Internal Evolution and Disequilibrium Crystallization of a Highly Fractionated, Sn-Nb-Ta-Bearing Granite-Pegmatite System: a Case Study from the Říčany Pluton, Czech Republic

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Abstract. This study investigates textural zoning of highly evolved boron-rich granitic systems, and discusses its implications for the differentiation and crystallization of volatile-rich silicic melts and the formation of Sn, Nb and Ta mineralization. We characterize a 5 km-long zone of tourmaline-bearing granitic aplites and pegmatites emplaced along the southern margin of the Říčany pluton, which is one of the youngest and most evolved units in the Variscan Central Bohemian Plutonic Complex (Czech Republic). The aplite-pegmatite suite has a layered structure, defined by grain-size and modal variations on centimeter scale, which includes massive aplites, tourmaline- and, rarely, garnet-bearing layered pegmatite concordant layers, discordant aplites. pegmatite dykes, pegmatite cavities as well as unidirectional solidification textures. We attribute these textures to disequilibrium crystallization and formation of a chemically distinct boundary melt layer ahead of the progressing solidification front, as a result of diffusioncontrolled oscillations. This oscillatory behaviour was responsible for the observed modal and textural variations. Pegmatite layers and dykes indicate sites, where these extremely evolved and mobile melts were formed. In principle, the solidification fronts can be the major sites of formation of extremely chemically evolved melts. which develop localized enrichments in incompatible elements and high-valence metals and point to potential presence of economic mineralization.

Keywords. pegmatite, cassiterite, columbite, tantalite, solidification textures, crystallization kinetics

1 Introduction

Mineral deposits of critical metals, in particular Sn, W, Nb and Ta, are frequently associated with highly evolved granitic systems (Černý 1991; Breiter et al. 2007). Metal enrichments are produced during protracted fractionation and are hosted by pegmatite differentiates, or form hightemperature hydrothermal stockworks and veins (Heinrich 1990; Linnen et al. 2012). The critical roles of magmatic enrichment, spatial distribution, efficacy of partitioning to hydrothermal fluids, mechanisms of fluid focusing and precipitation remain poorly understood (Černý et al. 1985; Rickers et al. 2006).

This study illustrates textural features of a highly evolved granitic system, which provides insights into differentiation and crystallization mechanisms of volatile-rich granitic melts and formation of disseminated Sn, Nb, and Ta mineralization. We emphasize the key role of disequilibrium solidification, describe the formation of chemically modified melt boundary layers and discuss implications of textural development for geochemical exploration. Broader implications include important insights into the crystallization kinetics and rheology of natural magma chambers.

2 Geological setting

The Říčany pluton (Czech Republic) is situated at the northernmost extremity of the Variscan Central Bohemian Plutonic Complex (~3200 km²), emplaced along the boundary between two major tectonic units in the Bohemian Massif. The Říčany granite pluton is one of the youngest and most evolved units in this complex and was emplaced as a post-tectonic, shallow-level intrusion (Fig. 1).

The interior of the pluton is formed by equigranular medium-grained or weakly porphyritic granite, whereas the marginal variety corresponds to densely porphyritic granite with abundant K-feldspar phenocrysts, 4 to 7 cm in size. The average modal composition of both intrusive units is nearly identical (vol %): quartz 26-29, K-feldspar 27-31, plagioclase An₁₆ 35-36, biotite 6-7, respectively (Janoušek et al. 2014).

The pluton shows a reverse compositional zoning, a less evolved central unit and a highly evolved peripheral unit, reflected nearly exclusively in the whole-rock traceelement composition (Janoušek et al. 1997). The composite intrusion has been intersected by numerous aplite dykes whose thickness ranges from few centimetres to several meters, but they are extremely rare in the volcanosedimentary hanging-wall sequence. Sedimentary units in the contact aureole were locally converted to cassiterite-bearing tourmaline-quartz metasomatic rocks.

