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**Pollucite and its alteration  
in Finnish pegmatites**

by David K. Teertstra, Seppo I. Lahti,  
Reijo Alviola and Petr Černý



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by

DAVID K. TEERTSTRA, SEPPO I. LAHTI, REIJO ALVIOLA,  
and PETR ČERNÝ

with 20 figures and 5 tables in the text



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The regional distribution, internal structure and mineralogy of pollucite-bearing pegmatites in Finland has been reviewed and combined with recent field work. The four localities of pollucite in the Finnish Precambrian (Luolamäki, Somero; Viitaniemi, Orivesi; Haapaluoma, Peräseinäjoki, and Oriselkä, Tohmajärvi) all belong to highly fractionated, complex pegmatites of the rare-element class; however, the geochemical features of two of the pegmatites are not particularly indicative of the presence of pollucite.

Pollucite and sequences of its alteration products were investigated using optical microscopy, the electron microprobe technique and X-ray powder diffraction. At most localities, primary pollucite (with  $CRK = 100 (Cs + Rb + K)/\Sigma$  cations = 78.9 — 87.2 and  $Si/Al = 2.26 — 2.52$ ) has re-equilibrated into a blebby matrix of Na-enriched ( $CRK \geq 76$  and  $Si/Al \leq 2.56$ ) and Cs-enriched ( $CRK \leq 88$  and  $Si/Al \geq 2.22$ ) domains, crosscut by Cs-rich veinlets  $\pm$  quartz ( $CRK \leq 93$  and  $Si/Al \geq 2.18$ ) with Na-rich pollucite adjacent to the veinlet margins ( $CRK \geq 76$  and  $Si/Al \leq 2.46$ ).

Sequences of alteration of pollucite are well-developed at Finnish localities, recording the evolution of late pegmatitic fluids. The stability and alteration of pollucite in these fluids are relevant to containment of nuclear waste in pollucite-related phases. Large pollucite bodies ( $> 1$  m) contain coarse fracture-filling veins with minerals possibly related to late primary phases: spodumene, petalite, quartz, albite, microcline and lepidolite.

All pollucite bodies ( $> 5$  cm) contain fine veins of microcrystalline mica, spodumene and cookeite. Adularia, locally with high Rb content, replaces pollucite at all localities. Cation exchange producing analcime is extensive. The analcimization of pollucite is closely associated with crystallization of clay minerals ( $\pm$  quartz), locally with high Cs contents.

Key words (GeoRef Thesaurus, AGI): pollucite, analcime, adularia, cesium, granitic pegmatites, Finland

*David K. Teertstra and Petr Černý*  
*Department of Geological Sciences, University of Manitoba, Winnipeg,*  
*Manitoba, Canada R3T 2N2.*

*Seppo I. Lahti and Reijo Alviola*  
*Geological Survey of Finland, SF-02150 Espoo, Finland.*

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## INTRODUCTION

Minerals in the analcime-pollucite series (ideally  $\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$  —  $\text{CsAlSi}_2\text{O}_6$ ) are of petrogenetic interest as indicators of advanced fractionation of complex, rare-element granitic pegmatites. Pollucite bodies represent valuable industrial sources of cesium. Their importance extends to problems of long-term containment of  $^{135}\text{Cs}$  and  $^{137}\text{Cs}$  nuclear wastes. Pollucite serves as a natural analogue of waste-bearing synthetic phases, and studies of its chemistry and alteration allow prediction of their probable behaviour.

The textural and compositional heterogeneity observed in primary pollucite from High Grade Dyke, Manitoba, Canada suggested a miscibility gap between Na- and Cs-enriched compositions, widening with decreasing temperature (Teertstra *et al.* 1992a). Neuvonen and Vesasalo (1960) reported the presence of analcime coexisting with pollucite at Luolamäki pegmatite, Finland and on this basis suggested immiscibility at low temperature. In addition, one of us (S.I.L.) discovered by X-ray powder diffraction the presence of analcime in pollucite at Haapaluoma and Viitaniemi pegmatites, Finland.

The discovery of analcime in pollucite specimens of Finnish pegmatites prompted us to study the pollucite in detail. The main aim of this study is to better characterize the crystallization history and subsolidus re-equilibration of pollucite in the four Finnish pegmatites that carry this mineral. Observations are based on field work, optical microscopy, and X-ray powder diffraction studies. The main emphasis, however, is on the

chemistry of analcime and pollucite, both thoroughly analysed by electron microprobe, and the relationships of pollucite to sequential products of alteration.

Alteration of pollucite has been noted by many authors (Landes 1925, Nel 1944, Ginsburg 1946, Melentyev 1961, Cooper 1964, Vlasov 1966, Nickel 1967, Aramakov *et al.* 1975, Khalili & von Knorring 1976, Smeds & Černý 1989); the sequence of alteration phenomena was examined by Quensel (1938, 1945), Shaub and Schenck (1954), Neuvonen and Vesasalo (1960), and Černý and Simpson (1978). On the basis of these investigations, Černý (1979, 1982) constructed a generalized sequence of alteration features. The consistency of this pattern of alteration allowed recognition of clay pods pseudomorphous after pollucite even at localities without relics of the original mineral (Černý 1978). Many of the alteration products are extremely fine-grained and occur within thin veins, thus limiting their characterization in these early studies. However, the electron microprobe technique provides access to the chemistry of the alteration products, including analcime generated by cation exchange. Results of this study aimed at the diverse products of alteration are also reported here. A thorough understanding of the secondary processes is essential from the economic viewpoint, as most of them tend to significantly disperse the Cs content of primary pollucite, and consequently reduce the ore grade in its deposits.

### Pollucite localities in the regional context

More than 30 local populations of rare-element granitic pegmatites have been so far encountered in the Finnish Precambrian (see Alviola 1989a). Most of them are distinctly peraluminous, with minerals of Li, Be, Sn,  $\text{Nb} \leq \text{Ta}$ , B and P, whereas the numbers of those with metaluminous to slightly peraluminous bulk mineralogy and Y-, REE-,  $\text{Nb} > \text{Ta}$ - and F-bearing accessory phases are limited (the LCT and NYF families, respectively; see the general pegmatite classification by Černý 1991a, 1991b). Cesium is enriched in the most fractionated lithium-bearing pegmatites; pollucite has so far been encountered in four of them.

Most of the Finnish LCT pegmatites, and all of the four pollucite-bearing bodies, are of Proterozoic age and situated as conformable sills,

discordant dikes, lenticular bodies or irregular intrusions in Svecokarelian schists and associated plutonic rocks. Most of the LCT pegmatite populations are associated with, and apparently derived from, synkinematic to late-kinematic granites of the 1.86 — 1.78 Ga age group (as defined by Simonen 1980a, 1980b). These granites commonly developed a pegmatitic facies along margins of their intrusions and in offshoots. Zoned pods segregated within the pegmatitic leucogranites locally contain assemblages of accessory minerals transitional to those typical of the surrounding pegmatitic aureoles.

The four pollucite-bearing pegmatites in Finland (Fig. 1) belong to the Li-rich complex type of rare-element pegmatites of Černý (1989, 1991 a, b) but to different subtypes of this category:

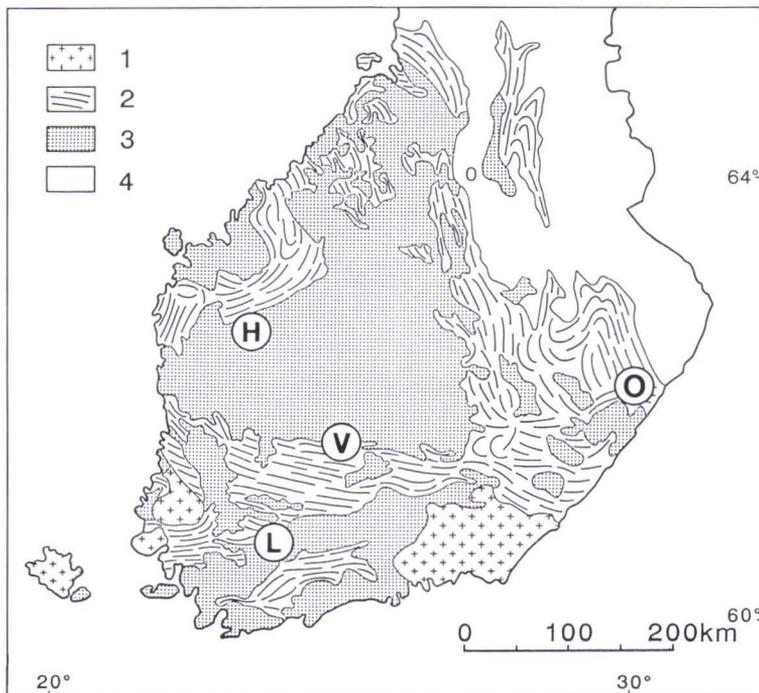


Fig. 1. Location of pollucite-bearing pegmatites of Finland, (L) = Luolamäki, (V) = Viitaniemi, (H) = Haapaluoma, (O) = Oriselkä. The map is schematized from Simonen (1980b). Symbols: (1) = Rapakivi granites, (2) = Svecokarelian schists, (3) = Svecokarelian igneous rocks, (4) = Archean basement.

1) The Luolamäki pegmatite, Somero, SW Finland and its pollucite were described and analysed by Neuvonen and Vesasalo (1960); abundant petalite, minor spodumene and a scarcity of Li- or (Fe, Mn)-bearing phosphates and lepidolite characterize this locality as the petalite subtype.

2) The Viitaniemi pegmatite in the Eräjärvi pegmatite area, Orivesi, central Finland was subject to a detailed examination by Volborth (1954) and Lahti (1981), and its pollucite was analysed by Erämetsä and Sihvonen (1973). Amblygonite-montebbrasite and triphylite-lithiophilite are the main Li-bearing minerals, late lepidolite is also relatively abundant, but neither petalite nor spodumene were found to date. This is one of the classic pegmatites of the amblygonite subtype.

3) The Haapaluoma pegmatite, Peräseinäjoki, W Finland was characterized by Haapala (1966).

Spodumene is the dominant mineral of Li, lepidolite is subordinate, and secondary eucryptite is rare (Lahti *et al.* 1982). Phosphates of Li have not been found, and the find of pollucite by S.I.L. in 1985 was so far only listed in a field trip guide (Alviola 1989a). This pegmatite conforms to the spodumene subtype.

4) The Oriselkä pegmatite, Tohmajärvi, E Finland was discovered (in 1971) and its pollucite identified (in 1978) by R.A., but it has not yet been introduced into the literature. Lepidolite dominates over spodumene here, Li-bearing phosphates are very subordinate, but elbaite (particularly rubellite) is abundant. In view of the rather limited outcrop, this pegmatite is only tentatively assigned to the lepidolite subtype.

Further details on the individual pegmatites are provided later, as introductions to the descriptions of individual occurrences of pollucite.

## EXPERIMENTAL METHODS

Polished thin sections of the analcime-pollucite minerals were analysed with a CAMEBAX SX-50 electron microprobe, using the »PAP» reduction procedure of Pouchou and Pichoir (1985). Backscattered electron imaging was used to select points for analysis.

For pollucite, major elements were measured using 15 kV, 20 nA, beam size 5  $\mu\text{m}$ , and counting times of 20 s (background 10 s). Standards used were pollucite (Al K $\alpha$ , Si K $\alpha$ , and Cs L $\alpha$ ), and albite (Na K $\alpha$ ). Minor elements were determined using 15 kV, 40 nA, beam size 5  $\mu\text{m}$ , and counting 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 elements were measured using 15 kV, 10 nA, beam size 20  $\mu\text{m}$ , and counting times of 20 s (background 10 s). These conditions were necessary to reduce sample burning

(volatilization of Na and H<sub>2</sub>O). Standards used were albite (Na K $\alpha$ , Al K $\alpha$ , and Si K $\alpha$ ), and pollucite (Cs L $\alpha$ ). Minor elements (Rb, Fe, K, Ca, and P) were determined using 15 kV, 20 nA, beam size 20  $\mu\text{m}$ , counting times of 50 s and the same standards as for pollucite.

Lower limits of detection for the minor oxides (in wt.%) were: 0.01 P<sub>2</sub>O<sub>5</sub>, K<sub>2</sub>O, CaO and MgO, 0.02 Fe<sub>2</sub>O<sub>3</sub> and 0.07 Rb<sub>2</sub>O.

Results of the analyses were recalculated to atomic contents on the basis of six atoms of oxygen per anhydrous subcell. The CRK index was calculated as  $100(\text{Cs} + \text{Rb} + \text{K})/\Sigma \text{ cations}$  (Černý 1974), where the cation sum = Li + Na + Ca + Mg + Cs + Rb + K. The CRK index is based on assignment of large cations Cs, Rb, and K to the 16*b* site which also hosts molecular H<sub>2</sub>O, and Li, Na, Ca, Mg and other small cations to the 24*c* site in Beger's (1969) model of the crystal structure of pollucite.

## LUOLAMÄKI, SOMERO

### The parent pegmatite

The Luolamäki pegmatite is a member of the Somero-Tammela pegmatite field, which is situated in SW Finland, about 100 km NW of Helsinki (Fig. 1). The field consists of more than 100 pegmatitic granites and pegmatites covering about 135 km<sup>2</sup>. Altogether 64 of them are known to contain rare-element minerals. Pegmatitic granites and pegmatites can be separated into 9 groups of spatially and mineralogically related bodies.

Almost all pegmatites are tourmaline-bearing with dominant muscovite and minor biotite. Four groups contain beryl- or chrysoberyl-bearing pegmatites with or without columbite and/or cassiterite. One group grades into the beryl-columbite-phosphate subtype (cf. Černý 1989, 1991a and 1991b for classification of granitic pegmatites). Three groups contain complex-type pegmatites: two dikes are of the petalite subtype (Vesasalo 1959, Neuvonen & Vesasalo 1960) and one of them is a pegmatite of the lepidolite or spodumene subtype. One group consists of complex-type pegmatites and also contains a single pegmatite which can be regarded as an albite-type pegmatite.

The updoming of microcline granites, east of the pegmatite field (Simonen 1956), caused folding and tilting of the supracrustal rocks into a broad synclinorium. Basic volcanics occupy the middle part of the synclinorium, whereas quartz-feldspar schists and mica schist crop out on the margins. Granodiorite, tonalite, diorite and gabbro intrusions are mainly in the central part but also on the flanks of the synclinorium. The rare-element pegmatites are situated within the synclinorium in the metavolcanics and in the basic and igneous rocks.

Porphyritic microcline granites from two localities, 40 — 50 km SSW of the Somero pegmatite field, have been dated at  $1830 \pm 14$  Ma (Muurla, zircon, Suominen 1991) and  $1840 \pm 8$  Ma (Ke-

miö, zircon, Suominen 1991). These are typical ages for late-kinematic granites in western Finland.

Outcrops of the Luolamäki pegmatite appear on the slopes of a small rounded hillock, 50 m in cross-section. The pegmatite is a flat concave sheet (Neuvonen & Vesasalo 1960) dipping inwards mostly at 10 — 20°, but locally up to 70 — 90°. Two holes were drilled in 1960 through the dyke; the thickness of the pegmatite sheet is 1 — 2 m at the NE corner and 8 — 10 m at the center of the dyke (Aurola 1963). The pegmatite is hosted by medium-grained equigranular and weakly schistose diorite.

