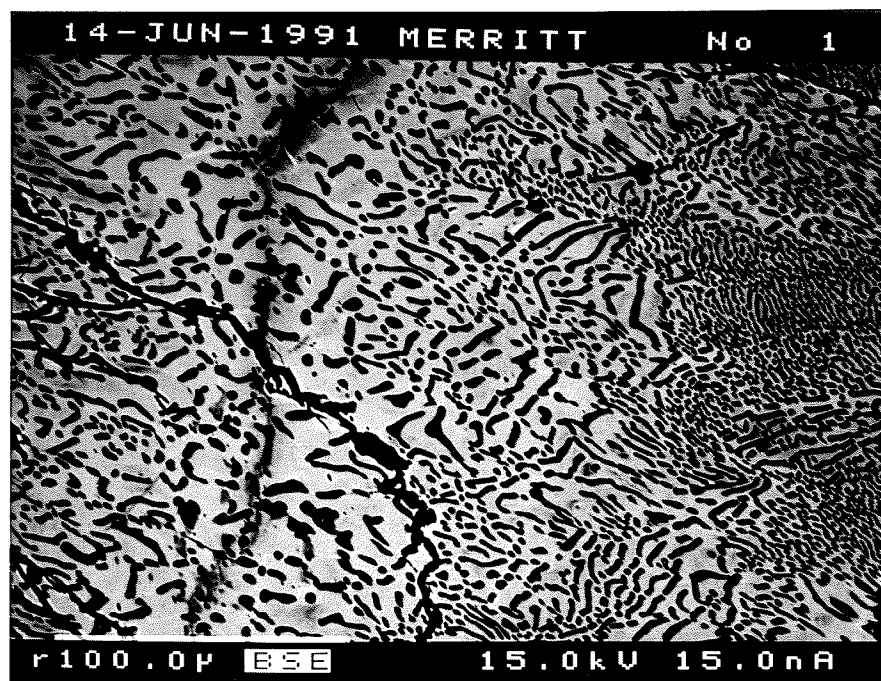


Fig. 5.8-1. BSE images of Merrill pollucite. a) and b) Cs-rich pollucite in a symplectitic intergrowth with quartz.

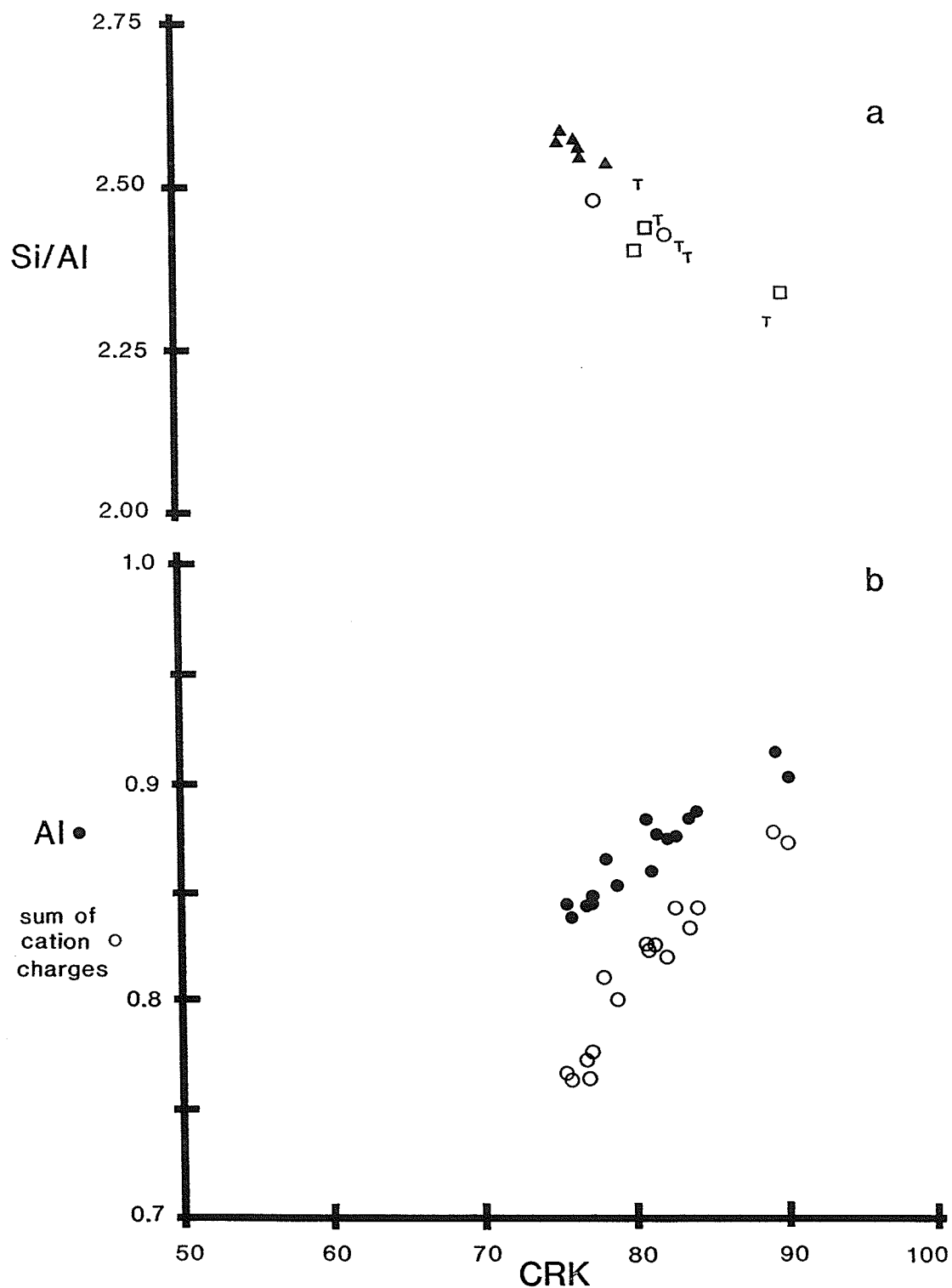


Fig. 5.8-2. Compositional trends for Merrill Mine, Paris, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Cs-rich pollucite in a symplectitic intergrowth with quartz (T).

Table 5.8-1: Composition of pollucite from Merrill Mine, Paris, Maine

No.	5	14	10	8	Average
SiO <sub>2</sub>	48.26	46.67	43.29	43.71	48.38 (.52)
Al <sub>2</sub> O <sub>3</sub>	15.98	15.84	16.08	15.88	16.03 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.02	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.22	0.07	0.07	0.01	0.19 (.08)
Na <sub>2</sub> O	2.00	1.74	1.01	0.91	2.04 (.07)
K <sub>2</sub> O	0.00	0.01	0.06	0.01	0.01
Rb <sub>2</sub> O	0.00	0.00	0.00	0.00	0.00
Cs <sub>2</sub> O	30.73	33.95	37.71	38.19	31.05 (.81)
CaO	0.02	0.01	0.00	0.03	0.04 (.04)
MgO	0.00	0.00	0.01	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.21	98.29	98.25	98.74	97.75 (.30)
Si	2.164	2.147	2.090	2.105	2.164 (.006)
Al	0.845	0.858	0.915	0.902	0.845 (.005)
Fe	0.000	0.000	0.001	0.000	0.000
P	0.009	0.003	0.003	0.000	0.007 (.003)
Na	0.174	0.155	0.095	0.085	0.177 (.005)
K	0.000	0.001	0.004	0.001	0.001
Rb	0.000	0.000	0.000	0.000	0.000
Cs	0.588	0.666	0.777	0.785	0.593 (.020)
Ca	0.001	0.001	0.000	0.001	0.002
Mg	0.000	0.000	0.001	0.000	0.000
Al + Fe	0.845	0.858	0.916	0.902	0.845 (.005)
Σ charges	0.764	0.824	0.878	0.872	0.774 (.014)
Si/Al	2.561	2.502	2.284	2.334	2.561 (.020)
CRK	77.1	81.0	89.1	90.1	76.9 (1.2)

5. Homogeneous.

14. Symplectitic with quartz.

10. Symplectitic with quartz.

8. Cs-rich veinlet.

Average of homogeneous pollucite (N = 6).

## 5.9 Dunton Gem Quarry, Newry, Maine

The pegmatite of the Dunton Gem Quarry (a.k.a. Neville tourmaline mine) was described by Fraser (1930). The pollucite is closely associated with spodumene and is fracture-filled by quartz (Fairbanks 1928). A 1.7 carat "brilliant-cut" gem of pollucite, described by Spencer (1934) with  $n = 1.51$  and  $D = 2.86$ , probably came from this popular collecting locality. The pollucite-bearing Crooker and Kinglet pegmatites are located 100 m north, and 100 m southwest, respectively, of the Dunton quarry (Shaub 1940). Sample No.: PA-038 (V. King); 2 thin sections, 30 analyses.

### 5.9.1 Properties, associations and alteration

In hand specimen, the pollucite ranges from white with a greasy lustre to clear and colourless with a sub-vitreous luster. Extremely thin sub-parallel white veins, spaced irregularly, cut the pollucite into slabs up to 3 mm thick.

In thin section, the white pollucite contains abundant micro-inclusions, not present in the clear colorless pollucite. An irregular vein of coarse mica + quartz cross-cuts the pollucite, as well as thin veins of fine-grained mica. Clay minerals, associated with fine-grained quartz, apatite and spodumene replace pollucite along fractures.



### 5.9.2 Textural relationships

The homogeneous pollucite has mm-sized areas with distinct compositions. Locally, the pollucite is highly enriched in  $\text{Na}^+$ , particularly along fractures and near veins. The most Na-enriched compositions are associated with small grains and veins of apatite (Fig. 5.9-1a). Locally, a well-developed blebby matrix consists of Na- and Cs-enriched domains. The blebs are elongate and lamellar (Fig. 5.9-1b). These are cross-cut by Cs-rich veinlets, which have a flame structure (Fig. 5.9-1c).

### 5.9-3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.9-2a. The homogeneous pollucite has Si/Al from 2.36 to 2.55 and CRK from 80.2 to 87.3; the average, and additional representative compositions are given in Tables 5.9-1 and 2. The highly Na-enriched compositions have an average Si/Al of 2.35. The Na-enriched blebs have Si/Al as high as 2.51 and CRK as low as 75.6. The Cs-enriched blebs have Si/Al as low as 2.35 and CRK as high as 85.3. The Cs-rich veinlets have Si/Al as low as 2.27 and CRK as high as 89.6.

A plot of Al (+ Fe), and sum of cation charges is shown in Figure 5.9-2b. The sum of cation charges follows a trend of near-constant Al with CRK for all the pollucite.

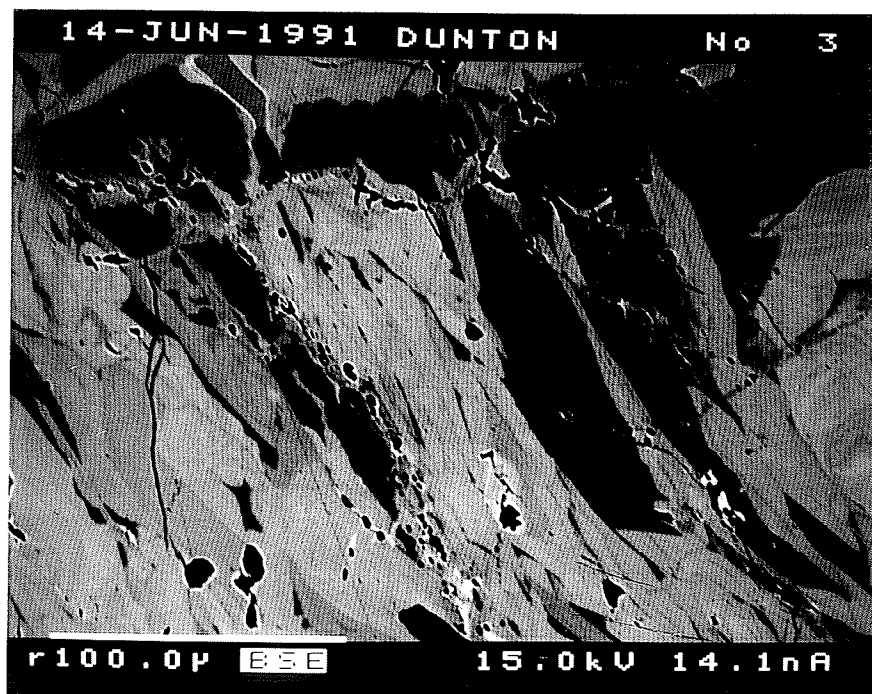
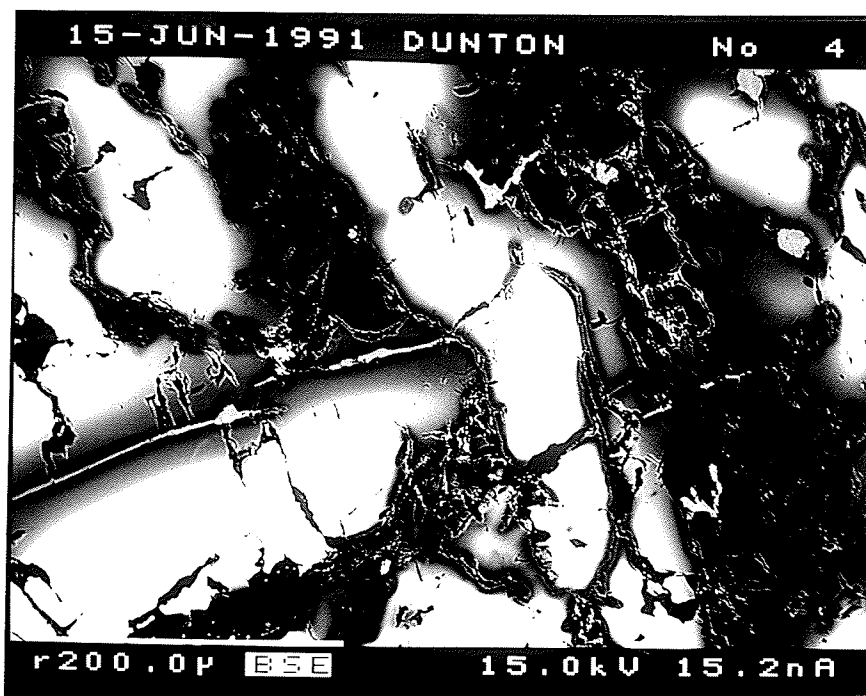


Fig. 5.9-1. BSE images of Dunton pollucite. a) Area of strong "analcimization" associated with clay minerals. The compositions most enriched in Na are near veinlets of apatite (bright). Note upper right: some of the clays have enrichment in Cs. b) Lamellar intergrowth of Na- and Cs-enriched blebs.

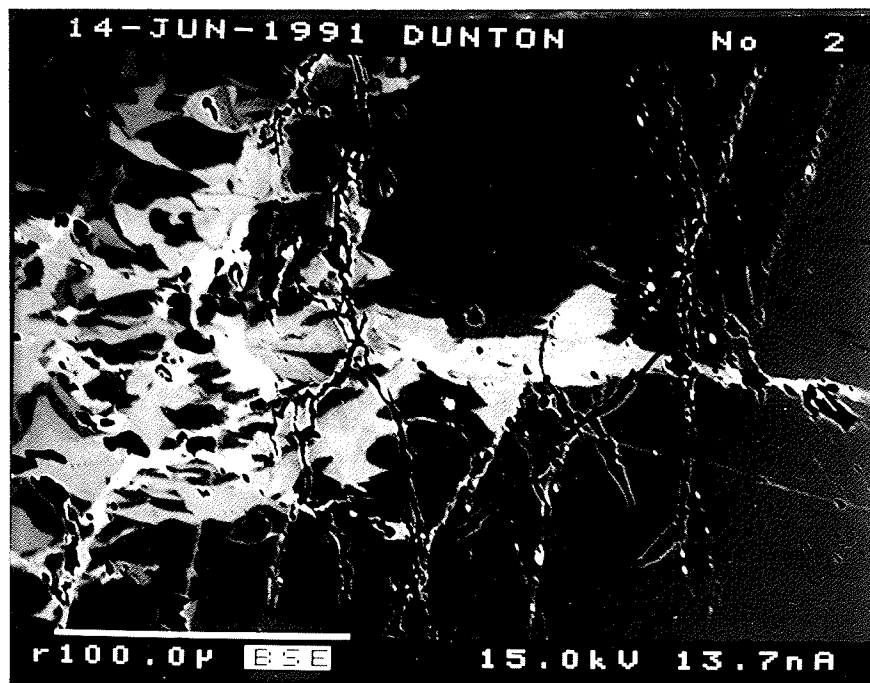


Fig. 5.9-1. c) Flame structure of Cs-rich veinlets replacing homogeneous pollucite from the surface of fractures. Note enrichment in Na along later fractures.

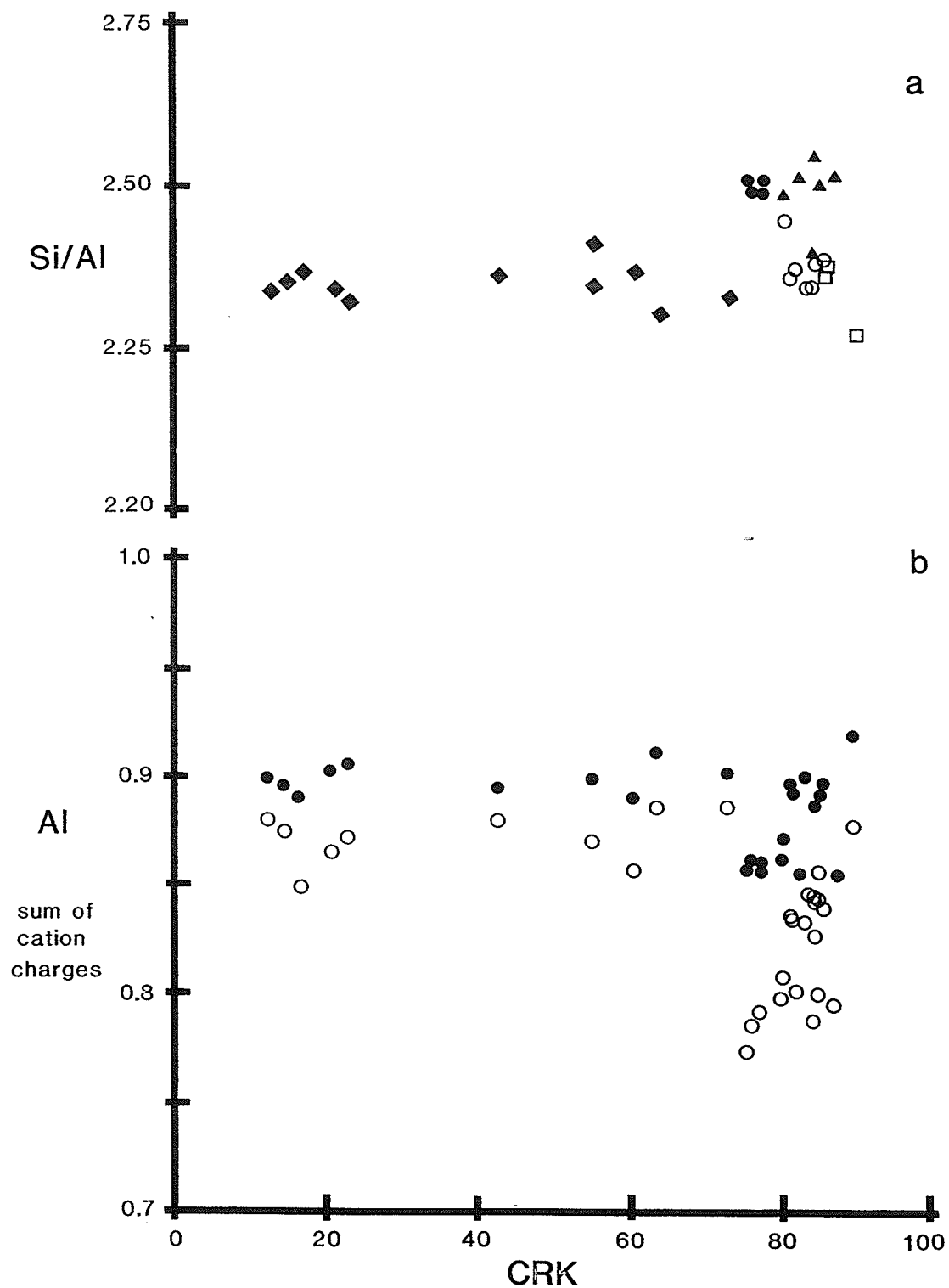


Fig. 5.9-2. Compositional trends for Dunton Gem Quarry, Newry, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 5.9-1: Composition of pollucite from Dunton Gem Quarry, Newry, Maine.

No.	6	9	10	27	21
SiO <sub>2</sub>	47.31	46.53	44.77	54.78	52.05
Al <sub>2</sub> O <sub>3</sub>	15.76	15.69	16.09	19.83	19.00
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.01	0.01	0.00	0.01
P <sub>2</sub> O <sub>5</sub>	0.37	0.34	0.17	0.00	0.07
Na <sub>2</sub> O	1.39	1.13	1.31	10.18	8.48
K <sub>2</sub> O	0.01	0.05	0.01	0.07	0.02
Rb <sub>2</sub> O	0.39	0.60	0.08	0.02	0.00
Cs <sub>2</sub> O	33.58	34.30	35.60	6.69	11.70
CaO	0.00	0.01	0.01	0.07	0.05
MgO	0.00	0.00	0.00	----	----
H <sub>2</sub> O	----	----	----	----	----
sum	98.83	98.64	98.04	91.63	91.37
Si	2.152	2.146	2.111	2.107	2.101
Al	0.845	0.853	0.894	0.898	0.905
Fe	0.001	0.000	0.000	0.000	0.000
P	0.014	0.013	0.007	0.000	0.002
Na	0.123	0.101	0.119	0.759	0.664
K	0.001	0.003	0.001	0.003	0.001
Rb	0.012	0.018	0.002	0.000	0.000
Cs	0.652	0.674	0.716	0.110	0.202
Ca	0.000	0.000	0.000	0.003	0.002
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.846	0.853	0.894	0.898	0.905
Σ charges	0.787	0.796	0.838	0.878	0.870
Si/Al	2.548	2.517	2.360	2.345	2.323
CRK	84.4	87.3	85.8	12.9	23.3

6. Homogeneous.  
 9. Homogeneous  
 10. Homogeneous.  
 27. Na-enriched.  
 21. Na-enriched.

Table 5.9-2: Composition of pollucite from Dunton Gem Quarry, Newry, Maine.

No.	25	12	15	4	Average	
SiO <sub>2</sub>	47.73	48.33	45.41	43.52	46.51	(.90)
Al <sub>2</sub> O <sub>3</sub>	16.78	16.38	16.34	16.27	15.87	(.16)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.02	0.00	0.00	0.01	
P <sub>2</sub> O <sub>5</sub>	0.06	0.28	0.18	0.22	0.32	(.07)
Na <sub>2</sub> O	4.54	2.19	1.74	0.98	1.43	(.24)
K <sub>2</sub> O	0.04	0.02	0.00	0.01	0.02	
Rb <sub>2</sub> O	0.00	0.01	0.00	0.00	0.35	(.21)
Cs <sub>2</sub> O	25.08	30.77	34.23	38.45	33.88	(1.0)
CaO	0.00	0.01	0.00	0.00	0.00	
MgO	----	0.00	0.00	0.00	0.00	
H <sub>2</sub> O	----	----	----	----	----	
sum	94.23	98.01	97.90	99.46	98.39	(.37)
Si	2.121	2.149	2.111	2.082	2.138	(.014)
Al	0.879	0.858	0.895	0.917	0.860	(.018)
Fe	0.000	0.001	0.000	0.000	0.000	
P	0.002	0.011	0.007	0.009	0.012	(.003)
Na	0.392	0.189	0.158	0.091	0.127	(.020)
K	0.002	0.001	0.000	0.001	0.001	
Rb	0.000	0.000	0.000	0.000	0.011	(.006)
Cs	0.475	0.584	0.679	0.785	0.648	(.021)
Ca	0.000	0.000	0.000	0.000	0.000	
Mg	-----	0.000	0.000	0.000	0.000	
Al + Fe	0.879	0.859	0.895	0.917	0.860	(.018)
Σ charges	0.869	0.774	0.836	0.877	0.803	(.018)
Si/Al	2.415	2.506	2.358	2.269	2.487	(.066)
CRK	54.9	75.6	81.2	89.6	84.2	(2.6)

25. Na-enriched.

12. Na-enriched bleb.

15. Na-enriched bleb.

4. Cs-rich veinlet.

Average of homogeneous pollucite (N = 6).

## 5.10 Locality X, Oxford County, Maine

Often Maine specimens were labelled in a general fashion to disguise the locality. This specimen probably came from the Dunton Gem mine, Newry, a popular collecting locality (C. Francis, personal communication 1991). Bulk ore from Oxford Co. was found to contain albite (52 %), muscovite (15%), spodumene (5%) and apatite (2%) (Dean & Nichols 1962).

Sample number: PA-030 (V. King); 2 thin sections, 1 grain mount, 14 analyses.

### 5.10.1 Properties, associations and alteration

In hand specimen, the pollucite is white to clear and colourless. It contains scattered grains of pink mica 1 mm in size. Clay minerals are present on fracture surfaces associated with small amounts of a (Mn-bearing ?) mineral.

In thin section, the colourless pollucite contains only mica grains. The white pollucite has scattered quartz, mica, and spodumene grains, and abundant micro-inclusions. Thin irregular mica veins contain later spodumene.

### 5.10.2 Textural relationships

The clear and colourless pollucite is almost entirely homogeneous, however, near fractures it is slightly enriched in  $\text{Na}^+$ . The pollucite which is white in colour has only local homogeneous areas, crosscut by Cs-enriched blebs associated with quartz grains (Fig. 5.10-1a). The blebs are cross-cut by Cs-rich veinlets + quartz. Cs-rich pollucite occurs near the mica veins, however, the veins seem to cross-cut the Cs-rich veinlets (Fig. 5.10-1b).