This study focuses on a \sim 5 km long and 500-600 m wide belt of tourmaline-bearing granitic aplites and pegmatites along the southern margin of the Říčany pluton (Fig. 1). The aplite-pegmatite suite has a layered structure defined by grain-size and modal variations in centimeter scale with tourmaline- and, rarely, garnetbearing line rocks (modally banded rocks – e.g. alternating tourmaline-rich and tourmaline-poor bands), pegmatite concordant layers, discordant dykes and devolatilization cavities as well as unidirectional solidification textures represented by comb layers of alkali feldspar. The orientation of a magnetic planar fabric, indicated by the anisotropy of magnetic susceptibility, shows subhorizontal emplacement of the aplite-pegmatite body with some steepening towards the

contact of the Říčany pluton. By contrast, the main granite units exhibit steep magmatic and magnetic foliations (Trubač et al. 2009). Spatial distribution of individual textural units and direct observations of the contact relationships indicate sharp intrusive contacts with all surrounding units. A thin lenticular unit of biotite granodiorite occurs along the contact interface of the aplite-pegmatite suite and the main pluton body.



Figure 1. Geological map of the Říčany pluton: (a) intrusive units of the pluton (types of rock are written within the image); (b) detailed geological map of the aplite-pegmatite suite. Symbols: 1 - massive leucogranite aplites; 2 - massive tourmaline aplites; 3 - biotite granodiorites; 4 - aplites with feldspar megacrysts; 5 - layered aplites; 6 - aplites with pegmatite cavities; 7 - tourmaline-biotite microgranites; 8 - biotite microgranites; 9 - secondary muscovite alteration; 10 - tourmaline-bearing pegmatite cavities; 11 - planar magmatic fabric; 12 - xenoliths of host rocks.

The granite-aplite-pegmatite suite represents a highly evolved, SiO₂-rich high-K to shoshonitic magmatic sequence, progressively enriched in Sn, W, Ta, Pb, Be, and B (Vejnar 1973; Němec 1978). The extreme enrichments in light elements and economic metals are indicated by occurrence of topaz, beryl, cassiterite, columbite and tantalite.

3 Textural evolution

The aplite-pegmatite body consists of texturally homogeneous (massive) aplites, heterogeneous layered aplites, concordant pegmatite layers, crosscutting pegmatite dykes and pegmatite cavities (Figs. 2 and 3).



Figure 2. Structures of layered aplites and pegmatites: (I) layered aplite ("line rock") with tourmaline (black) and garnet (pin) band; (II) layered tourmaline aplite ("line rock") with a concordant pegmatite layer and a crosscutting dyke of pegmatitic melt; (III) layered tourmaline aplite with a concordant pegmatite layer, comb layer representing unidirectional solidification texture and solitary K-feldspar megacrysts resulting from supressed nucleation rate.

Proportions of principal rock-forming minerals in the aplites vary about 30-35 vol. % quartz, 21-28 vol. % K-feldspar, and 33-40 vol. % plagioclase. Additional minerals are represented by biotite, muscovite, tourmaline, and garnet, with accessory cassiterite, columbite-tantalite, rutile, apatite, ilmenite, zircon, xenotime, zircon, and monazite. Tourmaline and muscovite are present in all textural types, whereas biotite is a rare constituent of some massive aplites. Spessartine-rich garnet is irregularly found in the layered aplites and all the other textural groups except massive aplites. Pegmatite dykes contain, in addition, topaz or beryl.



Figure 3. Megascopic magmatic structures: (I) layered tourmaline aplite with a comb layer consisting of solitary megacrysts, parallel to the rock layering, and with large K-feldspar megacrysts crosscutting and deforming the modal bands; (II) cross bedding and erosional structures during repetitive intrusive events; (III) layered aplite with a melt protrusion, offsetting the modal bands.

Massive and layered aplites have fine-grained equigranular texture with a mean grain size of 0.1-1.5 and 0.05-0.6 mm for homogeneous and layered varieties, respectively. Tourmaline is randomly disseminated in the massive aplites while it occurs in the form of tourmalinerich bands, rarely accompanied by garnet-rich bands, in the layered types (Figs. 2I-II). The bands are straight or show round protrusions that are reminiscent of advancing crystallization fronts and crystal growth controlled by diffusion. The tourmalines are internally zoned, from bluish grey, pale beige to richly brown pleochroic colours, in concentric growth arrangement or along secondary cracks, where the pleochroism has changed to green- blue zones due to hydrothermal reequilibriation.