The internal structure of the Luolamäki pegmatite is asymmetric. The hanging wall contact of the pegmatite is bordered by an albite-quartz aplite and the footwall by a fine-grained mica seam. The mica seam also contains schorl, chlorite and sillimanite. Drill holes reveal the upper half of the pegmatite to be composed of albite-rich pegmatite, and the lower half consisting in part to completely of K-feldspar pegmatite. The albitic pegmatite contains large quartz bodies and cleavelandite-bearing units near the core of the pegmatite. Cleavelandite, however, is missing and quartz pods are scarce farther away from the core. A few crystals of cassiterite, Nb,Ta-minerals, green elbaite, Li-mica and green alluaudite are found in the cleavelandite pegmatite. A few grains of lamellar alluaudite occur in the albite pegmatite with accessory schorl, garnet and muscovite.

The coarse-grained core is rather well exposed in an old quarry, which is an 11 m long, 5 m wide and 2 m deep pit and cave with large crystals of grey and water-clear petalite (up to 1.5 m long), quartz and pollucite. Petalite (Mikkola & Wiik 1947) and pollucite (Neuvonen & Vesasalo 1960) were identified for the first time in Finland from the Luolamäki pegmatite. The core is cut by veins

of white cleavelandite, quartz, schorl and muscovite. Some of the cleavelandite veins contain curvilamellar crystals of mauve lepidolite and locally green elbaite. About 6 m SE from the cave, in a drillhole, the core is represented by one petalite crystal, 5 cm in diameter, with its margins altered into a greenish mixture of clay minerals. Petalite crystals are commonly surrounded by a white alteration zone of spodumene + quartz intergrowth which penetrates them along cracks and cleavage fractures. Some petalite crystals are totally pseudomorphed this way.

### The pollucite

The pollucite is most closely associated with quartz, petalite, cleavelandite, K-feldspar and spodumene. Some cubic metres of pollucite-bearing rock have been excavated from the pit, and pollucite can be still observed on the cave walls. Coarse veins (0.5 cm) of albite and petalite cross-cut the pollucite masses (Neuvonen & Vesasalo 1960).

In hand specimen the pollucite is cloudy grey with a subvitreous lustre. Local small grains of the pollucite are water-clear, but usually the grey pollucite grades to white with a dull lustre. Scattered, blocky grains of pink K-feldspar up to 1 cm in size are embedded in the pollucite. Coarse veining by albite or petalite, most readily observed in outcrop, is poorly represented in hand specimen. The most abundant veins present in hand specimens are up to 5 mm wide and spaced at intervals of 3 to 5 cm. The veins are internally zoned, with rims of lepidolite or green muscovite

The Cs content in the muscovite and K-feldspar is low, 0.02 — 0.24 and  $\leq 0.09$  wt.%  $\text{Cs}_2\text{O}$ , respectively, although the Rb content in the primary blocky K-feldspar attains 1.50 wt.%, and increases in late nodules up to 2.98 wt.%  $\text{Rb}_2\text{O}$  (Neuvonen & Vesasalo 1960). The Li-mica and lepidolite contain 0.34 — 0.54 wt.%  $\text{Cs}_2\text{O}$ . No beryl has been spotted from the inner parts of the pegmatite. A greenish beryl from the marginal parts of the dyke contains 0.06 wt.%  $\text{Cs}_2\text{O}$ .

as coarse flakes perpendicular to a fine-grained, grey core. A second network of veins, at most 1 mm in width, is composed of white to pink fine-flaked mica or clay minerals. On the vein surfaces, and also fully embedded in the pollucite, are spherical brown to green feldspar grains 1 to 3 mm in size.

Margins of the pollucite bodies are heavily altered into a cryptocrystalline mixture of white, green and buff clay minerals. Earlier veining of the pollucite is preserved in the clayey matrix. Pollucite close to the altered rims is noticeably less dense than the central, fresh pollucite, even in hand specimen. Along fracture surfaces the pollucite is stained rusty yellow to brown, locally with a late spotty to dendritic coating of a black (Mn-bearing?) mineral. The products of alteration of pollucite are examined in greater detail below.

### Primary composition and subsolidus re-equilibration of pollucite

The pollucite locally contains areas which are compositionally and texturally homogeneous. These areas may be correlated with pollucite which appears most fresh in hand specimen, and optically homogeneous in thin section. In con-

trast, most of the pollucite which is white in hand specimen and with variable index of refraction in thin section, consists of Cs-enriched blebs, crosscut by Cs-rich veinlets containing small grains of quartz  $\pm$  spodumene  $\pm$  a

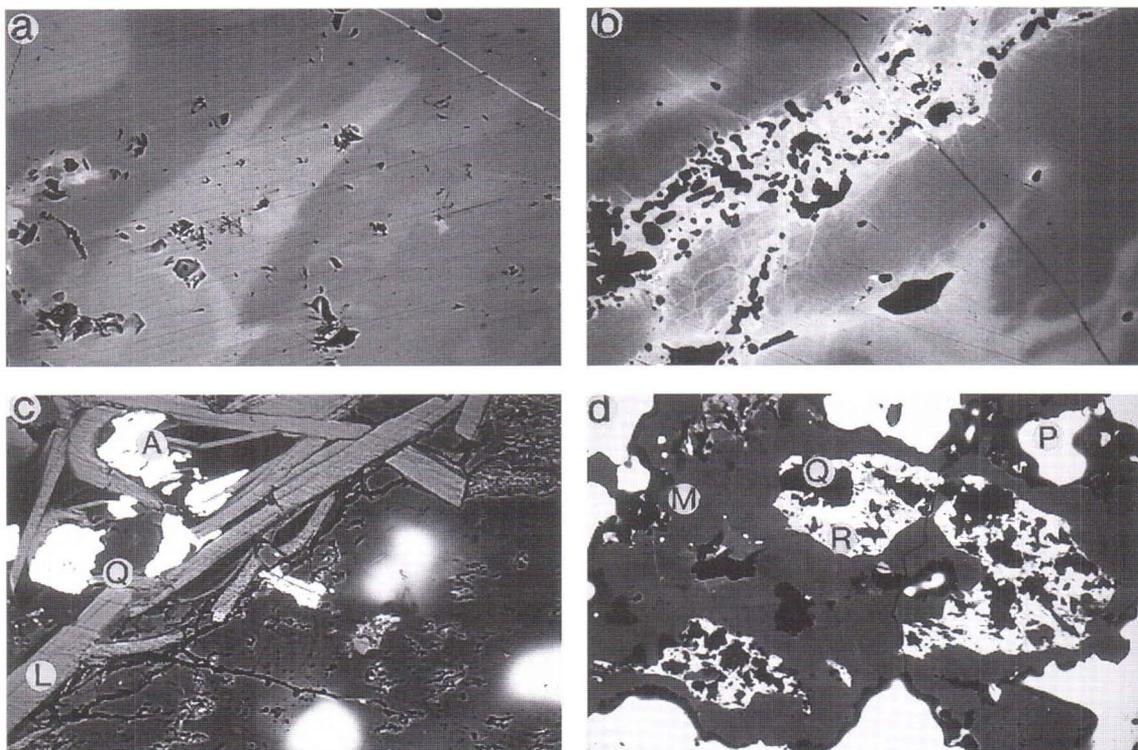


Fig. 2. BSE images of the Luolamäki pollucite. a) Cs-enriched blebs (lighter areas) within a matrix of primary pollucite intersected by a thin veinlet of Cs-rich pollucite (diagonal white line in the upper right corner). b) A relatively large veinlet of Cs-rich pollucite containing quartz and spodumene grains (black). c) Analcimization (black) leaving islands of pollucite (grey, grading to white) is most advanced near a vein of coarse lepidolite (L) + quartz (Q) ± apatite (A). d) RbAlSi-bearing inclusions (R) and quartz (Q) within a microcline vein (M). A thin rim of analcime surrounds the microcline and grades into pollucite (P). The horizontal edge of each image is 290  $\mu\text{m}$  long.

(Li?)AlP-bearing phase (Fig. 2a, b). Along fractures, veins, and around clay minerals, pollucite of all the above varieties is strongly analcimized (Fig. 2c, d). Compositions of the diverse types of the Luolamäki pollucite are listed in Table 1.

A plot of Si/Al vs CRK is shown in Fig. 3. The homogeneous pollucite has Si/Al from 2.44 to 2.52 and CRK from 79.2 to 82.2 (average, Table 1). The Cs-enriched blebs have Si/Al as low as 2.33 and CRK as high as 88.4. Cs-rich veinlets

have Si/Al as low as 2.18 and CRK as high as 92.8. The analcimized portions have CRK as low as 1.8, and Si/Al within the same range as the homogeneous pollucite, consistent with a cation exchange mechanism. However, at late stages of the cation exchange, the analcime also started crystallizing as fracture fillings. This late analcime with CRK < 10 can be recognized in Figure 3 by a wide variation in Si/Al from 2.14 to 2.91.

Table 1: Composition of pollucite from Luolamäki.

Oxide	Average		5	66	43	67
SiO <sub>2</sub> wt. %	46.93	(.32)	41.68	55.72	60.46	55.01
Al <sub>2</sub> O <sub>3</sub>	16.06	(.07)	16.21	19.28	17.61	21.87
Fe <sub>2</sub> O <sub>3</sub>	0.00		0.00	0.02	0.05	0.05
P <sub>2</sub> O <sub>5</sub>	0.35	(.01)	0.00	0.00	0.00	0.00
Na <sub>2</sub> O	1.78	(.08)	0.73	9.90	8.96	12.67
K <sub>2</sub> O	0.01		0.02	0.05	0.06	0.07
Rb <sub>2</sub> O	0.67	(.12)	0.18	0.00	0.00	0.00
Cs <sub>2</sub> O	32.10	(.35)	39.22	6.18	3.39	3.34
CaO	0.01		0.01	0.05	0.02	0.03
MgO	0.00		0.00	---	---	---
H <sub>2</sub> O	---		---	---	---	---
sum	97.91	(.35)	98.06	91.19	90.56	93.05
Si*	2.137	(.005)	2.064	2.136	2.245	2.040
Al	0.862	(.006)	0.946	0.871	0.771	0.956
Fe	0.000		0.000	0.000	0.002	0.001
P	0.013	(.001)	0.000	0.000	0.000	0.000
Na	0.157	(.007)	0.070	0.736	0.645	0.911
K	0.001		0.002	0.002	0.003	0.003
Rb	0.020	(.004)	0.006	0.000	0.000	0.000
Cs	0.623	(.008)	0.828	0.101	0.053	0.053
Ca	0.000		0.001	0.002	0.001	0.001
Mg	0.000		0.000	---	---	---
Al + Fe <sup>3+</sup>	0.862	(.006)	0.946	0.871	0.771	0.956
Σ charges	0.802	(.006)	0.908	0.843	0.704	0.969
Si/Al	2.479	(.023)	2.181	2.454	2.913	2.135
CRK	80.4	(0.9)	92.1	12.3	8.0	5.8

\* Atomic contents on the basis of six oxygen atoms per formula unit.

Average of homogeneous pollucite (N = 11).

5. Cs-rich veinlet.

66. Na-exchanged.

43, 67. Late cesian analcime.

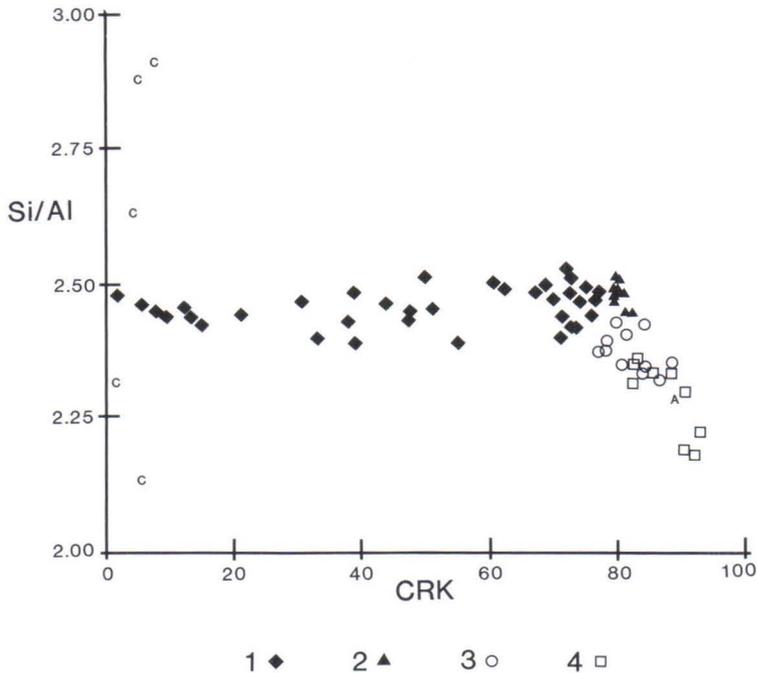


Fig. 3. Si/Al ratio vs CRK ( $= 100(\text{Cs} + \text{Rb} + \text{K})/\Sigma \text{ cations}$ ) for the Luolamäki pollucite. Symbols: (1) = Na-exchanged, (2) = primary homogeneous, (3) = Cs-enriched bleb, (4) = Cs-rich veinlet, (A) = inclusions in adularia, (C) = late cesian analcime.

### Alteration of the pollucite

Neuvonen and Vesasalo (1960) estimated the temperature of pollucite crystallization to be about  $450^{\circ}\text{C}$ , and described the alteration products of pollucite as follows. The pollucite contains thin needles and grains of apatite (up to 1 cm), and it is cut by veins of albite which are related to nearby bodies of cleavelandite + quartz + muscovite  $\pm$  tourmaline  $\pm$  columbite  $\pm$  beryl  $\pm$  laumontite. Rare thin veins of petalite, mostly altered to clay minerals, and wider veins of microcline were also found. Most abundant are thin veins (1 mm) of fine-grained micas, and fine-grained spodumene symplectitic with eucryptite(?).

The pollucite also contains spherical aggregates of K-feldspar (2.98 wt.%  $\text{Rb}_2\text{O}$ ) 1 to 4 mm in size. The pollucite is replaced, especially towards

its margins, by clay minerals consisting mostly of intergrown chlorite-montmorillonite with variable index of refraction (1.518 to 1.540), and kaolinite. Locally, the clay minerals fully replace

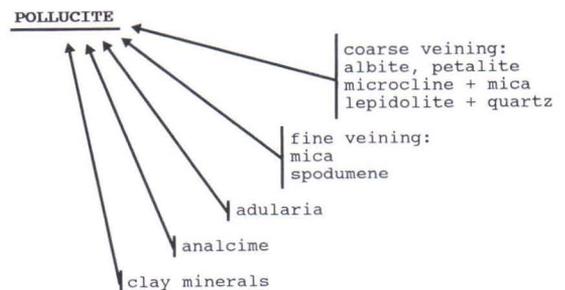


Fig. 4. Schematic sequence of alteration products of the Luolamäki pollucite. See text for details.

