Melt inclusions in quartz and topaz of the topaz granite from Eurajoki, Finland

Uzavřeniny magmatické taveniny v křemeni a topazu z topazového granitu od Eurajoki ve Finsku (Czech summary)

(3 text-figs, 1 tab.)

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Crystallized melt inclusions occur in quartz and topaz of the topaz granite (Väkkärä granite) of the 1.57 Ga Eurajoki rapakivi granite stock. Large silicate melt inclusions in quartz were fused to homogenization and the glass was analyzed by electron microprobe. The granitic melt inclusions are peraluminous and enriched in fluorine and tin (7.3 ± 0.4 wt % F, 0.03 wt % SnO_2), enabling magmatic crystallization of topaz. Together with recent corresponding observations from other highly evolved granites, the study demonstrates the important role of magmatic processes in formation of tin granites and other F-rich granites.

Key words: topaz granite, rapakivi granite, melt inclusions, cassiterite, magmatic evolution

Introduction

The origin of the mineralogically and geochemically anomalous tin granites has been a subject of controversy for decades. Many researchers have emphasized the role of postmagmatic metasomatic processes (albitization, topazization, muscovitization etc.) and concluded that the anomalous geochemical character of these specialized granites or "apogranites" is of secondary, metasomatic origin (Beus et al. 1962; Štemprok 1971), whereas others have preferred magmatic origin (e. g. Mackenzie et al. 1988; Taylor 1992). On the basis of detailed petrographic, mineralogical and geochemical studies of the tin-mineralized topaz granite from Eurajoki, southwestern Finland, Haapala (1977, 1997) concluded that this granite and its accessory minerals topaz and cassiterite are essentially magmatic in origin; autometasomatic fluid-rock interactions have only slightly modified the composition of the granite. In this paper we present additional evidence, based on studies of crystallized melt inclusions in quartz and topaz, for the magmatic origin of this mineralogically and geochemically specialized granite. Premilinary results of this study are presented in Thomas – Haapala (1998).

The topaz granite of the Eurajoki rapakivi stock

The topaz granite (Väkkärä granite) is the latest major intrusive phase of the 1.57 Ga Eurajoki stock, a satellite of the Laitila rapakivi granite batholith (Fig. 1). The earlier, less evolved granite phases are represented by a marginal medium-grained biotite-hornblende(-fayalite) granite (Tarkki granite) and a biotite granite, which varies in texture from fine-grained porphyritic to medium-grained equigranular. Rhyolite dykes, some of them topaz-bearing, cut the biotite-hornblende(-fayalite) granite. Similarly, dark porphyry dykes that were formed by mingling and mixing of rhyolitic crystal mush and mafic magma cut the earlier granites. Mineralogical and geochemical similarities indicate that the rhyolitic dykes were derived from the same magma chamber as the topaz granite. Greisen-type Sn-Be-W-Zn mineralization occurs as veins and lenses in different granites of the stock.

The topaz granite is typically slightly porphyritic, with 1–3 cm long alkali feldspar and 0.5–1 cm thick quartz megacrysts in fine- to medium-grained matrix, but locally the granite is nearly equigranular. The average mineral composition is: alkali feldspar 31.9, quartz 25.3, albite 23.9, biotite ± chlorite ± muscovite 4.8, topaz 2.1, and fluorite 1.4 vol %. Typical accessory heavy minerals are monazite, zircon, ilmenite, cassiterite, columbite, bastnäsite, xenotime and thorite. Miarolitic cavities are relatively common and grade locally into interconnected miarolitic texture as described from other places by Candela (1997). The miarolitic cavities indicate the presence of a separate fluid phase in the granite magma during final stages of crystallization.

In chemical composition, the topaz granite is a typical highly evolved F-rich, P-poor granite. It is peraluminous and shows high contents of F, Ga, Rb, Sn and Nb, and low concentrations of Ti, Mg, Ca, Ba, Sn, Zr and Eu (Haapala 1977, 1997; Table 1).

The alkali feldspar is perthitic with strings, veins and patches of albite in potassium feldspar (microcline). Swapped albite rims occur at the contact between two adjacent alkali feldspar grains, and turbid plagioclase grains have outer zones of water-clear albite against microcline grains. These albitic rims and zones are interpreted to result from exsolution of albite from alkali feldspar, migration to the grain boundaries, and growth on alkali feldspar or plagioclase grains (Haapala 1997).