### 5.10.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.10-2a. The homogeneous pollucite has Si/Al from 2.45 to 2.55 and CRK from 77.4 to 82.9; the average and additional representative compositions are given in Table 5.10-1. The Cs-enriched blebs have Si/Al as low as 2.34 and CRK as high as 89.5. The Cs-rich veinlets have Si/Al as low as 2.29 and CRK as high as 92.8.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.10-2b. The sum of cation charges follows a trend of increasing Al with CRK.



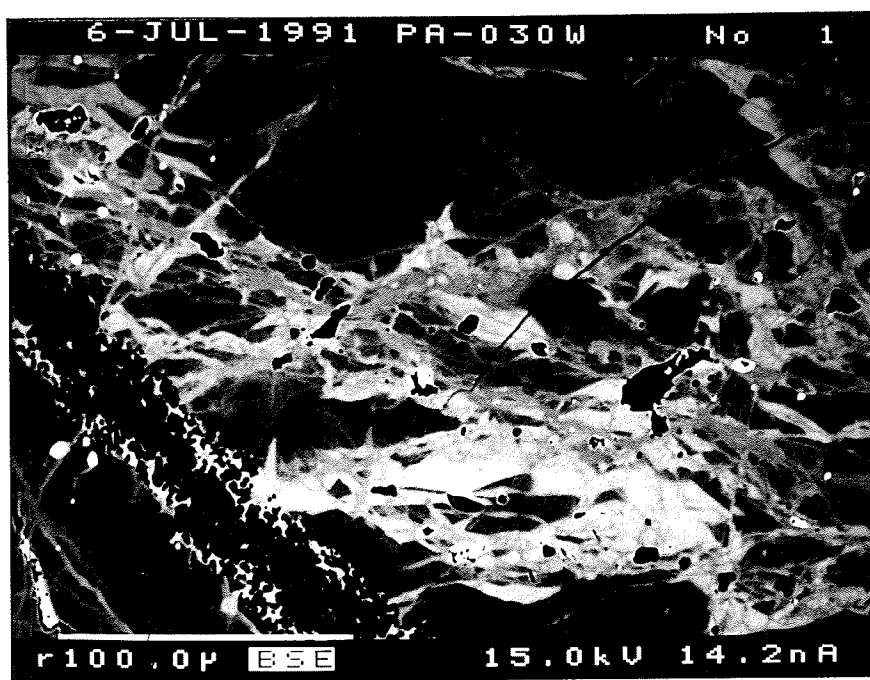
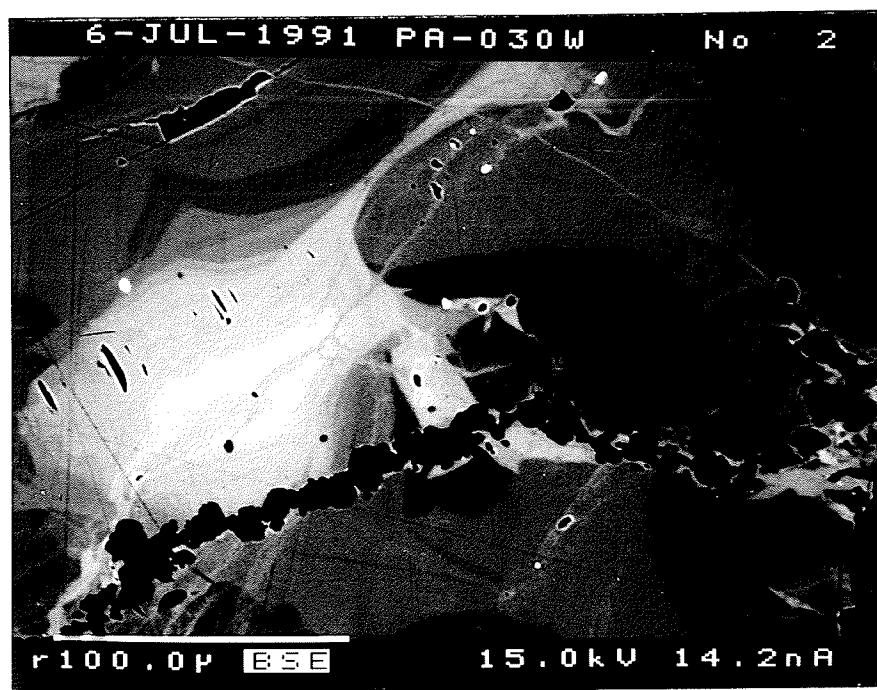


Fig. 5.10-1. BSE images of Locality X, Oxford Co.. a) Mica vein (dark) crosscutting Cs-enriched bleb. b) Mica vein with Cs-rich pollucite crosscutting(?) Cs-rich veinlets in homogeneous pollucite. Brightest specks are tin polish.

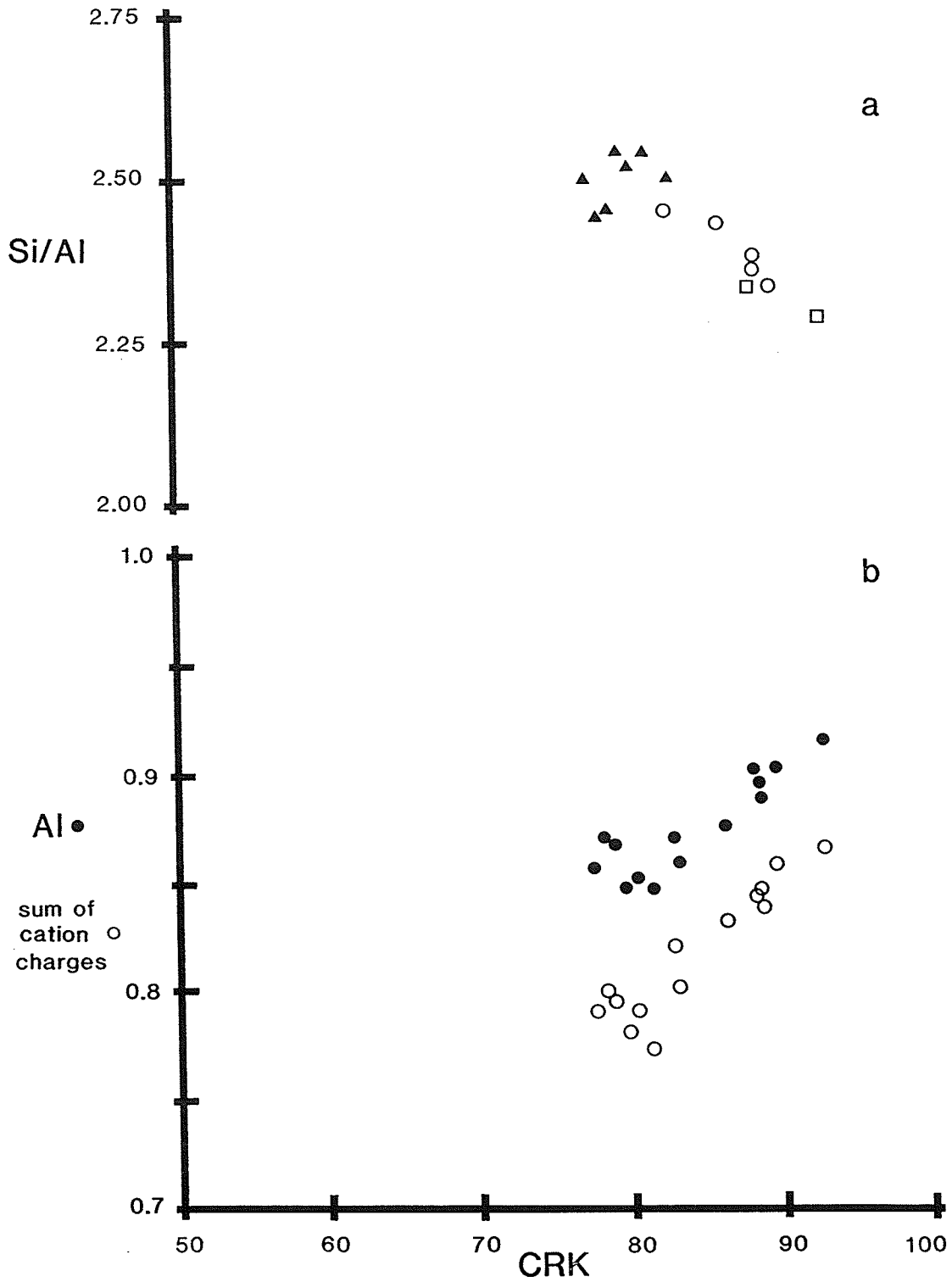


Fig. 5.10-2. Compositional trends for Locality X, Oxford Co., Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 5.10-1: Composition of pollucite from Locality X, Oxford County, Maine.

No.	3	13	12	7	Average
SiO <sub>2</sub>	46.84	47.36	44.40	43.31	47.65 (.45)
Al <sub>2</sub> O <sub>3</sub>	16.25	16.02	16.09	16.03	16.10 (.09)
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.00	0.01	0.00	0.01
P <sub>2</sub> O <sub>5</sub>	0.34	0.01	0.00	0.03	0.27 (.11)
Na <sub>2</sub> O	1.98	1.56	0.97	0.66	1.83 (.17)
K <sub>2</sub> O	0.02	0.03	0.03	0.01	0.02
Rb <sub>2</sub> O	0.10	0.55	0.54	0.54	0.36
Cs <sub>2</sub> O	32.02	33.37	36.89	38.10	32.11 (.60)
CaO	0.01	0.01	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.57	98.92	98.92	98.70	98.21 (.63)
Si	2.130	2.154	2.110	2.096	2.146 (.011)
Al	0.871	0.859	0.901	0.915	0.857 (.010)
Fe	0.001	0.000	0.000	0.000	0.000
P	0.013	0.001	0.000	0.001	0.011 (.004)
Na	0.175	0.137	0.090	0.062	0.160 (.015)
K	0.001	0.002	0.002	0.001	0.001
Rb	0.003	0.016	0.017	0.017	0.011 (.006)
Cs	0.621	0.647	0.748	0.786	0.624 (.017)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.872	0.859	0.901	0.915	0.857 (.010)
Σ charges	0.800	0.802	0.857	0.866	0.791 (.010)
Si/Al	2.446	2.507	2.342	2.292	2.505 (.040)
CRK	78.2	82.9	89.5	92.8	79.8 (1.9)

3. Homogeneous.

13. Homogeneous.

12. Cs-enriched bleb.

7. Cs-rich veinlet.

Average of homogeneous pollucite (N = 7).

### 5.11 BB # 7, Norway, Maine

Despite being termed "number 7", no other mines or pegmatites in the area are numbered (C. Francis, personal communication 1991).

Sample number: PA-026 (V. King); 1 thin section, 8 analyses.

#### 5.11.1 Properties, associations and alteration

In hand specimen, the pollucite is white to clear and colourless. Some thin grey veins are present, as is some buff clay. In thin section, the pollucite is crosscut by mica + spodumene veins. It is replaced by small granular aggregates of adularia.

#### 5.11.2 Textural relationships and composition

The pollucite is almost entirely homogeneous. Some Cs-rich veinlets are present. Na-enriched pollucite occurs along fractures, grading into the homogeneous pollucite over a distance of less than 5  $\mu\text{m}$ . The homogeneous pollucite has Si/Al from 2.30 to 2.33 and CRK from 84.1 to 86.1; the average and additional representative compositions are given in Table 5.11-1. The Cs-rich veinlets have Si/Al as low as 2.25 and CRK as high as 87.9.

Table 5.11-1: Composition of pollucite from BB # 7, Norway, Maine.

No.	1	8	3	4	Average
SiO <sub>2</sub>	44.69	44.90	44.73	43.84	44.76 (.17)
Al <sub>2</sub> O <sub>3</sub>	16.44	16.34	16.35	16.51	16.43 (.07)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.01	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	1.41	1.37	0.94	1.10	1.36 (.03)
Na <sub>2</sub> O	1.24	1.28	1.17	1.09	1.32 (.07)
K <sub>2</sub> O	0.02	0.01	0.01	0.01	0.01
Rb <sub>2</sub> O	1.05	1.00	0.65	0.53	0.93
Cs <sub>2</sub> O	33.07	33.10	34.88	35.28	32.99 (.07)
CaO	0.00	0.01	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.91	98.01	98.75	98.37	97.81 (.27)
Si	2.062	2.069	2.078	2.055	2.064 (.003)
Al	0.894	0.887	0.895	0.913	0.893 (.003)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.055	0.053	0.037	0.044	0.053 (.001)
Na	0.111	0.114	0.105	0.099	0.118 (.007)
K	0.001	0.001	0.000	0.001	0.001
Rb	0.031	0.029	0.020	0.016	0.028 (.004)
Cs	0.651	0.650	0.691	0.705	0.649 (.003)
Ca	0.000	0.001	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.894	0.887	0.895	0.913	0.893 (.003)
Σ charges	0.794	0.796	0.816	0.821	0.796 (.002)
Si/Al	2.306	2.332	2.321	2.252	2.311 (.012)
CRK	86.1	85.6	87.1	87.9	85.1 (0.8)

1. Homogeneous.
  8. Homogeneous.
  3. Cs-rich veinlet.
  4. Cs-rich veinlet.
- Average of homogeneous pollucite (N = 6).

### 5.12 Locality X, Norway, Maine

Besides BB # 7, the only other Norway pollucite locality is at the northwest corner of Cobble Hill about 3 km southwest of Mt. Marie (C. Francis, personal communication 1991). The composition of this specimen is different from BB # 7, so this may be a specimen from Cobble Hill.

Sample number: PA-031 (I. Cerná, M47); 1 thin section, 8 analyses.

#### 5.12.1 Properties, associations and alteration

In hand specimen, the pollucite is clear and colourless, and vitreous in lustre. It is crosscut by two sets of thin white veins, intersecting at an angle of about 30°. The veins are sub-parallel and spaced 1 to 3 mm apart.

In thin section, the pollucite contains a grain of quartz 4 mm in size. An irregular vein of microcline (+ mica ± tourmaline) is present. One set of the veins contain a core of medium-grained mica (± albite) grading to coarser-grained mica towards margins of the veins. The veins have a constant width of up to 1 mm and run for several cm in length before pinching out within the pollucite. The later, second set of veins contains only very fine-grained mica.

### 5.12.2 Textural relationships and composition

The pollucite is almost completely homogeneous. Some Cs-rich veinlets + quartz are present.

The homogeneous pollucite has Si/Al from 2.39 to 2.47 and CRK from 81.0 to 81.5; the average and additional representative compositions are given in Table 5.12-1. The Cs-rich veinlets have Si/Al as low as 2.37 and CRK as high as 84.4.

Table 5.12-1: Composition of pollucite from Locality X, Norway, Maine.

No.	22	26	20	25	Average
SiO <sub>2</sub>	46.29	47.05	44.90	45.53	47.77 (.14)
Al <sub>2</sub> O <sub>3</sub>	16.20	15.96	15.96	16.10	16.02 (.05)
Fe <sub>2</sub> O <sub>3</sub>	0.01	0.00	0.01	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.31	0.33	0.02	0.31	0.33 (.01)
Na <sub>2</sub> O	1.64	1.69	1.18	1.41	1.66 (.01)
K <sub>2</sub> O	0.04	0.01	0.03	0.02	0.02
Rb <sub>2</sub> O	0.30	0.26	0.29	0.27	0.29
Cs <sub>2</sub> O	32.65	32.37	35.42	34.33	32.52 (.20)
CaO	0.02	0.02	0.02	0.02	0.02
MgO	0.01	0.01	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	97.47	97.69	97.82	97.97	97.62 (.26)
Si	2.127	2.144	2.123	2.118	2.138 (.003)
Al	0.878	0.857	0.889	0.883	0.863 (.004)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.012	0.013	0.001	0.012	0.013 (.001)
Na	0.146	0.149	0.108	0.127	0.148 (.003)
K	0.002	0.000	0.002	0.001	0.001
Rb	0.009	0.008	0.009	0.008	0.009 (.002)
Cs	0.640	0.629	0.714	0.681	0.634 (.003)
Ca	0.001	0.001	0.001	0.001	0.001
Mg	0.001	0.000	0.000	0.000	0.000
Al + Fe	0.878	0.857	0.889	0.883	0.863 (.004)
Σ charges	0.813	0.801	0.836	0.831	0.807 (.003)
Si/Al	2.392	2.466	2.385	2.368	2.443 (.013)
CRK	81.5	81.0	86.9	84.4	81.2 (0.1)

22. Homogeneous.

26. Homogeneous.

20. Cs-rich veinlet.

25. Cs-rich veinlet.

Average of homogeneous pollucite (N = 6).



### 5.13 Locality Y, Norway, Maine

Sample number: PA-078 (I. Cerná, no M-number); 1 thin section, 18 analyses. The specific locality of this sample is not known. Its association, alteration, and composition are different from BB # 7 and Locality Y, Norway, Maine.

#### 5.13.1 Properties, associations and alteration

In hand specimen, the pollucite is colourless with a vitreous lustre. It is associated with 1 cm cleavelandite, and slightly altered by buff clays. It is crosscut by white veins 1 to 3 mm in width.

In thin section, the pollucite contains a 1mm vein of coarse-grained mica (lepidolite?). Later veins 0.5 to 3 mm wide of fine-grained mica, are partially replaced by spodumene. The spodumene is coarse-grained, and altered near grain margins to a symplectitic intergrowth with quartz (Fig. 5.13-1). Bands of pollucite 1 mm wide adjacent to the spodumene veins has a high index of refraction and abundant micro-inclusions. The veins are overgrown by adularia, light brown in thin section.

#### 5.13.2 Textural relationships

Most of the pollucite is homogeneous. It is slightly

enriched in  $\text{Na}^+$  near fractures. Some Cs-rich veinlets + quartz (Fig. 5.13-2a) are cross-cut by later veins of mica (+ spodumene). Pollucite adjacent to spodumene is Cs-rich and contains grains of quartz and a  $\text{KAlSi}$ -bearing phase, 1 to 3  $\mu\text{m}$  in size (Fig. 5.13-2b).

### 5.13.3 Compositional trends

A plot of  $\text{Si}/\text{Al}$  vs. CRK is shown in Figure 5.13-3a. The homogeneous pollucite has  $\text{Si}/\text{Al}$  from 2.49 to 2.55 and CRK from 84.5 to 86.6; the average and additional representative compositions are given in Table 5.13-1. The Na-enriched compositions have CRK as low as 81.7 and  $\text{Si}/\text{Al}$  in the same range as the homogeneous pollucite. The Cs-rich veinlets have  $\text{Si}/\text{Al}$  as low as 2.26 and CRK as high as 94.2. Pollucite adjacent to spodumene has  $\text{Si}/\text{Al}$  as low as 2.20 and CRK as high as 96.2.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.13-3b. The sum of cation charges follows a trend of Al increasing with CRK.

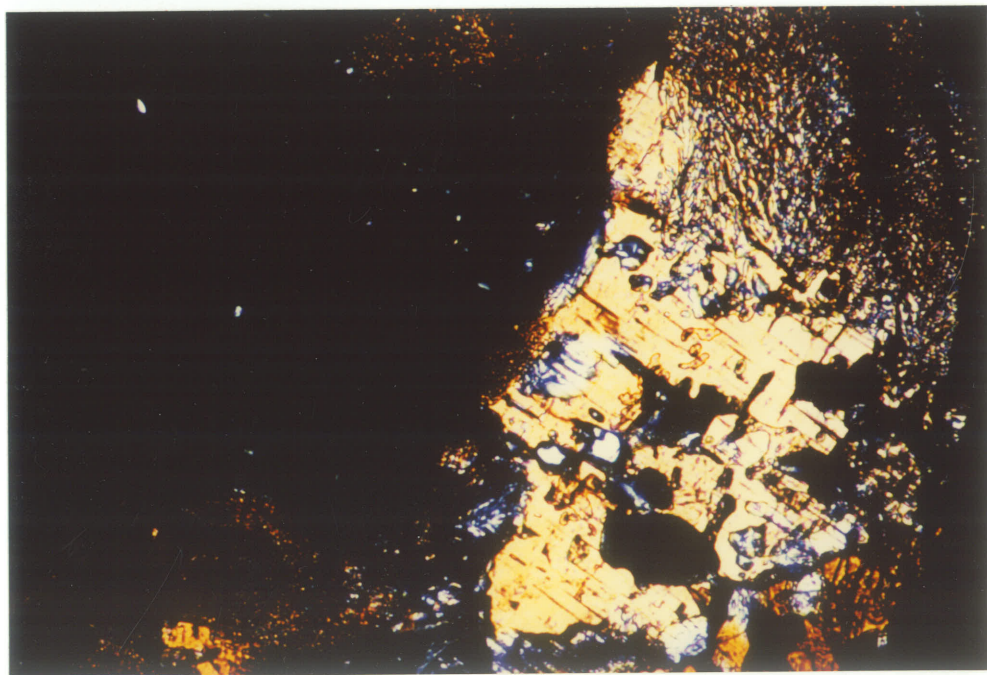
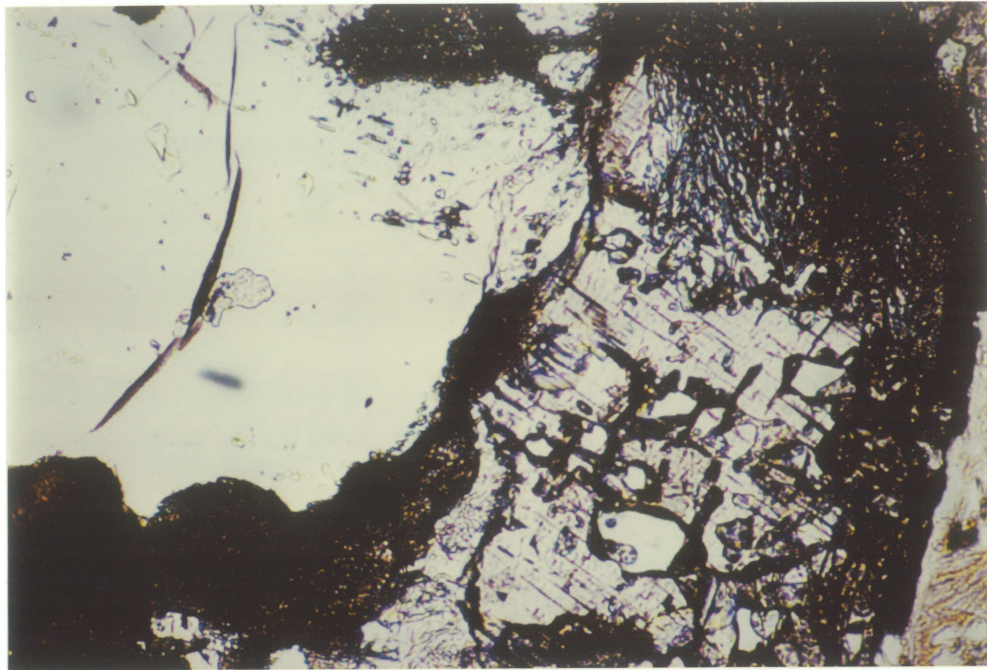


Fig. 5.13-1. Microphotographs of Locality Y, Norway, showing a crystal of spodumene (right), altered at its margins to a symplectitic intergrowth with quartz. The crystal is part of a vein which crosscuts pollucite. a) Plane-polarized light. b) Crossed-polarized light.

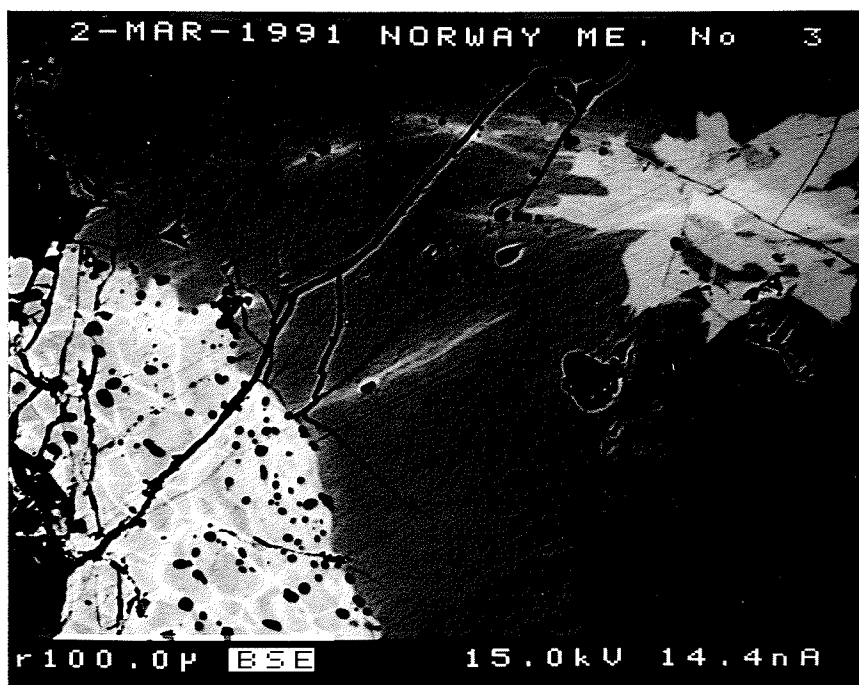
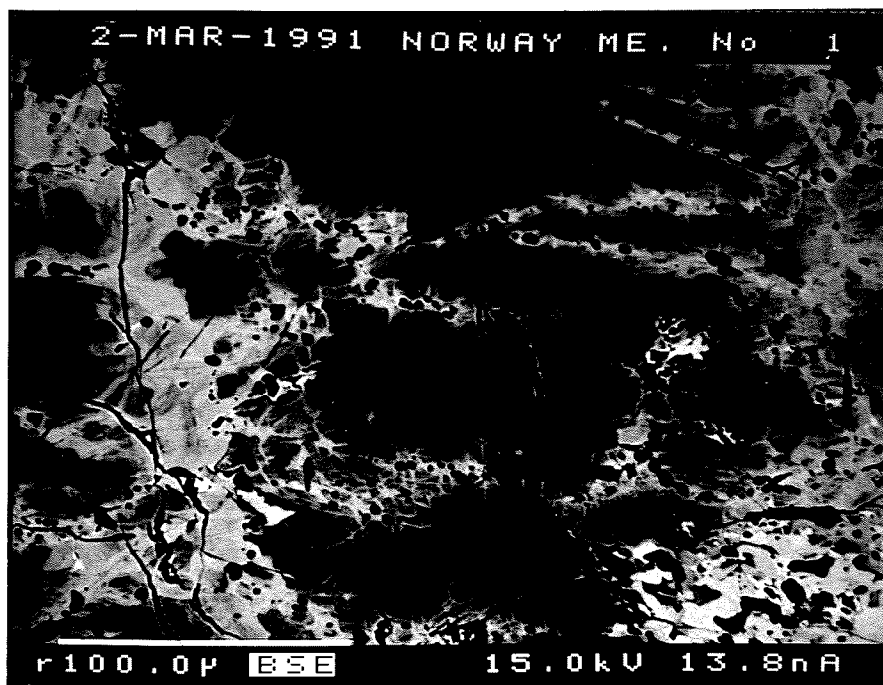


Fig. 5.13-2. BSE images of Locality Y, Norway pollucite. a) Veinlets of quartz-containing Cs-rich pollucite. b) Cs-rich pollucite containing quartz and a KAlSi-bearing mineral adjacent to lath spodumene.