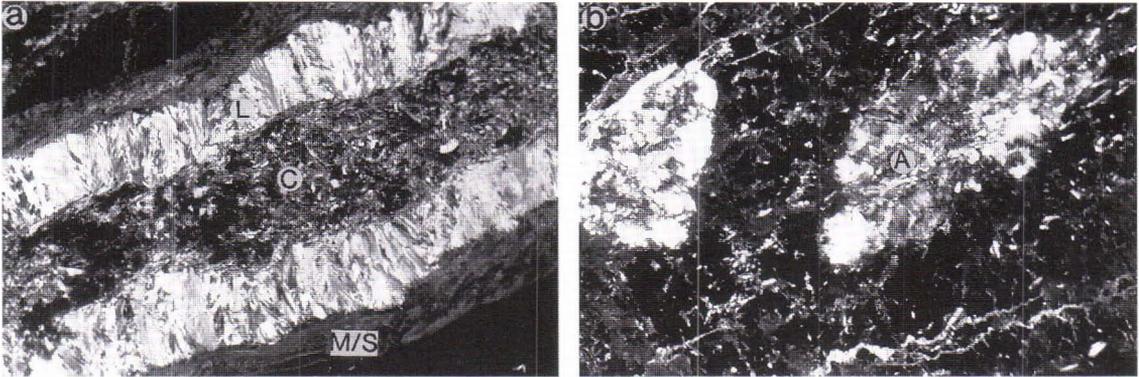


Fig. 5. Microphotographs of alteration products in the Luolamäki pollucite (black; crossed polars). a) A zoned vein, 6 mm in width. Core (C) consists of randomly oriented lepidolite in analcimized pollucite. Rims consist of radial flakes of coarse lepidolite (L) overgrown by fine-grained mica and spodumene (M/S) in a matrix of Cs-rich pollucite. Locally adularia follows the outer surfaces of the vein (not seen here). b) Fine mica veins and late granular aggregates of adularia (A). The adularia aggregate on the right is 3.3 mm long.

the pollucite, leaving intact the earlier veins and spherical aggregates.

Our observations described below agree in most respects with those of Neuvonen and Vesasalo (1960). Figure 4 shows the sequence of alteration products of the Luolamäki pollucite, reconstructed from the observations of Neuvonen and Vesasalo (1960) and our own.

The pollucite contains scattered grains of quartz, apatite and blocky K-feldspar. In our samples no veins of albite were found, but petalite veins (2 mm wide) are present, heavily replaced by clay minerals. Microcline veins (up to 1 cm wide) contain abundant mica and a RbAlSi-bearing phase (probably a feldspar). These veins seem to be fracture-filling rather than metasomatic.

Subparallel veins of relatively coarse-flaked lepidolite or muscovite + quartz  $\pm$  apatite are 1 to 2 mm in width. The flakes are perpendicular to the vein (Fig. 5a). Later irregular veins of very fine-grained mica less than 0.5 mm in width are locally replaced by fine-grained spodumene. These fine veins are abundant and seem to fol-

low a poor cubic parting of pollucite. They appear to be products of late crystallization rather than true replacements of pollucite. The veins do not cut entirely across the pollucite and adjacent mineral assemblages, but they pinch out within it.

Pollucite is replaced metasomatically by spherical granular aggregates of light green to brown adularia (up to 2 mm in size), completely embedded in the pollucite (Fig. 5b) or replacing pollucite along surfaces of all types of veins. The adularia locally replaces microcline. The Rb contents of the adularia are variable; in some cases they suggest atomic ratios of K/Rb slightly less than 1.0. Rb > K is typical of the feldspar phase coexisting with microcline. Abundant very thin »needles» are observed in optical microscopy, extending from the adularia (and veins) and also fully embedded in the pollucite. These are hollow capillaries associated with the analcimization, as indicated by a low index of refraction of their matrix relative to pollucite. Locally these capillaries are arranged in a three-dimensional rectangular grid. At a late stage, clay minerals invade pollucite as well as the analcime and capillaries.

### Cesian analcime veining in petalite

In altered petalite from Luolamäki, Neuvonen and Vesasalo (1960) identified analcime but found no Cs to be present. However, cesian analcime has been previously described from Greenwood, Maine and Tanco, Manitoba in association with the alteration of petalite (Richmond & Gonyer 1938, Černý 1972); consequently we also

examined the petalite at this locality. The petalite was found to contain widespread, but very thin ( $< 50 \mu\text{m}$  wide) veinlets of cesian analcime. The composition is variable within and between veins: Si/Al is scattered from 2.48 to 2.77 and CRK from 6.0 to 31.6 (Table 2).

Table 2: Composition of cesian analcime veins in petalite at Luolamäki.

Oxide	1	5	8	12	15
SiO <sub>2</sub> wt. %	52.71	57.59	56.87	58.42	55.69
Al <sub>2</sub> O <sub>3</sub>	17.11	19.00	17.41	17.98	19.06
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	0.04	0.00	0.02	0.00
Na <sub>2</sub> O	6.67	10.33	7.84	8.98	9.64
K <sub>2</sub> O	0.04	0.05	0.06	0.11	0.09
Rb <sub>2</sub> O	0.01	0.00	0.00	0.02	0.00
Cs <sub>2</sub> O	13.95	2.88	9.61	4.41	5.42
CaO	0.01	0.01	0.00	0.02	0.00
MgO	0.00	0.00	0.00	0.00	0.00
H <sub>2</sub> O	---	---	---	---	---
sum	90.51	89.91	91.78	89.97	89.92
Si*	2.179	2.167	2.213	2.212	2.147
Al	0.834	0.842	0.799	0.802	0.866
Fe	0.000	0.000	0.000	0.000	0.000
P	0.000	0.001	0.000	0.001	0.000
Na	0.535	0.754	0.592	0.659	0.721
K	0.002	0.002	0.003	0.005	0.004
Rb	0.000	0.000	0.000	0.001	0.000
Cs	0.246	0.046	0.159	0.071	0.090
Ca	0.001	0.000	0.000	0.001	0.000
Mg	0.000	0.000	0.000	0.000	0.000
Al + Fe <sup>3+</sup>	0.834	0.842	0.799	0.802	0.866
Σ charges	0.785	0.802	0.754	0.738	0.814
Si/Al	2.613	2.573	2.770	2.757	2.479
CRK	31.6	6.0	21.5	10.5	11.5

\* Atomic contents on the basis of six oxygen atoms per anhydrous formula unit.

- 1. High CRK.
- 5. Low CRK.
- 8 and 12. High Si/Al ratios.
- 15. Low Si/Al ratio.

## VIITANIEMI, ORIVESI

### The parent pegmatite

The Viitaniemi pegmatite is located about 50 km NE of the town of Tampere in the eastern part of the Tampere schist belt, which was folded and intruded by a variety of magmatic rocks during the Svecokarelian orogeny (Fig. 1). The bedrock of the area consists of diverse mica schists with intercalations of black schist, sericitic quartzite, quartz-feldspar gneiss and intermediate volcanic rocks. Several intrusions of intermediate plutonic rocks, granites, pegmatitic granites and pegmatites outcrop in the area.

During the orogeny, the schists were metamorphosed under amphibolite-facies conditions. Andalusite, garnet and locally staurolite form porphyroblasts in metapelites. The geology of the area is shown by the map sheets 2141 Kangasala (Matisto 1964) and 2142 Orivesi (Laitakari 1986). The most important granitic pegmatite occurrences are marked on these geological maps.

Close to 70 pegmatites with a diversity of accessory minerals of Li, Be, Nb, Ta, Sn, B and P and hundreds of barren pegmatites are known within an area of about 250 km<sup>2</sup> around Lake Eräjärvi. A great part of the pegmatites are distributed in schists along the contacts with pegmatitic granites and granitic intrusions. Most of the rare-element pegmatites are beryl pegmatites (beryl-columbite and beryl-columbite-phosphate subtypes), but a number of complex pegmatites of spodumene, lepidolite and amblygonite subtypes have also been observed (Lahti 1981, 1986, 1989, see the classification of the granitic pegmatites by Černý 1991a and b).

The pegmatites are postmetamorphic and cut the Svecokarelian igneous rocks of the area. Two radiometric age determinations are available from rare-element pegmatite dykes. According to the U-Pb dating at the Unit of Isotope geology of the Geological Survey of Finland, the age of the metamict zircon from the Viitaniemi pegmatite is  $1794 \pm 15$  Ma (O.Kouvo, personal com-

munication 1992), and likewise the preliminary lead isotope analysis on uranothorite from the Eräpyhä pegmatite indicates a similar age (Lahti 1981).

The age determinations indicate that the Viitaniemi pegmatite and possibly the other pegmatite dykes belong to the group of late-orogenic Svecokarelidic igneous rocks. A preliminary U-Pb age determination of three zircon fractions of the large, several kilometres long Rappuvuori granite intrusion, which is situated in the western part of the Eräjärvi pegmatite area, indicates, however, that the granites of the region are markedly older, possibly ranking with the syn-orogenic Svecokarelian granitoids (Matti Vaasjoki, personal communication 1992).

The Viitaniemi pegmatite forms a subhorizontal, quasi-lenticular body 100 — 130 m wide, up to 10 m thick and several hundred meters long, emplaced in mica schists. The pegmatite is exposed in two outcrops about 200 m apart, both extensively opened by quarries. The internal structure and mineralogy of the pegmatite were described in detail by Volborth (1954) and Lahti (1981).

The Viitaniemi pegmatite is well-zoned, but the original zoning is locally masked by albitic units and crystallization of late- and postmagmatic replacement assemblages and fracture fillings. The border zone, usually about 10 cm thick, is composed of fine-grained albite, quartz and muscovite with accessory schorl, intermediate microcline, beryl and apatite. The wall zone is coarse-grained, with albitic plagioclase, microcline, quartz and muscovite as the main minerals. The feldspars reach up to several decimeters in length, and muscovite locally forms large books. Black to dark green tourmaline, yellowish beryl, columbite and greenish or blue-green apatite are typical accessory minerals.

The intermediate zone gradually evolves from

the wall zone by coarsening of grain size. It contains microcline, quartz, albitic plagioclase (usually the platy albite, cleavelandite), large books of muscovite locally rimmed by brown lepidolite, and dark green tourmaline. The crystals of K-feldspar are up to several metres long and commonly subperpendicular to the outer contacts of the zone. Quartz forms a discontinuous "core" of irregularly branching to lenticular masses, reaching locally several metres in size.

Late- or postmagmatic units and fracture fillings form large branching bodies in the central parts of the dike, or crosscut the early zones. They consist mainly of saccharoidal albite or cleavelandite, quartz, muscovite, brownish lepidolite, topaz, green elbaite tourmaline, microcline, blue-green Mn-rich fluorapatite and diverse phosphates of Li, Fe-Mn and Be: amblygonite-montebbrasite, triphylite-lithiophilite, triplite, hurlbutite, herderite, beryllonite and väyrynenite are widespread. Amblygonite-montebbrasite occurs locally as the main mineral in the phosphate-rich units and fracture fillings.

Both the amblygonite-montebbrasite and triphy-

lite-lithiophilite are partly altered into numerous secondary phosphates. Besides the phases listed above, columbite-tantalite, cassiterite, zircon, microlite, wadginite, stibnite, native antimony and bismuth, and some other Sb-bearing minerals are found in accessory quantities in the Li-rich units. The detailed list of the minerals identified is in Lahti (1981).

The cesium content of the K-feldspar from the internal zones and units varies from 0.02 to 0.10 wt. %  $\text{Cs}_2\text{O}$ , along with the K/Rb ratio of 70 to 35 (Lahti 1981, Smeds & Černý 1989). Electron microprobe determinations indicate that the brownish Fe- and Mn-rich lepidolite, common in the intermediate zone and late cleavelandite-rich units, contains up to 0.73 wt. %  $\text{Cs}_2\text{O}$ . Volborth (1956) reported the Cs content of beryl increasing from 0.03 wt. % in yellow beryl from near pegmatite contacts with the country rocks to 0.66 wt. % in the pink variety associated with Li-enriched albitic units. An electron microprobe analysis (by the present authors) of similar pink beryl gave an even higher value, 0.90 wt. %  $\text{Cs}_2\text{O}$ .

### The pollucite

Pollucite has so far been found only in the smaller eastern quarry. It forms rounded granu-

lar aggregates within central parts of late units consisting mainly of cleavelandite, lepidolite,

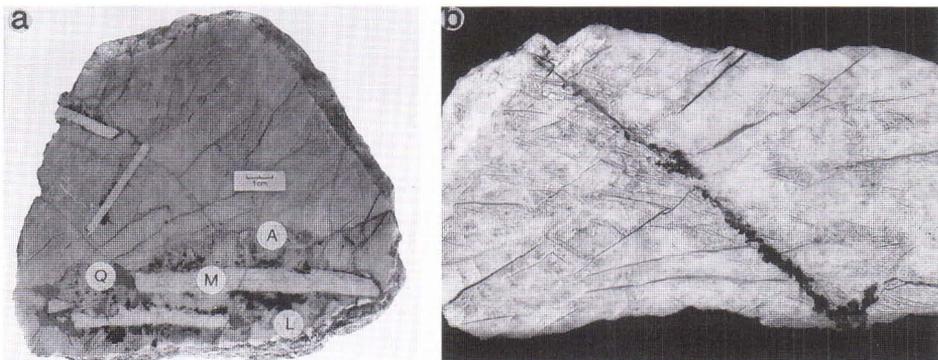


Fig. 6. Polished sections of the Viitaniemi hand specimens. a) Margins of pollucite aggregates contain laths of montebbrasite (M) overgrown by quartz (Q), albite (A) and lepidolite (L, dark); fine veins of alteration products criss-cross the pollucite interior. b) A coarse vein of lepidolite and later fine veins of fine-scaled green muscovite cross-cutting pollucite. The horizontal edge of the figure a is 19 cm and that of b 10 cm.

quartz and elbaite. Yellowish or pinkish beryl, montebrasite, topaz, columbite-tantalite, cassiterite, Mn-rich blue-green apatite and lithiophilite also are associated with pollucite. The pollucite aggregates are normally several centimetres to one decimetre in size; however, in 1963 a large

pod of pollucite weighing several tons was found in the central part of the eastern quarry.