The biotite is a lithian siderophyllite and occurs typically as anhedral flakes between feldspar and quartz grains

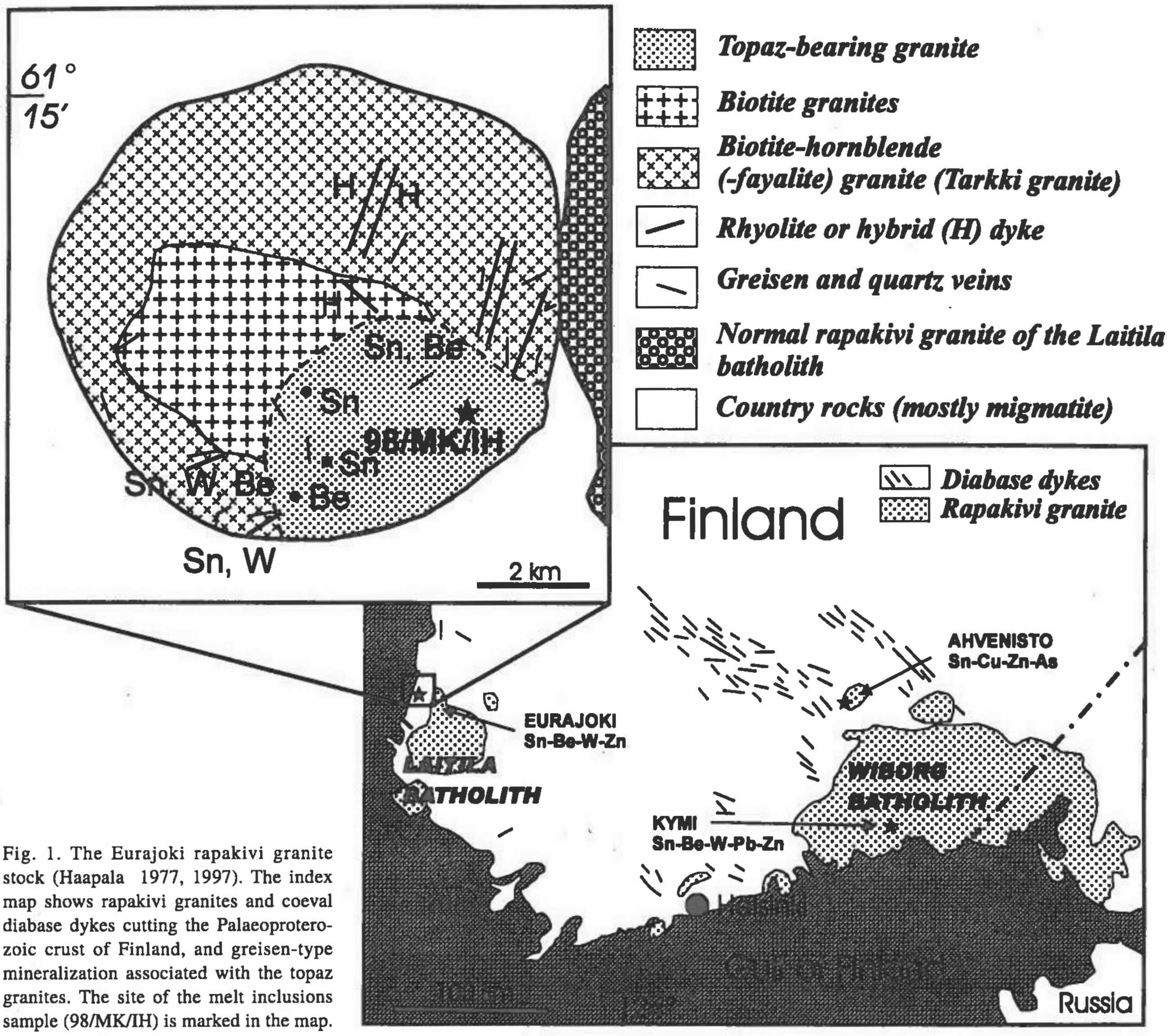

Fig. 1. The Eurajoki rapakivi granite stock (Haapala 1977, 1997). The index map shows rapakivi granites and coeval diabase dykes cutting the Palaeoproterozoic crust of Finland, and greisen-type mineralization associated with the topaz granites. The site of the melt inclusions sample (98/MK/IH) is marked in the map.

in small amount as zone-controlled inclusions in quartz crystals. An unaltered biotite of the same locality (outcrop 98/MK) as the melt inclusion sample of this study gave composition: SiO_2 40.30, TiO_2 0.64, Al_2O_3 20.13, Fe_2O_3 4.04, FeO 16.53, ZnO 0.40, MnO 0.95, MgO 0.10, CaO 0.08, Li_2O 1.63, Na_2O 0.18, K_2O 9.22, Rb_2O 0.90, Cs_2O 0.024, F 4.90, P_2O_5 0.06, H_2O^+ 1.55 and H_2O^- 0.01 wt %; Σ after fluorine correction 99.58 wt % (Haapala 1977).