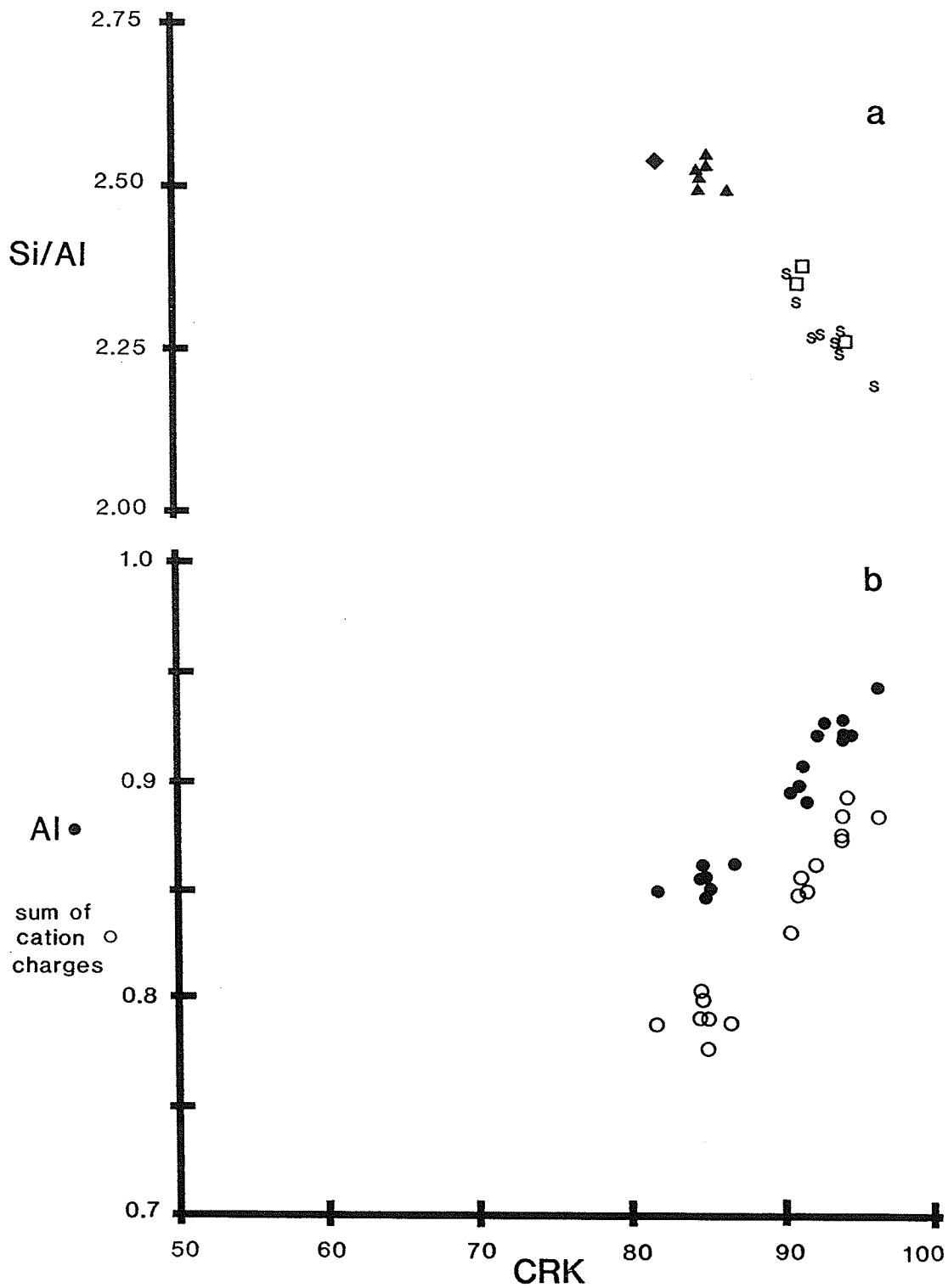


Fig. 5.13-3. Compositional trends for Locality Y, Norway, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Cs-rich pollucite adjacent to spodumene (S).

Table 5.13-1: Composition of pollucite from Locality Y, Norway, Maine.

No.	1	2	6	18	Average
SiO <sub>2</sub>	46.72	47.39	42.71	42.00	46.76 (.35)
Al <sub>2</sub> O <sub>3</sub>	15.77	15.80	16.01	16.24	15.76 (.04)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.03	0.01	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.22	0.21	0.00	0.00	0.21 (.05)
Na <sub>2</sub> O	1.37	1.63	0.54	0.36	1.32 (.08)
K <sub>2</sub> O	0.02	0.02	0.02	0.01	0.02
Rb <sub>2</sub> O	0.24	0.10	0.21	0.24	0.26 (.02)
Cs <sub>2</sub> O	34.13	32.94	39.97	40.04	33.82 (.28)
CaO	0.00	0.01	0.00	0.00	0.01
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.46	98.13	99.46	98.90	98.15 (.29)
Si	2.148	2.157	2.086	2.071	2.151 (.006)
Al	0.854	0.848	0.922	0.943	0.854 (.006)
Fe	0.000	0.001	0.000	0.001	0.000
P	0.009	0.008	0.000	0.001	0.008 (.002)
Na	0.123	0.144	0.052	0.034	0.118 (.007)
K	0.001	0.001	0.001	0.001	0.001
Rb	0.007	0.003	0.007	0.008	0.008 (.001)
Cs	0.669	0.639	0.833	0.841	0.657 (.024)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.854	0.849	0.922	0.944	0.854 (.006)
Σ charges	0.799	0.787	0.892	0.884	0.807 (.032)
Si/Al	2.514	2.543	2.263	2.195	2.518 (.075)
CRK	84.7	81.7	94.2	96.2	85.1 (0.8)

1. Homogeneous.
  2. Na-enriched.
  6. Cs-rich veinlet.
  18. Cs-rich adjacent to spodumene.
- Average of homogeneous pollucite (N = 6).

#### 5.14 Old Tom Mt., Greenwood, Maine

The specific locality for this sample from Old Tom Mt. (a.k.a. Uncle Tom Mt.) is the Emmons mine.

Sample number: PA-134 (M. Wise, U.S.N.M. 126152); 2 thin sections, 14 analyses.

##### 5.14.1 Properties, associations and alteration

In hand specimen, the pollucite is white to colourless. It is crosscut by very thin, sub-parallel white veins.

In thin section, the veins are irregular and contain mica. Round aggregates of adularia replace the pollucite, adjacent to the mica veins.

##### 5.14.2 Textural relationships

The pollucite is mostly homogeneous. It is slightly Na-enriched near fractures. Sporadic quartz-containing Cs-rich veinlets are present (Fig. 5.14-1). Pollucite adjacent to and included within the adularia is Cs-rich.

### 5.14.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.14-2a. The homogeneous pollucite has Si/Al from 2.47 to 2.53 and CRK from 85.2 to 87.8; the average and additional representative compositions are given in Table 5.14-1. The Na-enriched compositions have CRK as low as 76.3 and the same Si/Al as the homogeneous pollucite. The Cs-rich veinlets have Si/Al as low as 2.35 and CRK as high as 92.3. Pollucite inclusions in adularia have Si/Al as low as 2.10 and CRK as high as 97.2.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.14-2b. The sum of cation charges follows a trend of increasing Al with CRK, except for the Na-enriched pollucite, in which Al is constant with CRK.





Fig. 5.14-1. BSE image of Old Tom pollucite. Veinlet of Cs-rich pollucite within homogeneous pollucite. Pollucite is enriched in Na along a fracture.

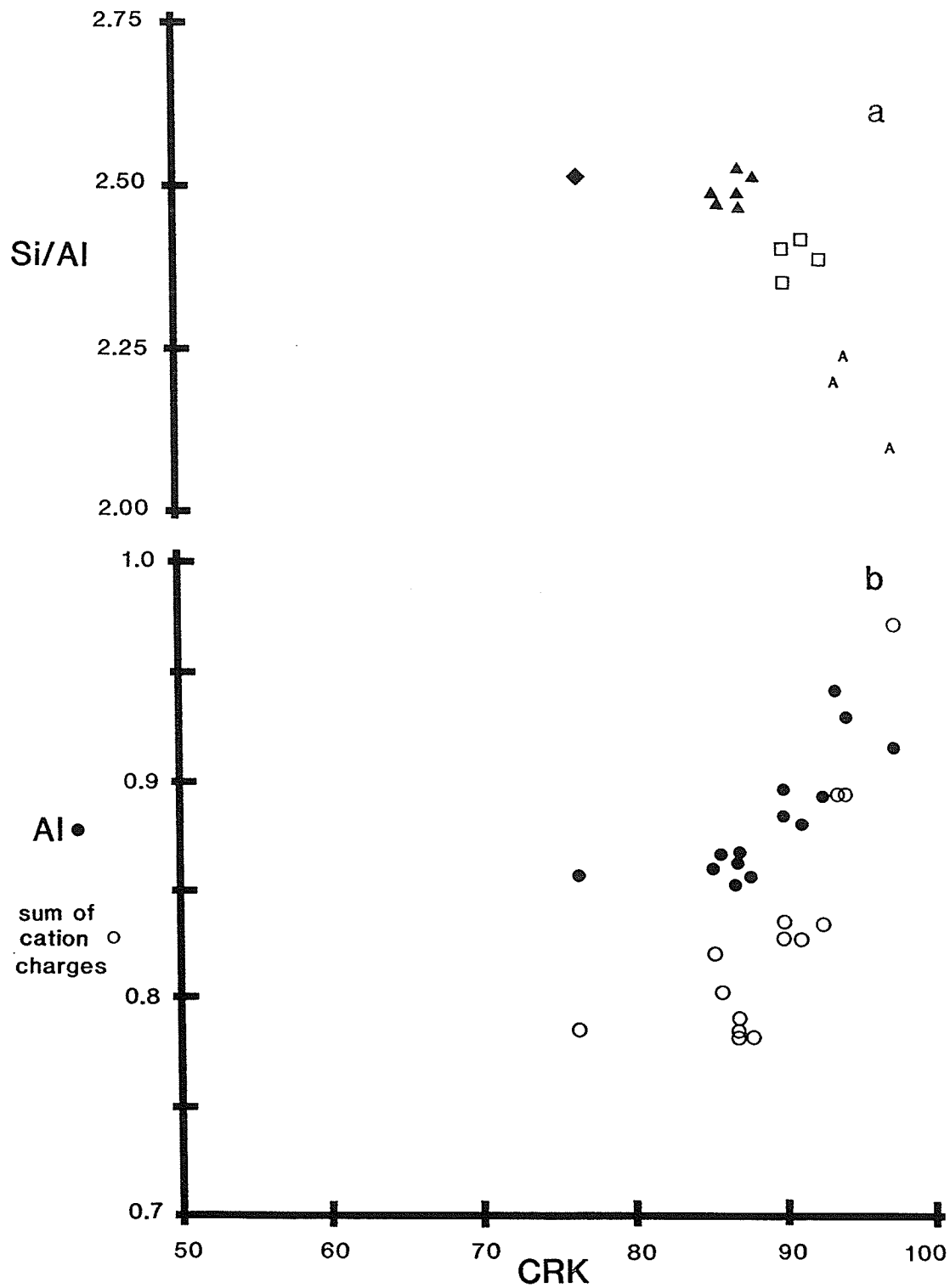


Fig. 5.14-2. Compositional trends for Old Tom Mt., Greenwood, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Inclusions of Cs-rich pollucite in adularia (A).

Table 5.14-1: Composition of pollucite from Old Tom Mt, Greenwood, Maine.

No.	2	5	14	9	Average
SiO <sub>2</sub>	46.52	47.56	44.20	40.36	46.60 (.41)
Al <sub>2</sub> O <sub>3</sub>	15.87	16.05	15.73	16.27	15.86 (.08)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.35	0.32	0.00	0.00	0.31 (.03)
Na <sub>2</sub> O	1.17	2.13	0.69	0.26	1.20 (.09)
K <sub>2</sub> O	0.05	0.01	0.04	0.06	0.09
Rb <sub>2</sub> O	0.61	0.02	0.55	0.00	0.60 (.03)
Cs <sub>2</sub> O	33.66	31.04	36.64	41.01	33.80 (.29)
CaO	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.22	97.13	97.85	97.97	98.47 (.47)
Si	2.141	2.148	2.124	2.043	2.142 (.007)
Al	0.860	0.856	0.891	0.886	0.859 (.006)
Fe	0.000	0.000	0.000	0.000	0.000
P	0.014	0.012	0.000	0.000	0.012 (.001)
Na	0.104	0.186	0.064	0.026	0.107 (.009)
K	0.003	0.000	0.002	0.004	0.005
Rb	0.018	0.001	0.017	0.000	0.018 (.006)
Cs	0.661	0.598	0.751	0.886	0.663 (.004)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.860	0.856	0.891	0.886	0.859 (.007)
Σ charges	0.785	0.785	0.833	0.915	0.793 (.015)
Si/Al	2.489	2.514	2.385	2.104	2.493 (.024)
CRK	86.8	76.3	92.3	97.2	86.5 (1.0)

2. Homogeneous.

5. Na-enriched.

14. Cs-rich veinlet.

9. Inclusion in adularia.

Average of homogeneous pollucite (N = 6).

### 5.15 Locality X, Greenwood, Maine

This sample is likely from Old Tom, Greenwood, as the alteration and composition are so similar to that locality.

Sample number: PA-006 (C. Frondel, Harvard 92864); 1 thin section, 12 analyses.

#### 5.15.1 Properties, associations and alteration

In hand specimen, the pollucite is colourless and has a vitreous lustre. It contains a mica grain, and a 2 mm white grain.

In thin section, the white grain is identified as a round granular aggregate of adularia, embedded in the pollucite. The pollucite contains a very thin vein of fine-grained mica.

#### 5.15.2 Textural relationships

The pollucite is mostly homogeneous. It contains some Cs-enriched blebs and Cs-rich veinlets (+ quartz). Pollucite adjacent to and enclosed in the adularia grain is Cs-rich.

### 5.15.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.15-1a. The homogeneous pollucite has Si/Al from 2.49 to 2.56 and CRK from 87.7 to 88.4; the average and additional representative compositions are given in Table 5.15-1. The Cs-enriched blebs have Si/Al as low as 2.46 and CRK as high as 93.0. The Cs-rich veinlets have Si/Al as low as 2.40 and CRK as high as 92.8. Pollucite inclusions in adularia have Si/Al as low as 2.16 and CRK as high as 99.0.

A plot of Al (+ Fe), and sum of cation charges vs. CRK is shown in Figure 5.15-1b. The sum of cation charges follows a trend of increasing Al with CRK.

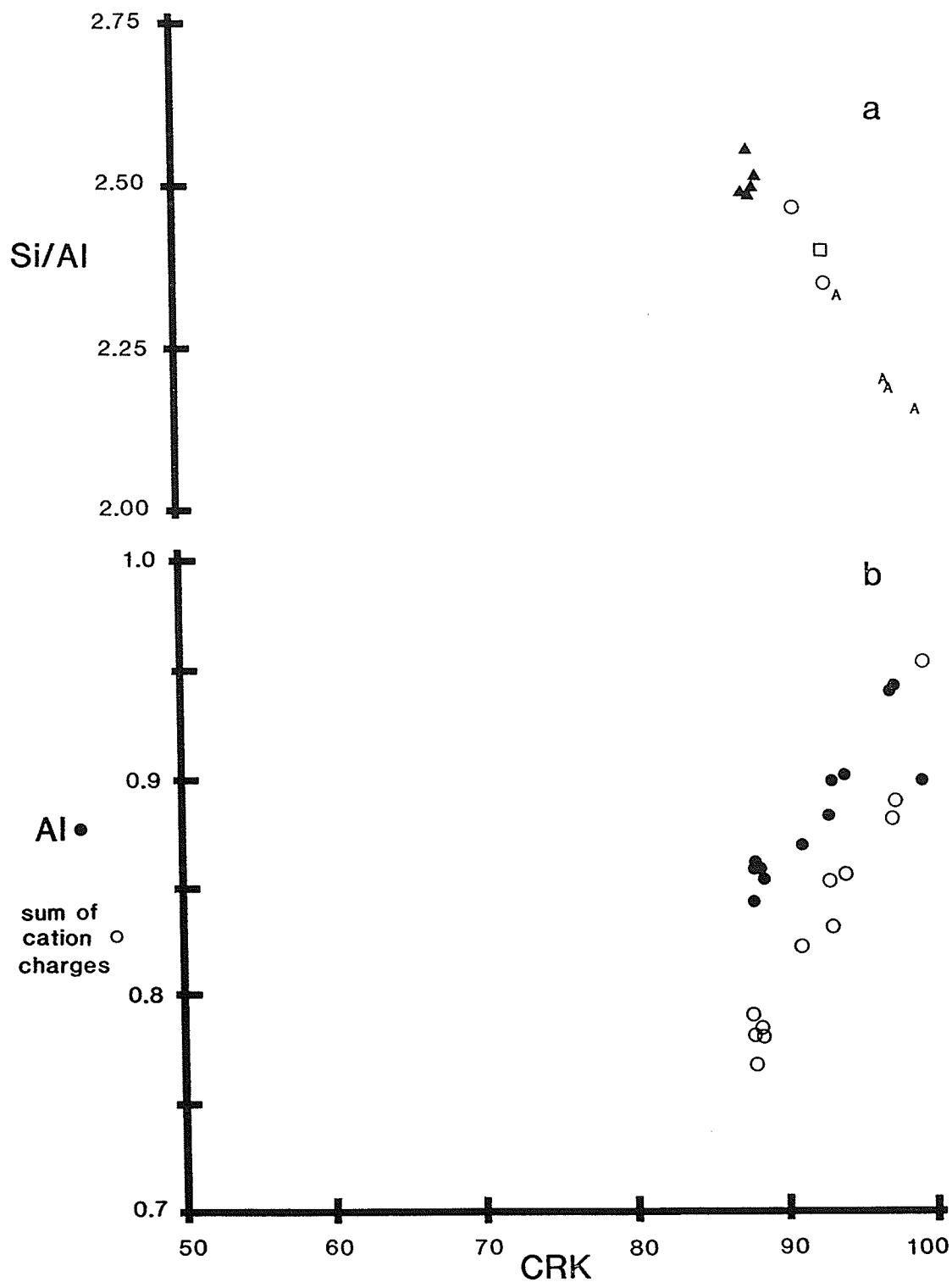


Fig. 5.15-1. Compositional trends for Locality X, Greenwood, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3. Inclusions of Cs-rich pollucite in adularia (A).

Table 5.15-1: Composition of pollucite from Locality X, Greenwood, Maine.

No.	17	22	21	27	Average
SiO <sub>2</sub>	46.36	44.47	44.36	41.05	46.72 (.48)
Al <sub>2</sub> O <sub>3</sub>	15.66	16.06	15.69	16.17	15.82 (.11)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.02	0.00	0.02	0.00
P <sub>2</sub> O <sub>5</sub>	0.36	0.00	0.02	0.00	0.34 (.01)
Na <sub>2</sub> O	1.01	0.62	0.66	0.10	1.06 (.03)
K <sub>2</sub> O	0.01	0.01	0.01	0.04	0.01
Rb <sub>2</sub> O	0.52	0.46	0.56	0.37	0.53 (.05)
Cs <sub>2</sub> O	34.14	37.49	37.92	41.05	34.33 (.24)
CaO	0.00	0.01	0.00	0.00	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.06	99.14	99.21	98.82	98.81 (.47)
Si	2.147	2.116	2.122	2.058	2.146 (.007)
Al	0.854	0.900	0.884	0.879	0.856 (.008)
Fe	0.000	0.001	0.000	0.001	0.000
P	0.014	0.000	0.001	0.000	0.013 (.001)
Na	0.091	0.057	0.061	0.009	0.094 (.002)
K	0.001	0.000	0.000	0.002	0.001
Rb	0.016	0.014	0.018	0.012	0.016 (.001)
Cs	0.674	0.760	0.774	0.879	0.672 (.008)
Ca	0.000	0.001	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.854	0.901	0.884	0.880	0.856 (.008)
Σ charges	0.782	0.833	0.853	0.901	0.782 (.008)
Si/Al	2.513	2.350	2.400	2.155	2.507 (.030)
CRK	88.4	93.0	92.8	99.0	88.0 (0.3)

17. Homogeneous.

22. Cs-enriched bleb.

21. Cs-rich veinlet.

27. Inclusion in adularia.

Average of homogeneous pollucite (N = 5).

### 5.16 Locality Y, Greenwood, Maine

The Tamminen quarry (Ch. 5.17) is only 150 m north of the Harvard quarry at the base of Noyes Mt.. Landes (1925) described a quartz pseudomorph after pollucite from the top of Noyes Mt.. From the base of Noyes Mt., Richmond & Gonyer (1938) described massive pollucite and crystals of "pollucite" replacing petalite. The crystals are a secondary generation, in this sample not directly contacting the primary pollucite is described here. This sample is probably from the same material analysed by Richmond & Gonyer (1938) (No. 62 in Table 2.1k). Ahrens (1947) determined in pollucite from Greenwood (as wt.%): (0.16)  $K_2O$ , (0.68)  $Rb_2O$ , (0.008)  $Li_2O$ , (0.0019)  $Tl_2O$ , (0.0005)  $Ga_2O_3$  and trace Sr.

Sample number: PA-005 (C. Frondel, Harvard 94630); 1 thin section, 1 grain mount, 16 analyses.

#### 5.16.1 Properties, associations and alteration

In hand specimen, the pollucite is white to colourless. In thin section, the pollucite is cut by an irregular vein of microcline, 1 to 2 mm wide. Later veins of fine-grained mica are partly replaced by fine-grained spodumene.



### 5.16.2 Textural relationships

The pollucite contains relatively large homogeneous areas. Near fractures and veins the pollucite is enriched in  $\text{Na}^+$ . Locally the pollucite contains a blebby matrix of Na- and Cs-enriched domains. Rarely, very thin Cs-rich veinlets are present.

### 5.16.3 Compositional trends

A plot of Si/Al vs. CRK is shown in Figure 5.16-1a. The homogeneous pollucite has Si/Al from 2.46 to 2.51 and CRK from 75.8 to 80.6; the average and additional representative compositions are given in Table 5.16-1. The areas enriched in  $\text{Na}^+$  have CRK as low as 67.0 and Si/Al in the same range as the homogeneous pollucite. Na-enriched blebs have Si/Al as high as 2.44 and CRK as low as 69.9. Cs-enriched blebs have Si/Al as low as 2.18 and CRK as high as 82.1.

A plot of Al (+ Fe), and sum of cation charges is shown in Figure 5.16-1b. The sum of cation charges follow a trend of Al constant with CRK, except for the Cs-enriched bleb which has high Al. The Na-enriched pollucite shows an apparent alkali deficit due to sample burning.

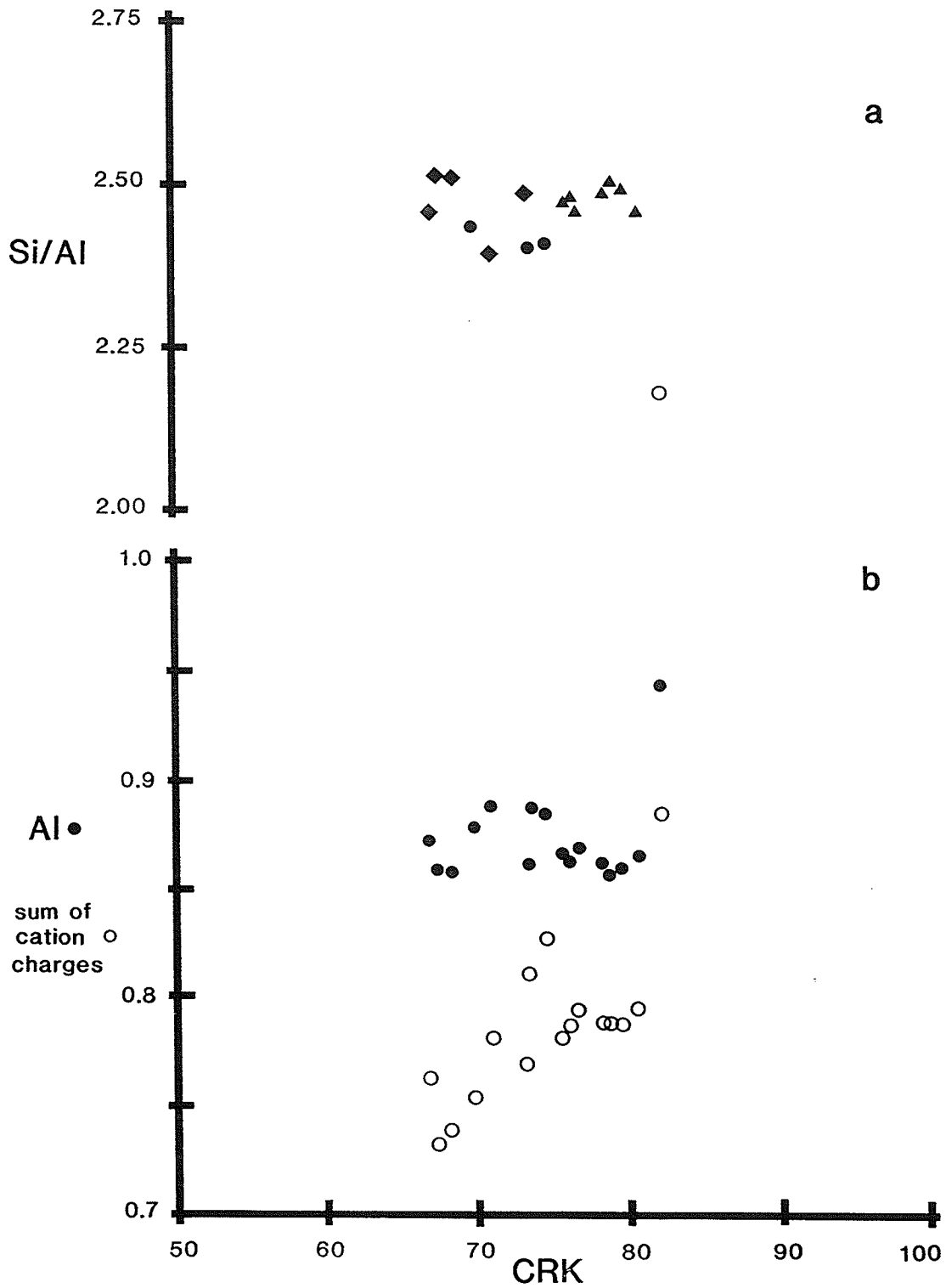


Fig. 5.16-1. Compositional trends for Locality Y, Greenwood, Maine. a) Si/Al vs. CRK, and b) Al (+ Fe), and sum of cation charges vs. CRK. Symbols as in Figure 3.1-3.