The pollucite, largely white and opaque in hand specimen, is locally clear and colourless. Inclusions of quartz and deep blue apatite are observed; 10 cm long lath-shaped montebrasite crys-

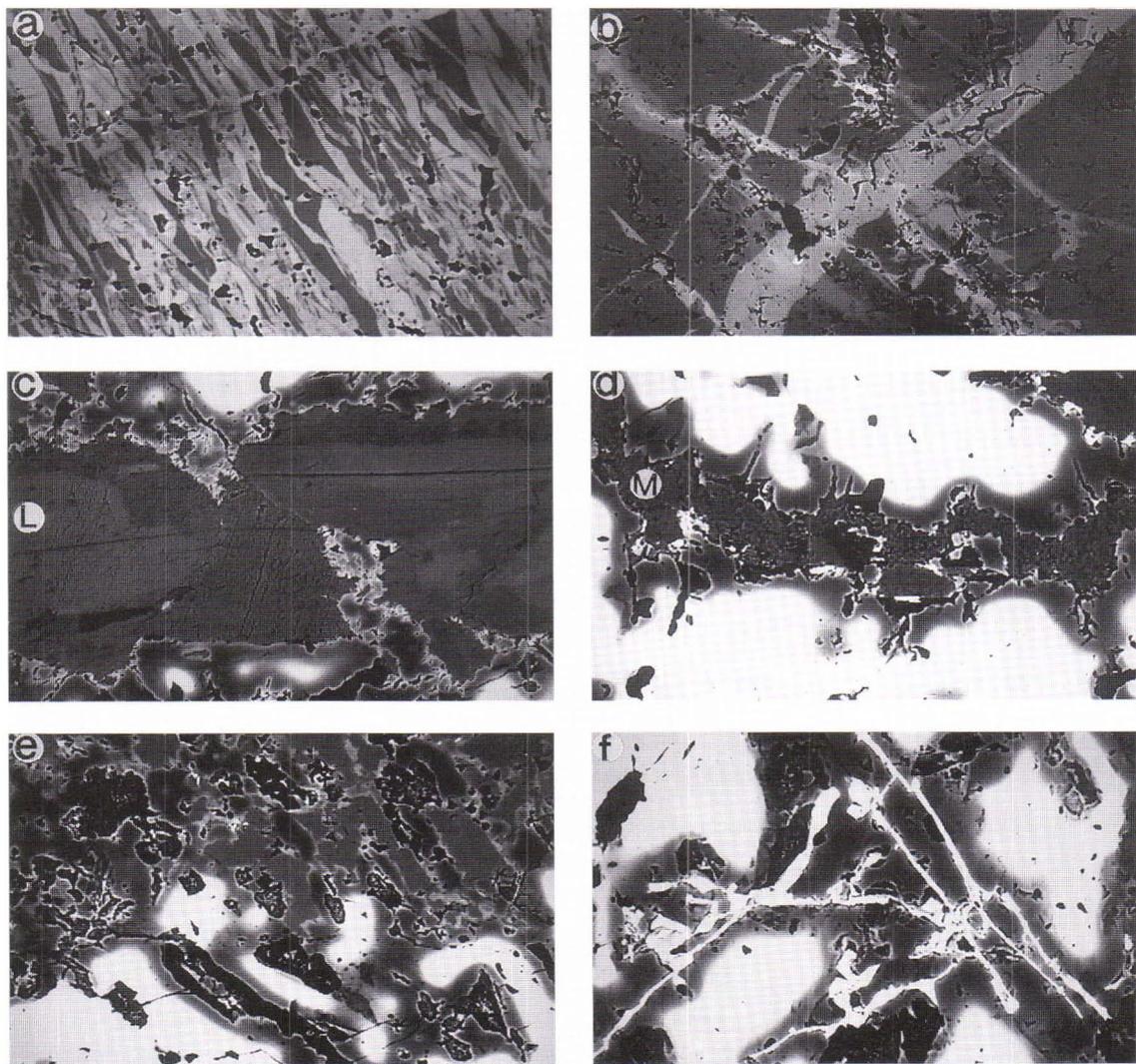


Fig. 7. BSE images of the Viitaniemi pollucite. a) Lamellar Cs-enriched blebs (white), associated with quartz grains (black) alternating with Na-enriched blebs. b) Multiple generations of Cs-rich veinlets cutting primary pollucite. Na-rich margins to the Cs-rich veinlets are rather diffuse and rarely obvious in BSE images. With advanced stages of Na-exchange (c to f), cesian analcime developed along silicate mineral surfaces: c) near a coarse vein of lepidolite (L), d) near a fine vein of mica (M), e) in association with clay minerals (black,  $\pm$  quartz) and f) in association with veins of apatite (white). The horizontal edge of each image is 280  $\mu\text{m}$  long.

tals are rimmed by albite, quartz and lepidolite (Fig. 6a). Veins of lepidolite and a network of thin green muscovite veins (1 mm wide) are obvious in a cut slab of pollucite (Fig. 6b). The separate pollucite specimens encountered in the pegmatite show various degrees of alteration. In

partly altered types the alteration products appear as a light brown network of veins in white pollucite; the strongly altered marginal parts of the specimens are light brown in their entirety. Fractures of the pollucite are stained rusty yellow to red.

### Primary composition and subsolidus re-equilibration of pollucite

Very little primary homogeneous pollucite is present in the bulk of the pollucite samples; however, some seems to have been preserved as an inclusion within amblygonite and albite. In contrast, blebby mosaics of Na- and Cs-enriched domains are well developed. The domains have a constant width of up to 10  $\mu\text{m}$ , are 100  $\mu\text{m}$  long, and locally have a lamellar structure. The »lamellae» occasionally bifurcate into pointed ends (Fig. 7a).

The blebby mosaic and the homogeneous pollucite are cross-cut by a dense network of several generations of Cs-rich veinlets ( $\pm$  quartz), locally rimmed on either side by Na-rich pollucite (Fig. 7b). A compositional traverse across such a veinlet and the adjacent homogeneous pollucite is given in Figure 8. Early veinlets seem to follow fractures, do not contain quartz, and may be locally offset relative to later veinlets. Cross-cutting veinlets are usually, though not necessarily, more Cs-rich; the latest veinlets have the highest Cs content and contain quartz grains.

Along fractures, veins, and near clay minerals the pollucite is strongly enriched in Na (Fig. 7c — f). Compositions of the diverse types of Viitaniemi pollucite are given in Table 3.

A plot of Si/Al vs CRK is shown in Figure 9. As the figure shows, the homogeneous pollucite has Si/Al from 2.38 to 2.49 and CRK from 78.9

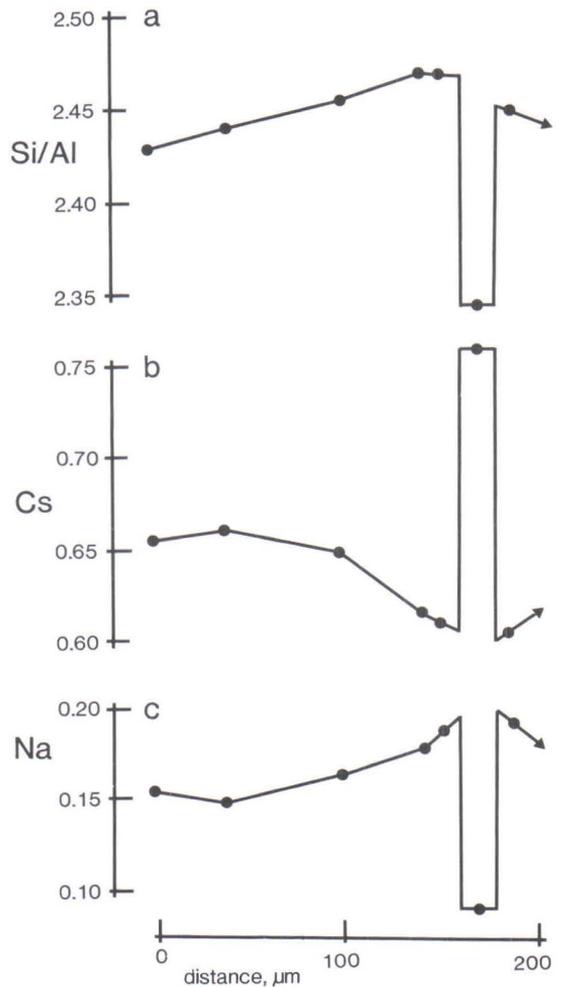


Fig. 8. Traverse from primary homogeneous pollucite of Viitaniemi to Na, Si-rich pollucite adjacent to a Cs-rich veinlet. Veinlet width and compositional trends were derived from BSE observations of the Viitaniemi pollucite. a) The highest values of Si/Al are found in the Na-rich margins, and the lowest in the Cs-rich veinlet. b) The veinlet margins have the lowest Cs content, in apparent mass balance with c) contents of Na, both as atoms p.f.u..

Table 3: Composition of pollucite from Viitaniemi.

Oxide	Average		49	29	62	78
SiO <sub>2</sub> wt. %	46.61	(.60)	49.16	47.38	42.56	56.29
Al <sub>2</sub> O <sub>3</sub>	15.94	(.06)	16.28	16.08	16.05	20.67
Fe <sub>2</sub> O <sub>3</sub>	0.00		0.01	0.01	0.01	0.01
P <sub>2</sub> O <sub>5</sub>	0.30	(.05)	0.09	0.33	0.01	0.13
Na <sub>2</sub> O	1.82	(.07)	2.09	2.12	0.88	11.92
K <sub>2</sub> O	0.02		0.00	0.00	0.01	0.15
Rb <sub>2</sub> O	0.04		0.00	0.00	0.00	0.00
Cs <sub>2</sub> O	32.89	(.43)	32.27	31.86	39.06	1.83
CaO	0.01		0.00	0.00	0.01	0.01
MgO	0.00		0.00	0.00	0.00	—
H <sub>2</sub> O	—		—	—	—	—
sum	97.64	(.51)	99.89	97.77	98.58	91.02
Si*	2.137	(.009)	2.166	2.142	2.082	2.092
Al	0.861	(.009)	0.846	0.856	0.925	0.906
Fe	0.000		0.000	0.000	0.000	0.000
P	0.012	(.002)	0.003	0.013	0.000	0.004
Na	0.161	(.005)	0.178	0.185	0.083	0.859
K	0.001		0.000	0.000	0.000	0.007
Rb	0.002		0.000	0.000	0.000	0.000
Cs	0.643	(.012)	0.607	0.614	0.815	0.029
Ca	0.000		0.000	0.000	0.000	0.000
Mg	0.000		0.000	0.000	0.000	—
Al + Fe <sup>3+</sup>	0.861	(.009)	0.846	0.856	0.925	0.906
Σ charges	0.820	(.012)	0.788	0.812	0.898	0.895
Si/Al	2.447	(.036)	2.552	2.464	2.252	2.310
CRK	79.9	(0.7)	77.3	76.8	90.7	4.0

\* Atomic contents on the basis of six oxygen atoms per anhydrous formula unit.

Average of homogeneous pollucite (N = 14).

49. Na-enriched bleb.

29. Na-rich margin to Cs-rich veinlet.

62. Cs-rich veinlet.

78. Na-exchanged.

to 81.6 (average, Table 3). The Na-enriched blebs have Si/Al as high as 2.56 and CRK as low as 76.2. The Cs-enriched blebs have Si/Al as low as 2.22 and CRK as high as 88.2. The Cs-rich veinlets have Si/Al as low as 2.26 and CRK as high as 91.2. The Na-rich flanks of the veinlets have Si/Al as high as 2.42 and CRK as low as

76.2.

Compositions enriched in Na due to analcimi- zation (see alteration below) have CRK as low as 4.0. Si/Al ratios are in the same range as those of the homogeneous pollucite, consistent with a cation exchange mechanism for their origin.

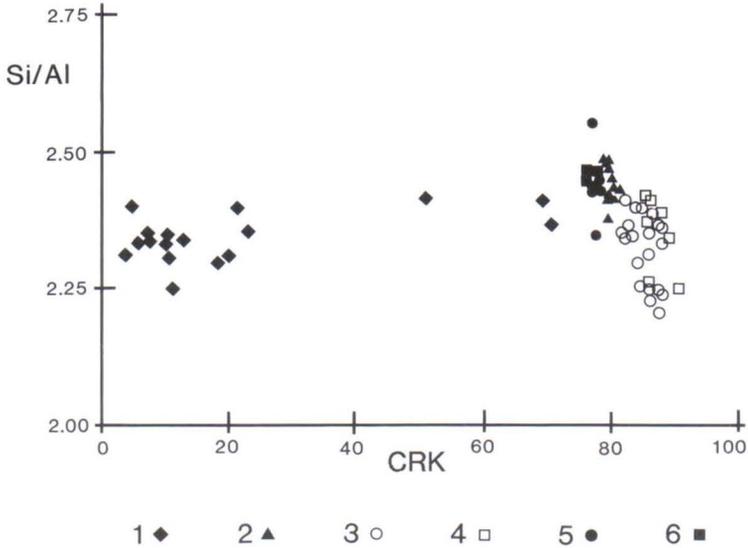


Fig. 9. Si/Al vs CRK for the Viitaniemi pollucite. Symbols: (1) = Na-exchanged, (2) = primary homogeneous, (3) = Cs-enriched bleb, (4) = Cs-rich veinlet, (5) = Na-enriched bleb, (6) = Na-rich margin to Cs-rich veinlet.

### Alteration of pollucite

A schematic alteration sequence is shown in Figure 10 as a guide to the following discussion. Rare thin veins of quartz cross the pollucite. Later veins of albite are also relatively rare. Locally these veins are overgrown by microcline + mica. These veins are relatively more abundant near the margins of the pollucite bodies. Veins of coarse-grained lepidolite + quartz ± apatite are common, both near the margins and within the pollucite (Fig. 6a, b, Fig. 7c and Fig. 12a, b, c). The flakes of mica in some veins are oriented normal to the attitude of the vein. Most common are thin veins of fine-grained mica (Fig. 6a, b, Fig. 7d, and Fig. 12b, c, d).

The veins typically follow fractures or a cube parting of the pollucite (Fig. 12c, d). The veins are overgrown by adularia (Fig. 12d), usually as spherical granular aggregates near the surface of the veins or fully embedded in the pollucite, but also as aggregates following the surface of the veins (Fig. 12c). The adularia metasomatically

replaces the pollucite. Neither adularia nor microcline show heterogeneity due to variable Rb content, in contrast to their counterparts at Luolamäki.

Several density fractions ranging from 2.20 to 3.05 g/cm<sup>3</sup>, separated from crushed pollucite in heavy liquids, were examined by X-ray powder

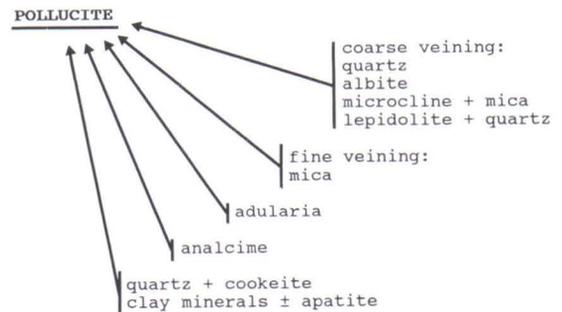


Fig. 10. Schematic sequence of alteration products of the Viitaniemi pollucite. See text for details.