Topaz occurs in the granite in three different generations: 1) euhedral, commonly zone-controlled inclusions in quartz crystals, 2) subhedral crystals in the matrix and 3) as irregular, usually small grains replacing plagioclase and more rarely alkali feldspar. The two first generations are interpreted to be magmatic. The second generation predominates, indicating that the majority of topaz crystallized at a relatively late magmatic stage. Some euhedral fluorite crystals in the matrix appear to be primary, late magmatic minerals; however, fluorite is in part clearly secondary, formed in connection of de-anorthitization of plagioclase.

In the rhyolite dykes, topaz occurs 1) as phenocrysts and euhedral inclusions in quartz phenocrysts, 2) as small prismatic crystals in the matrix, locally showing a flow texture, and 3) locally, as metasomatic, irregular grains in the matrix. The occurrence of magmatic topaz in two generations is probably related to crystallization at different pressures. According to the experimental studies of Weidner – Martin (1987), high pressure favors early crystallization of topaz in fluorine-rich granitic systems.

High Nb and Ta contents of cassiterite occurring as an accessory mineral in the topaz granite, and in pegmatite veins and pockets, have been used as an evidence of their magmatic origin (Haapala 1977, 1997). The hydrothermal cassiterite of greisen has very low contents of Nb and Ta.

The sample 98/MK/IH, used for the melt-inclusion studies of this paper, is a typical well-preserved porphyritic topaz granite that contains alkali feldspar and quartz megacrysts in a fine-grained matrix. Photomicrographs

Table 1. Mean chemical composition (electron-microprobe analyses) of silicate melt inclusions in quartz of the topaz granite from Eurajoki (sample 98/MK/IH), compared to the whole rock composition of the host granite. Presented are also mean compositions of crystallized melt inclusions in quartz of some highly evolved granitic rocks and compositions of their host granites from Erzgebirge: Zinnwald microgranite (a quartz-phenocrystic albite microgranite; Thomas – Klemm 1997), Podlesí stock granite (F-rich, P-poor melt inclusions; Breiter et al. 1997; Breiter 1995), and Ehrenfriedersdorf pegmatite (Webster et al. 1997).

	Eurajoki (samp. 98/MK/IH)		**Zinnwald**	**Podlesí stock granite**		**Ehrenfriedersdorf**	
	Melt inclusions	Whole rock	Melt inclusions	Melt inclusions (samp. 2358b)	Whole rock (samp. 2358)	Melt inclusions (pegmatite quartz)	Whole rock (aplitic granite)
SiO_2	67.8 ± 0.5	74.63	66.2 ± 1.6	66.0 ± 1.4	76.11	67.5 ± 3.8	70.4
TiO_2	0.02	0.06	0.01	0.1	0.03	0.04 ± 0.02	0.06
Al_2O_3	14.8 ± 0.2	13.82	15.2 ± 0.7	16.9 ± 0.2	12.76	15.8 ± 1.7	17.2
$FeO_{tot.}$	0.7 ± 0.1	0.89	0.7 ± 0.1	1.2 ± 0.1	0.77	0.41 ± 0.2	1.48
MnO	0.1	0.04	0.1	0.1	0.02	0.05 ± 0.03	0.02
MgO	d. l.	0.01	d. l.	d. l.	0.05	0.01 ± 0.02	0.08
CaO	0.9 ± 0.04	0.68	0.2	0.4 ± 0.01	0.36	0.24 ± 0.68	0.36
Na_2O	5.6 ± 0.2	3.72	3.2 ± 0.2	3.7 ± 0.2	3.35	3.0 ± 1.0	2.84
K_2O	5.3 ± 0.2	4.90	4.8 ± 0.3	4.8 ± 0.4	4.35	4.3 ± 1.5	4.41
P_2O_5	0.02	0.01	< 0.02	0.3 ± 0.05	0.47	3.6 ± 2.0	0.36
F	7.3 ± 0.4	0.06	4.9 ± 0.3	8.9 ± 0.3	1.25	3.1 ± 1.0	2.2
Cl	0.03	n. a.	0.1 ± 0.01	0.2 ± 0.01	n. a.	0.1 ± 0.07	0.05
Rb_2O	n. a.	0.12	0.4 ± 0.04	0.6 ± 0.08	0.18	0.40	0.15
SnO_2	0.03	0.004	0.05	0.02	0.005	0.101	0.003
H_2O+	n. a.	0.4	4.2 ± 0.2	(2.8 ± 0.6)	1.16	1.2	0.4
$O=F_2$	–3.07	–0.45	–2.06	–3.75	–0.53	–1.31	–0.93
Σ	99.53	99.89	98.02	102.27	100.34	98.54	99.08
n	10		24	15		19	