Table 5.16-1: Composition of pollucite from Locality Y, Greenwood, Maine.

No.	32	30	36	35	Average
SiO <sub>2</sub>	47.57	50.54	48.78	43.32	47.64 (.34)
Al <sub>2</sub> O <sub>3</sub>	16.19	17.05	16.99	16.82	16.30 (.16)
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.02	0.00	0.00
P <sub>2</sub> O <sub>5</sub>	0.34	0.35	0.36	0.19	0.34 (.02)
Na <sub>2</sub> O	1.86	2.87	2.67	1.71	1.99 (.17)
K <sub>2</sub> O	0.01	0.01	0.02	0.01	0.01
Rb <sub>2</sub> O	0.29	0.00	0.00	0.00	0.10
Cs <sub>2</sub> O	32.15	27.13	28.20	35.79	31.95 (.52)
CaO	0.00	0.01	0.00	0.00	0.00
MgO	0.01	0.01	0.00	0.00	0.00
H <sub>2</sub> O	----	----	----	----	----
sum	98.41	97.96	97.02	97.84	98.35 (.28)
Si	2.143	2.158	2.137	2.061	2.140 (.005)
Al	0.859	0.858	0.878	0.943	0.863 (.005)
Fe	0.000	0.000	0.001	0.000	0.000
P	0.013	0.013	0.013	0.008	0.013 (.001)
Na	0.162	0.238	0.227	0.158	0.174 (.014)
K	0.001	0.000	0.001	0.001	0.001
Rb	0.009	0.000	0.000	0.000	0.003
Cs	0.618	0.494	0.526	0.726	0.612 (.013)
Ca	0.000	0.000	0.000	0.000	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe	0.859	0.858	0.878	0.943	0.863 (.005)
Σ charges	0.789	0.732	0.754	0.886	0.789 (.005)
Si/Al	2.494	2.515	2.435	2.186	2.480 (.018)
CRK	79.5	67.5	69.9	82.1	78.0 (1.8)

32. Homogeneous.

30. Na-enriched.

36. Na-enriched bleb.

35. Cs-enriched bleb.

Average of homogeneous pollucite (N = 7).

### 5.17 Tamminen Quarry, Greenwood, Maine

Pollucite in this locality is fully altered (M. Wise, personal communication 1991). An earlier pit is present to the north, also called the Tamminen, but is very small and barren.

Sample number: PA-135 (M. Wise, U.S.N.M. 126627); 1 thin section, 1 grain mount.

#### 5.17.1 Associations and alteration

In hand specimen, the pollucite pseudomorphs are associated with light blue albite grains, unaltered and 1cm in size. Thin near-parallel veins less than 0.5 mm wide spaced 2 to 4 mm apart are preserved in a matrix of white to buff clays.

In thin section, the veins consist of fine-grained spodumene and mica(?). They are locally overgrown by adularia(?). Between the veins, extremely fine-grained radial clusters of spodumene (1 to 3  $\mu\text{m}$  size), quartz, and clay minerals pseudomorph the original pollucite.

## 5.18 Locality X, Maine

Sample number: PA-012 (C. Guillemin); 1 thin section.

### 5.18.1 Associations and alteration

The pollucite is associated with quartz, and veined by quartz. Later veining by microcline is partly replaced by albite (+ mica), and fine-grained spodumene. This early veining is preserved within clay pseudomorphs after pollucite. As well, a 35  $\mu\text{m}$  sized round grain of K-feldspar in the associated quartz seems to be a pseudomorph after pollucite, because it bears the sub-rounded morphology of a trapezohedral(?) crystal of pollucite.

## CHAPTER 6

## DISCUSSION

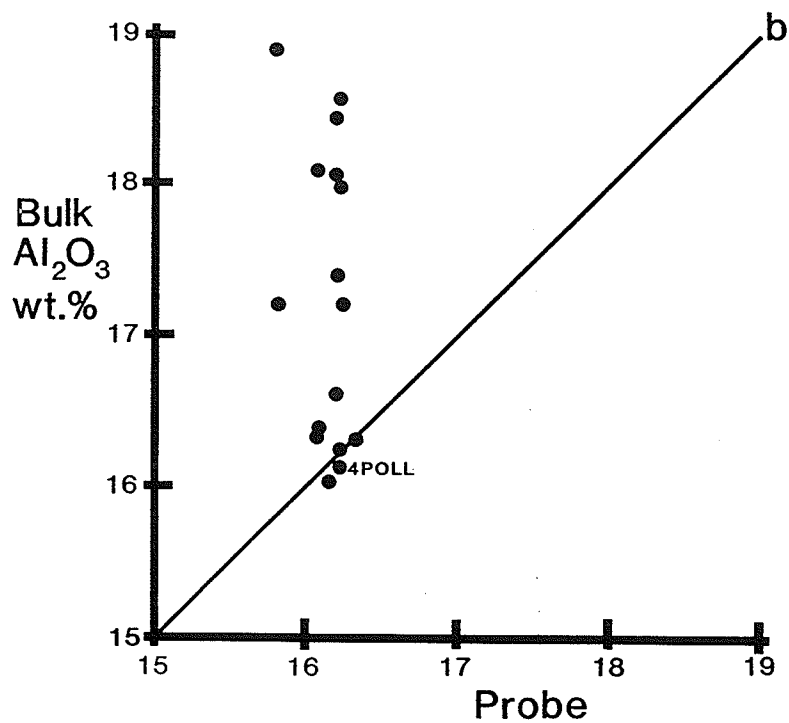
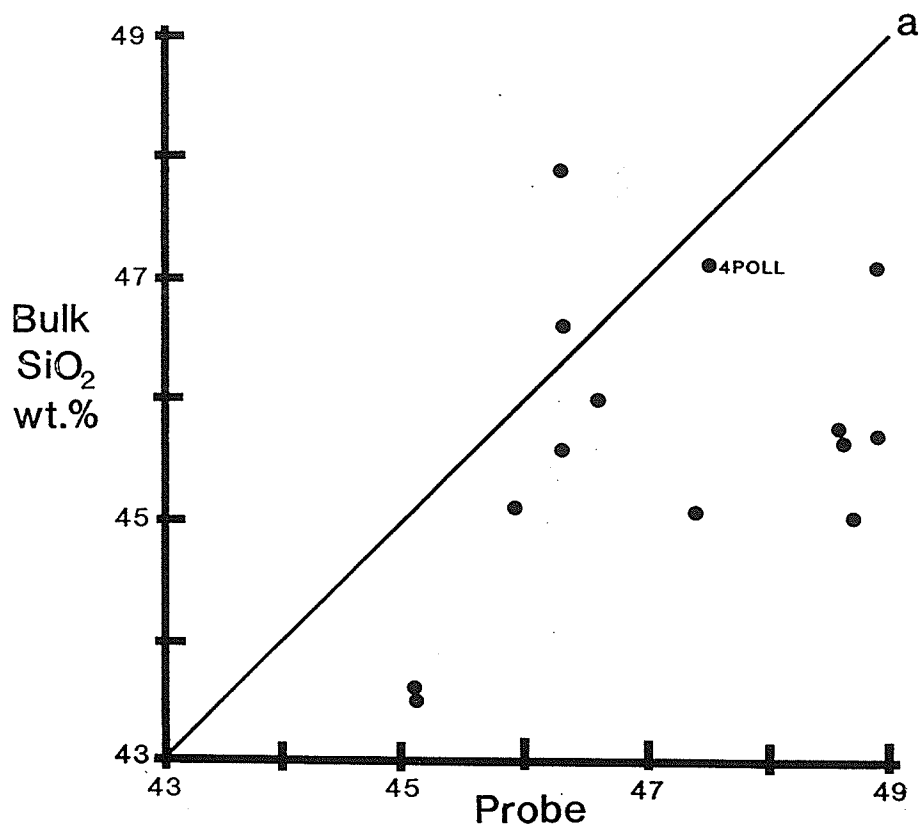
## 6.1 Quality of the results

The quality of individual probe analyses may be judged by comparison to results of bulk analyses of pollucite from the same locality. Knowledge of  $\text{Li}^+$  contents (from bulk compositions) is useful for estimates of alkali cation charge balance relative to Al. Comparison of individual oxide weights is also useful, but generally not as valuable as the sum of oxide weights (near 100). The probe results, however, do not include either  $\text{Li}^+$  or  $\text{H}_2\text{O}$  contents. In these analyses, no strong relation of the oxide total to  $\text{Na}^+$  content or CRK is found (e.g. see Helikon data); this could be due, however, to different volatilization dependent upon the composition and history of the sample. The most Cs-rich pollucite has totals near 100 suggesting it is anhydrous.

The overall quality of the probe results may be judged by comparison with bulk analyses from a number of localities. Average oxide weights of homogeneous pollucite (as determined by electron microprobe) are plotted against bulk (wet chemical) analyses reported in the literature. Relative to bulk compositions, probe  $\text{SiO}_2$  is systematically higher by 1 to 2 wt.% (Fig. 6.1-1a).  $\text{Al}_2\text{O}_3$  is lower by 0.5

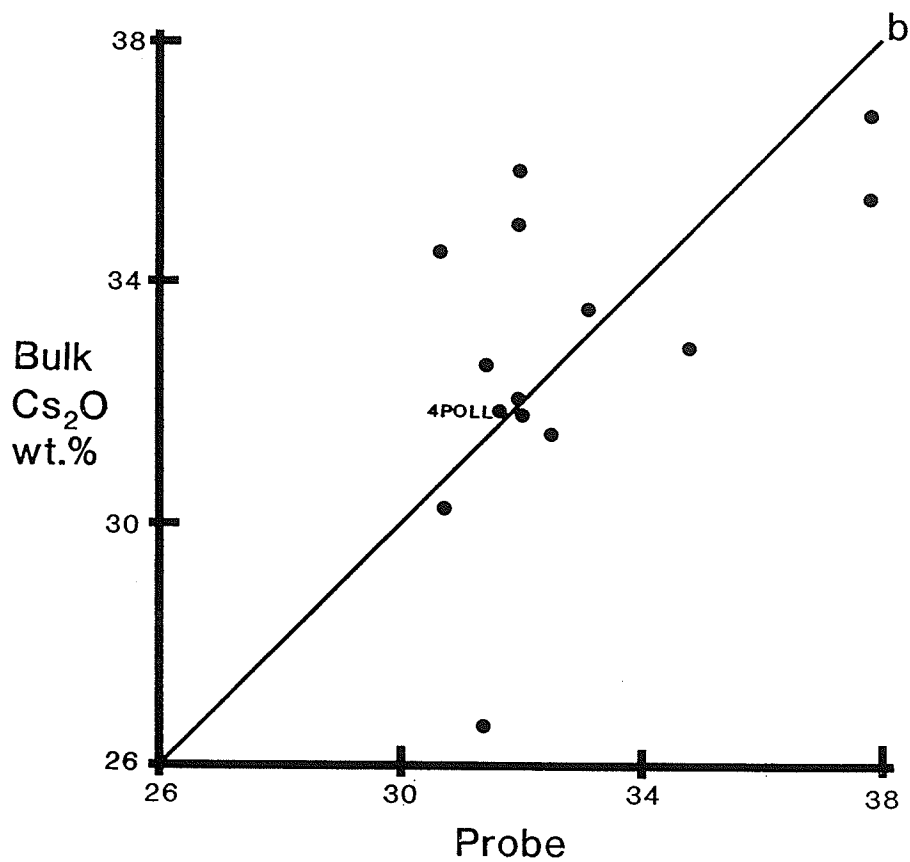
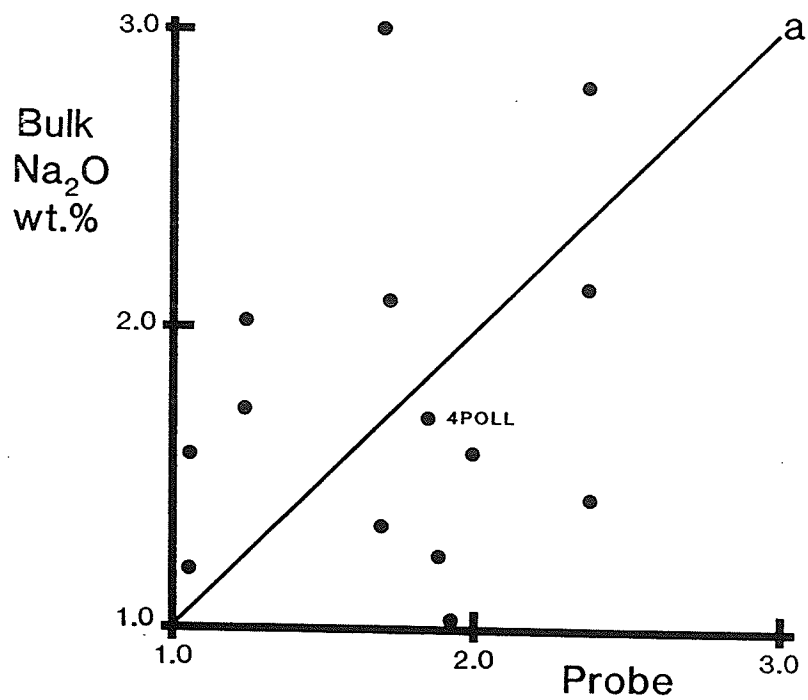
to 1.0 wt.% (Fig. 6.1-1b); however, a very limited weight range was found in the primary pollucite ( $16.0 \pm 0.2$ ) suggesting that the wet chemical methods exaggerate  $\text{Al}_2\text{O}_3$  contents. The determination of Al by wet chemical methods is dependent upon complete reaction in the method used, among other uncertainties. If these assumptions are correct, probe Si/Al ratios should be preferred to those based on wet chemical determinations of bulk samples.

Results for both  $\text{Cs}_2\text{O}$  and  $\text{Na}_2\text{O}$  generally agree with bulk compositions (Fig. 6.1-2a and b) suggesting no systematic shift in CRK. Each analytical approach has inherent difficulties in determination of accurate quantities (bulk wet determination of  $\text{Al}_2\text{O}_3$ , probe volatilization of sample); thus it is difficult to decide which method yields better results. It should be stressed, however, that despite any error in absolute determination, the data of this study are internally comparable to the 4POLL standard within the ranges of precision quoted in Table 3.1-1.



6.1-1. Bulk compositions from the literature vs. average probe compositions of the homogeneous pollucite, as wt.% oxide. a)  $\text{SiO}_2$ , and b)  $\text{Al}_2\text{O}_3$ .





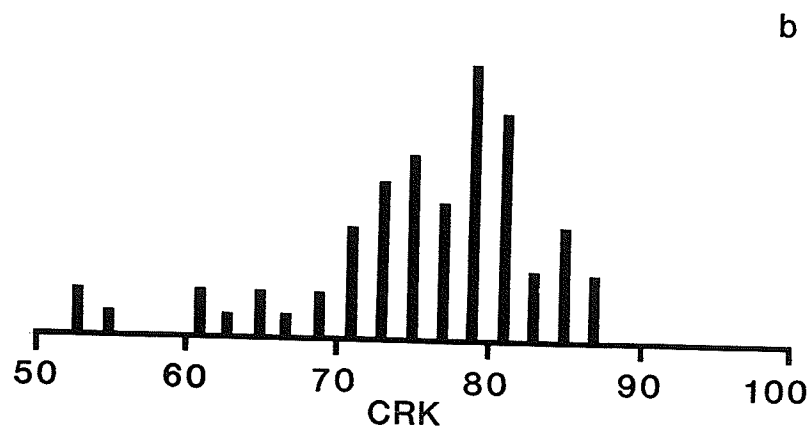
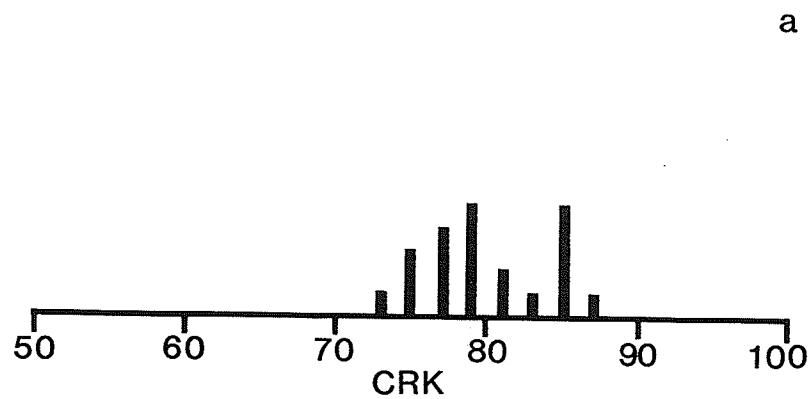
6.1-2. Bulk compositions from the literature vs. average probe compositions of the homogeneous pollucite, as wt.% oxide. a)  $\text{Na}_2\text{O}$ , and b)  $\text{Cs}_2\text{O}$ .

## 6.2 Composition of the primary pollucite

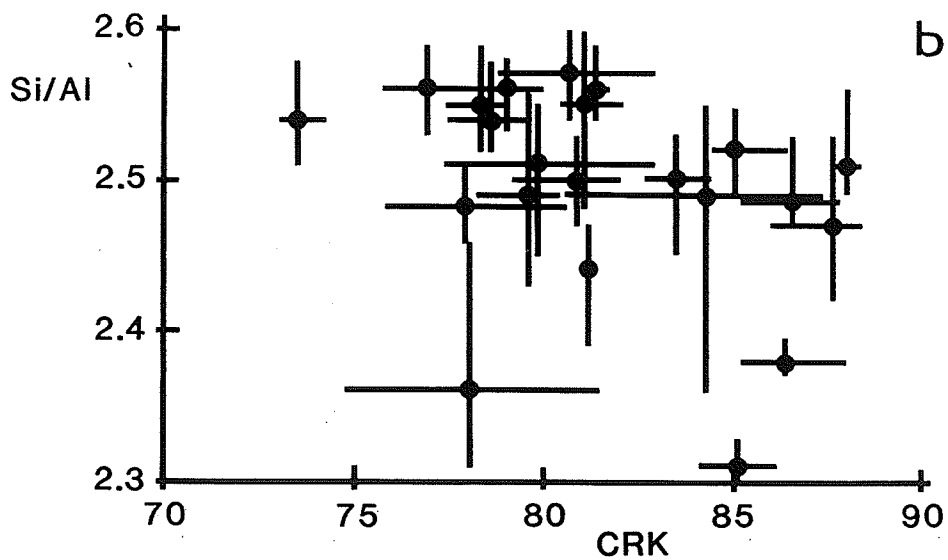
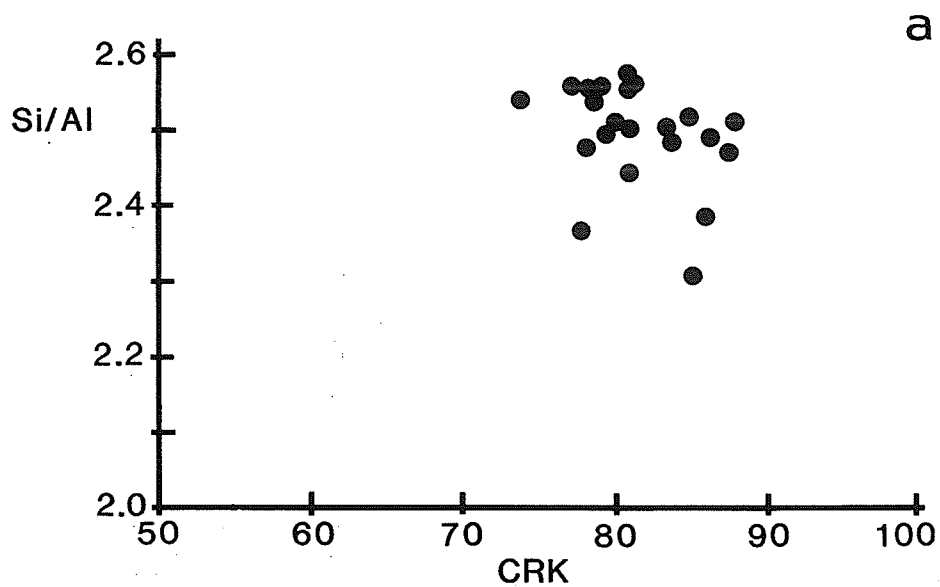
The range in CRK of the homogeneous pollucite is given in Figure 6.2-1a for the African and U.S. localities. Most localities have CRK near 80, however, the distribution seems to be bimodal. One group has CRK from 74 to 82 with most localities nearest 80. A second group has CRK centering on 85 and as high as 88. No primary pollucite with CRK less than 70 was found. The histogram of bulk compositions is duplicated (from Fig. 2.1-1a) in Figure 6.2-1b. Extensive enrichment in  $\text{Na}^+$  (such as observed at Tin Mt.) could have affected some of the bulk determinations, particularly those with CRK less than 70.

A plot of Si/Al vs. CRK is given in Figure 6.2-2a. The range of Si/Al (2.3 to 2.6) is smaller than that found in bulk analyses (2.0 to 2.6). Within individual localities the homogeneous pollucite has a range of Si/Al ratios (Fig. 6.2-2b) up to 0.19 and a range of CRK up to 6.6. There does not appear to be any zoning within individual samples of pollucite (or variation among samples from the same locality); however, mm-sized homogeneous areas of distinct composition are observed at some localities. This could be a feature of primary growth, or at some localities incipient breakdown to a very coarse blebby mosaic.

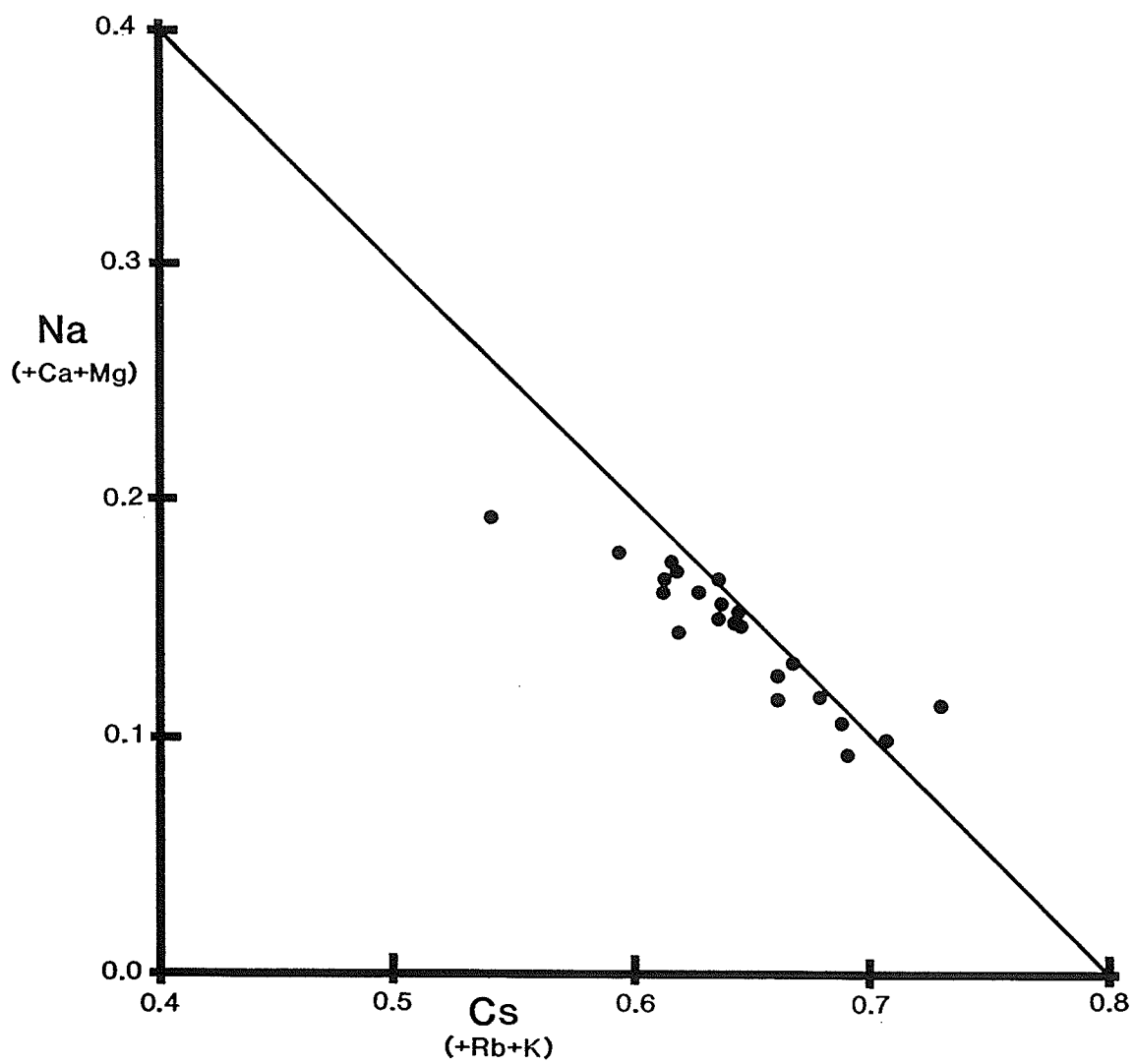
A plot of Na (+ Ca + Mg) vs. Cs (+ Rb + K), both atoms p.f.u., is shown in Figure 6.2-3. Most of the data are



6.2-1. Histogram of pollucite compositions. a) Averaged probe compositions of the homogeneous pollucite, and b) wet chemical bulk determinations from the literature.