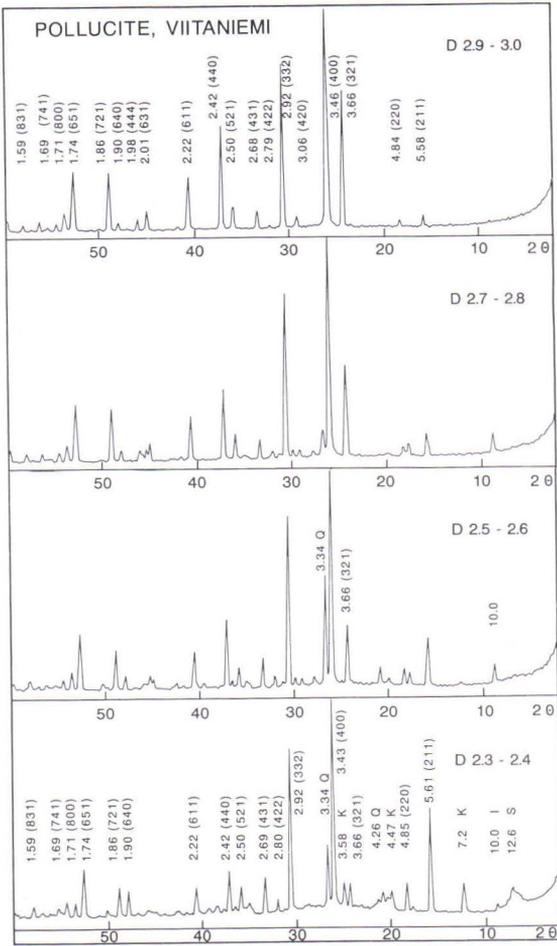


Fig. 11. X-ray powder diffraction patterns of the various density fractions of pollucite plus its alteration products from Viitaniemi. Varying proportions of analcime (low density) and pollucite are indicated by variations in intensity of certain peaks (see text). Symbols: (Q) = quartz, (K) = kaolinite, (I) = illite, (S) = smectite.

diffraction. The results showed analcime with subordinate micas and/or clay minerals (kaolinite, smectite, illite), quartz and minor apatite in the low-density fractions, whereas pollucite proper dominated in the high-density material (Fig. 11). The analcime component can be easily recognized in the X-ray powder diffraction patterns by a prominent increase of intensities of the 211 (5.61 Å), 220 (4.85 Å), 422 (2.80 Å), 431 (2.69 Å) and 640 (1.90 Å) diffractions, as indicated by Černý (1978).

Microscopic observations indicate that analcime is an alteration product of pollucite. Grains of pollucite entrained or enclosed within mica veins, and pollucite adjacent to the veins are strongly analcimized (Fig. 7c, d). In thin section, the index of refraction of the pollucite decreases noticeably adjacent to the veins. The analcimization post-dates vein emplacement; vein/pollucite interfaces and fractures seem to have acted as conduits for later Na-rich fluids. Analcimization is also strongly associated with replacement of the pollucite by clay minerals. An unusual feature of the Viitaniemi analcimization is the extremely thin rims of cesian analcime that separate inclusions and veinlets of silicates and quartz from the Cs-poor analcime proper and its transition into pollucite (Fig. 7c, d, e).

Locally the pollucite is fully replaced by a mixture of quartz and cookeite. Less altered parts are composed of analcime to cesian analcime and K-bearing clay minerals (Fig. 7e). Rare veins of apatite are also present in analcime (Fig. 7f).

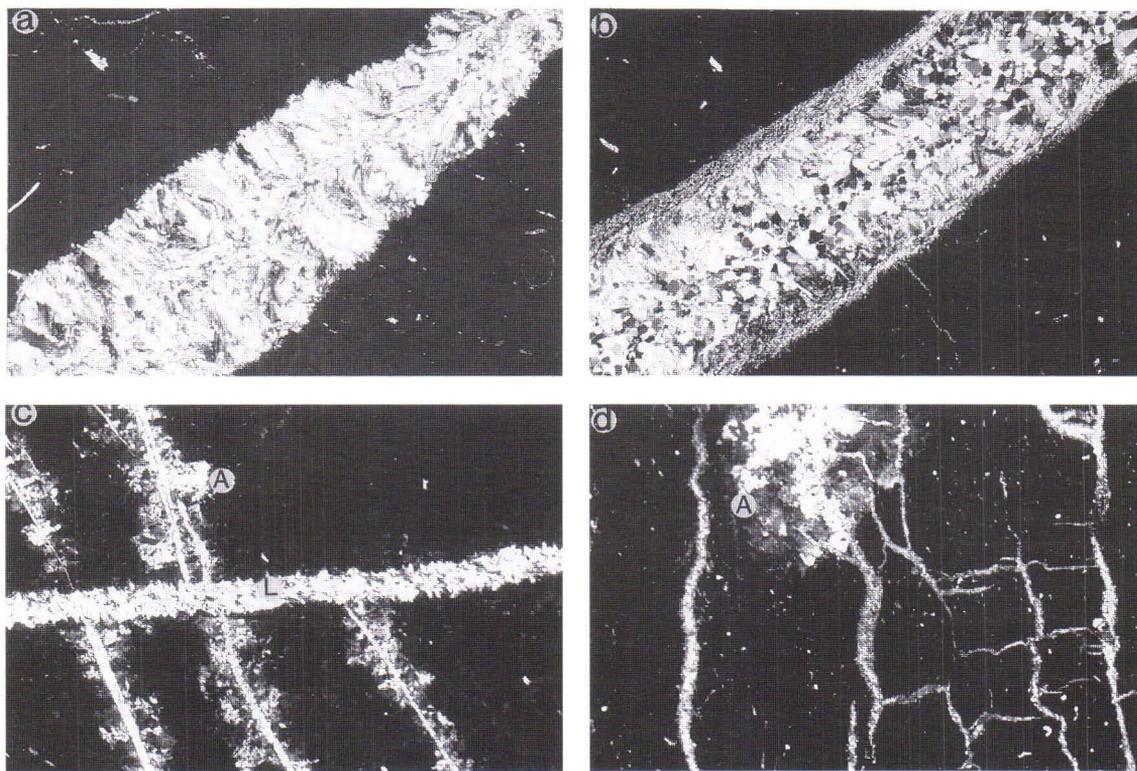


Fig. 12. Microphotographs of alteration products of the Viitaniemi pollucite (black; crossed polars). a) Coarse vein of lepidolite. b) Coarse vein of lepidolite + quartz with overgrowth of fine mica. c) Coarse vein of lepidolite (L) and later fine veins overgrown by adularia (A). d) Fine veins filling a cubic parting of pollucite, and a later granular aggregate of adularia (A). The horizontal edge of the figure a, b, c and d is 4.0, 3.4, 5.2 and 5.4 mm, respectively.

## HAAPALUOMA, PERÄSEINÄJOKI

### The parent pegmatite

The Haapaluoma pegmatite is situated 30 km SE from the town of Seinäjoki, near the contact of the Ostrobothnian schist belt within the central Finland granitoid complex (Fig. 1). Several other mineralogically interesting Li-, Be-, Nb-Ta- and Sn-mineral pegmatites, some of them economically important, have been located in the vicinity (Alviola 1989b). Both beryl (beryl-columbite and beryl-columbite-phosphate subtypes) and complex pegmatites (spodumene and lepidolite subtypes) have been observed (Haapala

1966, Mäkitie & Lahti 1991). The regional geology is shown on the map sheet of Pre-Quaternary rocks 2223 Alavus (Tyrväinen 1970), 2221 Jalasjärvi (Lahti & Mäkitie 1990) and Seinäjoki (Mäkitie *et al.* 1991). The most important rare-mineral pegmatites are marked on the aforementioned geological maps.

Haapala (1966) reported a  $^{207}\text{Pb} - ^{206}\text{Pb}$  age of 1800 Ma for Haapaluoma xenotime. The pegmatite dike sharply cuts its host rock, the syn-tectonic Svecokarelian tonalite. The U-Pb zircon

age of a similar tonalite in the same formation some 30 km W of the Haapaluoma pegmatite is  $1881 \pm 9$  Ma (sample from Luopajärvi in Jalasjärvi, see Mäkitie & Lahti 1991).

The pegmatite dike is vertical, several hundred metres long and 10 — 30 m wide, and is distinctly zoned. A thin aplitic border zone, locally absent, contains microcline perthite, albite, and quartz, with accessory schorl and garnet. The border zone grades rapidly to a 5 to 10 m wide wall zone of microcline perthite (in part with graphic quartz rods), quartz and schorl. Micas are rare, accessory minerals include garnet, apatite and beryl. The intermediate zone develops by gradual transition from the wall zone; it contains giant blocks of microcline perthite, subordinate quartz and accessory schorl plus beryl. Several lenses of quartz, 1 — 15 m long, form a discontinuous »core« along the boundaries of the wall and intermediate zones.

Albitic units with subordinate quartz and schorl form irregular bodies, in part metasomatic, along the wall zone/intermediate zone transition, and fracture fillings within the blocky microcline of the intermediate zone. The albitic units carry accessory beryl, columbite, cassiterite, lepidolite, spodumene, apatite, spessartine,

monazite, xenotime and brockite. Both irregular and fracture-filling albitic units grade into late Li-rich units containing mainly spodumene, quartz, lepidolite, green and red elbaïtes and cleavelandite. Some of the Li-rich units also contain rare accessory minerals such as löllingite, microlite, bertrandite, zircon and cookeite. Spodumene is typically lilac-coloured, and locally grades into purple, gem-quality kunzite, as reported by Lahti and Saikkonen (1986). Lahti *et al.* (1982) described eucryptite as a breakdown product of spodumene.

Haapala (1966) gives the concentrations of  $\text{Cs}_2\text{O}$  from 0.05 wt. % (wall zone) to 0.07 wt. % (Li-stage replacement unit) for the K-feldspar of the Haapaluoma pegmatite. He also noticed an increase in  $\text{Cs}_2\text{O}$  from 0.1 wt. % in green beryl of the intermediate zone to 1.64 wt. % in the pink variety from the Li-rich units. The present authors encountered a small white crystal of beryl, associated with Li-bearing minerals, which is exceptionally rich in Cs: the crystal is zoned and locally contains up to 6.62 wt. %  $\text{Cs}_2\text{O}$ . Albite, spodumene and red elbaïte contain less than 0.1 wt. %  $\text{Cs}_2\text{O}$ , but flakes of lepidolite closely associated with pollucite have 0.45 — 0.70 wt. %  $\text{Cs}_2\text{O}$ .

### The pollucite

Pollucite is found as granular aggregates 5 — 10 cm in maximum dimension. It is white to translucent in hand specimen and appears greenish upon pervasive alteration. The intensity of alteration seems to increase toward the margins of the pollucite aggregates. Pollucite nodules are surrounded by big crystal laths of grey to slight-

ly purple spodumene, prismatic crystals of rubellite, and quartz and cleavelandite. The same minerals also may occur as inclusions in pollucite. The pollucite is cut near its margins by 5 mm wide veins of white spodumene; however, branching veins of fine-flaked lepidolite about 1 mm wide are common throughout the pollucite.

### Primary composition and subsolidus re-equilibration of pollucite

The pollucite is mostly compositionally and texturally homogeneous and usually free from in-

clusions; thin sections of the pollucite seem to exhibit a poor cubic parting (Fig. 13a). A blebby

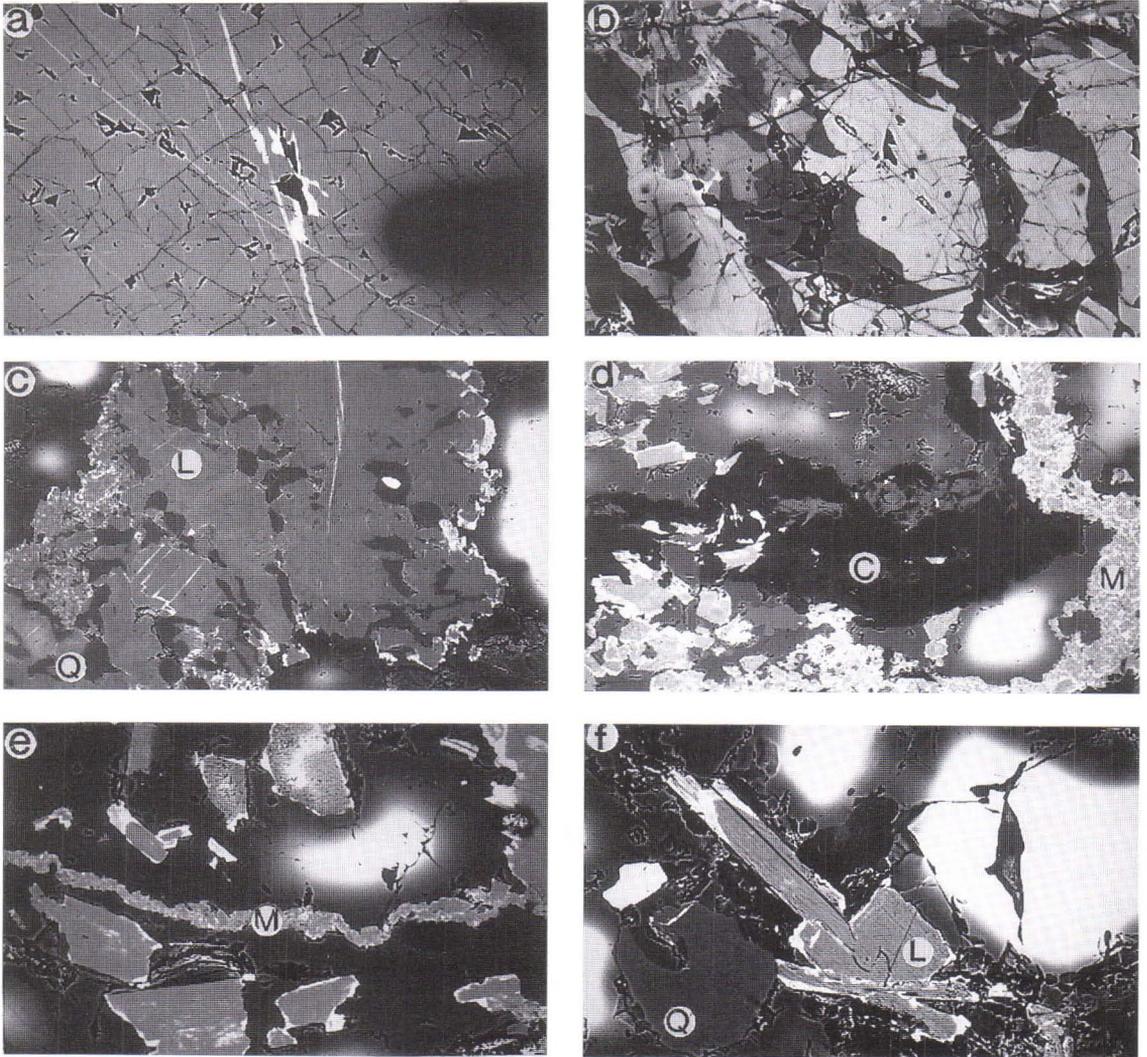


Fig. 13. BSE images of the Haapaluoma pollucite. a) Cubic parting of primary, homogeneous pollucite cut by thin Cs-rich veinlets (+ quartz) and penetrated by lobes of analcime. b) Locally well-developed blebby mosaic of pollucite associated with quartz (black) and rare Cs-rich veinlets. c) Vein of lepidolite (L) and quartz (Q) with enrichment in Cs along grain margins and cleavage traces. The quartz includes blebs of pollucite (white). Analcimization follows the vein contacts with pollucite. d) Vein of cookeite (C) and later fine vein of mica (M) in analcimized pollucite. e) Analcimization near fine veins of mica (M) are closely associated with f) scattered grains of lepidolite (L), the margins of which are enriched in Rb and Cs. Associated quartz (Q). The horizontal edge of each image is 280  $\mu\text{m}$  long.

texture appears locally: Na-enriched blebs are roughly elliptical in shape (20  $\mu\text{m}$  wide, 50  $\mu\text{m}$  long); Cs-enriched blebs are larger in size than the Na-enriched blebs, up to 100  $\mu\text{m}$  in diameter

(Fig. 13b). Immediately adjacent to some Cs-enriched blebs, the homogeneous pollucite shows minor enrichment in Na. Only rarely are the homogeneous and blebby pollucite intersected by

Table 4: Composition of pollucite from Haapaluoma.