d. l. = below detection limit
n. a. = not analyzed

of this granite are presented in Haapala (1997, Figs 3a and 4a–c), whole-rock and mineral compositions in Haapala (1977).

The melt inclusions

Primary crystallized melt inclusions in topaz of the Väkkärä granite from Eurajoki were first described by Haapala (1977) who used them as an evidence of magmatic origin of the topaz. The melt inclusions occur as fine-grained aggregates of minerals together with gas bubbles, and range in size from less than one µm to several tens of µm (Fig. 2). For this study, the nature and composition of melt inclusions in one topaz granite specimen (98/MK/IH) were studied by Rainer Thomas in GeoForschungsZentrum, Potsdam, using the methods described by Thomas (1994), and Thomas – Klemm (1996). Doubly polished chips of quartz and topaz grains containing melt inclusions were heated to homogenization in a tubular furnace, then quenched and polished down until the inclusions were exposed on the surface. Electron-microprobe analyses were carried out using a Cameca Camebax SX50 electron microprobe. Only completely homogenized melt inclusions with diameter ≥ 30 µm were used. Some quenched inclusions contained a shrinkage bubble (Fig. 3). The wavelength-dispersive analyses were conducted at 15 kV acceleration voltages, 10 nA beam current and a beam diameter of 10 to 30 µm. Defocused beam was used because of potential problems with alkali and F mobility during electron-microprobe measurements (see Hanson et al. 1996) The counting time was 40 s for F (using a multilayer PCI crystal), 60 s for Sn, 30 s for P and 20 s for other elements. Synthetic oxides and minerals were used as standards.

The mean composition of ten well-homogenized melt inclusions in quartz of the topaz granite (sample 98/MK/IH) is presented in Table 1, compared with the whole-rock composition of the host granite, and with compositions of crystallized melt inclusions in quartz of two topaz granites and one pegmatite from the Erzgebirge area: the Zinnwald microgranite (Thomas – Klemm 1997), the Podlesí stock granite (Thomas – Klemm 1997; Breiter et al. 1997; Breiter 1995) and a pegmatite from Ehrenfriedersdorf (Webster et al. 1997). The granites of Zinnwald and Podlesí are peraluminous topaz-bearing granites, and the Ehrenfriedersdorf pegmatite contains a number of rare minerals including Li-bearing mica, topaz, triplite, wolframite, beryl, and columbite.

The melt inclusions from the Eurajoki topaz granite differ in composition to some extent from their host granite. The melt inclusions are enriched in Al_2O_3, F and Sn (probably also in H_2O) and impoverished in SiO_2 compared to the granite. The melt inclusions contain in average 7.3 wt % F and 0.03 % SnO_2, whereas the granite contains "only" 1.06 % F and 0.004 % SnO_2. The mean tin content, based on 31 analyses, is about 80 ppm in the topaz granite (Haapala 1977). Some melt inclusions in quartz showed lower fluorine contents, 0.5 to 3.1 wt % F, but they were discarded because they appeared to con-