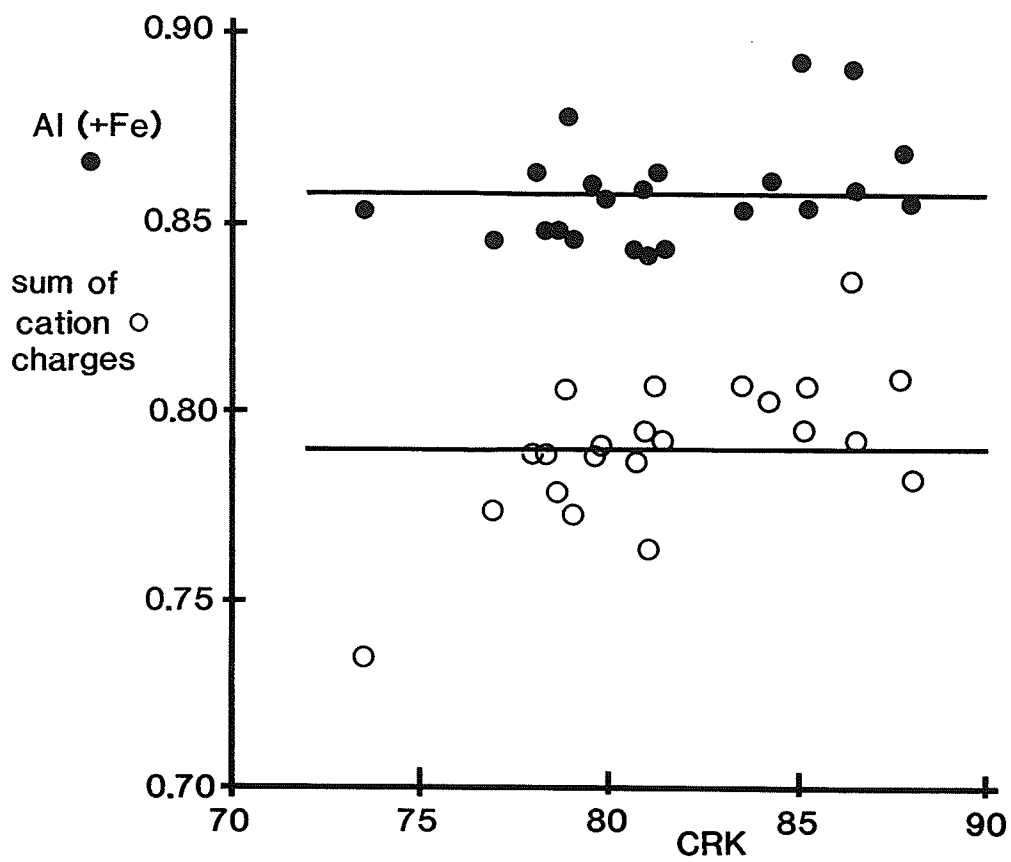
6.2-2. Plots of Si/Al vs. CRK for the homogeneous pollucite. a) average values shown as points, and b) expanded scale showing the ranges in Si/Al and CRK within each locality.



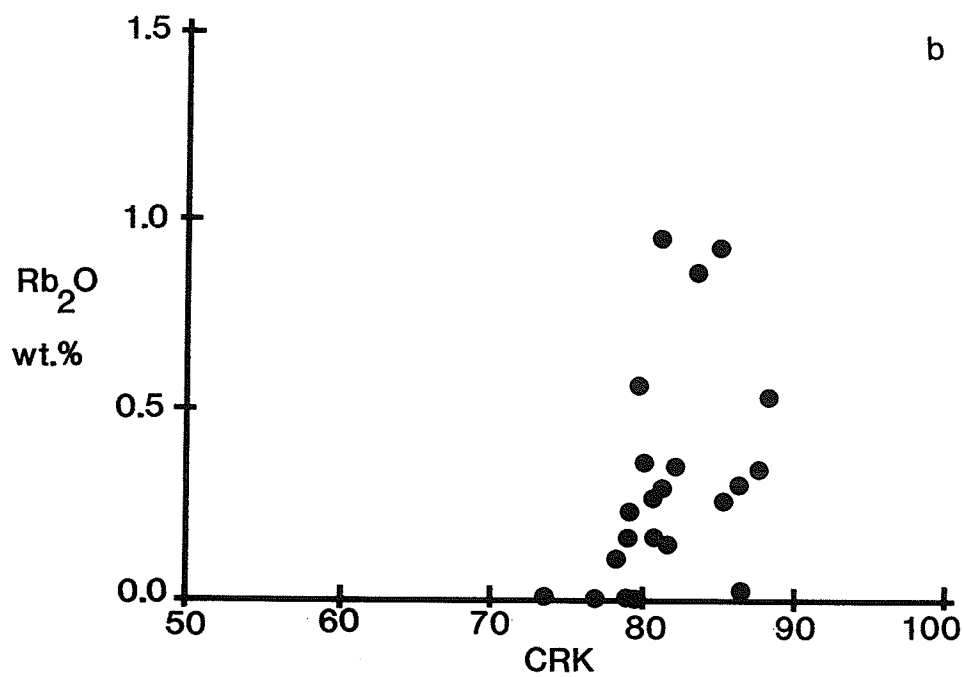
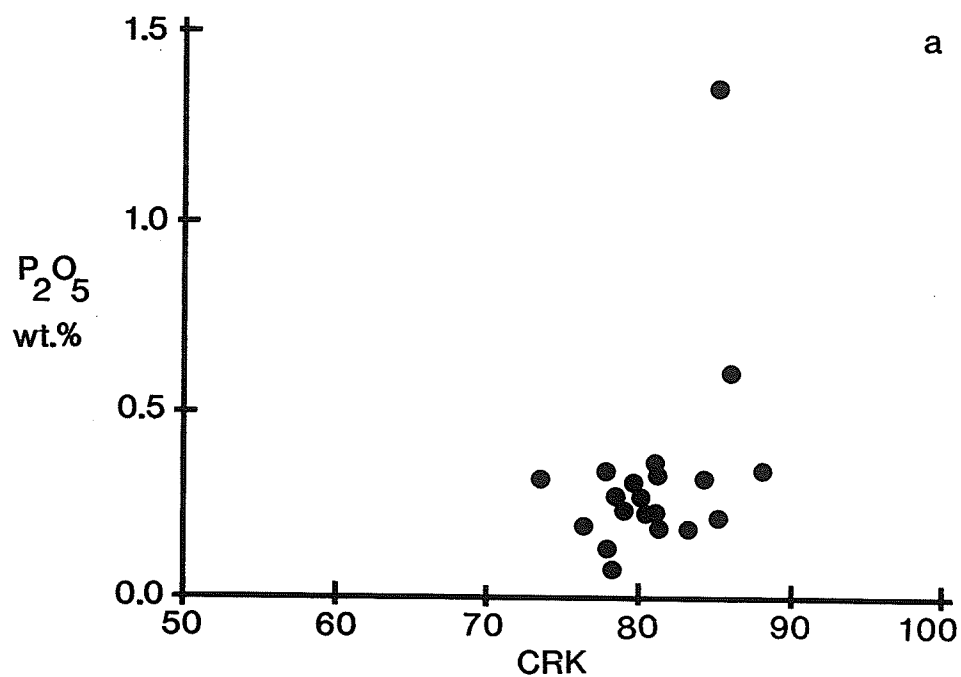
6.2-3. Plot of Na (+ Ca + Mg) vs. Cs (+ Rb + K) for the homogeneous pollucite (average of probe values).

distributed near a line of constant Al. A plot of Al (+ Fe), and sum of cation charges, both atoms p.f.u. vs. CRK is shown in Figure 6.2-4. Further evidence for a bimodal distribution is found here: the pollucite with CRK greater than 82 has higher Al than the less Cs-rich pollucite. The sum of cation charges follows this trend of Al. The average Al is 0.858 and the sum of cation charges is 0.791. The average alkali cation deficit relative to Al is 7.8% (or 0.067 monovalent atoms p.f.u.). This deficit may be accounted for by only 0.4 wt.%  $\text{Li}_2\text{O}$ , possible presence of some form of  $\text{H}^+$ , presence of cations not sought, and/or feature(s) of the analytical method.

Compared to bulk analyses, probe results show significant differences in the amounts of minor cations.  $\text{P}_2\text{O}_5$  substitution was found to be significant (Fig. 6.2-5a), up to 1.4 wt.%. The mechanism is likely  $\text{P}^{5+} + \text{Al}^{3+} = 2\text{Si}^{4+}$  (the berlinite substitution in feldspars).  $\text{Rb}_2\text{O}$  contents (Fig. 6.2-5b) were up to 0.93 wt.%. Fe, K, Ca, and Mg were rarely above detection limits. Fe could have been introduced in bulk analyses during sample preparation. Pollucite was observed in thin section to contain abundant fine-grained inclusions of other minerals (mainly quartz, spodumene, micas, feldspars, and apatite), and these must have influenced some of the bulk determinations. Cerny & Simpson (1978) noted at Tanco abundant thin veins were "an obstacle for obtaining reliable (bulk) compositional data".



6.2-4. Plot of Al (+ Fe), and sum of cation charges vs. CRK for the homogeneous pollucite. The average values are shown as horizontal lines.



6.2-5. Contents of minor cations as wt.% oxide vs. CRK for the homogeneous pollucite. a)  $P_2O_5$ , and b)  $Rb_2O$ .



### 6.3 Associations and compositional buffering

The most common association of pollucite is with spodumene and quartz. Less intimate associations are with albite, microcline, lepidolite, and amblygonite. Even less frequent are direct contacts with petalite, tourmaline, mica, apatite, and beryl. Pollucite is always anhedral and interstitial to these minerals and has not been observed to replace them. Rarely, pollucite is observed to replace margins of included grains of spodumene. Segregations of pollucite are usually monomineralic; however, they are penetrated by alteration assemblages. Pollucite is thus considered a late product of primary, near-solidus crystallization, in agreement with its overall location within pegmatite bodies, and with experimental evidence (cf. Henderson & Martin 1985).

Textural and other evidence for a primary rather than a late metasomatic origin of Tanco pollucite was discussed in Cerny & Simpson (1978). In contrast, the euhedral crystals which replace petalite (Greenwood B of Richmond & Gonyer 1938) and pollucite (Afghanistan, Rossovskiy 1977) are probably cesian analcime, a late hydrothermal product (Cerny 1972). The consistent paragenetic position of pollucite with other primary phases, and the limited compositional range of homogeneous pollucite, suggests a constrained range of conditions consistent with a primary

rather than late metasomatic origin.

Primary, single-phase pollucite contains an average 20 mol.% analcime (range 12 to 26), and Si/Al ratios are within the range 2.3 to 2.6. This limited range suggests that the compositions of primary pollucite are strongly buffered.

The Na<sup>+</sup> contents of pollucite may be controlled by an equilibrium with co-precipitating albite, and with Na-rich fluids which later crystallize fine-grained albite. Sebastian & Lagache (1990) suggest that in an equilibrium between pollucite and albite the Cs/Na ratio of the coexisting fluid is buffered, to 0.11 at 450°C, 0.22 at 600°C and 0.23 at 750°C. Their data (reported in bulk analyses of pollucite + quartz), however, suggest a complete solid solution in pollucite at 600 and 750°C, and a miscibility gap at 450°C. The consistency of their Cs/Na (fluid) ratio at 600 and 750°C seems to indicate that the composition of the pollucite is not strongly dependent upon temperature, but rather, is more dependent upon the Cs/Na ratio of the co-existing fluid.

The Si/Al ratios of primary pollucite may be buffered to a maximum value in an equilibrium with quartz. In laboratory synthesis of pollucite at 450 to 250°C from gel compositions of varying Si/Al ratio, Barrer et al. (1953) showed that the starting gel has a maximum Si/Al ratio of 2.5 before quartz coexists with pollucite (for Si/Al less

than 2.0,  $\text{CsAlSiO}_4$  appears). This roughly corresponds with the maximum Si/Al ratio (2.6) of primary pollucite. Compositions with Si/Al ratios greater than 2.0 are favoured by a disordered (cubic) framework (Teertstra et al. 1992b). This factor may induce pollucite to approach a limit of thermodynamic instability and the observed maximum Si/Al ratio before liberation of  $\text{SiO}_2$ .

#### 6.4 Petrological aspects of primary pollucite

Pollucite was estimated to crystallize in the range 600 to 300°C (Cerny 1979), and was found to be a solidus phase in experiments with granitic bulk compositions at 620 to 540°C (Henderson & Manning 1984, Henderson & Martin 1985). The temperature of precipitation of primary pollucite in natural B-, F-, Li-,  $\text{H}_2\text{O}$ -enriched pegmatitic melts is probably somewhat below this range.

Pollucite most commonly occurs in spodumene pegmatites, but also in pegmatites containing petalite, and petalite and spodumene, suggestive of a relatively low pressure of formation (cf. phase diagram of London 1990). Pollucite-bearing lepidolite pegmatites also tend to intrude shallow, low-pressure loci; within a pegmatite group or field, pollucite is found in the low-pressure pegmatites (Smeds & Cerny 1989). Based upon paragenesis of lithium aluminosilicates in relatively well-studied

pegmatites (e.g. Bikita, Zimbabwe or Tanco, Manitoba), a P-T crystallization path can be defined, and the pressure during magmatic crystallization may be estimated to be in the range 2 to 4 kbar (London 1990). The pressure during crystallization of primary pollucite is probably in the lower end of this range.

At some stage during the evolution of a pegmatitic melt, eventual attainment of minimum Cs/Na and Cs/K ratios may be necessary before pollucite crystallization is triggered. Alkali fractionation is also indicated by a decreasing K/Rb ratio as Cs content of K-feldspars increase (Smeds & Cerny 1989), indicating fractionation of Rb with Cs in pollucite-bearing pegmatites.

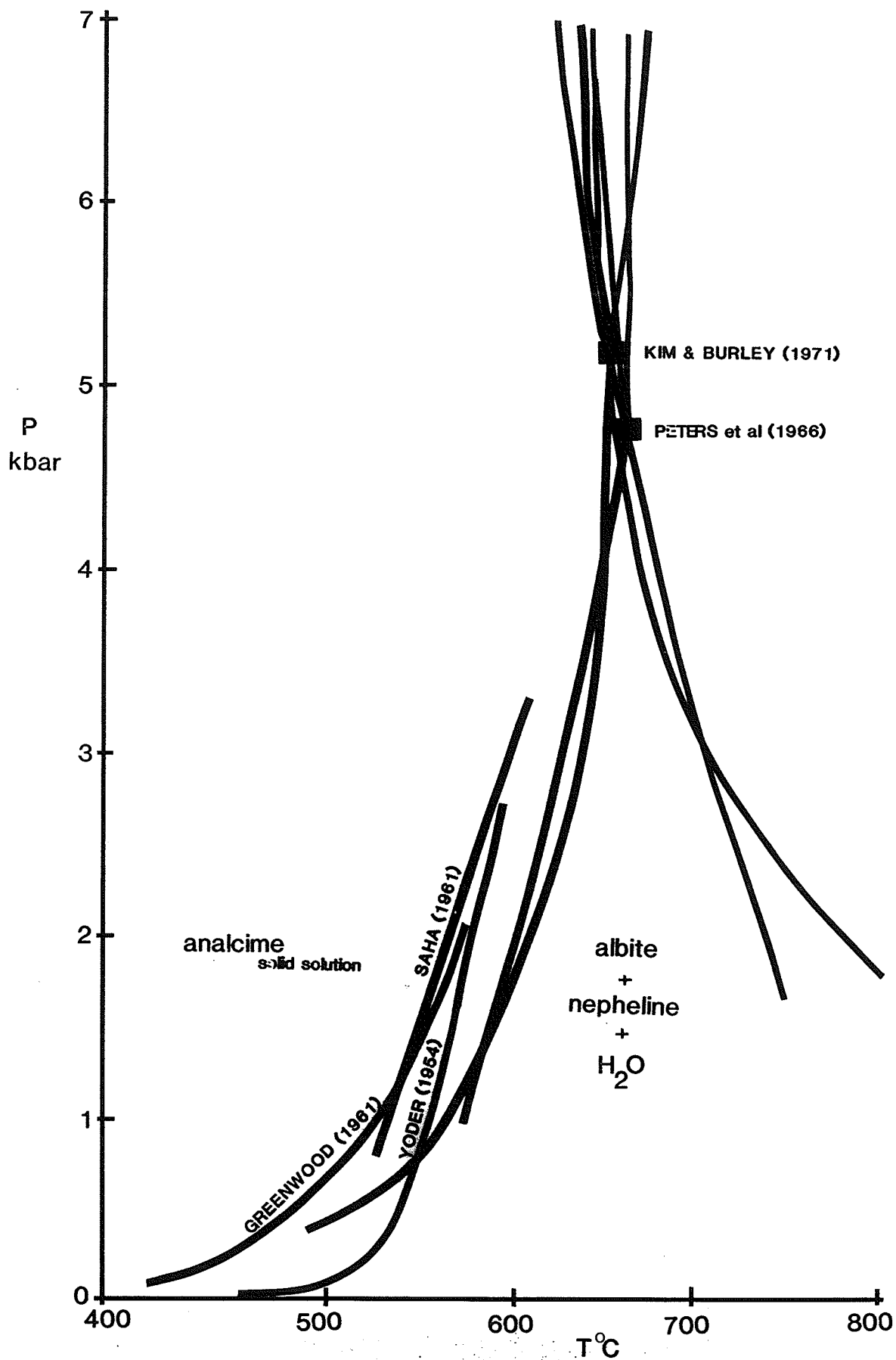
In the view of Solodov (1966) an initial concentration of 1.0 wt.% cesium is required in a bulk pegmatitic melt in order to crystallize pollucite. Very small amounts of pollucite, however, may crystallize from a late, highly fractionated fluid derived by internal evolution of the pegmatitic melt. In this view, the presence of pollucite does not necessarily indicate a high overall Cs<sub>2</sub>O content of the starting pegmatitic melt. If a minimum Cs/Na (fluid) ratio must be attained (eventually) to trigger pollucite crystallization, then the mere presence of pollucite, while still indicating advanced fractionation, does not suggest that an "ultimate" degree of fractionation has been attained in the starting melt. Instead, this may be

indicated by the amount of pollucite crystallized relative to the bulk of the pegmatite.

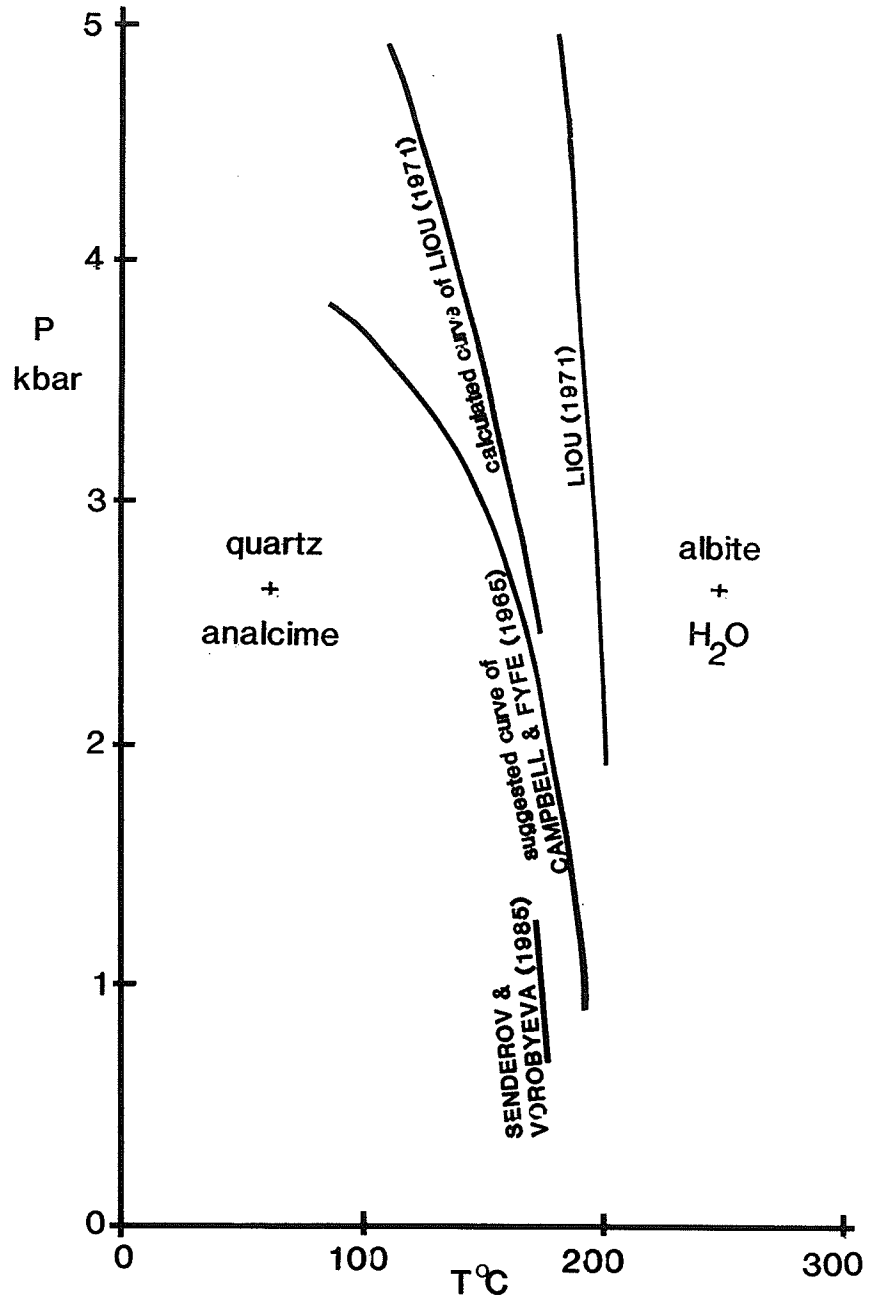
In this study, the pollucite bodies larger than 1m are Benson # 1 (CRK 83.5), Bikita (81.0), Helikon, Brown Derby (78.3), Tin Mt. (78.8), and Dunton (84.2). The smallest segregations are Leominster (80.9), Hebron (86.4), and probably also many of the other U.S. localities. There is no obvious relationship between the size of the pollucite body and its CRK. Small amounts of relatively Cs-rich pollucite perhaps crystallized from very Na-rich parent fluids, if the size of later fine-grained albite units is any estimate of low Cs/Na ratios. In contrast, large pollucite bodies are not necessarily Cs-rich; however, the parent fluids probably had high Cs/Na ratios. No studies are available on these pegmatites which report the bulk compositions. If the above assumptions are correct, then the Cs/Na ratio of the parent fluid does not exert a strong influence on the Cs content of the pollucite. Conversely, the composition of the pollucite does not seem to indicate the overall Cs/Na of the pegmatitic melt.

## 6.5 Stability relations of pollucite

Analcime melts incongruently to albite, nepheline and liquid above an invariant point at 665°C and 4.75 kbar (Peters et al. 1966) to 657°C and 5.15 kbar (Kim & Burley 1972) (Fig. 6.5-1). The stability field of analcime is considerably decreased when in equilibrium with quartz (Campbell & Fyfe 1965; Liou 1971; Senderov & Vorob'yeva 1983) and by increasing temperature (Saha 1961) (Fig. 6.5-2). Increased contents of Cs may expand the stability field (Fig. 6.5-3). If this is the case, then cesian analcime probably has a larger stability field than analcime, and sodian pollucite is the preferred (stable) phase under the pressure and temperature conditions of a pegmatitic melt. The more Cs-rich primary pollucite compositions may have crystallized at higher pressure than the less Cs-rich pollucite.