Oxide	Average		8	21	28	36
SiO <sub>2</sub> wt. %	44.40	(.45)	43.65	48.00	48.05	58.80
Al <sub>2</sub> O <sub>3</sub>	16.19	(.10)	16.34	15.77	15.68	20.91
Fe <sub>2</sub> O <sub>3</sub>	0.00		0.01	0.01	0.01	0.00
P <sub>2</sub> O <sub>5</sub>	0.01		0.00	0.06	0.04	0.00
Na <sub>2</sub> O	1.33	(.15)	1.21	1.98	1.90	11.95
K <sub>2</sub> O	0.02		0.00	0.02	0.03	0.07
Rb <sub>2</sub> O	0.00		0.00	0.00	0.11	0.00
Cs <sub>2</sub> O	35.59	(.69)	35.90	32.18	32.02	0.20
CaO	0.00		0.01	0.02	0.02	0.01
MgO	0.00		0.00	0.00	0.00	—
H <sub>2</sub> O	—		—	—	—	—
sum	97.55	(.55)	97.12	98.04	97.87	91.94
Si*	2.108	(.007)	2.095	2.169	2.174	2.122
Al	0.906	(.009)	0.924	0.840	0.836	0.890
Fe	0.000		0.000	0.000	0.000	0.000
P	0.000		0.000	0.002	0.001	0.000
Na	0.123	(.014)	0.113	0.174	0.167	0.837
K	0.000		0.000	0.001	0.002	0.003
Rb	0.000		0.000	0.000	0.003	0.000
Cs	0.721	(.016)	0.735	0.620	0.618	0.003
Ca	0.000		0.000	0.001	0.001	0.000
Mg	0.000		0.000	0.000	0.000	—
Al + Fe <sup>3+</sup>	0.906	(.009)	0.924	0.840	0.836	0.890
Σ charges	0.845	(.011)	0.848	0.798	0.792	0.843
Si/Al	2.325	(.032)	2.268	2.577	2.596	2.384
CRK	85.5	(1.6)	86.7	78.1	78.8	0.7

\* Atomic contents on the basis of six oxygen atoms per anhydrous formula unit.

Average of homogeneous pollucite (N = 14).

8. Cs-enriched bleb.

21. Na-enriched bleb.

28. Inclusion in quartz.

36. Na-exchanged.

1 to 2 μm wide veinlets of Cs-rich pollucite (Fig. 13a).

Na-metasomatism generating almost pure analcime is widespread, replacing more than 50% of the pollucite (Fig. 13a, c — f). Compositions representative of the analcime, and of the above varieties of pollucite, are given in Table 4.

In Figure 14, variations in Si/Al ratios are plotted as a function of CRK. The homogeneous pollucite has Si/Al 2.26 to 2.41, and CRK 80.3 to 87.2 (average, Table 4). The Cs-enriched blebs

have lower Si/Al, and slightly higher CRK than the homogeneous pollucite. Similarly, the Na-enriched blebs tend to have higher Si/Al, but only slightly lower CRK than the homogeneous pollucite. Pollucite enclosed within quartz grains has Si/Al 2.5 to 2.6 and CRK 76 to 78. As a result of analcimization, Si/Al ratios remain constant from the primary homogeneous pollucite to nearly pure analcime, with CRK reaching values as low as 0.7.

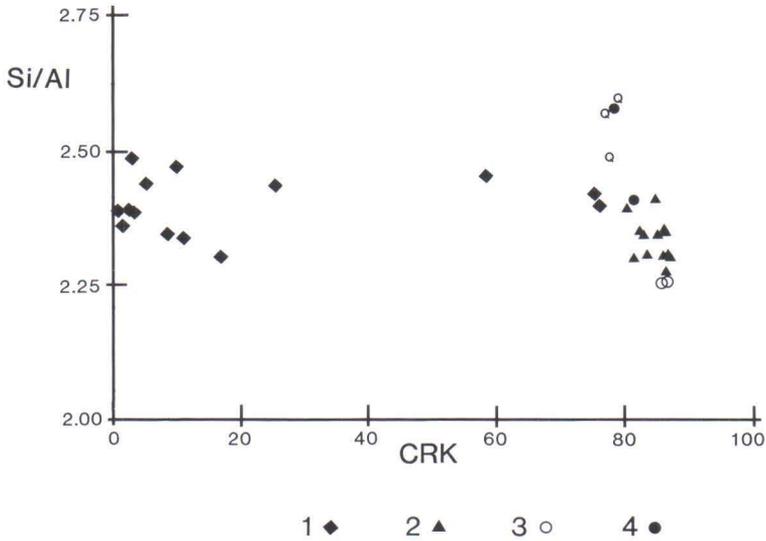


Fig. 14. Si/Al vs CRK for the Haapaluoma pollucite. Symbols: (1) = Na-exchanged, (2) = primary homogeneous, (3) = Cs-enriched bleb, (4) = Na-enriched bleb, (Q) = pollucite inclusions in quartz.

### Alteration of pollucite

In Figure 15 a schematized alteration sequence is given for the Haapaluoma pollucite, as a guide to the following discussion. Margins of the pollucite are crosscut by veins 5 mm wide containing a spodumene-quartz intergrowth. Laths of spodumene adjacent to the pollucite also have a symplectitic intergrowth with quartz (Fig. 16a, b). The pollucite contains subparallel veins of lepidolite + quartz  $\pm$  apatite 0.5 to 1.0 mm in

width (Fig. 16b), which pinch out towards the margins of the pollucite aggregates. Margins and cleavage traces of the lepidolite are locally enriched in Cs, with atomic ratios of K/Cs < 1.0 (Fig. 13c).

Sinuuous veins of cookeite up to 1 mm wide are followed in time by veins of very fine-grained mica (Fig. 16b, c). The fine mica is enriched in Cs (Fig. 13d, e). The enrichment in Cs of all generations of sheet silicates occurred during analcimization, as described below. Homogeneous adularia replaces pollucite as spherical aggregates fully embedded in the pollucite, up to 1 mm in diameter, and as overgrowths on the veins (Fig. 16c).

X-ray powder diffraction of density fractions from 2.20 to 3.05 g/cm<sup>3</sup>, separated from crushed samples of pollucite, gave the same results as for the Viitaniemi pollucite: low-density fractions showed the presence of substantial analcime with micas and/or clay minerals, where-

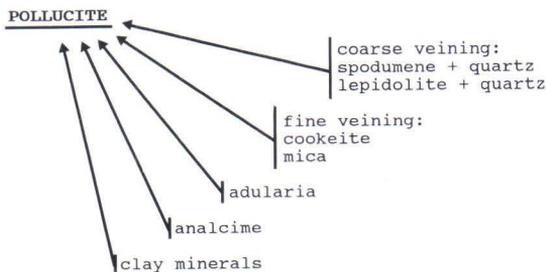


Fig. 15. Schematic sequence of alteration products of the Haapaluoma pollucite. See text for details.

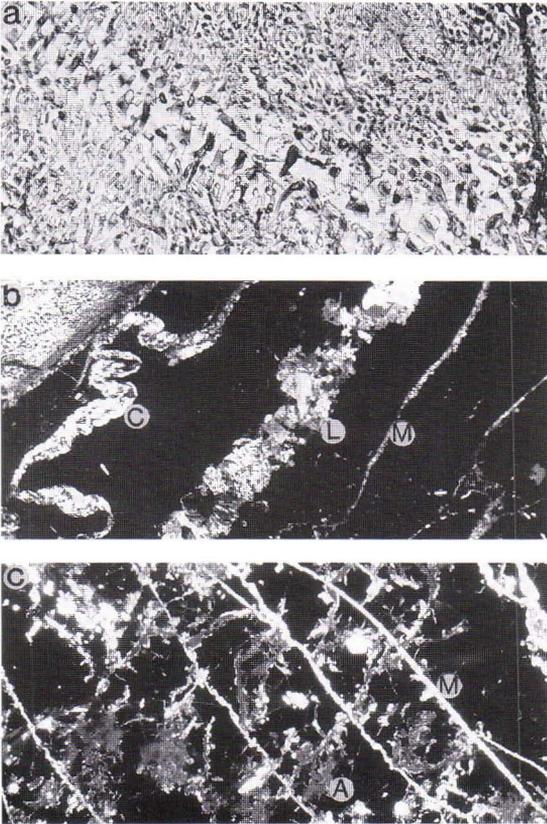


Fig. 16. Microphotographs of alteration products of the Haapaluoma pollucite. a) Laths of spodumene (adjacent to pollucite aggregates) showing a typical symplectitic intergrowth with quartz. Plane-polarized light. b) A sinuous vein of cookeite (C) along the surface of the spodumene lath of a; coarse veins of lepidolite (L) and fine veins of mica (M) are located further away from this contact. Crossed polars. c) Fine veins of mica (M) overgrown by adularia (A). Crossed polars. The horizontal edge of the figure a, b and c is 1.3, 4.0 and 1.3 mm, respectively.

as the high-density material corresponded to pollucite proper.

The clay minerals associated with analcime are enriched in Cs, and also contain appreciable Rb, Mg and Fe. Smectite and illite phases, identified

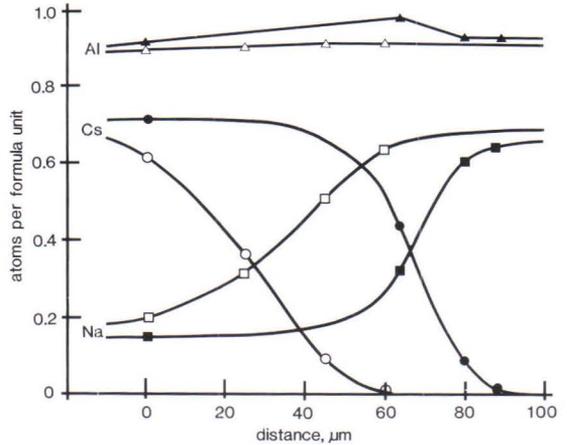


Fig. 17. Two traverses (one marked by open symbols, the other by solid symbols) from the primary Haapaluoma pollucite to extreme analcime. Symbols: circles = Cs (+ Rb + K), squares = Na (+ Ca + Mg) and triangles = Al (+ Fe), all as atoms p.f.u..

by X-ray powder diffraction, are particularly abundant along the margins of pollucite nodules in the form of branching green veins. The smectite, suspected to be the main carrier of Cs, shows a peak at 12.5 Å corresponding to Na-dominant montmorillonite, or a group of broad peaks at < 12.5 Å that indicates the presence of mixed-layer clay minerals. Fine-flaked, Cs-rich mica minerals also are present; they are most abundant in the density fraction from 2.60 to 2.65 g/cm<sup>3</sup>.

In thin sections, fully isotropic analcime can be recognized by its low refractive index. It replaces pollucite, postdating the veining and adularia. Analcimization is extensive. The pollucite is replaced by curvilinear to lobate embayments of analcime, with diffuse gradational boundaries about 100 μm wide (Fig. 17, compositional traverse). The pollucite is locally preserved only as relic islands 50 to 300 μm in size.

## ORISELKÄ, TOHMAJÄRVI

### The parent pegmatite

The Oriselkä pegmatite belongs to the Kitee pegmatite group which consists of about 800 pegmatites or pegmatitic granites covering an area of some 300 km<sup>2</sup> (Fig. 1). Virtually all pegmatites of this group are tourmaline- and muscovite-bearing, with potash feldspar dominant over albite. About 100 of them are beryl-bearing, 22 contain Li-phosphates, 6 have spodumene, lepidolite, cookeite or elbaite, and only one of them carries pollucite. They range from beryl-columbite through beryl-columbite-phosphate to spodumene subtype of rare-element pegmatites (Černý 1989, 1991a, 1991b). Pegmatites are hosted by Karelidic medium-grade andalusite- and staurolite-bearing pelitic metasediments, volcanics, diabases and plutonic rocks (Nykänen 1968, 1971). The NE-SW running contact between the Svecokarelidic rocks and the Archean basement is about 20 km from the Kitee pegmatites.

Numerous pegmatitic granite and pegmatite bodies surround a 50 km long ovoidal granite cupola called Puruvesi granite, conformable to the regional metamorphic fabric on a large scale but cutting it in detail. The granite itself contains occasionally pegmatitic patches and veins mainly in the southwestern part of the body, which alternate locally with white, fine- to medium-grained aplitic granite. The bulk of the pegmatite granites and pegmatite dykes is situated E and NE of the granite, and ranges from barren and primitive beryl-bearing bodies in the east to extremely fractionated ones in the NE corner of the pegmatite field.

The Puruvesi granite intrusion is late-kinematic. The radiometric U-Pb zircon age of the granite is  $1797 \pm 19$  Ma (Nykänen 1983), whereas that of the pegmatitic granite parental to the pegmatite group is 1780 Ma (Rb-Sr age on muscovite, see Nykänen 1968; recalculated 1977 using new decay constants). A complex-type pegmatite of the Kitee group, with mineralization similar to

that of the Oriselkä pegmatite, was dated at 1770 Ma (U-Pb age on zircon and uraninite, Nykänen 1975; recalculated 1977 using new decay constants).

The Oriselkä pegmatite, examined during a regional exploration program of the Geological Survey of Finland in the nineteen seventies, has not yet been introduced into the literature. It is therefore reported and illustrated here in some detail.

The Oriselkä pegmatite dike is about 60 m long and 5 — 10 m wide in outcrop, with a couple of swellings which widen it up to 30 m (Fig. 18). The host rock is a graphite- and sulphide-bearing amphibolite which locally grades into amphibolite-bearing black schist or sulphide-bearing mica schist; the pegmatite crosscuts the general trend of foliation.

Given the poor exposure of the body, the northeastern part of the dike seems to be a rather simple, muscovite- and schorl-bearing microcline pegmatite. At the tip of the eastern swelling of the dike, the pegmatite consists of blue apatite, black tourmaline, quartz and K-feldspar. The black schist near the tip was altered into coarse-grained muscovite-tourmaline schist.

The southwestern part of the dike is strongly albitized and complex, with minerals of Be, Nb-Ta, Sn, Li and Cs. No zoning was visible at the time of detailed mapping, because of poor exposure and considerable damage to the best outcrop by blasting. The Li-mineralization is best developed on the northern flank, well developed at the southwestern end and poorly developed at the southern side of the dike.