In pegmatite layers (Figs. 2II-III), ~3-X0 mm thick, the grain size increases abruptly (~0.1-X mm). These layers often have internal comb texture (Figs. 2III and 3I), that is, the minerals are elongated in the direction of growth, which is perpendicular to the magmatic layering. Some of the larger (up to several cm) phenocrysts extend from a pegmatite layer into an adjacent aplite layer, and cut the tourmaline-rich bands (Fig. 3I). The pegmatite layers are generally associated with tourmaline-, garnet-or cassiterite-rich bands.

The pegmatite dykes, 3 mm to several cm thick, crosscut the aplite layering in a semi-brittle manner (Figs. 2II and 3III). These dykes can be locally internally zoned and often consist of a quartz core, feldspar margin, and a very fine-grained plagioclase-rich rim. Tourmaline, garnet, muscovite, beryl, topaz, rutile, cassiterite, apatite and columbite-tantalite are frequently concentrated in the marginal zone.

Pegmatite cavities occur in the aplite-pegmatite suite as well as in the Říčany granite, and are characterized by very large grain size (up to several centimetres) and no central hollow (complete crystal infill). They contain the same principal minerals as the surrounding aplitepegmatite host.

4 Implications for crystallization and Sn-Nb-Ta mineralization

Magmatic textures of the aplite-pegmatite suite strongly suggest crystallization under disequilibrium conditions (e.g., Lofgren and Donaldson 1975; London 2009). Such conditions are promoted by rapid cooling following emplacement of highly evolved melt and propagation of solidification fronts. Chemical constituents, which are not incorporated into the principal crystallizing minerals (mainly quartz, K-feldspar and albite) are accumulating ahead of the rapidly advancing crystallization front, thus forming a chemically distinct melt boundary layer. This compositional departure leads to localized and intermittent undersaturation in solid phases, which constitute the solidification front, and a supersaturation in volatile-rich and peraluminous phases as documented by periodic nucleation and growth of tourmaline or spessartine-rich garnet. Fluxing components, mainly H₂O and B, accumulate in the boundary layer, lower the melt viscosity, thus enhance the lateral diffusivity of essential structural components and inhibit the crystal nucleation. This is responsible for variations in the

crystal size, that is, the formation of textural layering and solitary megacrysts or megacryst fronts.

The aplite-pegmatite suite bears all similarities to silicic systems with diffusion-controlled oscillatory nucleation (e.g., Webber et al. 1997; London 2008, 2009). These phenomena further imply variations in the rates of nucleation and growth. Large K-feldspar megacrysts preserved in a fine-grained, aplitic matrix can be crosscut by, or deform a modal band (Figs. 2III and 3I); and locally nucleate from a single planar surface, which is parallel to the rock layering (Fig. 3I). These unidirectional solidification textures represent analogues of comb layer crystallization.

Modally layered aplites are locally truncated by erosional surfaces (Fig. 3II), which together with the flow fabrics (Fig. 3II), indicate multiple injections of low-viscosity magmas. The role of crystal settling or density currents was, however, very limited due to short inferred solidification time. However, fractions of a more fluid melt, possibly boundary layer melt enriched in fluxing components, could flow parallel to the solidification fronts or be expelled by compaction and crosscut the crystal mush or compact crystal frameworks. The pegmatite dykes provide example of such a melt, which probably meets requirements for accumulation of incompatible and economic elements. In this example, the amount of these melts is limited by the centimetre scale of the diffusive distance and the size of the intrusive body. In principle, the solidification fronts can be major sites of formation of extremely chemically evolved melts and, in combination, with suitable extraction mechanisms, can provide explanation for the metal pre-enrichment and formation of late-stage magmatic deposits of high-valence metals.

In summary, the aplite-pegmatite suite of the Říčany pluton represents subhorizontal tabular intrusion of very highly evolved, H_2O -B-rich melts, which have solidified far from equilibrium, possibly driven by undercooling. This is documented by the evolved chemical composition of the rocks, unidirectional solidification textures, modal and textural layering of the rocks, abrupt changes in grain size, and occurrences of late, extremely evolved and probably mobile melts.

This study provides insights into differentiation and crystallization mechanisms of volatile-rich granitic melts and proposes the formation of Sn, Nb, and Ta deposits *via* the origin of very evolved melts under transient and oscillatory disequilibrium conditions. The textural features and their spatial zoning represent potential exploration vectors for the Sn, Nb and Ta exploration.

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