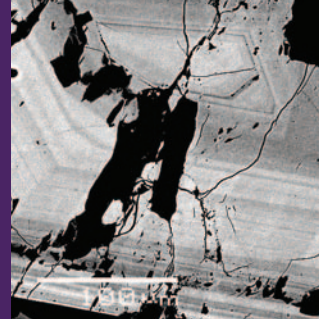


Granitic Pegmatites as Sources of Strategic Metals



Backscattered electron image of a wodginite crystal

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1811-5209/12/0008-0275\$2.50 DOI: 10.2113/gselements.8.4.275

Rare-element granitic pegmatites are well recognized for the diversity and concentrations of metal ores that they host. The supply of some of these elements is of concern, and the European Commission recently designated metals such as tantalum and niobium as “critical materials” or “strategic resources.” Field relationships, mineral chemistry, and experimental constraints indicate that these elements are concentrated dominantly by magmatic processes. The granitic melts involved in these processes are very unusual because they contain high concentrations of fluxing compounds, which play a key role at both the primary magmatic and metasomatic stages. In particular, the latter may involve highly fluxed melts rather than aqueous fluids.

KEYWORDS: strategic, rare element, fractionation, metasomatism, ore genesis

INTRODUCTION

Strategic elements, generally metals, are those that are of greatest risk to supply disruptions or are important to a country's economy or defence. The list varies with country but includes Ta, Nb, Be, Sb, W, rare earth elements (REEs), and Co. Rare-element pegmatites are enriched in incompatible elements (high-field-strength and large-ion lithophile elements), and many of these elements are also strategic metals. A particularly important class of rare-element pegmatites is the complex-type pegmatites of the LCT (Li–Cs–Ta) family (Černý and Ercit 2005; Černý et al. 2012 this issue). This type of pegmatite contains extremely high concentrations of Rb, Cs, Be, Ta, Nb, and Sn, as well as elevated levels of fluxing components (Li, P, F, and B). For example ores in the Tanco pegmatite (Manitoba, Canada) contain up to 13,900 ppm Li, 236,000 ppm Cs, and 28,900 ppm Rb, as well as >360 ppm Be and >1200 ppm Ta (Stilling et al. 2006). These values contrast with the bulk continental crust, which contains 16 ppm Li, 2 ppm Cs, 49 ppm Rb, 1.9 ppm Be, and 0.7 ppm Ta (Rudnick and Gao 2004); that is, some enrichment factors are greater than 100,000. One of the greatest challenges in the study of pegmatites is to explain how such extreme elemental enrichment occurs. A different family of pegmatites, the NYF pegmatites, is enriched in Nb, Y, and F. Their current economic importance is much less than that of the LCT family, but these pegmatites may produce rare earth and other strategic metals in the future; an example is the Strange Lake deposit, Canada, which contains 140.3 million tonnes grading 9330 ppm total REEs, 19,300 ppm

ZrO₂, 1800 ppm Nb₂O₅, and 800 ppm BeO (www.questrareminerals.com; accessed December 2011).

STRATEGIC METALS AS COMMODITIES

The uses of strategic metals are not widely known to the general public, but these metals play a vital role in society. Tantalum capacitors are utilized in computers, smart phones, and automobiles (e.g. ABS and airbag-activation devices). Technical-grade lithium is used in ceramics and glasses, whereas chemical-grade Li is used in lubricants and is a major

component of rechargeable batteries in, for example, electric vehicles. Cesium formate is fabricated for applications in high-pressure and high-temperature drilling in petroleum exploration. Because of its photoemissive properties, Cs is also used in solar photovoltaic cells. Beryllium and copper alloys are in components of aerospace, automotive, and electronic devices.

Tantalum is not a publicly traded commodity, and information on production and prices is difficult to obtain. However, Schwela (2010) presents a “most likely resource base”: South America has reserves of approximately 130,000 tonnes of Ta₂O₅ (40% of the known global resource), followed by Australia (21%), China and Southeast Asia (10%), Russia and the Middle East (10%), and Central Africa (10%). The rest of Africa, North America, and Europe make up the final 10%. Currently exploited pegmatites of note are the Mibra operation in Brazil, Kenticha in Ethiopia, and Marropino in Mozambique. Historically much of the world's Ta production has come from complex-type pegmatites (e.g. Tanco, Canada; Greenbushes, Australia; Altai #3, Mongolia; Bikita, Zimbabwe), albite-type pegmatites (Wodgina, Australia), and albite–spodumene pegmatites (Mt. Cassiterite, Australia) (Černý and Ercit 2005). A geopolitical concern specific to tantalum is the mining of columbite–tantalite (termed *coltan*) from Central Africa, particularly the Democratic Republic of Congo; this tantalum has been sold by illegal militias implicated in human rights abuses to fund a civil war. The U.S. government signed the Financial Stability Act into law in 2010 to stem the trade of “conflict tantalum,” and a system that includes mineral chemistry, geochronology, and mineral-liberation analysis is being developed to trace the origin of coltan ore (Melcher et al. 2008).