At the exocontact of the pegmatite, the schist is rich in brownish-black dravite. Near the contact, the pegmatite contains grey albite, quartz, K-feldspar, and muscovite. The accessory minerals are deep-blue tourmaline, pyrite, marcasite, arsenopyrite, löllingite, cassiterite and vivianite,

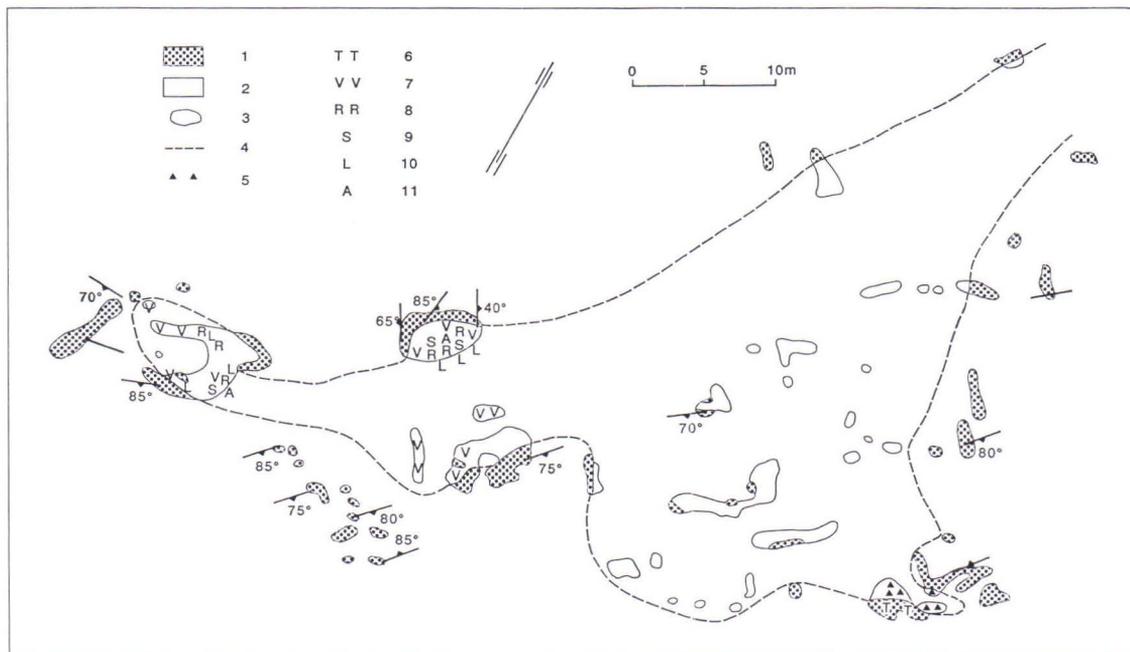


Fig. 18. Detailed map of the Oriselkä pegmatite. Symbols: (1) = country rock, (2) = pegmatite, (3) = outcrop, (4) = contacts, (5) = apatite-tourmaline-quartz-feldspar pegmatite, (6) = tourmaline-muscovite schist, (7) = green elbaite, (8) = red, yellow and colourless elbaite, (9) = spodumene, (10) lepidolite, (11) = amblygonite-montebrasite.

the last as thin fracture coatings. The first Li-rich mineral in the crystallization sequence of the pegmatite is dark green elbaite, occurring farther away from the contacts. The internal parts of the pegmatite consist, in addition to bluish cleavelandite and quartz, mainly of deep purple lepidolite, spodumene and multicoloured elbaite, with minor amounts of pinkish montebrasite, gray and pink beryl, Mn-columbite and cassiterite.

Reddish lithiophilite and its alteration products, sicklerite and purpurite, are the most common among accessory phosphates. Aggregates of blackish rockbridgeite and small masses of brown hureaulite have been observed locally. Secondary minerals include illite, pinkish chlorite, and rare, white bityite. Bityite occurs as small aggregates (1 cm in cross section) in gray

albite and quartz with accessory green elbaite and blue apatite.

At the southwestern end of the dike the zoning in elbaite and muscovite-lepidolite aggregates is reversed relative to the other parts of the dike: the elbaite has a dark green crust and pink core, and the lepidolite is covered by yellow muscovite. Beryl is altered into spongy bertrandite. Lepidolite is present in two generations: a book lepidolite and a fine-grained massive variety.

The Cs content of the muscovite and K-feldspar is low,  $\leq 0.05$  and  $\leq 0.17$  wt.%  $\text{Cs}_2\text{O}$ , respectively. In the Li-micas and two generations of lepidolite, the Cs contents range from 0.44 to 1.87 wt.%  $\text{Cs}_2\text{O}$ . Gray beryl from the Li-bearing inner parts of the pegmatite contains 0.6 to 3.77 wt.%  $\text{Cs}_2\text{O}$ .

### The pollucite

Pollucite was spotted in 1978 by R.A. in a pile of boulders around the northern flank of the pegmatite. Pollucite is gray to white in color with a subvitreous lustre; it fills interstices between spodumene laths in a cleavelandite + quartz +

lepidolite pegmatite. Pollucite is a rather rare mineral in the Oriselkä pegmatite and it forms only small aggregates. The pollucite is cut by fine veins which are difficult to detect by the naked eye.

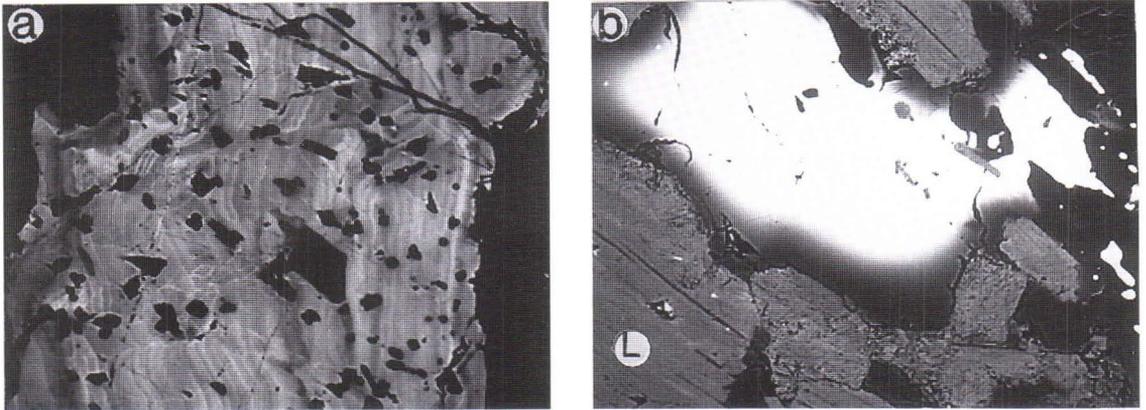


Fig. 19. BSE images of the Oriselkä pollucite. a) Layered Cs-rich pollucite contained within fractures of lath spodumene (black). b) Advanced stages of analcimization near lepidolite (L) and spodumene (black). The horizontal edge of each image is 260  $\mu\text{m}$  long.

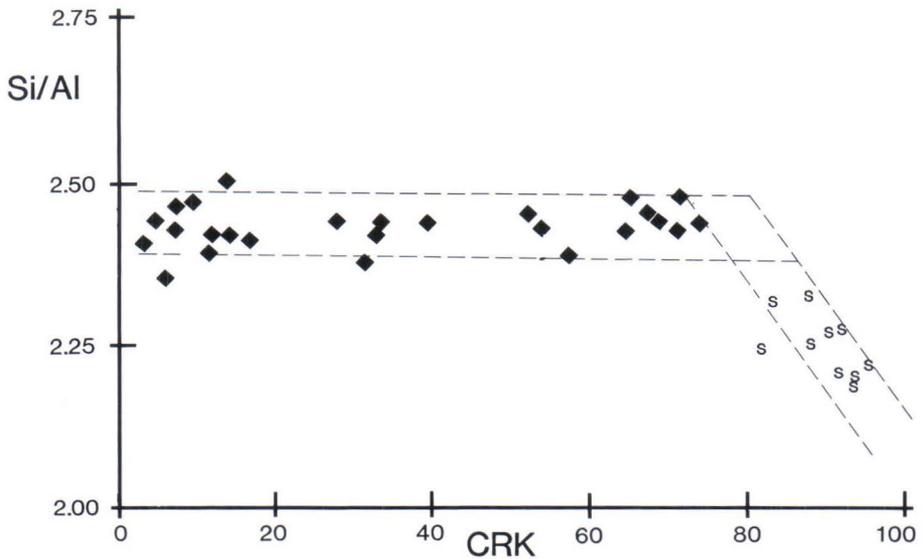


Fig. 20. Si/Al vs CRK for the Oriselkä pollucite. Symbols: diamonds = Na-exchanged pollucite and (s) = layered pollucite inclusions in spodumene. The probable composition of the primary pollucite is indicated by the large diamond-shaped area.

Within one hand specimen a crystallization sequence may be recognized from spodumene through quartz, pollucite, and lepidolite to albite: each phase has mostly segregated from the other to form individual masses. Laths of spodumene (1 — 2 cm long) are fresh near the pollucite and altered to clay minerals further away. The spodumene is strongly red luminescent in

ultraviolet light — an unusual feature more typical of eucryptite. In thin section, some crystals are twinned, some are symplectitic with quartz near their margins, and some seem to contain thin lammellae of an exsolved(?) phase. The lepidolite is dark mauve, and associated with the fine-grained albite and rubellite.

### Primary composition and subsolidus re-equilibration of pollucite

Analcimization has affected most of the pollucite. No undisturbed primary compositions were found in the samples examined. The pollu-

cite contains rare, thin (less than 2  $\mu\text{m}$ ) Cs-rich veinlets. Pollucite interstitial to spodumene grains is in part fracture-filling, and consists of bands

Table 5. Composition of pollucite from Oriselkä.

Oxide	2	18	5	34	39
SiO <sub>2</sub> wt. %	44.77	42.07	47.30	47.96	57.92
Al <sub>2</sub> O <sub>3</sub>	16.41	16.11	16.45	16.73	20.48
Fe <sub>2</sub> O <sub>3</sub>	0.00	0.00	0.00	0.04	0.00
P <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.04	0.05	0.09
Na <sub>2</sub> O	1.56	0.45	2.38	4.08	11.46
K <sub>2</sub> O	0.04	0.02	0.00	0.09	0.46
Rb <sub>2</sub> O	0.04	0.01	0.00	0.00	0.05
Cs <sub>2</sub> O	35.51	40.43	31.60	23.14	0.37
CaO	0.02	0.02	0.05	0.66	0.06
MgO	0.00	0.00	0.01	—	—
H <sub>2</sub> O	—	—	—	—	—
sum	98.34	99.11	97.84	92.74	90.89
Si*	2.104	2.074	2.135	2.127	2.122
Al	0.908	0.936	0.875	0.875	0.884
Fe	0.000	0.000	0.000	0.001	0.000
P	0.000	0.000	0.002	0.002	0.003
Na	0.142	0.043	0.208	0.350	0.814
K	0.003	0.001	0.000	0.005	0.022
Rb	0.001	0.000	0.000	0.000	0.001
Cs	0.712	0.850	0.609	0.437	0.006
Ca	0.001	0.001	0.003	0.032	0.002
Mg	0.000	0.000	0.001	0.000	0.000
Al + Fe <sup>3+</sup>	0.908	0.936	0.875	0.876	0.884
$\Sigma$ charges	0.860	0.893	0.825	0.855	0.846
Si/Al	2.316	2.216	2.440	2.432	2.400
CRK	83.3	95.1	74.1	53.7	3.4

\* Atomic contents on the basis of six oxygen atoms per anhydrous formula unit.

2, 18. Cs-rich bands in spodumene.  
5, 34, 39. Na-exchanged.

of Cs-rich pollucite which are parallel to, and follow around uneven fractures of the spodumene (Fig. 19a). This Cs-rich pollucite does not seem to have been affected by the pervasive analcimization of the main pollucite (Fig. 19b).

A plot of Si/Al vs CRK is shown in Figure 20. No compositions near CRK 80 were found. The Cs-rich pollucite in spodumene has Si/Al as low as 2.22 and CRK as high as 95.1. The Na-enriched pollucite has Si/Al from 2.35 to 2.50 and

CRK from 74.1 to as low as 3.4 in the most analcimized material. Representative compositions are given in Table 5. Based on the compositional relationships of primary, Cs-enriched and analcimized pollucite at numerous other localities, the composition of the primary phase can be estimated by interpolation of the Na- and Cs-enrichment trends at about CRK 76–83 and Si/Al 2.38 — 2.49 (Fig. 20).

### Alteration of the pollucite

As observed in thin sections, the pollucite is crosscut by thin veins (up to 1 mm wide) of fine-grained mica, in part replaced by fine-grained spodumene. The veins follow fractures or grain boundaries of pollucite with other minerals. Most

of the pollucite is Na-exchanged to analcime compositions, particularly in the vicinity of fractures and veins (Fig. 19b). The pollucite is replaced by minor amounts of clay minerals associated with analcime.

## DISCUSSION

### Evolution of pollucite

Primary, homogeneous pollucite from the four Finnish localities has Si/Al ratios within the range 2.26 to 2.52 and  $CRK = 100 (Cs + Rb + K) / \Sigma$  cations from 78.9 to 87.2. These are relatively restricted ranges, considering the broad regional dispersion and different bulk compositions of the parent pegmatites. The most abundant minor elements are Rb (up to 0.34 wt.%  $Rb_2O$ ) and P (up to 0.68 wt.%  $P_2O_5$ ); Ca, Mg, K and Fe are typically near the limits of detection by electron microprobe.

The homogeneous pollucite has locally evolved to a blebby mosaic of alternating Na-enriched and Cs-enriched domains. They are locally strikingly similar to the familiar perthitic textures of alkali feldspars (Viitaniemi pollucite in Figure 7a). This mosaic aggregate is commonly crosscut by Cs-rich veinlets ( $\pm$  quartz) that are local-

ly rimmed with Na-rich pollucite. Both these textural styles of re-equilibration seem to proceed with a local mass-balance preservation of Na, Cs, Al and Si, and they may be interpreted as features of subsolvus exsolution.

Details of the re-equilibration mechanisms — breakdown into Na, Si-enriched and Cs, Al-enriched compositions, and liberation of  $SiO_2$  leading to alkali cation- and Al-rich pollucite with Si/Al approaching 2.00 — have been previously documented. This suggests the existence of a miscibility gap between analcime and pollucite, widening with decreasing temperature (Teertstra *et al.* 1992a). In Finnish pollucite, Si/Al ratios drop as low as 2.18 and CRK reaches as high as 95.1; in contrast, the Na, Si-enriched component attains Si/Al of 2.46 and CRK of 76.

The textural features of subsolidus re-

equilibration are more pronounced in the pegmatites with larger pollucite bodies (Luolamäki and Viitanieniemi) than in the smaller pollucite segrega-

tions (Haapaluoma and Oriselkä). Also, sequences of alteration are more complex in the larger pegmatites.