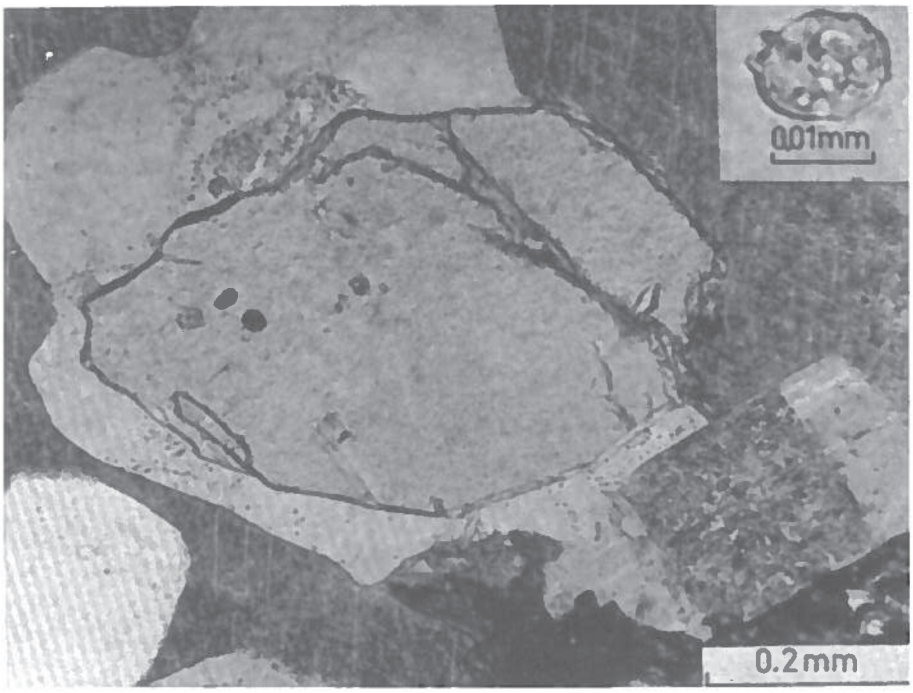

Fig. 2. Crystallized melt inclusions in a topaz grain surrounded by quartz and microcline (Haapala 1977). The inset shows enlargement of one of the inclusions. Sample 301/PL/68, one nicol.

tain accidentally trapped mineral phases. Similar relations between the composition of melt inclusions and their host granites have also been observed in the highly evolved topaz granites of Erzgebirge (Table 1). Ion-microprobe analyses of the melt inclusions from the pegmatite quartz of Ehrenfriedersdorf showed enrichment in several other incompatible trace elements (Be, P, Rb, Ga, Nb, Ta) and depletion in Sr and Zr (Webster et al. 1997). Minerals of the Podlesí stock granite contain, in addition to the F-rich, P-poor melt inclusions, also late-stage melt inclusions with 2.6 ± 0.4 wt % P_2O_5 (Breiter et al. 1997).

Discussion and conclusions

The primary magmatic melt inclusions in quartz and topaz of the topaz granite (Väkkärä granite) from Eurajoki provide very strong evidence for magmatic origin of these minerals, and confirm the conclusions based on petrographic observations (Haapala 1977, 1997). The exact time of trapping of the melt inclusions in relation to the emplacement and crystallization history of the granite magma is not known. It is, however, obvious that the fluorine-rich melt inclusions represent the highly

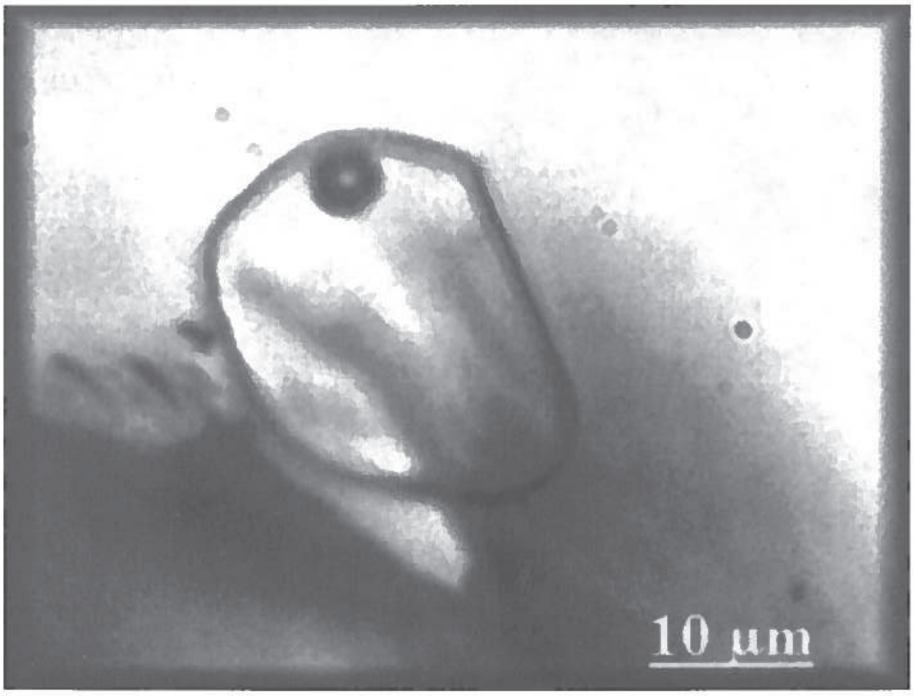

Fig. 3. A crystallized melt inclusion in quartz after homogenization and quenching to glass. The inclusion shows a small shrinkage bubble. Sample 98/MK/IH.

