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Stable isotope (B, H, O) and mineral-chemistry constraints on the magmatic to hydrothermal evolution of the Varuträsk rare-element pegmatite (Northern Sweden)

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ABSTRACT

The internal evolution of the Varuträsk rare-element pegmatite (Skellefte District, Northern Sweden) has been investigated using stable isotope (B, H, O) geochemistry of tourmaline and coexisting micas, feldspar and quartz. Varuträsk is a classic and typical example of highly fractionated LCT-type pegmatites, with a pronounced concentric zoning pattern composed of well-developed border, wall and intermediate zones and a quartz core. The pegmatite displays considerable rare-element enrichment, culminating in the formation of albite-lepidolite and pollucite units in the innermost zones. Major and trace element variations in tourmaline from the main pegmatite zones correlate well with the internal zoning pattern. Mineral compositions record an abrupt change in fractionation trends between the barren outer and intermediate zones and the inner, late-stage assemblages that carry rare-element mineralization. This change is also shown by the B-isotope variations of tourmaline. Early and mid-stage tourmalines record a systematic increase in δ^{11}B from −14.6‰ to −6.2‰ which can be explained by closed-system melt-mineral isotope fractionation whereby crystallization of large amounts of muscovite preferentially removes ^{10}B from the residual melt. In contrast, tourmaline from late-stage assemblages in the inner zones and cross-cutting veinlets shows a reversal in the B isotope trend, with a decrease in δ^{11}B from −8‰ to −14.1‰. This reversal cannot be explained by mineral-melt isotope fractionation, but requires fluid-melt partitioning and partial fluid loss. Hydrogen isotope variations in mica support this model. The systematic increase in δD from −75‰ in the outer zones (muscovite) to −63‰ and −53‰ in the inner zones (Li-micas) cannot be explained by closed-system variations in temperature or melt-mica fractionation, but it is consistent with late fluid exsolution. Oxygen isotope compositions of tourmaline (δ^{18}O from 9.7‰ to 11.6‰), quartz (13.3‰ to 14‰) and mica (10.3‰ to 11.3‰) show good agreement with equilibrium partitioning and yield temperatures in the range 450 °C to 600 °C. Combining this with the stability fields of Li-aluminosilicates petalite and spodumene indicates crystallization pressures of 2–3 kbar. Taken together, the stable isotope and mineral chemistry data demonstrate that rare-element enrichment in the innermost fractionated assemblages in the Varuträsk pegmatite was associated with the transition from purely magmatic crystallization to conditions where a separate aqueous fluid phase became important.

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1. Introduction

Geochemically specialized granitic pegmatites like Varuträsk form from the residual melts of large felsic intrusions, and they represent the most highly fractionated end-members of granitic suites. Pegmatites are therefore important for understanding the processes of magma evolution and enrichment in incompatible elements. Granitic pegmatites can also be economically important sources of rare elements such as Ta, Nb, Li, Cs, Be, Y and the REE (Linnen and Cuney, 2005; London, 2005a; Linnen et al., 2012). The pegmatites of the so-called rare-element class (see Černý, 1991a, 1991b; Černý and Ercit, 2005; Černý et al., 2012) are separated into two families according to contrasting mineralogical-geochemical features and the nature of associated granite suites. The lithium-cesium-tantalum (LCT) family is associated with peraluminous S-type leucogranites, whereas the niobium-yttrium-fluorine (NYF) family is associated with peralkaline granites.

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Further subdivisions within the two families are defined by distinctive mineral assemblages (Černý and Ercit, 2005). According to this scheme, the Varuträsk pegmatite belongs to the petalite subtype of the LCT family, along with other well-known and economically important pegmatites like Tanco in Canada (Černý, 2005; Stilling et al., 2006), Bikita in Zimbabwe (Cooper, 1964) and Greenbushes in Australia (Partington et al., 1995).

Early research on the internal evolution and petrogenesis of pegmatites based on field studies and experimental work (Jahns and Burnham, 1969; Jahns, 1982) resulted in the concept that the coexistence of a granitic melt and an aqueous fluid phase was an essential factor for their internal textural and geochemical-mineralogical features. Recent work on the late-stage processes in pegmatites responsible for element enrichments, mineral segregation and rare-element mineralization has challenged the classic Jahns-Burnham model. Major new insights into the understanding of the evolution of the melt and fluid phase(s) in pegmatites has come from experimental petrology (London, 1992, 2005b, 2008; Linnen and Keppler, 1997) and from studies of natural fluid and melt inclusions (e.g., London, 1986; Thomas et al., 1988; Thomas et al., 2000; Sirbescu and Nabelek, 2003; Kamenetsky et al., 2004; Badanina et al., 2006; Nabelek et al., 2010; Thomas et al., 2012). The inclusion studies, particularly, point out the potential importance of liquid immiscibility in flux-rich pegmatite melts, a phenomenon which has been confirmed experimentally (e.g., Veksler and Thomas, 2002; Veksler, 2004).