### Evolution of alteration processes

Three consecutive stages of alteration of pollucite were recognized by Černý and Simpson (1978) at Tanco, Manitoba: (i) coarse veining, usually polygonal, fracture-filling, and more abundant near the margins of pollucite bodies, (ii) pervasive fine veining, braided to subparallel, and (iii) replacement assemblages. These stages were also recognized in the High Grade Dyke, Manitoba (Teertstra *et al.* 1992a) and are confirmed here for the Finnish localities. However, the style and products of the alteration are different in some respects: it is particularly the extensive replacement of pollucite by analcime which sets the Finnish localities apart from any others that have so far been described.

(i) *Coarse veining* is present only in large pollucite bodies, greater than about 1 m in size. The veining is fracture-filling rather than metasomatic; only locally do the veins seem to replace pollucite. The mineralogy of the coarse veining is variable, resembling that of late-crystallizing assemblages in the surrounding pegmatite (Figs. 4, 11 and 15), but genetically distinctive.

The only mineral consistently present as coarse veining is lepidolite (+ quartz  $\pm$  apatite). Lepidolite veining is never physically connected to lepidolite units, and it pinches out within the pollucite. In contrast, albite veins cutting across margins of pollucite bodies may usually be traced in outcrop to nearby albite units. Rather than having a source external to the pollucite bodies, lepidolite may have formed by crystallization of residual fluids excluded by crystallization of pollucite. The same may apply to the veins of microcline, which is largely non-perthitic and also seems to be restricted to the pollucite bodies alone.

(ii) *Fine veining*, subparallel to braided, is

characteristic of most localities where pollucite has segregated into masses greater than about 5 cm in size. The veins contain mica (muscovite?)  $\pm$  quartz, and typically decrease in grain size from vein core to margin. The mica is usually followed by crystallization of fine-grained spodumene. The spodumene may be symplectitic with quartz, and locally forms its own set of veins. Fine veins which contain cookeite are a rare and unusual part of this sequence of alteration. They are reported here for the first time, in pollucite from Haapaluoma.

The fine veining tends to follow fractures in pollucite, a cubic parting, or grain boundaries. Rarely the veins are sinuous and meander through the pollucite. The veins do not seem to replace pollucite, and always pinch out within the pollucite. Unlike coarse veining, the abundance of fine veins does not seem to increase towards margins of the pollucite. The veins vary in time from continuous with uniform width (early veins up to 5 mm, later veins up to 1 mm) to dispersed and irregular.

Černý and Simpson (1978) noted that at Tanco the veins were so pervasive as to be »an obstacle for obtaining reliable compositional data» in bulk chemical analysis of pollucite. They interpreted this veining as a product of replacement. In contrast, Shaub and Schenck (1954) observed in thin section veins of spodumene and mica that were strictly fracture-filling, without any replacement of pollucite. Cooper (1964) suggested that at Bikita the (mica) veining is a result of »crystallization of fluids excluded from pollucite crystallization».

The characteristic mica and spodumene veining of pollucite results from action of late fluids which are typical of late pegmatite evolution. It

seems probable that hydrolytic effects in the presence of K and Li caused the »sericitization» of pollucite and the growth of spodumene, leading to dispersal of liberated Cs. Mobilization and transport of the reactants generated the fracture-filling veins.

The style of the veining (subparallel and perpendicular) seems to be controlled by a cube parting of pollucite. The parting may be induced by a thermal contraction of pollucite (which has its maximum rate at temperatures below about 200°C; Taylor & Henderson 1968, Richerson & Hummel 1972), and enhanced by stress.

The sequence of alteration can be ambiguous and difficult to determine as a result of the parting-controlled texture of alteration. The veins are commonly parallel (Fig. 16b), or overgrow each other sequentially (Fig. 12b, c). In outcrop, hand specimen and thin section, coarse veins seem to cut across fine veins (Fig. 6b, 12c). However, fine veins locally continue across coarser veins, and may penetrate them slightly at the junction. The real sequence is consistent with two evolutionary features: an overall decrease in grain size with time, and an overall decrease in the size of fractures but an increase in their number, making the veining more pervasive. Opening of fractures continuous with the growth of mica is probably responsible for the flake orientation normal to the vein length.

(iii) *Adularia* metasomatically replaces pollucite, crystallizing along surfaces of the earlier veins and as spherical granular aggregates fully embedded in the pollucite. The aggregates are usually inclusion-rich, containing albite, K-bearing micas, cookeite and Cs-rich pollucite. Pollucite adjacent to adularia grains can be very Cs-rich, suggesting in some localities a potential conservation of Cs during metasomatism.

The habit of the adularia crystals and the form of their aggregates indicate high nucleation and crystallization rates. These conditions, combined with low crystallization temperatures (about 300 to 250°C) favour metastable precipitation of highly disordered crystals; at many localities, the

adularia has the structure of high sanidine (Černý & Chapman 1986, unpublished data of the authors).

Grains of the adularia may be heterogeneous, locally with atomic contents of Rb > K. Analytical results indicate a constant Si/Al ratio near 3.0. This, combined with thin section observations, indicates the presence of an adularian rubidium feldspar. A rubidium feldspar was also found to coexist with microcline. Both these potential new mineral species are currently being studied. The exceptionally high content of Rb in these phases suggests that the fluids which generated the adularia probably are residual, and are not derived from typically Rb-poor country rocks.

(iv) *Isotropic analcime* replaces pollucite by cation exchange (Fig. 17). The Si/Al ratio is maintained from the primary pollucite, but it tends to decrease slightly in advanced stages. Analcimite penetrates by at most 1 mm from any vein or fracture into the pollucite. Rb contents (and rarely P) are decreased relative to the pollucite; however, K and Ca contents (and rarely Fe) are increased (Tables 1 — 5).

Analcimite is most advanced near margins of the pollucite aggregates; however, at some localities primary composition of the pollucite may be fully obscured by pervasive Na-exchange. The cation exchange penetrating mainly along veins and fractures is associated with formation of clay minerals.

The cation exchange is accompanied by the formation of thin hollow capillaries that extend into analcime from vein margins and adularia surfaces. The capillaries are locally arranged into a three-dimensional rectangular grid, suggesting dependence on preferred crystallographic directions of the cubic structure. This orientation suggests dissolution of the pollucite-analcime matrix. However, the subradial aggregation of the capillaries on the surfaces of veins and adularia could be better explained by dissolution of acicular crystals of a preexisting mineral.

Barrer (1950) treated pollucite with various salt

solutions and inferred from index of refraction measurements that there was little or no  $\text{Cs}^+$  exchange. Komarneni *et al.* (1978) showed a very low cation exchange capacity at 25°C (similar to that of feldspars) that was probably due to surface phenomena and not to classic ion exchange processes. More recently, however, Komarneni and White (1981) demonstrated »substantial cation exchange« at 300°C using brine solutions with high activity of  $\text{Na}^+$ ,  $\text{K}^+$  and  $\text{Ca}^{2+}$ .

In natural pollucite, however, only exchange of  $\text{Na}^+$  and  $\text{Cs}^+$  is observed (Tables 1 — 5).  $\text{K}_2\text{O}$  contents in both analcime and pollucite are low: rather than K-exchange, adularia or K-bearing micas form. There is no indication of Li-exchange; if  $\text{Li}^+$  is present it seems to be contained by cookeite and AIP-bearing or AlSi-bearing phases. Only low  $\text{Ca}^{2+}$  contents seem to have been present in the metasomatic fluids, and these are concentrated into apatite.

Loss of  $\text{Cs}^+$  by cation exchange mechanisms is important at lower temperatures, probably around 150 to 250°C (*i.e.* less than estimated temperatures of adularia crystallization). The depth of cation exchange in natural pollucite appears limited to about 1 mm before dissolution of the aluminosilicate framework starts. At this stage minerals replacing pollucite may crystallize, including late analcime. Below about 100°C Na-metasomatism becomes less important (*i.e.* the rate of cation exchange becomes slow), and framework dissolution (with Al and Si presumably taken into clay minerals) is the dominant mechanism for loss of  $\text{Cs}^+$ . Such a process is close to the conditions under which weathering occurs. Further studies are required to identify the weathering mechanisms important for the loss of  $\text{Cs}^+$  to solution and formation of clay minerals.

New experimental evidence (Mimura *et al.* 1990) agrees in part with our observations on natural pollucite. On exposure of pollucite (synthetic  $\text{CsAlSi}_2\text{O}_6$ ) to concentrated  $\text{MgCl}_2$  solutions at 300°C, layers of clinocllore form. With  $\text{CaCl}_2$ , spherical aggregates of anorthite form; KCl

generates clusters of leucite; and NaCl produces cube-shaped crystals of analcime. Only with Na and K did a cation-exchanged layer form beneath the surface-covering crystals. The result of the NaCl experiment corresponds to the observed analcimization, but with a significant reversal in abundances and types of reaction products.

The cation exchange is widespread at the Finnish and some other localities, but growth of non-metasomatic, fracture-filling analcime was observed only at Luolamäki, and at Puklice, Czechoslovakia (Teertstra *et al.* 1992b).

(v) *Replacement by clay minerals* is most extensive near the margins of pollucite bodies. This replacement leaves the earlier veining and inclusions intact, and local clay pseudomorphs after pollucite are formed. The clay minerals are associated with quartz and rare apatite, and generally form after analcimization. Illite, chlorite, expanding-type smectites (possibly Na montmorillonite) or mixed-layer sequences are the possible hosts for large interlayer cations. Cs-rich clay minerals were discovered; however, more detailed studies are required for their unambiguous identification.

At Luolamäki and Oriselkä, no Cs-rich layer silicates were found: mica and clay minerals were in close contact with analcime. Cs-rich layer silicates were not found at Viitaniemi either; however, cesian analcime forms thin seams along silicate veins and inclusions, possibly due to adsorption of Cs on their surfaces. At Haapaluoma, margins of sheet silicates are enriched in Cs (locally with atomic contents of  $\text{Cs} > \text{K}$ ) possibly by cation exchange. These represent potential new mica and/or clay mineral species. A progression from complete dispersal of Cs through adsorption(?) to apparent Cs-exchanged sheet silicates could be postulated, indicative of differences in conditions of analcimization and clay mineral formation among these four localities.

The small-scale evolution of Cs-bearing clay minerals after the Haapaluoma pollucite resembles the process that led to extensive alteration of pollucite into illite, kaolinite and smectite in

several pegmatites of Manitoba, and ultimately to its total destruction (Černý 1978). Compositional data from Manitoba, however, indicate that only limited residual Cs is preserved in the bulk of the clay minerals; most of the Cs is dispersed in this type of alteration.

The source of fluids involved in analcimization and clay formation is ambiguous. Assemblages involving only Na, K and/or acid leaching could be generated by strictly residual pegmatitic fluids.

In contrast, London (1990) emphasizes open bi-metasomatic exchanges between pegmatite- and wallrock-derived fluids. In either case, low-temperature, hydrothermal fluids are undoubtedly responsible. A possible influence of supergene weathering processes cannot be ruled out (W.L. Roberts and K.A. Vlasov, quoted in Černý 1978); however, claims that some pollucite weathers rather easily require documentation and experimental investigation of the mechanism(s).

## CONCLUSIONS

The composition of primary homogeneous pollucite in the Finnish pegmatites, and the overall textural style plus compositional divergence of its exsolution, are comparable to the behavior of pollucite at other localities. The same applies to the veining and alteration processes but with significant quantitative differences. Coarse veining by quartz, feldspars and/or lepidolite is developed at the two Finnish localities containing large pollucite bodies — Luolamäki and Viitaniemi — but only to a very modest extent when compared to the giant pollucite bodies of Tanco (Černý & Simpson 1978), Bikita (Cooper 1964), or even Varuträsk (Quensel 1938, 1945).

In contrast, low-temperature sodium metasomatism of pollucite generating Cs-poor to nearly Cs-free analcime is widespread at all four localities examined here. Most other occurrences of pollucite usually show restricted analcimization, if any at all (e.g. High Grade Dyke, Teerstra *et al.* 1992a). The regional geochemical significance of this process is, however, uncertain. The extensive action of late fluids with a high activity of  $\text{Na}^+$  at all four localities seems to be a coincidence rather than the result of a broad regional hydrothermal event that would equally disturb all the widely dispersed pollucite occurrences.

The four localities of pollucite in the Finnish Precambrian all belong to highly fractionated

complex pegmatites of the rare-element class, as is typical for occurrences of pollucite worldwide. However, the geochemistry of at least two of the pegmatites is peculiar. Primary blocky potassium feldspar at Luolamäki, and particularly Viitaniemi, shows a relatively high K/Rb and low Cs, compared to K-feldspar in most other pollucite-bearing pegmatites (Smeds & Černý 1989). This distinguishing feature is currently under a more detailed study. Nevertheless, it seems already obvious that the geochemical criteria for identification of pollucite-bearing pegmatites defined by, e.g., Gordiyenko (1976) and Trueman and Černý (1982) should be applied with caution.

From the viewpoint of petrology and geochemistry, the mosaic and veining breakdown of pollucite into Na, Si- and Cs, Al-enriched phases ( $\pm$  quartz) suggests exsolution along a pollucite-analcime solvus. Such a process is controlled by decreasing temperature, possibly catalysed by the presence of a fluid phase, and it probably proceeds largely with mass-balance and local preservation of Na, Cs, Al and Si. Coarse veining by quartz, feldspar and micas is evidently generated by late internal pegmatitic fluids, as is the lower-temperature fine veining by »sericite» micas and spodumene ( $\pm$  cookeite), plus the relatively closely associated adularia.

As to the analcimization and argillization, unambiguous interpretation of the fluid source does

not seem to be feasible at present. The Na-exchange may be possible via pristine low-temperature pegmatitic fluids, but the high Ca and Mg contents of the clay minerals (Černý 1978) indicate significant involvement of pore fluids from regional lithologies. Effects of weathering under surface conditions should be considered and experimentally investigated.

Features of the re-equilibration of pollucite and its sequence of alteration are significant records of the late history and geochemical evolution of granitic pegmatites. As well, the importance of these observations to the long-term stability and containment of nuclear wastes should not be underestimated.

Implications for the stability vs breakdown of synthetic pollucite in geological storage of nuclear wastes are obvious. Primary homogeneous pollucite is affected by at least five different processes, all of which involve mobilization and transport of Cs. The migration and dispersal of Cs increase with decreasing temperature: early exso-

lution seems to preserve cation mass balance and early coarse veining is largely fracture-filling, but fine veining and adularia are in part metasomatic, and the analcimization plus argillization mobilize and disperse the Cs at a large scale.

As a final comment, we would like to point out the similarity of minerals in the alteration sequence of pollucite (excluding the coarse veining) to those found in association with cesian analcime. At Tanco, Manitoba cesian analcime crystallizes in low-temperature hydrothermal »alpine-vein« assemblages of quartz, spodumene, adularia, albite, cookeite, apatite, calcite and clay minerals in leaching cavities remote from the primary bodies of pollucite (Černý 1972). Except for apatite and calcite, all of these minerals are abundant members of the alteration sequence of Finnish pollucite. This mode of occurrence of cesian analcime emphasizes the liberation and relatively long-range migration of Cs from pollucite affected by alteration processes.

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