**First natural occurrences of end-member pollucite: A product of low-temperature reequilibration**

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**Abstract:** Sodium-poor pollucite extremely close to the ideal end-member composition CsAlSi_2O_6 was discovered on a microscopic scale in five specimens from localities in Europe, Africa and North America. It crystallized at subsolidus temperatures of about 300–200 °C under hydrothermal conditions and is closely associated with the latest minerals in a typical sequence of alteration of pollucite. Wide veins of coarse-grained lepidolite + quartz ± spodumene cut the pollucite, followed by thin veins of fine-grained muscovite and spodumene; both types of veins are overgrown by granular aggregates of (K,Rb)-feldspar and albite. End-member CsAlSi_2O_6 occurs with these feldspars. The CRK = 100(Cs+Rb+K)/L cations of the latest pollucite ranges from 97.2–99.9 with Si/Al near 2.0. A mechanism of dissolution and reprecipitation is suggested for its origin, which could remarkably increase its Cs,Al content relative to its Na,Si-rich precursor. Later events (at less than 150–100 °C) caused only analcimization and replacement of pollucite by clay minerals.

**Key-words:** pollucite, cesium, CsAlSi_2O_6, granitic pegmatites.

**Introduction**

The aluminosilicate of cesium, pollucite occurs only in complex, rare-element granitic pegmatites as a product of extreme alkali-metal fractionation. The abundance of Cs in the continental crust is about 2 ppm (McDonough *et al.*, 1992) yet CsAl-Si_2O_6 contains ~ 45 wt.% Cs_2O. Pollucite concentrations in pegmatites are economically significant as the main industrial source of Cs. Also, synthetic pollucite is an important long-term containment phase of cesium radioisotopes which are prominent nuclear fission products. For example, analcime-pollucite phases commonly crystallize at low temperatures during hydrothermal alteration of various nuclear waste forms (as reviewed by Teertstra & Černý, 1992). The composition and crystallization conditions of natural pollucite are thus of considerable technological interest.

Historically, it was established by wet, bulk analytical methods that pollucite contains 70–80 mol.% of its end-member component, the remainder being complemented by solid solution with analcime, ideally NaAlSi_2O_6·H_2O (Fig. 1A). An important feature which has received less attention is the Si/Al ratio, generally greater than the ideal value of 2.0, typically 2.1–2.5 (Fig. 1B). Consequently, a natural occurrence of end-member pollucite of ideal formula was not expected on statistical grounds. Spurred on by poor correlations between the bulk composition of pollucite and some of its physical properties (Fig. 2A, 2B), which were noted by several authors (Černý, 1974; Feklichev, 1975; Fel'dman & Pleskova, 1978), we began a detailed investigation of this mineral in 1988.

Pollucite specimens representing at least 55 different localities were analysed in detail by electron microprobe. They were found to be slightly to extensively heterogeneous, exhibiting relics of a primary phase in a microscopic network of secondary phases modified in terms of both Na/Cs and Si/Al ratios (Teertstra, 1991; Teertstra *et al.*, 1989, 1992, 1993). Three principal