The characteristic textural development and internal zoning of pegmatites, which involve extreme and abrupt variations in crystal sizes and mineral modes, was explained in the classic Jahns-Burnham model by the coexistence of hydrous melt with a separate aqueous fluid, with the latter playing a key role in the growth of large crystals and differential transport of alkali elements. Experimental observations and thermal models that attest to rapid cooling of pegmatites have led to an alternative model championed by London (2005b, 2009) and Simmons and Webber (2008). This explains pegmatite textures by special conditions of slow nucleation and rapid crystal growth caused by liquidus undercooling and the establishment of a low-viscosity crystal-melt boundary layer enriched in H_2O and other fluxing elements such as B, Li, P and F. Nabelek et al. (2010) underscored the importance of undercooling in pegmatite textural development but emphasized the additional factor that a separate aqueous fluid phase must be present to allow for rapid growth of large crystals. There is no disagreement in the pegmatite literature that late-stage melts have high concentrations of water and other flux components. The important open questions are whether and at what stage in pegmatite evolution does fluid phase separation take place, particularly in reference to rare-element mineralization. For example, it has been suggested that the extremely fractionated albite-lepidolite inner units, which typically host the highest concentration of ore minerals like Nb-Ta oxides are crystallization products of boundary layer melts, with no phase separation implied (London, 2008). However, this concept was challenged by Thomas et al. (2012) as inconsistent with the results of fluid and melt inclusion studies.

Fluid exsolution is indisputable in the shallow-level, miarolitic pegmatites that contain crystal-lined open cavities and show abundant fluid inclusions with the typical signature of aqueous fluids (Taylor et al., 1979; London, 1986), but in deeper pegmatites including Varuträsk and most others of the rare-element class, direct evidence for fluid separation is subtle and restricted to the late-stage assemblages (London, 2008). The role of magmatic vs. hydrothermal processes in pegmatite mineralization continues to be debated. Mounting experimental evidence for high solubility of many rare-elements in high-temperature hydrothermal fluids including the REE and HFSE (Mayanovic et al., 2007, 2009; Migdisov et al., 2009, 2011; Loges et al., 2013) warrants renewed consideration of the role of aqueous fluids in the mineralization potential of rare-element pegmatites. One of the potentially most successful ways to detect the magmatic-hydrothermal transition in pegmatites is by analysis of stable isotope variations in pegmatite minerals, which is the subject of this study of the Varuträsk pegmatite in NE Sweden.

The Varuträsk pegmatite is a classic example of a complex LCT-type pegmatite with well-developed internal zoning and advanced rare-element enrichment culminating in the formation of Nb-Ta oxide-bearing albite-lepidolite and pollucite units. The pegmatite has been well characterized mineralogically (Quensel, 1952, 1956; Smeds, 1990; Černý et al., 2004) and it can be sampled systematically from surface and underground exposures complemented by an extensive archive of diamond drillcores. This paper presents stable isotope (B, O, H) data of tourmaline, mica, quartz and feldspar from the Varuträsk pegmatite. The results are combined with relevant major and trace element data from Matalin (2010), which document the essential variation trends of these minerals and their relationships to the internal units of the pegmatite. We show that the trends in boron and hydrogen isotope composition of tourmaline and mica from outer zones to the inner albite-lepidolite units hosting Nb-Ta and Cs mineralization are best explained if saturation with an aqueous fluid phase was reached.

2. Geological setting

The Varuträsk pegmatite is situated approximately 11 km west of Skellefteå, in Västerbotten County, Northeastern Sweden (Fig. 1). It is located within the Paleoproterozoic Skellefte District, a 150 by 50 km region of greenschist- to amphibolite-facies metasedimentary and metavolcanic rock series intruded by several generations of granitoids. According to lithostratigraphic analyses, geochemistry and geochronology, the geologic setting of the Skellefte District can be briefly summarized as follows (Bergström, 2001). The oldest units are Skellefte Group rhyolites and basalts with minor intercalated sediments (1.90–1.88 Ga), which were deposited in an extensional continental margin setting. Overlying the Skellefte Group are units of the Vargfors Group (1.88–1.87 Ga), which consist mainly of clastic sediments (mudstone, siltstone and greywackes), with MORB-like mafic volcanic rocks in the upper Varuträsk Formation. The latter indicates a possible rift basin in the eastern part of the Skellefte District (Allen et al., 1996; Bergström, 2001). During the Svecokarelian orogeny, the sedimentary and volcanic rocks were deformed and metamorphosed at greenschist to upper amphibolite facies conditions (Billström and Weihed, 1996; Bergström, 2001). Also related to the Svecokarelian orogeny are various syn-to post-tectonic granitoid intrusions (Kathol and Weihed, 2005, and references therein). Of interest to this study are the intrusions of the peraluminous, S-type Härnö-Skellefte granitic suite, which have been dated at 1.82–1.80 Ga and interpreted as late orogenic (Claesson and Lundquist, 1995). Weihed et al. (2002) reported a U-Pb titanite age of 1798 ± 4 Ma for the Skellefte granite proper, which crops out close to the Varuträsk pegmatite (Fig. 1). A second suite, the Revsund I-type granites intruded after the main phase of deformation and were dated at 1.80–1.78 Ga, (Skiöld, 1988). However, the age of the Revsund suite is somewhat uncertain because older ages were obtained for Revsund suite granites located southwest of the Skellefte District by Högdahl (2000). Several post-orogenic pegmatite dikes with a LCT-type mineral affiliation, including the Varuträsk pegmatite, occur in the eastern part of the Skellefte District (Smeds, 1990; Romer and Smeds, 1994). Based on the geochemical similarities, age association and physical proximity, the pegmatites are considered to be genetically related with the Härnö-Skellefte peraluminous granitic suite (Smeds, 1990).

3. The Varuträsk pegmatite

The Varuträsk pegmatite is mainly hosted by an amphibolite body within metasediments (metapelite, metagreywacke) of the Varuträsk Formation (Fig. 1). The southern margin of the pegmatite straddles the contact between the amphibolite and metasediments. The pegmatite was discovered in 1933 and mined by Boliden Mineral AB (New Boliden) for lithium and cesium. Today, an underground visitors mine

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