evolved melt from which quartz and topaz of the latest granitic phase of the Eurajoki stock crystallized. The melt inclusions are characterized by elevated contents of Al_2O_3, F and Sn when compared to the composition of the host granite. This is characteristic also of melt inclusions in the highly evolved Younger Granites of Erzgebirge (Zinnwald, Podlesí, Ehrenfriedersdorf) (Breiter et al. 1997; Thomas – Klemm 1997; Webster et al. 1997).

The conclusion that the accessory cassiterite in the topaz granite is a magmatic mineral was based on mode of occurrence and mineral chemical data (Haapala 1977), and this conclusion is not in contradiction with recent experimental studies of Linnen et al. (1996). The observed increased oxygen fugacity – an obvious precondition for the crystallization of cassiterite – at the end of magmatic evolution (Rieder et al. 1996; Haapala 1997) may be related to the separation of the aqueous fluid phase and partial escape of hydrogen, or to contamination of the late-stage melt with more oxidized water or other components from the surrounding country rocks.

The existence of primary magmatic topaz and cassiterite in the topaz granite of Eurajoki is a clear evidence that the late-stage granite magma was enriched in F and Sn, and the composition of the melt inclusions confirms this conclusion. Ion and electron microprobe analyses of crystallized melt inclusions from other localities (e. g., the Ehrenfriedersdorf pegmatite; Webster et al. 1997) further show that the parent melt was strongly enriched in Li, Rb, Be, B, Nb and several other lithophile elements (Webster et al. 1997). Enrichment of these elements is observed also in the glass inclusions in quartz from the Mexican tin rhyolites (Webster et al. 1996).

During magmatic evolution, the lithophile elements were concentrated in the residual melt. When the aqueous fluid phase separated, the trace elements and volatiles (F, Cl) partitioned between solid, melt and vapor phases according to their fractionation rules. The main F-bearing solid phases are micas, especially biotite, and, at a late stage of crystallization, fluorite and topaz. The available experimental studies suggest that in coexisting evolved melt-vapor systems fluorine generally concentrates into the melt phase relative to the aqueous fluid ($D_F^{vapor/melt}$ is ~ 0.3 when the melt contains 1–4 wt % F; London 1988; Webster et al. 1989), but at very high fluorine contents (≥ 7 wt % F in the melt), fluorine partitions in favor of the fluid phase (Webster – Holloway 1990).

It is evident that the role of metasomatic processes (albitization, topazization etc.) has often been overemphasized in attempts to explain the anomalous character of tin granites and topaz granites in general. However, postmagmatic mineral alterations (growth of secondary topaz, fluorite and muscovite, de-anorthitization of plagioclase) are common in the late-stage granites. Our conclusion is, that the anomalous geochemical character of the topaz granites and tin granites is primarily the results of magmatic crystallization of highly evolved melts. Postmagmatic fluid-rich reactions have, however, often modified the mineral and chemical composition of the granites.

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Uzavřeniny magmatické taveniny v křemeni a topazu z topazového granitu od Eurajoki ve Finsku

Vykrystalizované uzavřeniny magmatu se vyskytují v křemeni a topazu topazového granitu Väkkärä v intruzi 1,57 Ga rapakivi granitu Eurajoki. Velké uzavřeniny silikátové taveniny z křemene byly homogenizovány roztavením a výsledné sklo bylo analyzováno elektronovou mikrosondou. Inkluze granitické taveniny jsou peraluminózní a obohacené na F a Sn (7,3 ± 0,4 hm. % F, 0,03 hm. % SnO_2), což umožňuje magmatickou krystalizaci topazu. Společně s dalšími současnými studiemi jiných vysoce diferencovaných granitů, které vedou ke stejným výsledkům, naše pozorování dokumentují důležitou roli magmatických procesů při konsolidaci cínonosných a jiných fluorem bohatých granitů.