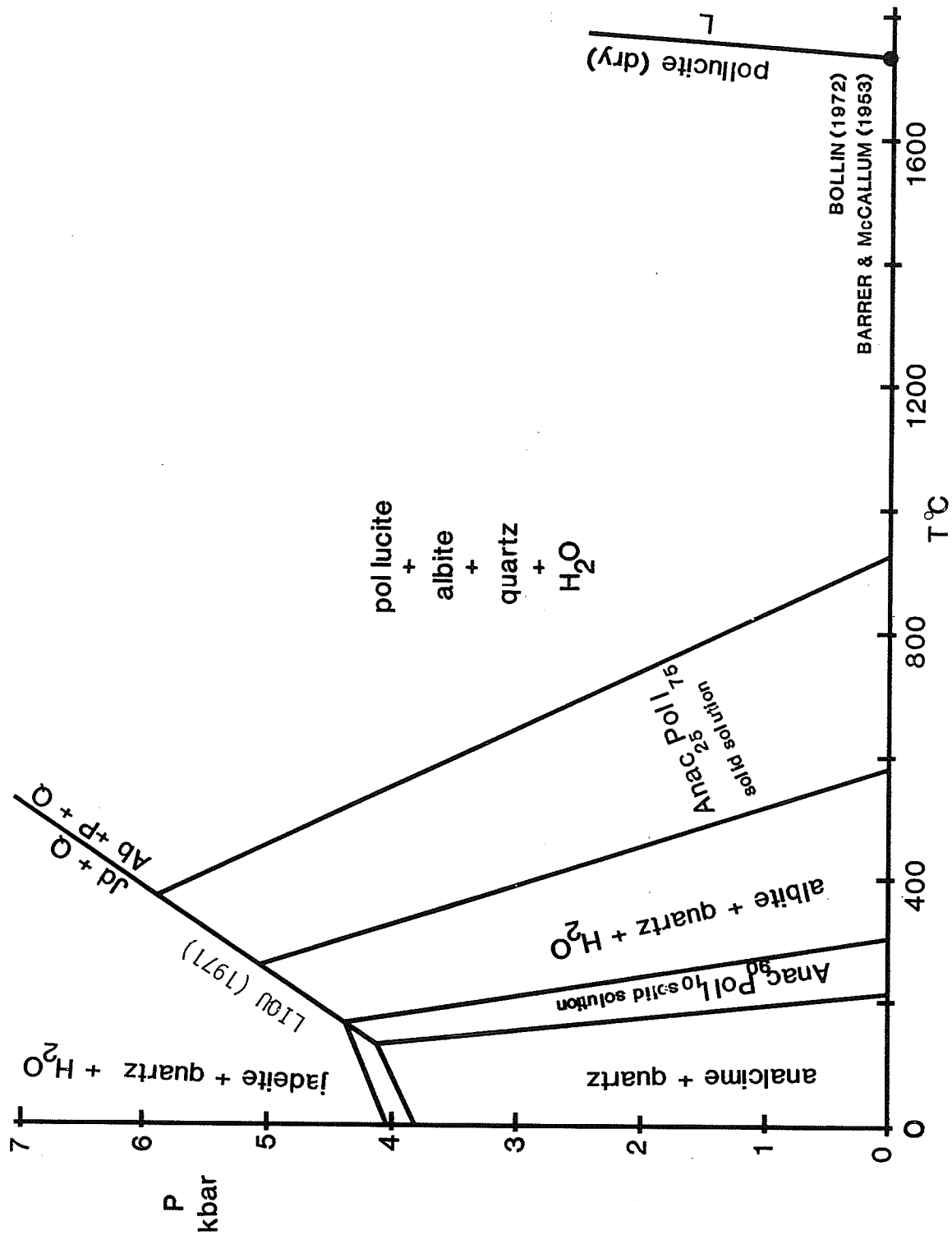


6.5-1. Stability field of analcime, from the literature.



6.5-2. Stability field of analcime, in the presence of quartz, from the literature.





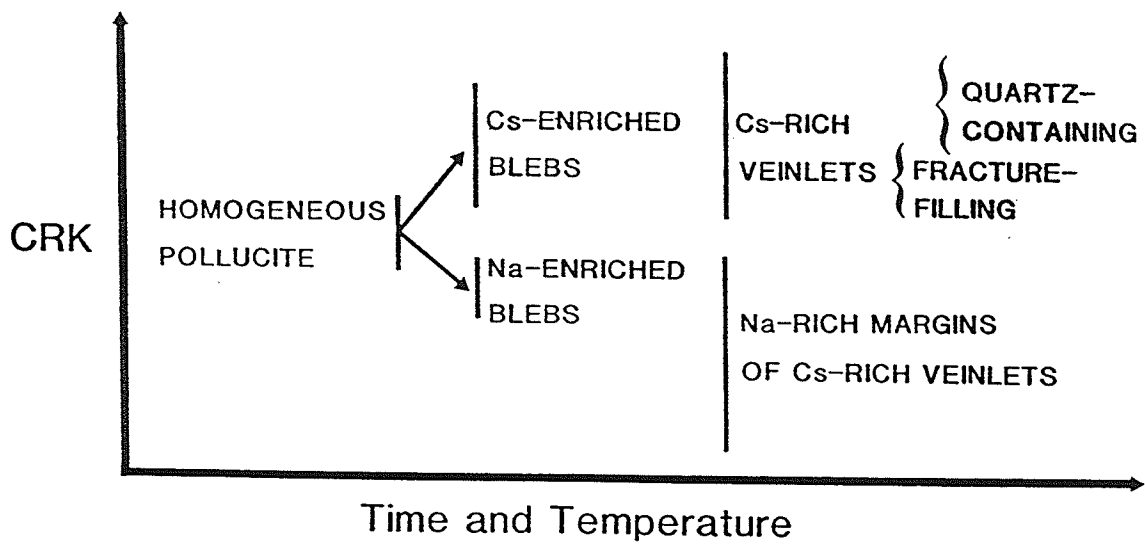
6.5-3. Proposed stability field of cesian analcime and sodian pollucite in the presence of quartz.

## 6.6 Features of the subsolidus re-equilibration

After crystallization, the primary homogeneous pollucite has locally re-equilibrated to Na- and Cs-enriched compositions, manifested as a variety of crosscutting textural features. The features of the compositional heterogeneity are described below, first in terms of the typical textures, then in terms of unusual features and the relationships to alteration products.

The degree of re-equilibration varies markedly between localities; however, a consistent sequence is observed: the homogeneous pollucite has locally evolved to a blebby mosaic of alternating Na-enriched and Cs-enriched domains, crosscut by veinlets of Cs-rich pollucite ( $\pm$  quartz) rimmed by Na-rich pollucite (Fig. 6.6-1).

The domains are 20 to 300  $\mu\text{m}$  in size and range from ovoid or hour-glass in shape to lammellar with bifurcating tips (similar to the perthite texture of feldspars). The compositions of the Na-enriched domains do not range far from that of homogeneous pollucite. In some localities the blebs are all Cs-enriched relative to the homogeneous pollucite. Some blebs have a complex internal zonation, mostly consisting of slight variation in Si/Al ratio with constant CRK; these seem to be features of growth. Later generations of blebs are usually (but not always) more Cs-enriched; successive replacement creates a brecciated to



6.6-1. Schematized general sequence of exsolution products of primary, homogeneous pollucite, as a function of CRK and decreasing temperature

schollen texture containing rafts of Na-enriched or homogeneous pollucite.

Early generations of Cs-rich veinlets are mostly fracture-filling and do not contain quartz. They are 10 to 20  $\mu\text{m}$  wide, and are crosscut by later generations of quartz-containing Cs-rich veinlets usually less than 10  $\mu\text{m}$  wide. The later veinlets are usually (but not always) more Cs-rich, and appear to have developed in situ. Both generations of veinlets are continuous in length on a scale of cm; the maximum width observed is 50  $\mu\text{m}$ . Na-rich pollucite rims both generations of veinlets, but the rims are particularly pronounced adjacent to the later Cs-rich veinlets. Na contents and Si/Al of the Na-rich rims are highest adjacent to the margins of the Cs-rich veinlets, and decrease over 30 to 300  $\mu\text{m}$  into the homogeneous pollucite (Fig. 4.3-4).

In the blebs and veinlets, contents of minor cations (Rb, P, Ca, and K when present originally in the homogeneous pollucite) tend to decrease or remain constant but only rarely increase. No clearly defined trend has been found for the redistribution of the minor cations relative to the composition, type, or generation of the textural feature.

The larger veinlets of Cs-rich pollucite, besides containing quartz grains, also locally contain feldspars, micas, and AlSi-bearing minerals (which are also generally

associated with Cs-rich blebs). Grains of quartz 10 to 30  $\mu\text{m}$  in size frequently have rims of Cs-rich pollucite, and are surrounded by Na-rich pollucite, grading over 5 to 50  $\mu\text{m}$  into the homogeneous pollucite. Cs-rich pollucite has been observed in a symplectitic relationship with quartz, and as Cs-rich bands (up to 1mm wide) adjacent to spodumene grains. An unusual feature observed at only a single locality is one Na- and Si-enriched veinlet within Tin Mt. pollucite (Figs. 5.2-2g and 5.2-3).

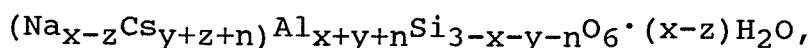
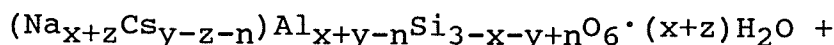
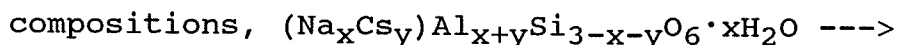
Pollucite occurs as inclusions in adjacent primary phases. This pollucite is usually more Cs-rich than the homogeneous pollucite. Fracture-fillings in spodumene are common, and pollucite is also present along cleavage traces of micas. In quartz, round inclusions are common, particularly nearest to the contact with the main pollucite. Some crystals of pollucite in quartz seem euhedral, showing several flat faces in cross-section.

The subsolidus re-equilibration occurs before most of the alteration of pollucite, as the alteration products generally cross-cut the blebs and veinlets. Veins of lepidolite and quartz, fine-grained mica and spodumene, and overgrowths of adularia usually contain inclusions of pollucite. The inclusions (as well as pollucite adjacent to the alteration products) are progressively more Cs-rich with later alteration (e.g. Fig. 5.5-1a).

## 6.7 Petrological aspects of the re-equilibration

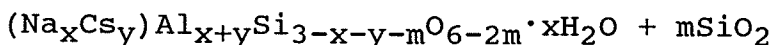
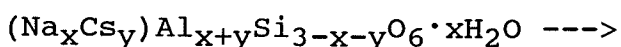
The blebby mosaic appears to have developed from homogeneous pollucite strictly by local migration of H<sub>2</sub>O, Na, Cs, Al, and Si. Likewise, veinlets of Cs-rich low Si/Al pollucite appear to be involved in a mass-balance with adjacent Na-rich high Si/Al pollucite. This mass balance is controlled by three principal mechanisms (Fig. 6.7-1), the first two being dominant:

(i) exsolution into Na, Si-enriched and Cs, Al-enriched



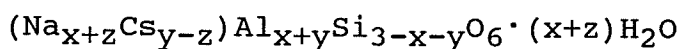
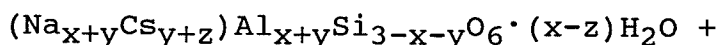
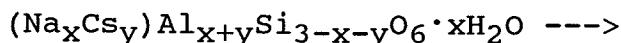
where  $0 < x + y + z \leq 1$ ;

(ii) liberation of SiO<sub>2</sub> as quartz, leading to Si-poor, but alkali cation- and Al-enriched pollucite,



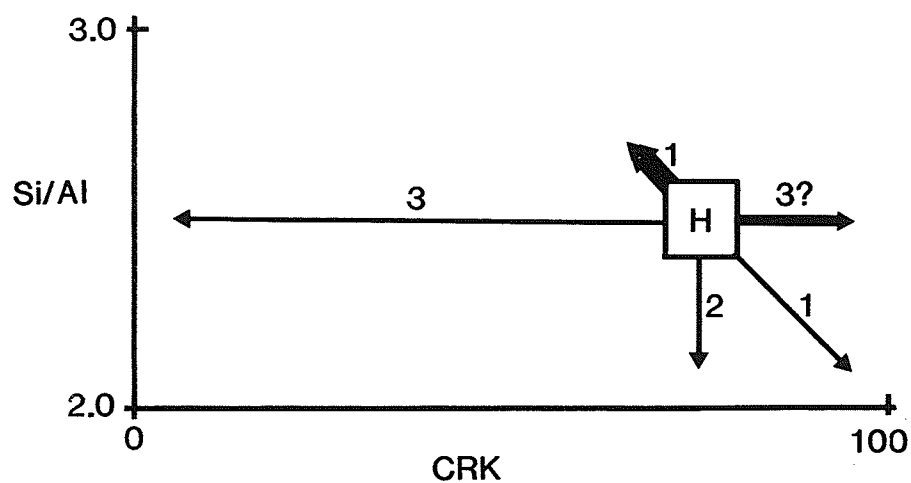
where  $0 < y + m \leq 1$ , Si/Al  $\geq 2$ , and Na/Cs is constant;

(iii) cation exchange which modifies the Na/Cs ratio but does not affect Si/Al ratios,

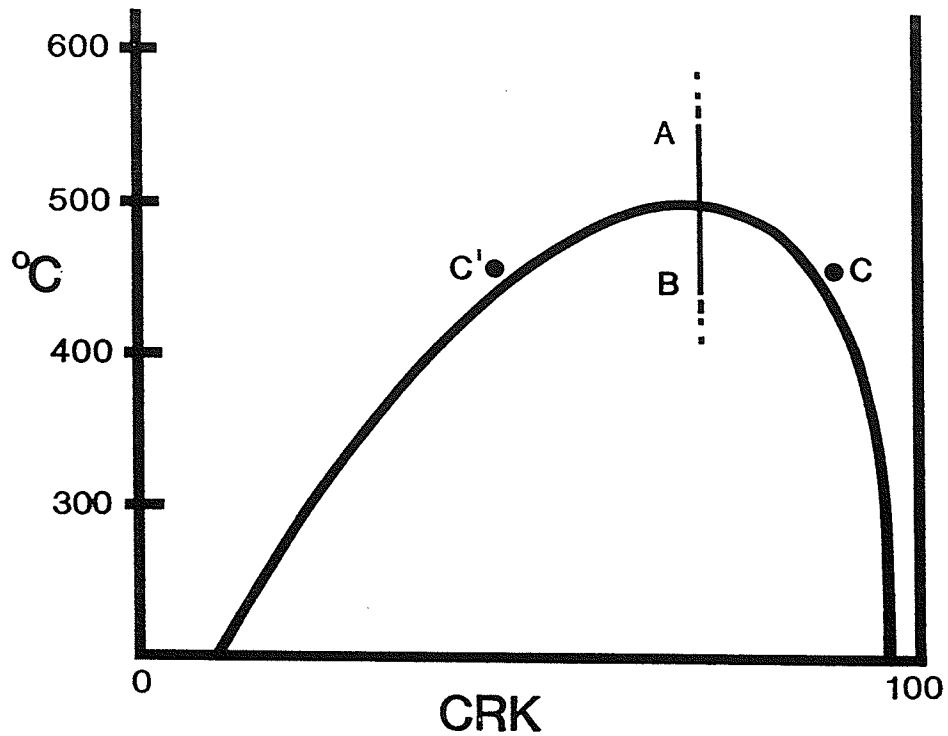


where  $0 < z \leq 1$ .

The above features are compatible with an exsolution



6.7-1. Schematized representation of the compositional trends of Si/Al with CRK, for homogeneous pollucite with compositional range (H). Numbers 1, 2, 3 correspond with substitution mechanisms (i), (ii), (iii); equations for which are given in Chapter 6.7.



6.7-2. Schematic solvus in the analcime-pollucite series:  
 A- temperature range of possible stable, homogeneous, hypersolvus pollucite. B- temperature range of possible metastable, homogeneous, subsolvus pollucite.  
 C- Synthetic composition of Sebastian & Lagache (1990) relative to composition of natural primary pollucite A, B, and possible co-existing Na-rich composition C<sup>1</sup>.



process involving a miscibility gap between Na- and Cs-enriched compositions which broadens with decreasing temperature (Fig. 6.7-2). Immiscibility is suggested by the data of Sebastian & Lagache (1990) in their experiments at 450°C. At 600 and 750°C their data indicate a complete solid solution between analcime and pollucite.

In Fig. 6.7-3 the trends of Si/Al with CRK are plotted for all the localities. In individual localities, the relation between Si/Al and CRK has been noted as a trend towards a Si/Al ratio of 2.0 as CRK approaches 100. Localities in which the primary pollucite has a high CRK have a steeper trend towards this composition. A trend of Al to a value of 1.0 near CRK 100 predicts end-member pollucite with a simple formula of  $\text{CsAlSi}_2\text{O}_6$  as reported in most high-temperature synthesis (e.g. Martin & Lagache 1975). In this study, compositions near this formula have been found at Locality Y, Norway, Maine, and Locality X and Old Tom, Greenwood, Maine (Teertstra & Cerny, in prep.).

The associations of Cs-rich pollucite with a variety of silicates (described above) also suggests a local equilibrium between pollucite and silica. The Si/Al ratio of the pollucite seems to have a maximum value which is strongly dependent upon the CRK. Studies of analcime by Khundadze et al. (1970) and Senderov & Khitarov (1971) showed that crystals with Si/Al near 2.0 were most thermodynamically stable. The most Cs-rich pollucite which



has Si/Al near 2.0 may, like analcime, be the most thermodynamically stable. The composition fixed at integral stoichiometry suggests that the crystals are potentially ordered.

#### 6.8 The alteration of pollucite

Pollucite, which is compositionally homogeneous, tends to not have any inclusions and is colorless with a vitreous to sub-vitreous lustre in hand specimen. Compositionally heterogeneous pollucite has abundant inclusions, is white in colour (termed a "porcelanous variety" in some Russian literature), and has a greasy to earthy or dull lustre in hand specimen. It is also optically inhomogeneous in thin section. Pollucite from bodies larger than 1 m were all found to be compositionally heterogeneous, even though small fresh-looking grains may be extracted from crushed specimens. Fresh, homogeneous pollucite was found mainly in small (less than 5 cm) segregations which contained only small amounts of alteration products. Na-exchanged, analcimized pollucite has a greasy lustre, and is associated with clay minerals. A significantly lower density may be noticed, even in hand specimen, compared to fresh pollucite. In thin section the index of refraction decreases noticeably towards veins, fractures, and clays.

Three consecutive stages of alteration were recognized

by Cerny & Simpson (1978) at Tanco, Manitoba: (i) coarse veining, usually polygonal, fracture-filling and more abundant near the margins of bodies, (ii) pervasive fine veining, braided to subparallel, and (iii) replacement assemblages. This pattern holds for all other localities of pollucite that were examined and described in sufficient detail. Aspects of one or more alteration styles could be observed even on the material available for the present study, limited as it was in some cases. The coarse veining is not usually represented in hand specimens, but has been well-described in the literature at out-crop scale.

Coarse veining is present only in large pollucite bodies, greater than 1 m in size. The veining is fracture-filling and related to sequences of late crystallization of otherwise mostly primary phases (quartz, fine-grained albite, lepidolite, microcline, amblygonite). Only locally is the veining metasomatic and replacing pollucite (particularly in the case of microcline). The microcline is non-perthitic, compositionally homogeneous, usually untwinned, and free from inclusions. Compositions of the microcline from different localities are similar: Locality X, Maine has no Fe, Mg, Sr, Ba or Ti, but (as wt.%), Na<sub>2</sub>O 1.08, CaO 0.00, Cs<sub>2</sub>O 0.04, P<sub>2</sub>O<sub>5</sub> 0.26, Rb<sub>2</sub>O 0.00; Locality X, Greenwood, Maine (0.91, 0.00, 0.30, 0.50, 0.81); and Helikon (0.21, 0.00, 0.30, 0.41, 1.13). The microcline from Helikon

has local extreme enrichment in Rb, with atomic contents of Rb > K. Homogeneous pollucite seems to have co-existed with albite veins and not to have cation-exchanged to Na-enriched cesian analcime compositions as suggested by Cerny & Simpson (1978) at Tanco. Lepidolite veining is fracture-filling and does not seem to have replaced pollucite, but rather may be a product of late crystallization.

Fine braided to subparallel veining is characteristic of most localities where pollucite has segregated into masses greater than 5 cm in size. The veins follow fractures and fill interstices along grain boundaries, and only rarely replace pollucite from these surfaces. They vary from continuous with even width (early veins up to 5 mm, later veins up to 1 mm) to dispersed and irregular. They are always observed to pinch out within the pollucite, and do not connect with external bodies (such as lepidolite). The veins contain mica (lepidolite?) ± quartz which decreases in grain size from core to margin. Later veins consist of fine-grained mica which either crystallized with, or was replaced by, fine-grained spodumene. Grains of mica (or spodumene) are often oriented perpendicular to the vein. The spodumene is symplectitic with quartz, and locally retains outlines of earlier euhedra, and may form its own set of veins. Some of the veins contain Cs-rich pollucite, some contain analcime, and some neither, separated from pollucite by a sharp contact.

The above observations suggest that the veins are not true replacements but instead are products of late crystallization, in possible equilibrium with pollucite.

Pollucite is most commonly replaced by adularia, analcime ( $\pm$  apatite), and clay minerals. These are described in sequence below.

Adularia metasomatically replaces pollucite along surfaces of the earlier veins and as spherical aggregates (0.5 to 1.0 mm) fully embedded in the pollucite. The adularia contains abundant inclusions, most commonly of Cs-rich pollucite, micas, albite and AlSi-bearing phase(s). Adularia concentrates Rb, especially towards the margins of its grains where it has atomic contents of  $Rb > K$ . Qualitative analysis of the micas indicates equal atomic contents of Al and Si. At localities where the adularia is enriched in Rb, the micas usually are as well; the enriched micas have atomic contents of  $Rb > K$ .

Analcime replaces pollucite by cation exchange (termed here enrichment in  $Na^+$  or analcimization). This analcime has Si/Al ratios in the same range or slightly lower than in the homogeneous pollucite.  $Rb_2O$  and  $P_2O_5$  contents are decreased; however, contents of  $CaO$  and  $K_2O$  (and rarely  $Fe_2O_3$ ) are increased. The cation exchange is most advanced near fractures and clay minerals. It may extend into the pollucite over a scale of cm, leaving no pollucite with original composition, or may only be local. It is

associated with the leaching (or dissolution) of pollucite near clays, locally in the form of thin hollow capillaries. The capillaries are fully embedded in the analcime or extend from vein and adularia surfaces. Observed in thin section, they locally form a three-dimensional, rectangular (grid) relationship to each other, suggesting dissolution along preferred crystallographic directions (probably {111} along 6-membered rings, or possibly {100} along 4-membered rings).

Argillic replacement of pollucite by white to buff clays is most extensive near the margins of pollucite bodies. It leaves the earlier veining and inclusions intact, producing clay pseudomorphs after pollucite. It replaces also the analcime containing hollow capillaries. Some of the clays are Cs-rich; however, the bulk of the Cs content of the pollucite has been dispersed. Based upon powder X-ray diffraction, some of the clays are of the illite group, and expanding-type smectites (perhaps montmorillonite). These observations are partly confirmed by electron microprobe examination, showing presence of K, Al and Si in a hydrous phase; however, in some samples there is additional presence of Na and Mg, suggesting solid solution towards chlorite. Which of these clays are Cs-rich is not known - they may be polymineralic mixed-layer sequences of the above clay minerals, in any case future study of individual grains is required for identification.

## 6.9 Petrological interpretations of the alteration

The coarse veining is probably related to relatively high-temperature fluids associated with crystallization of late primary phases. These phases include fine-grained albite which may have buffered the primary compositions of pollucite. Most of the veining cannot be considered a true alteration of pollucite, as there is only marginal metasomatic replacement of pollucite. Only some of the lepidolite veining may be fracture-filling, and is therefore only tentatively grouped with the coarse veining.

Most of the lepidolite veining may be considered a product of late crystallization after much or all of the pollucite has crystallized. The veins become thinner with time and more fine-grained, and the veining does not leave the pollucite body. It is followed closely by spodumene crystallization, either initially fine-grained or later altering to a fine spodumene-quartz symplectitic intergrowth.

Bodies of pollucite are often brecciated in appearance, however, the fractures do not extend into adjacent or included assemblages. The fractures contain minerals of the early, coarse veining. On a microscopic scale, however, even the fine veining follows fractures, grain boundaries, and traces of a poor cubic cleavage.

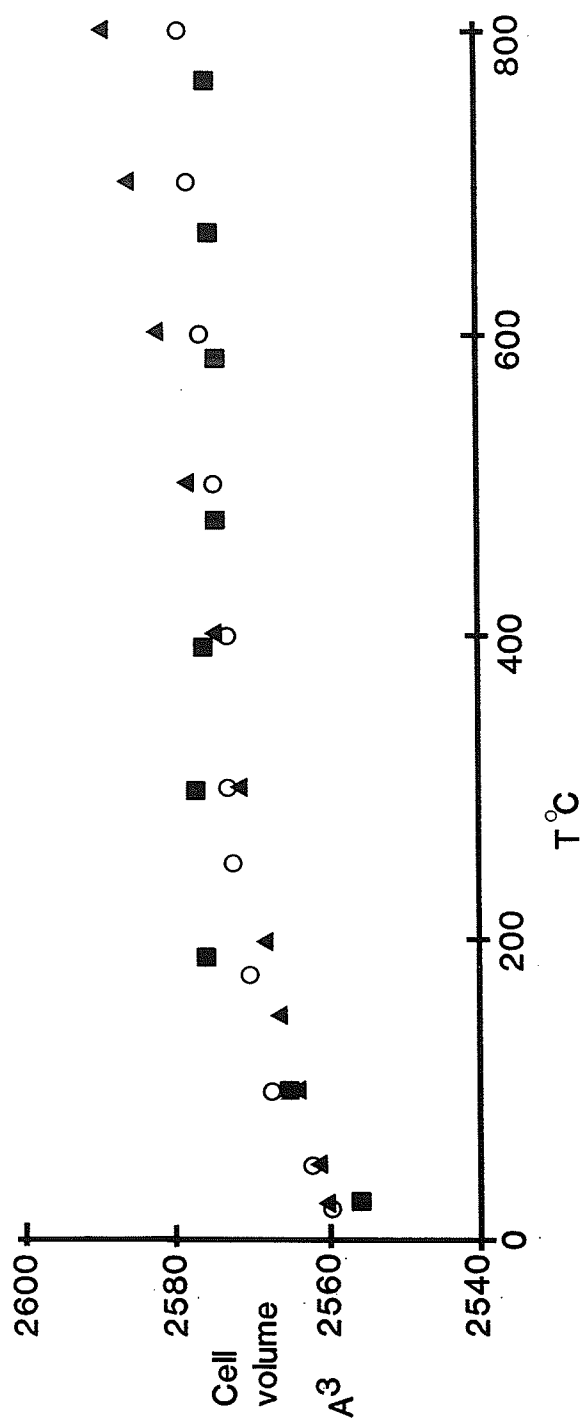
One reason for this common style of alteration could



be a contraction of pollucite with decreasing temperature. Taylor & Henderson (1968) found a maximum rate of thermal expansion of synthetic  $\text{CsAlSi}_2\text{O}_6$  up to  $200^\circ\text{C}$ , above which there was no increase in cell volume. Richerson & Hummel (1972) gave similar results and found a greater thermal expansion in cubic analcime than in pollucite. Based upon these experiments, thermal contraction of bodies of natural sodian pollucite may initiate internal fracturing, probably at temperatures below  $200^\circ\text{C}$  (Fig.6.9-1).

Crystallization of adularia may, like the fine braided veining, be considered a product of crystallization of residual pegmatitic fluids. In contrast to the veining, the adularia does not follow fractures, cleavage traces or grain boundaries, but instead metasomatically replaces pollucite. The habit of the crystals indicates high nucleation and crystallization rates. These conditions, combined with low crystallization temperatures (of about  $300$  to  $250^\circ\text{C}$ ) favour metastable precipitation of highly disordered crystals (Cerny & Chapman 1986).

The cation exchange of pollucite to Na-enriched compositions (analcimization) generally occurs before, but is associated with, the alteration of pollucite to clays. Barrer (1950) treated pollucite with various salt solutions and inferred from index of refraction measurements that there was little or no  $\text{Cs}^+$  exchange. Later, Komarneni et al. (1978) showed a low cation exchange capacity at  $25^\circ\text{C}$



6.9-1. Thermal expansion of analcime and pollucite, cell volume vs. temperature, from the literature.  $(Al, Si)O_4$  tetrahedra initially rotate to form a less-collapsed framework; once the greatest extension is reached further expansion rates are limited. Symbols: Taylor & Henderson 1968  $CsAlSi_2O_6$  (squares); Richerson & Hummel 1972  $NaAlSi_2O_6$  (circles), and  $CsAlSi_2O_6$  (triangles).

(similar to that of feldspars) that was probably due to surface phenomena and not classic ion exchange processes. More recently, however, Komarneni & White (1981) demonstrated substantial cation exchange at 300°C using brine solutions with high activity of Na<sup>+</sup>, K<sup>+</sup>, and Ca<sup>2+</sup>.

Cerny (1974) studied clay pods of illite + kaolinite + smectites pseudomorphous after pollucite and inferred that this association formed at very low temperatures, likely under hydrothermal conditions. Under these conditions, the source of the fluids could be an open bimetasomatic exchange between pegmatite- and wall-rock-derived fluids (London 1990). Even though this may be the dominant mechanism of both analcimization and clay formation of most pollucite localities, the mechanism by which weathering and supergene weathering degrades pollucite remains to be examined both experimentally and in natural specimens.

### 6.10 Relevance of pollucite to nuclear waste containment

Analcime-pollucite minerals are natural analogues to phases involved in the long-term containment of nuclear wastes. In studies so far, only synthetic pollucite (with formula  $\text{CsAlSi}_2\text{O}_6$ ) has been considered as the host for  $^{137}\text{Cs}$  (Strachan & Schulz 1979, Haaker & Ewing 1981), and only specifically for containment of high-level reprocessing wastes. A number of methods for synthesis of pollucite are available (Hogan & Risbud 1991, Gallagher & McCarthy 1981, Martin & Lagache 1975, and Richerson & Hummel 1972 contain additional references). Firing of gels, sludges, and calcined sludges also crystallize pollucite as a common product.  $\text{Cs}^+$  reacts directly with a number of AlSi-bearing precursors at high temperature to produce pollucite (e.g. Mukerji & Kayal 1975, Plyushchev 1959). At high temperature, however, the high volatility of  $\text{Cs}^+$  creates problems of loss and contamination (Odoj et al. 1979). Pollucite has appeared in SYNROC formulations (titanite-based high-level waste ceramics; McCarthy & Davidson 1975, McCarthy 1977, Buykx et al. 1990) which are supposed to incorporate  $\text{Cs}^+$  in a limited solid solution within hollandite-type phases.

Phases with intermediate compositions within the analcime-pollucite series are common products of hydration and hydrothermal alteration of nuclear waste glasses (Bates

et al. 1982 and 1983, Caurel et al. 1987 and 1990, McCarthy et al. 1978, and many others). As well, the phases crystallize in hydrothermal reactions of calcined sludges and gels, Cs-loaded zeolites (Mimura et al. 1990) and clays, and from a variety of waste forms in reactions with basalt and shales.

Cesian analcime may have certain advantages over pollucite as a waste form. A variety of low-temperature methods exist for synthesis (Barrer & Hinds 1953, Barrer & McCallum 1953, Saha 1959, Balgord & Roy 1971, Borer & Meier 1971, Barrer & Sieber 1977, Ueda & Koizumi 1979). Analcime is readily crystallized, and stable under a wide range of conditions, and concentrates Cs during crystallization (Keith et al. 1983, Berg 1968, Barrer et al. 1953). The resulting waste form will have lower "activity" because of low Cs/Na ratios, and may have a decreased volume compared to other low-level or medium-level waste forms.

This study of natural pollucite has revealed many features relevant to the use of analcime-pollucite phases as nuclear waste hosts. Si/Al ratios are typically 2.3 to 2.6. The crystals are disordered (required by the cubic structure for the above ratios) and are probably thermodynamically less stable than crystals with Si/Al ratios near 2.0. Pollucite has a maximum Si/Al ratio near 2.6 in the presence of quartz or other silicates, which decreases with CRK upon re-equilibration. End-member

pollucite has Si/Al near 2.0, and may therefore be thermodynamically stable, and is potentially ordered.

A high-temperature solid solution exists between analcime and pollucite above about 500°C. Subsolvus re-equilibration produces pollucite with Na-enriched and Cs-enriched compositions ( $\pm$  SiO<sub>2</sub>) and does so in an apparent mass balance which seems to preserve Cs within the resulting mineral assemblage.

Of greatest concern to nuclear waste containment using analcime-pollucite host phases, are the replacement assemblages: (i) replacement by adularia, (ii) cation exchange to analcime locally accompanied by leaching (dissolution) and (iii) replacement by clay minerals.

At some localities, adularia replacement has been extensive. Locally, the pollucite is Cs-rich around the adularia and the Cs<sup>+</sup> content may have been preserved, as Na<sup>+</sup> seems to be mobilized preferentially. Replacement of pollucite by adularia can be presumed to be a relatively high-temperature process, associated with fine braided veining of mica + spodumene (300 to 250°C; Cerny & Chapman 1986). This temperature is within the upper range estimated for a waste package: about 250°C for spent fuel and 150°C for reprocessed waste packages (McCright 1991). The peak temperature depends upon many factors; age and amount of the waste, isotopic content, degree of (re)processing, and depth and thermal characteristics of the site.

Most localities of pollucite are, at least slightly, Na-enriched along fractures. The extent of cation exchange varies considerably among different localities: sodium metasomatism may advance locally to total cation exchange, generating analcime. Early experiments showed that pollucite has no capacity for ion exchange, similar to that of feldspars (Barrer 1950), or a very low capacity, probably due to surface phenomena and not classic ion exchange processes (Komarneni et al. 1978). More recent studies have, however, demonstrated cation exchange of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$  (Komarneni & White 1981).

Leaching rates of the pollucite are of a similar order of magnitude as the cation exchange, as dissolution is closely associated with the analcimization. Locally, dissolution creates a three-dimensional rectangular grid of hollow capillaries within the pollucite.

Replacement of pollucite by clays acts to disperse most of the Cs content (this study, and Cerny 1978). Even residual Cs contents on the clays are significant, however, as these indicate a mechanism for some degree of retardation of migrating  $\text{Cs}^+$ . These clays could be studied as natural analogues to minerals used as backfill, and present in the host rock of waste sites. For example, illite has been shown to be a sorbent for  $\text{Cs}^+$  in grouted waste injection into shales, but there are no studies of Cs-rich clay minerals in the literature. The conditions of

replacement of pollucite by clay minerals, whether alkaline or acidic, needs to be established - indeed, conditions of all the above processes require experimental investigation.



## CHAPTER 7

## CONCLUSIONS

This study has advanced understanding of minerals in the analcime-pollucite series in several key areas. The subject was approached from a variety of perspectives: literature reviews, synthesis, X-ray diffraction studies, MAS NMR spectroscopy, optical petrology, crystal chemistry and morphology. Results from use of the electron microprobe technique, however, proved most useful in revealing the essential features of the series. The following sections emphasize the new finds while summarizing what is known about the minerals.

## 7.1 The primary pollucite

The occurrence of pollucite is restricted to complex rare-element granitic pegmatites which have attained a high degree of alkali fractionation. The most fractionated pegmatites had high Cs/Na and Cs/K ratios in the parent melt and crystallized large amounts of pollucite. Other less fractionated pegmatites eventually attained high enough Cs/Na and Cs/K ratios during late stages of internal evolution of the parent melt to crystallized small amounts of pollucite. Pollucite occurs late in primary consolidation of the pegmatites, and forms near the central

parts of the pegmatites, interstitial to spodumene and quartz and closely associated with albite, microcline, lepidolite, amblygonite and petalite. The conditions of pollucite crystallization are about 400 to 550°C and 2 to 4 kbar.

Primary, single-phase pollucite has a composition within the range CRK 70 to 90 (most at 80) and Si/Al ratios of 2.3 to 2.6 (most near 2.6). Within a locality CRK ranges by up to 6.6 and Si/Al by up to 0.2. Of the minor cations, only  $\text{Rb}_2\text{O}$  (up to 1.0 wt.%) and  $\text{P}_2\text{O}_5$  (1.5 wt.%), and probably also  $\text{Li}_2\text{O}$  (0.4 wt.%) are significant. Contents of  $\text{K}_2\text{O}$ ,  $\text{CaO}$ ,  $\text{MgO}$ ,  $\text{Fe}_2\text{O}_3$  are less than 0.1 wt.%. Pollucite may also contain small amounts of Ba, Sr, Pb, Ce, Ti, Tl and Ga. Contents of these cations are mostly limited by geochemical scarcity, however, amounts of K and Ca may also be limited by solubility and stability relations with pollucite.

$\text{Na}^+$  contents of primary pollucite are buffered to an average 20 mol.% analcime (range 12 to 26) by Na-rich fluids in equilibrium with albite. The  $\text{Cs}^+$  content of the pollucite does not seem to be controlled by Cs/Na ratios of the parent fluid, but more by P-T conditions. Analcime is not stable in a pegmatitic melt in the presence of quartz, however, suggesting that incorporation of significant amounts of  $\text{Cs}^+$  enlarges the stability field of analcime to higher temperatures and pressures.

Si/Al ratios of primary pollucite are buffered to near 2.6 (range 2.3 to 2.6) by Si-rich fluids in equilibrium with quartz (and probably also with other Si-rich phases). Compositions with high Si/Al ratios are favoured by a disordered framework (a feature required by the cubic structure), even though Si/Al ratios of 2.0 are most thermodynamically stable.

## 7.2 The subsolidus re-equilibration

Primary, single phase pollucite has locally evolved to a mosaic of alternating Na-enriched and Cs-enriched 20 to 300  $\mu\text{m}$  domains, crosscut by 1 to 50  $\mu\text{m}$  wide veinlets of quartz-containing Cs-rich pollucite rimmed by Na-rich pollucite. An overall positive correlation of Al contents with CRK is noted, such that the most Cs-rich compositions have low Si/Al. The blebby mosaic appears to have developed by strictly local migration of  $\text{H}_2\text{O}$ , Na, Cs, and Al from the homogeneous pollucite. Likewise, veinlets of Cs-rich low Si/Al pollucite appear to be involved in a mass balance with adjacent Na-rich high Si/Al pollucite. The mass balance is controlled by three principal mechanisms, the first two being dominant: (i) exsolution into Na, Si-enriched and Cs, Al-enriched compositions, (ii) liberation of  $\text{SiO}_2$  as quartz, leading to Si-poor, but cation and Al-enriched pollucite, and (iii) cation exchange which

modifies the Na/Cs ratio but does not affect Si/Al.

The above features indicate a re-equilibration of primary pollucite with decreasing temperature into a two-phase subsolvus assemblage of Na- and Cs-rich compositions. Very Na-rich (analcime) compositions will be stable only with decreasing pressure and (thus) have not been observed. Very Cs-rich compositions have CRK near 100 and Si/Al near 2.0. The integral stoichiometry of  $\text{CsAlSi}_2\text{O}_6$  is reported in high-temperature synthesis but a natural occurrence has not been reported until now. This composition may be most thermodynamically stable and is potentially ordered.

### 7.3 The alteration of pollucite

Pollucite does not seem to replace any of the minerals it is associated with, but instead contains alteration assemblages. In fact veining is so common that it is used as a diagnostic field tool to distinguish pollucite from massive quartz. The alteration occurs in three consecutive stages: (i) coarse veining, usually polygonal, fracture-filling and more abundant near the margins of bodies, (ii) pervasive fine veining, braided to subparallel, and (iii) replacement assemblages.

The coarse veining is present only in pollucite bodies larger than 1 m, the veining is fracture-filling and only locally metasomatic, and is related to sequences of late

crystallization of the primary phases quartz, fine-grained albite, microcline, lepidolite, and amblygonite. The fractures may be related to thermal contraction of the pollucite below 200°C.

The fine braided veining is characteristic of localities where the pollucite has segregated into masses larger than about 5 cm; it follows fractures, grain boundaries and a poor cubic cleavage of pollucite. Early veins consist of coarse-grained lepidolite and quartz (+ spodumene); later veins consist of finer-grained mica and spodumene. The veins are not true replacements but instead are products of late crystallization, in possible equilibrium with pollucite.

Pollucite is most commonly replaced by adularia, analcime, and clay minerals. Adularia (+ mica + albite + AlSi-bearing phases) metasomatically replaces pollucite along surfaces of veins and as (0.5 to 1.0 mm) spherical aggregates fully embedded in the pollucite. The adularia is enriched in Rb, locally with atomic contents of  $Rb > K$ . The associated mica follows the Rb-enrichment trend of the adularia and also locally has  $Rb > K$ . Analcime (+ apatite) replaces pollucite by cation exchange and is associated with clay minerals and leached areas of pollucite and hollow acicular channels. The rate of framework dissolution is thus similar to the rate of cation exchange, both of which are mechanisms for the dispersal of Cs from

pollucite. Margins of pollucite bodies are replaced by clay minerals (mainly illite, montmorillonite, kaolinite) which also disperse most of the Cs. Some of the clay minerals are Cs-rich, however, and require further investigation.

#### 7.4 Implications for nuclear waste containment

Phases within the analcime-pollucite series may be useful as hosts for nuclear wastes, particularly  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$ . The synthetic phases are prone to metastable formation with high Si/Al ratios, especially in Si-undersaturated systems (i.e. no quartz present), so should be crystallized with Si/Al ratios near 2.0 for maximum thermodynamic stability. For crystallization at low temperature, either cesian analcime (low Cs/Na ratios) or sodian pollucite (near end-member with high Cs/Na ratios) should be used, as mid-solvus compositions are probably less stable. Long-term containment should be within a compatible host rock in which the minerals are stable, to prevent formation of replacement assemblages. To prevent cation exchange the groundwater should have only low cationic contents, especially of Na. As a "built-in" deterrent to cation exchange, a waste crystal could have a core of pollucite and a rim of analcime. This is potentially stable and reduces a high geochemical gradient

favouring dispersal of  $\text{Cs}^+$ .

#### 7.5 Recommendations for future research

Verification of compositional data produced by the electron microprobe technique, are required for  $\text{SiO}_2$  contents and possibly also for  $\text{Al}_2\text{O}_3$ . The cause of the possible alkali cation deficiency relative to Al requires investigation; these are the presence of other cations including  $\text{Li}^+$ , possible role of some form of  $\text{H}^+$ , or Al over-estimates. Comparison to synthetic phases may be useful. Judging quality of the analytical results depends, in part, upon determination of  $\text{H}_2\text{O}$  contents. Calculation of index of refraction is greatly influenced by measurements of  $\text{H}_2\text{O}$  or the model used to estimate  $\text{H}_2\text{O}$ , and the  $K_n$  value of specific refractive energy. Determinations of  $\text{H}_2\text{O}$ , density, and index of refraction are required to better establish determinative curves for the analcime-pollucite series.

Synthesis of pollucite is required to answer many of the questions posed by this study. What are the maximum Si/Al ratios as related to the CRK, in possible equilibrium with quartz or other silicates? Is pollucite with Si/Al of 2.0 most thermodynamically stable? Is there in fact no solid solution towards wairakite (Ca) or leucite (K), as suggested by the compositions of natural pollucite? Are

contents of Mg or Fe limited only by geochemical scarcity at the time of pollucite crystallization? What are the extents of  $\text{Li}^+$ ,  $\text{Rb}^+$ , and  $\text{P}^{5+}$  substitution? Complete definition of the solvus is required, as are studies of the stability of pollucite. No synthesis of sodian pollucite has been published, and the composition of this pollucite in equilibrium with albite as a function of temperature, pressure, and Cs/Na of co-existing fluid needs to be established.

The conditions of leaching, cation exchange, and alteration to clays needs to be established. The mechanisms of weathering of pollucite under surface conditions requires investigation as well. Additional studies of the alteration products are urgently required. This study has shown the existence of Rb-dominant adularia and associated Rb-dominant mica, so far not investigated in detail but representing potential new mineral species. The structure and composition of Cs-bearing clay minerals needs further study. These are essential to the containment of radioisotopes in waste package backfill, subseabed disposal, grouted waste injection, contaminated soils, host rock, and direct disposal in clays. Among the alteration products was noted KMn-bearing (melane) minerals which are natural analogues to hollandite-type phases used to contain  $\text{Cs}^+$  in some SYNROC formulations; though in small amounts, a careful study of these minerals could be rewarding.



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## APPENDIX 1

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Table 2.1a: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	1	2	3	4	5	6
SiO <sub>2</sub>	51.27	48.81	49.23	47.88	48.08	44.03
Al <sub>2</sub> O <sub>3</sub>	19.70	18.29	18.61	17.96	17.20	15.97
Fe <sub>2</sub> O <sub>3</sub>	----	0.11	0.45	0.14	0.32	0.68
P <sub>2</sub> O <sub>5</sub>	----	----	----	----	----	----
Li <sub>2</sub> O	0.80	0.34	0.43	0.12	0.42	----
Na <sub>2</sub> O	3.58	3.62	3.20	3.58	3.10	3.88
K <sub>2</sub> O	----	0.50	0.59	1.06	1.00	----
Rb <sub>2</sub> O	----	1.30	0.00	----	----	----
Cs <sub>2</sub> O	22.90	25.69	22.80	23.46	26.61	34.07
CaO	0.76	0.37	0.23	1.04	0.57	0.68
MgO	trace	0.19	0.63	0.38	trace	----
H <sub>2</sub> O <sup>+</sup>	0.93	1.10	3.28	3.45	2.79	2.40
H <sub>2</sub> O <sup>-</sup>	----	0.00	0.00	0.37	----	----
sum	100.24	100.32	99.51	99.44	100.04	101.71
Si	2.082	2.075	2.076	2.071	2.095	2.051
Al	0.943	0.916	0.925	0.923	0.883	0.877
Fe	-----	0.003	0.014	0.004	0.010	0.023
P	-----	-----	-----	-----	-----	-----
Li	0.131	0.058	0.073	0.021	0.074	-----
Na	0.282	0.298	0.262	0.300	0.262	0.351
K	-----	0.027	0.032	0.059	0.056	-----
Rb	-----	0.036	0.000	-----	-----	-----
Cs	0.397	0.466	0.410	0.433	0.494	0.677
Ca	0.033	0.017	0.011	0.048	0.027	0.034
Mg	-----	0.012	0.040	0.025	-----	-----
Al + Fe	0.943	0.920	0.939	0.920	0.894	0.900
Σ charges	0.849	0.942	0.880	0.958	0.939	1.095
Si/Al	2.208	2.264	2.244	2.262	2.372	2.279
CRK*	47.0	52.8	53.3	55.5	60.3	61.8
n	-----	-----	-----	-----	1.527	-----
D	-----	-----	-----	-----	2.89	-----

\* CRK = 100(Cs + Rb + K)/cation sum; atomic contents based on a subcell with 6 oxygens in anhydrous formula.

1. Unknown locality, U.S.S.R. (also Pb<sub>2</sub>O 0.30 wt.%; Saltykov 1959)

2. Far East, U.S.S.R. (Vlasov 1964)

3. Siberia, U.S.S.R. (also MnO 0.06 wt.%; Vlasov 1964)

4. Tin Mt., Custer Co., South Dakota (Connolly & O'Hara 1929; quoted in Schwartz 1930 and in Roberts & Rapp 1965)

5. Ungursay # 3, Kalbin Range, eastern Kazakhstan, U.S.S.R. (also trace Be, Sr, Sn, Mn, Ga; Ginsburg 1946)

6. San Piero in Campo, Elba, Italy (Pisani 1864)

Table 2.1b: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	7	8	9	10	11	12
SiO <sub>2</sub>	48.00	47.92	47.09	40.71	46.30	45.66
Al <sub>2</sub> O <sub>3</sub>	19.60	18.71	17.41	19.69	18.00	16.64
Fe <sub>2</sub> O <sub>3</sub>	----	trace	----	0.32	trace	0.22
P <sub>2</sub> O <sub>5</sub>	----	----	0.30	----	0.00	----
Li <sub>2</sub> O	0.25	----	0.41	0.80	trace	0.22
Na <sub>2</sub> O	2.56	3.15	3.02	1.09	~3.0	2.12
K <sub>2</sub> O	----	----	2.00	0.74	trace	0.13
Rb <sub>2</sub> O	----	----	0.91	0.81	trace	0.00
Cs <sub>2</sub> O	27.10	27.20	26.60	31.35	30.50	31.80
CaO	0.50	----	0.00	1.46	trace	0.36
MgO	----	----	0.00	0.43	----	0.29
H <sub>2</sub> O <sup>+</sup>	1.83	2.55	2.20	2.21	2.30	2.17
H <sub>2</sub> O <sup>-</sup>	----	----	----	----	----	0.42
sum	100.19	99.53	99.94	99.67	99.00	100.13
Si	2.058	2.087	2.069	1.895	2.076	2.088
Al	0.990	0.960	0.901	1.080	0.951	0.897
Fe	-----	-----	-----	0.011	-----	0.007
P	-----	-----	0.011	-----	0.000	-----
Li	0.043	-----	0.072	0.150	-----	0.041
Na	0.213	0.266	0.257	0.098	0.261	0.188
K	-----	-----	0.112	0.044	-----	0.008
Rb	-----	-----	0.025	0.024	-----	0.000
Cs	0.495	0.505	0.498	0.622	0.583	0.620
Ca	0.023	-----	0.000	0.073	-----	0.018
Mg	-----	-----	0.000	0.030	-----	0.020
Al + Fe	0.990	0.960	0.901	1.091	0.951	0.904
Σ charges	0.799	0.771	0.966	1.149	0.844	0.938
Si/Al	2.078	2.173	2.295	1.754	2.182	2.328
CRK	63.8	65.5	65.9	66.2	69.1	69.9
n	-----	-----	-----	1.522	-----	-----
D	-----	-----	-----	2.881	-----	-----

7. Unknown locality, U.S.S.R. (also Pb<sub>2</sub>O 0.35 wt.%; Saltykov 1959)

8. Sayan, U.S.S.R. (Fel'dman & Pleskova 1978; mineralogy of this occurrence is described in Filippova 1970)

9. Bikita, Fort Victoria area, Zimbabwe (Cooper 1964)

10. Siberia, U.S.S.R. (also MnO 0.06 wt.%; Feklichev 1975)

11. An American locality (Kastler 1927)

12. Tin Mt., Custer Co., South Dakota (also FeO 0.10 wt.%, no Ti; Wells & Stevens 1937)

Table 2.1c: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	13	14	15	16	17	18
SiO <sub>2</sub>	46.03	46.00	45.70	46.48	45.98	47.29
Al <sub>2</sub> O <sub>3</sub>	17.76	19.00	17.20	17.24	17.23	16.00
Fe <sub>2</sub> O <sub>3</sub>	0.02	----	0.00	----	0.08	0.60
P <sub>2</sub> O <sub>5</sub>	----	----	----	----	----	----
Li <sub>2</sub> O	0.16	----	trace	----	0.34	----
Na <sub>2</sub> O	2.08	2.01	2.80	2.25	2.09	2.21
K <sub>2</sub> O	1.81	----	as Rb	0.60	0.39	----
Rb <sub>2</sub> O	0.61	----	1.30	----	0.22	----
Cs <sub>2</sub> O	28.88	31.41	30.20	30.62	31.45	30.77
CaO	0.03	0.39	0.00	----	0.09	0.32
MgO	----	0.72	0.00	----	----	----
H <sub>2</sub> O <sup>+</sup>	2.74	----	2.66	2.32	2.19	1.45
H <sub>2</sub> O <sup>-</sup>	----	----	----	----	0.14	0.03
sum	100.13	99.53	99.86	99.51	100.20	99.58
Si	2.071	2.034	2.087	2.104	2.085	2.133
Al	0.942	0.990	0.926	0.920	0.921	0.851
Fe	0.001	-----	0.000	-----	0.003	0.020
P	-----	-----	-----	-----	-----	-----
Li	0.029	-----	-----	-----	0.062	-----
Na	0.181	0.172	0.248	0.198	0.184	0.193
K	0.104	-----	as Rb	0.034	0.023	-----
Rb	0.018	-----	0.038	-----	0.006	-----
Cs	0.554	0.592	0.588	0.591	0.608	0.592
Ca	0.001	0.019	0.000	-----	0.004	0.016
Mg	-----	0.047	0.000	-----	-----	-----
Al + Fe	0.942	0.990	0.926	0.920	0.923	0.870
Σ charges	0.879	0.896	0.874	0.823	0.892	0.856
Si/Al	2.199	2.054	2.254	2.287	2.264	2.508
CRK	70.6	71.3	71.6	71.8	71.8	72.1
n	1.520	-----	1.517	-----	-----	-----
D	-----	-----	2.865	-----	-----	-----

13. Nagatare, Fukuoka Prefecture, Japan (also MnO 0.01 wt.%; Sakurai et al. 1972; mineralogy of this occurrence is described in Kuwano 1970)

14. Unknown locality (Plyushchev & Shakhno 1958)

15. Helikon, near Karibib, Namibia (also trace Tl, Ga; no B, Mn, Sr, Pb, Ti, V, Cr; Nel 1944)

16. San Piero in Campo, Elba, Italy (Rammelsberg 1880, quoted in Wells 1891)

17. Myokenzan, Ibaraki Prefecture, Japan (Sakurai et al. 1977)

18. Northwestern European U.S.S.R. (also FeO 0.53 wt.%; Fel'dman & Pleskova 1978)

Table 2.1d: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	19	20	21	22	23	24
SiO <sub>2</sub>	43.76	45.56	45.25	45.81	47.80	46.09
Al <sub>2</sub> O <sub>3</sub>	16.75	16.15	16.38	17.38	15.70	16.38
Fe <sub>2</sub> O <sub>3</sub>	0.15	0.06	0.77	0.06	0.01	0.01
P <sub>2</sub> O <sub>5</sub>	----	----	----	0.21	----	----
Li <sub>2</sub> O	----	0.15	----	0.049	0.28	0.43
Na <sub>2</sub> O	2.05	2.30	2.28	1.97	1.98	1.74
K <sub>2</sub> O	----	0.50	0.19	0.32	0.17	0.16
Rb <sub>2</sub> O	0.53	0.36	----	0.82	0.77	0.74
Cs <sub>2</sub> O	33.03	30.91	32.31	30.38	30.60	32.20
CaO	0.90	0.05	0.72	1.02	0.04	0.01
MgO	0.28	0.08	----	0.12	0.003	0.003
H <sub>2</sub> O <sup>+</sup>	2.46	2.36	2.66	2.19	2.11	1.73
H <sub>2</sub> O <sup>-</sup>	----	0.02	----	----	0.08	0.04
sum	99.96	98.60	100.56	100.39	99.54	99.53
Si	2.053	2.110	2.084	2.065	2.160	2.112
Al	0.926	0.881	0.889	0.924	0.836	0.885
Fe	0.005	0.002	0.026	0.002	0.000	0.000
P	-----	-----	-----	0.008	-----	-----
Li	-----	0.028	-----	0.009	0.051	0.079
Na	0.186	0.207	0.204	0.172	0.174	0.155
K	-----	0.030	0.011	0.019	0.010	0.010
Rb	0.016	0.011	-----	0.024	0.022	0.022
Cs	0.661	0.610	0.635	0.584	0.590	0.629
Ca	0.045	0.003	0.036	0.049	0.002	0.001
Mg	0.020	0.006	-----	0.009	0.000	0.000
Al + Fe	0.931	0.883	0.915	0.926	0.837	0.885
Σ charges	0.995	0.901	0.920	0.915	0.850	0.896
Si/Al	2.217	2.394	2.344	2.236	2.583	2.388
CRK	72.9	72.9	73.0	73.1	73.3	73.8
n	1.518	-----	-----	----	1.517	1.520
D	2.916	-----	2.896	2.88	2.84	2.85

19. Kazakhstan, U.S.S.R. (also MnO 0.05 wt.%; Feklichev 1975)

20. Henan # 2, China (Liu & Shan 1990)

21. San Piero in Campo, Elba, Italy (Gossner & Reindl 1932)

22. Kazakhstan, U.S.S.R. (Aramakov et al. 1975)

23 and 24. Tanco, Bernic Lake, Manitoba (Cerny & Simpson 1978)

Table 2.1e: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	25	26	27	28	29	30
SiO <sub>2</sub>	46.22	46.22	45.73	48.22	46.50	45.48
Al <sub>2</sub> O <sub>3</sub>	15.97	15.84	17.98	17.94	16.76	16.01
Fe <sub>2</sub> O <sub>3</sub>	0.14	0.01	----	----	0.25	0.11
P <sub>2</sub> O <sub>5</sub>	----	----	----	----	----	----
Li <sub>2</sub> O	0.15	0.15	0.015	0.538	----	0.15
Na <sub>2</sub> O	2.30	2.20	1.37	1.07	2.23	2.10
K <sub>2</sub> O	0.50	0.50	0.03	0.04	----	0.59
Rb <sub>2</sub> O	0.27	0.45	0.21	0.34	----	0.45
Cs <sub>2</sub> O	31.15	32.17	32.57	30.41	32.62	31.60
CaO	----	0.09	0.19	0.20	0.30	----
MgO	----	----	----	----	----	0.09
H <sub>2</sub> O <sup>+</sup>	2.58	2.28	2.50	2.07	0.54	2.32
H <sub>2</sub> O <sup>-</sup>	0.24	0.08	0.01	0.02	----	0.10
sum	99.58	100.11	100.59	100.31	99.20	99.09
Si	2.125	2.126	2.072	2.110	2.108	2.112
Al	0.865	0.859	0.960	0.925	0.896	0.876
Fe	0.005	0.000	-----	-----	0.008	0.004
P	-----	-----	-----	-----	-----	-----
Li	0.028	0.028	0.003	0.095	-----	0.028
Na	0.205	0.196	0.120	0.091	0.196	0.189
K	0.029	0.029	0.002	0.002	-----	0.035
Rb	0.008	0.013	0.006	0.010	-----	0.013
Cs	0.611	0.631	0.629	0.567	0.631	0.626
Ca	-----	0.004	0.092	0.009	0.015	-----
Mg	-----	-----	-----	-----	-----	0.006
Al + Fe	0.870	0.881	0.960	0.925	0.904	0.880
Σ charges	0.881	0.906	0.945	0.784	0.856	0.903
Si/Al	2.456	2.476	2.158	2.281	2.354	2.410
CRK	74.5	74.7	74.7	74.8	75.0	75.1
n	-----	-----	1.5177	1.5111	-----	-----
D	-----	-----	2.880	2.923	-----	-----

25. Hubei, China (Liu & Shan 1990)

26. Henan # 3, China (Liu & Shan 1990)

27. Bikita mine, Fort Victoria area, Zimbabwe (Khalili & von Knorring 1976)

28. Benson # 3, M'toko area, Zimbabwe (Khalili & von Knorring 1976)

29. Northwestern European U.S.S.R. (Fel'dman & Pleskova 1978)

30. Henan # 1, China (Liu & Shan 1990)



Table 2.1f: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	31	32	33	34	35	36
SiO <sub>2</sub>	44.33	46.90	46.28	45.82	46.92	43.54
Al <sub>2</sub> O <sub>3</sub>	18.44	17.28	16.71	16.00	17.27	16.90
Fe <sub>2</sub> O <sub>3</sub>	0.20	0.33	0.01	0.05	0.08	----
P <sub>2</sub> O <sub>5</sub>	----	----	0.25	----	----	----
Li <sub>2</sub> O	0.13	0.18	0.25	0.018	0.20	0.13
Na <sub>2</sub> O	1.76	1.95	1.87	2.23	1.27	2.11
K <sub>2</sub> O	0.15	0.82	0.51	0.43	----	0.42
Rb <sub>2</sub> O	----	2.13	1.60	1.38	0.85	trace
Cs <sub>2</sub> O	32.14	29.10	30.77	31.20	30.80	36.03
CaO	0.298	0.14	0.00	0.05	0.45	----
MgO	0.20	0.08	0.00	----	0.18	----
H <sub>2</sub> O <sup>+</sup>	1.77	1.06	1.80	2.30	1.53	1.59
H <sub>2</sub> O <sup>-</sup>	----	----	0.00	----	0.02	----
sum	99.42	99.97	100.02	99.48	99.57	100.72
Si	2.030	2.090	2.098	2.104	2.106	2.054
Al	0.995	0.908	0.893	0.866	0.914	0.940
Fe	0.007	0.011	0.000	0.002	0.003	----
P	-----	-----	0.010	-----	-----	-----
Li	0.024	0.032	0.046	0.003	0.036	0.025
Na	0.156	0.169	0.164	0.199	0.111	0.193
K	0.009	0.047	0.295	0.038	-----	0.025
Rb	-----	0.061	0.047	0.041	0.025	-----
Cs	0.628	0.553	0.595	0.611	0.590	0.725
Ca	0.015	0.032	0.000	0.003	0.022	-----
Mg	0.014	0.005	0.000	-----	0.012	-----
Al + Fe	1.002	0.918	0.893	0.868	0.916	0.939
Σ charges	0.873	0.885	0.881	0.898	0.828	0.968
Si/Al	2.040	2.303	2.350	2.430	2.305	2.186
CRK	75.3	75.6	76.2	77.1	77.3	77.5
n	-----	-----	1.5218	-----	1.524	-----
D	-----	-----	2.917	2.849	2.94	2.98

31. Koktokay # 3, Xinjiang region, China (Liu & Shan 1990)  
 32. Unknown locality (Panasenko & Goroshchenko 1970)  
 33. Varuträsk, Sweden (Quensel 1938)  
 34. South Ghana (Agamor et al. 1986)  
 35. Sayan, U.S.S.R. (also trace Tl, Cu, Ti, Ga, Ge; Melentyev 1961)  
 36. Black Mt., Rumford, Maine (Foote 1896)

Table 2.1g: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	37	38	39	40	41	42
SiO <sub>2</sub>	45.26	45.04	44.60	44.50	43.59	45.40
Al <sub>2</sub> O <sub>3</sub>	15.95	15.88	16.36	15.82	16.39	15.28
Fe <sub>2</sub> O <sub>3</sub>	----	0.06	0.01	0.01	----	0.01
P <sub>2</sub> O <sub>5</sub>	----	----	----	0.14	----	0.07
Li <sub>2</sub> O	0.25	0.15	0.17	0.21	0.04	0.03
Na <sub>2</sub> O	1.70	1.81	1.70	1.63	2.03	1.77
K <sub>2</sub> O	0.68	0.59	0.50	0.08	0.51	0.12
Rb <sub>2</sub> O	0.45	0.62	0.22	0.34	0.00	0.36
Cs <sub>2</sub> O	32.71	31.43	31.57	35.73	35.36	32.05
CaO	0.08	0.09	0.03	0.28	0.22	0.20
MgO	----	----	----	0.03	----	0.04
H <sub>2</sub> O <sup>+</sup>	1.86	1.74	2.19	1.62	----	1.92
H <sub>2</sub> O <sup>-</sup>	0.16	0.20	0.24	0.06	----	0.06
sum	99.19	97.70	98.02	100.26	98.14	97.38
Si	2.111	2.115	2.101	2.096	2.071	2.144
Al	0.877	0.879	0.908	0.878	0.918	0.851
Fe	-----	0.002	0.000	0.000	-----	0.000
P	-----	-----	-----	0.006	-----	0.003
Li	0.047	0.028	0.032	0.040	0.008	0.006
Na	0.154	0.165	0.155	0.149	0.187	0.162
K	0.041	0.035	0.030	0.005	0.031	0.007
Rb	0.014	0.019	0.007	0.010	0.000	0.011
Cs	0.651	0.629	0.634	0.719	0.716	0.645
Ca	0.004	0.005	0.002	0.014	0.011	0.010
Mg	-----	-----	-----	0.002	-----	0.003
Al + Fe	0.877	0.881	0.908	0.878	0.918	0.851
Σ charges	0.913	0.886	0.861	0.955	0.964	0.857
Si/Al	2.408	2.407	2.313	2.387	2.257	2.521
CRK	77.5	77.6	78.0	78.2	78.4	78.6
n	-----	-----	-----	1.523	1.525	1.518
D	-----	-----	-----	2.954	2.97	2.922

37. Shanxi, China (Liu & Shan 1990)

38. Henan # 4, China (Liu & Shan 1990)

39. Xinjiang, China (Liu & Shan 1990)

40. Tot Lake, near Dryden, Northwestern Ontario (Ucakuwun 1981)

41. Hebron, Maine (Wells 1891)

42. Tot Lake, near Dryden, Northwestern Ontario (Ucakuwun, 1981)

Table 2.1h: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	43	44	45	46	47	48
SiO <sub>2</sub>	45.10	45.20	45.02	46.60	47.42	43.75
Al <sub>2</sub> O <sub>3</sub>	15.44	16.98	16.32	18.50	16.02	16.77
Fe <sub>2</sub> O <sub>3</sub>	0.17	-----	0.08	-----	0.01	-----
P <sub>2</sub> O <sub>5</sub>	-----	-----	0.22	-----	-----	-----
Li <sub>2</sub> O	0.085	-----	-----	0.006	0.29	0.03
Na <sub>2</sub> O	1.84	2.04	1.94	2.00	1.30	2.06
K <sub>2</sub> O	0.24	0.61	0.22	1.00	0.10	0.33
Rb <sub>2</sub> O	0.18	-----	0.20	0.25	0.68	trace
Cs <sub>2</sub> O	34.00	33.02	32.34	32.00	32.57	36.25
CaO	0.19	-----	-----	-----	0.06	-----
MgO	0.02	-----	-----	-----	0.01	-----
H <sub>2</sub> O <sup>+</sup>	3.00	2.04	3.10	3.45	1.37	1.57
H <sub>2</sub> O <sup>-</sup>	-----	-----	-----	0.37	0.04	-----
sum	100.27	99.89	99.47	103.36	99.87	100.76
Si	2.125	2.089	2.103	2.065	2.150	2.065
Al	0.858	0.925	0.899	0.966	0.856	0.933
Fe	0.006	-----	0.003	-----	0.000	-----
P	-----	-----	0.009	-----	-----	-----
Li	0.016	-----	-----	0.001	0.053	0.006
Na	0.168	0.183	0.176	0.174	0.114	0.189
K	0.014	0.036	0.013	0.057	0.006	0.020
Rb	0.006	-----	0.006	0.007	0.020	-----
Cs	0.712	0.651	0.644	0.605	0.630	0.729
Ca	0.010	-----	-----	-----	0.003	-----
Mg	0.001	-----	-----	-----	0.001	-----
Al + Fe	0.863	0.925	0.899	0.966	0.856	0.933
Σ charges	0.938	0.870	0.839	0.842	0.830	0.944
Si/Al	2.478	2.259	2.341	2.137	2.512	2.214
CRK	78.9	79.0	79.1	79.3	79.3	79.4
n	1.522	1.520	-----	-----	1.523	-----
D	2.92	2.89	-----	-----	2.87	2.98

43. Kulam field, Afghanistan (Rossovskii 1977)  
 44. Leominster, Massachusetts (Richmond & Gonyer 1938)  
 45. Altai region (Fel'dman & Pleskova 1978)  
 46. Tin Mt., Custer Co., South Dakota (Roberts & Rapp 1965; values for Li<sub>2</sub>O and Rb<sub>2</sub>O from Ahrens 1947)  
 47. Tanco, Bernic Lake, Manitoba (Cerny & Simpson 1978)  
 48. Black Mt., Rumford, Maine (Foote 1896)

Table 2.1i: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	49	50	51	52	53	54
SiO <sub>2</sub>	46.79	45.37	44.32	47.65	46.18	45.60
Al <sub>2</sub> O <sub>3</sub>	17.99	16.96	17.13	17.14	16.92	15.70
Fe <sub>2</sub> O <sub>3</sub>	0.02	0.06	0.10	0.08	0.03	----
P <sub>2</sub> O <sub>5</sub>	----	----	0.16	----	----	----
Li <sub>2</sub> O	0.13	----	0.05	0.08	----	----
Na <sub>2</sub> O	1.74	1.90	1.64	1.36	1.70	1.90
K <sub>2</sub> O	2.10	0.61	0.42	----	1.11	0.10
Rb <sub>2</sub> O	1.59	----	0.07	----	0.22	0.50
Cs <sub>2</sub> O	27.54	32.51	33.85	32.37	29.00	34.50
CaO	0.03	----	0.22	0.29	0.03	----
MgO	trace	----	0.02	0.11	0.03	----
H <sub>2</sub> O <sup>+</sup>	2.77	2.39	1.90	1.06	2.30	1.50
H <sub>2</sub> O <sup>-</sup>	----	0.12	0.11	0.80	----	----
sum	100.73	99.92	100.11	100.26	97.52	99.80
Si	2.078	2.095	2.063	2.125	2.115	2.131
Al	0.942	0.923	0.936	0.901	0.913	0.865
Fe	0.001	0.002	0.003	0.001	0.001	----
P	----	----	0.006	----	----	----
Li	0.023	----	0.009	0.014	----	----
Na	0.150	0.170	0.148	0.118	0.151	0.172
K	0.119	0.036	0.025	----	0.065	0.006
Rb	0.045	----	0.002	----	0.007	0.015
Cs	0.552	0.640	0.672	0.615	0.566	0.688
Ca	0.001	----	0.011	0.014	0.002	----
Mg	----	----	0.001	0.007	0.002	----
Al + Fe	0.942	0.925	0.943	0.902	0.914	0.865
Σ charges	0.862	0.846	0.890	0.790	0.796	0.881
Si/Al	2.207	2.270	2.195	2.359	2.316	2.464
CRK	79.7	79.9	80.1	80.1	80.5	80.5
n	----	1.521	----	----	1.520	----
D	----	2.896	----	----	2.88	----

49. Nagatare, Fukuoka Prefecture, Japan (Sakurai et al. 1977)

50. Jeclov, Czechoslovakia (Miskovsky 1960; described in Miskovsky 1955)

51. Viitaniemi, Eräjärvi, Finland (also TiO<sub>2</sub> 0.01, FeO 0.10, MnO 0.01 and CeO 0.22 wt.%; Erämetsä & Sihvonen 1973)

52. Northwestern European U.S.S.R. (Sosedko 1954, quoted in Melentyev 1961)

53. Central Asia (Babaev & Okulov 1972; described in Okulov & Vinner 1973)

54. Buckfield, Maine (Cerny 1974; the occurrence is described in Landes 1925)

Table 2.1j: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	55	56	57	58	59	60
SiO <sub>2</sub>	43.48	44.89	45.46	45.60	43.90	44.70
Al <sub>2</sub> O <sub>3</sub>	16.41	16.29	16.30	17.60	16.60	14.90
Fe <sub>2</sub> O <sub>3</sub>	----	0.06	0.08	0.01	0.20	----
P <sub>2</sub> O <sub>5</sub>	----	0.28	----	0.41	----	----
Li <sub>2</sub> O	0.03	----	----	0.009	----	0.05
Na <sub>2</sub> O	1.72	1.76	1.74	1.65	1.52	1.74
K <sub>2</sub> O	0.47	0.33	0.35	0.01	----	0.06
Rb <sub>2</sub> O	0.00	0.64	0.78	0.73	----	1.00
Cs <sub>2</sub> O	36.77	33.13	32.46	32.43	35.63	36.20
CaO	0.21	----	----	0.00	0.45	----
MgO	----	----	----	0.00	----	----
H <sub>2</sub> O <sup>+</sup>	1.53	2.87	2.84	1.91	1.49	1.44
H <sub>2</sub> O <sup>-</sup>	----	----	----	0.00	----	----
sum	100.62	100.29	100.35	100.39	99.79	100.09
Si	2.070	2.097	2.116	2.073	2.075	2.135
Al	0.921	0.897	0.894	0.943	0.925	0.839
Fe	-----	0.002	0.003	0.000	0.007	-----
P	-----	0.011	-----	0.016	-----	-----
Li	0.006	-----	-----	0.002	-----	0.010
Na	0.159	0.159	0.157	0.145	0.139	0.161
K	0.005	0.020	0.021	0.001	-----	0.036
Rb	0.000	0.019	0.023	0.021	-----	0.031
Cs	0.746	0.660	0.644	0.629	0.718	0.737
Ca	0.011	-----	-----	0.000	0.023	-----
Mg	-----	-----	-----	0.000	-----	-----
Al + Fe	0.921	0.899	0.897	0.943	0.932	0.839
Σ charges	0.937	0.858	0.844	0.800	0.903	0.942
Si/Al	2.248	2.338	2.366	2.198	2.244	2.546
CRK	81.1	81.4	81.4	81.5	81.6	81.9
n	1.525	-----	-----	1.522	-----	1.520
D	2.97	-----	-----	2.832	-----	2.88

55. Hebron, Maine (Wells 1891)

56. Sayan, U.S.S.R. (Fel'dman & Pleskova 1978)

57. Altai region (Fel'dman & Pleskova 1978)

58. Luolamäki, Somero, Finland (Neuvonen & Vesasalo 1960)

59. Northwestern European U.S.S.R. (Fel'dman & Pleskova 1978)

60. Tanco, Bernic Lake, Manitoba (Nickel 1961)

Table 2.1k: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	61	62	63	64	65	66
SiO <sub>2</sub>	46.11	44.28	45.65	42.53	44.95	45.00
Al <sub>2</sub> O <sub>3</sub>	16.09	16.32	16.16	17.15	17.44	17.15
Fe <sub>2</sub> O <sub>3</sub>	0.01	----	----	0.19	----	----
P <sub>2</sub> O <sub>5</sub>	0.32	----	----	----	----	----
Li <sub>2</sub> O	0.27	----	0.018	0.006	0.075	0.056
Na <sub>2</sub> O	1.52	1.59	1.44	1.15	1.20	1.20
K <sub>2</sub> O	0.07	0.38	0.03	0.20	0.03	0.03
Rb <sub>2</sub> O	0.95	----	0.23	0.14	0.33	0.18
Cs <sub>2</sub> O	32.90	35.83	34.49	36.98	34.01	34.59
CaO	0.01	0.13	0.06	0.61	0.05	0.04
MgO	0.01	----	----	0.08	----	----
H <sub>2</sub> O <sup>+</sup>	1.69	1.62	2.46	1.36	2.17	2.07
H <sub>2</sub> O <sup>-</sup>	0.01	----	0.01	0.03	0.02	0.34
sum	99.95	100.15	100.53	100.42	100.20	100.58
Si	2.116	2.092	2.125	2.024	2.083	2.092
Al	0.870	0.909	0.886	0.978	0.953	0.940
Fe	0.000	----	----	0.007	----	----
P	0.012	----	----	----	----	----
Li	0.050	----	0.003	0.001	0.014	0.011
Na	0.135	0.146	0.130	0.106	0.108	0.108
K	0.004	0.023	0.002	0.012	0.002	0.002
Rb	0.028	----	0.007	0.004	0.010	0.005
Cs	0.644	0.722	0.694	0.751	0.672	0.686
Ca	0.001	0.007	0.003	0.031	0.003	0.002
Mg	0.001	----	----	0.006	----	----
Al + Fe	0.870	0.909	0.886	0.985	0.953	0.940
Σ charges	0.813	0.904	0.842	0.948	0.810	0.814
Si/Al	2.432	2.302	2.397	2.069	2.187	2.226
CRK	82.9	83.0	83.8	84.2	84.6	85.2
n	----	1.52	1.5173	1.522	1.5169	1.5143
D	----	2.97	2.848	2.95	2.867	2.869

61. Tanco, Bernic Lake, Manitoba (Bennington et al. 1983)  
 62. Greenwood A, Maine (Richmond & Gonyer 1938)  
 63. Helikon, near Karibib, Namibia (Khalili & von Knorring 1976)  
 64. Pamirs (Durnev et al. 1973)  
 65. Luolamäki, Somero, Finland (Khalili & von Knorring 1976)  
 66. Varuträsk, Skelleftea, Sweden (Khalili & von Knorring 1976)

Table 2.11: Compositional data, unit cell contents and physical properties of pollucite from the literature.

No.	67	68	69	70	71	4POLL
SiO <sub>2</sub>	45.03	45.14	45.12	45.10	43.56	47.06
Al <sub>2</sub> O <sub>3</sub>	17.35	16.13	18.87	18.07	18.53	16.06
Fe <sub>2</sub> O <sub>3</sub>	----	----	----	----	----	----
P <sub>2</sub> O <sub>5</sub>	----	----	----	----	----	----
Li <sub>2</sub> O	0.022	0.037	0.037	0.037	0.028	0.43
Na <sub>2</sub> O	1.24	1.20	1.20	1.05	1.11	1.75
K <sub>2</sub> O	0.03	0.03	0.03	0.04	0.03	0.12
Rb <sub>2</sub> O	0.11	0.25	0.29	0.41	0.08	0.77
Cs <sub>2</sub> O	34.89	35.52	32.93	33.49	35.00	31.81
CaO	0.03	0.04	0.06	0.11	0.02	----
MgO	----	----	----	----	----	----
H <sub>2</sub> O <sup>+</sup>	1.78	2.02	1.72	1.89	1.94	1.79
H <sub>2</sub> O <sup>-</sup>	0.10	0.10	0.11	0.24	0.05	----
sum	100.56	100.34	100.33	100.40	100.32	99.79
Si	2.086	2.120	2.037	2.071	2.033	2.136
Al	0.947	0.893	1.004	0.978	1.019	0.859
Fe	----	----	----	----	----	----
P	----	----	----	----	----	----
Li	0.004	0.007	0.007	0.007	0.005	0.079
Na	0.111	0.109	0.105	0.094	0.101	0.154
K	0.002	0.002	0.002	0.002	0.002	0.007
Rb	0.003	0.008	0.084	0.012	0.002	0.023
Cs	0.689	0.711	0.634	0.656	0.697	0.616
Ca	0.002	0.002	0.003	0.005	0.001	----
Mg	----	----	----	----	----	----
Al + Fe	0.942	0.893	1.004	0.978	1.019	0.859
Σ charges	0.813	0.841	0.838	0.781	0.809	0.878
Si/Al	2.202	2.374	2.029	2.118	1.995	2.486
CRK	85.6	85.9	86.3	86.4	86.8	73.5
n	1.5149	1.5162	1.5153	1.5135	1.5145	----
D	2.908	2.886	2.924	2.886	2.898	----

- 67. Muiane Mine, Alto Ligonha area, Mozambique (Khalili & von Knorring 1976)
- 68. Namacotche Mine, Alto Ligonha area, Mozambique (Khalili & von Knorring 1976)
- 69. Morrua mine, Alto Ligonha area, Mozambique (Khalili & von Knorring 1976)
- 70. Benson # 1, M'toko area, Zimbabwe (Khalili & von Knorring 1976)
- 71. Viitanemi, Eräjärvi, Finland (Khalili & von Knorring 1976)
- 4POLL. Unpublished ASTM 4POLL standard, locality unknown.