Lithium is mined from brine deposits in Chile and Argentina, granites in China, and pegmatites in Australia, China, and Zimbabwe. An important aspect of the lithium

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market is that lithium carbonate is one of the lowest-cost components in a lithium-ion battery. In addition to the mining cost, a secure supply is important to the sale of chemical-grade Li. Lithium pegmatites remain a viable source of the metal because of its high concentration in the ores, and production from these deposits mitigates concerns about supplies from brine producers in Chile and Argentina. Chemical-grade Li refers to spodumene that is converted to lithium carbonate or lithium hydroxide. By far the most important Li pegmatite is the Greenbushes deposit, which produced roughly one-third of the world's lithium in 2009 and contains 70.4 million tonnes grading 2.6 wt% Li₂O (www.talisonlithium.com; accessed December 2011). The Greenbushes pegmatite is also the major source of technical-grade Li (USGS 2011 Mineral Commodities Summary), where spodumene is used directly in the ceramics or glass industries without processing.

Significant past production of Li has come from albite–spodumene-type pegmatites (e.g. Kings Mountain, USA), and other pegmatites are currently under evaluation (e.g. Whabouchi, Canada). These pegmatites lack internal zoning and contain large tonnages of high-grade Li mineralization [26 million tonnes of 1.5 wt% Li₂O at Kings Mountain (Černý 1989); 25 million tonnes of 1.54 wt% Li₂O at Whabouchi (www.nemaskalithium.com; accessed March 2012), but their origin has received little scientific investigation and they remain poorly understood. Lithium has also been produced from petalite in complex-type pegmatites in the past (e.g. Bikita, Zimbabwe), and other deposits, such as Separation Rapids (Ontario, Canada) are potential future resources, particularly for technical-grade Li.

The world's supply of **cesium** is almost exclusively from the Tanco pegmatite, Manitoba, Canada, although other pegmatites, such as Bikita, contain significant reserves. **Beryllium** production is dominated by the rhyolite-hosted epithermal bertrandite [Be₄Si₂O₇(OH)₂] deposit at Spor Mountain, Utah, USA. However, beryl from pegmatites continues to be a lesser, but local, source of beryllium (USGS 2011 Mineral Commodities Summary).

RARE-ELEMENT PEGMATITES

Fractionation Trends

The LCT pegmatites are typically associated with late-tectonic peraluminous granites, and their emplacement is controlled at least in part by shear zones (FIG. 1; Černý 1989; London 2008). Quartz and feldspars are the dominant minerals that crystallize from granitic melts, and the rare elements are highly incompatible in these minerals. Thus, extreme fractionation resulting from extended crystallization of quartz and feldspars can generate very high concentrations of rare elements in residual melts. Similarly, individual pegmatites also consist largely of quartz and feldspar, and the rare elements are concentrated in small volumes (Stilling et al. 2006). Cesium is highly incompatible because of its very large ionic radius, except in micas, feldspars, and cordierite, where it is moderately incompatible. Rubidium behaves similarly, except that its ionic radius is closer to that of K, and is compatible in micas and alkali feldspar. The ionic radius of Li is much smaller than that of the other alkali metals, and Li partitions into micas, cordierite, and amphiboles via coupled substitution reactions (London 2005). These elements are commonly interrelated in minerals; for example, Li–Rb–Cs-rich micas are also fluorine rich. Beryllium is normally incompatible because of its small ionic radius, but is compatible in cordierite and muscovite (London 2005). Partition coefficients will change with the various substitutions; therefore it is

challenging to estimate melt compositions from mineralogical and partitioning data alone. However, the number of common minerals in which these rare elements are compatible is relatively small; even where oxides and micas crystallize, the abundance of these minerals is generally low, such that they reflect the concentrations of rare elements in the melt rather than control them. Several key minerals record (but don't control) the trace element behavior in the melt: with fractionation, the K/Rb ratio in blocky K-feldspar crystals decreases; the Li, Rb, and Cs contents of K-feldspar and muscovite increase; the Na/Li ratio decreases and the Cs content increases in beryl; and the Nb/Ta ratio in columbite-group minerals decreases (Černý 1989). These parameters vary continuously from barren through Be–Nb-rich to complex LCT pegmatites (FIG. 1). The same trends appear in early to late assemblages within individual pegmatites. However, crystal–melt fractionation may not be the sole process, and a number of alternate explanations for these trends have been proposed (cf London 2005, 2008).

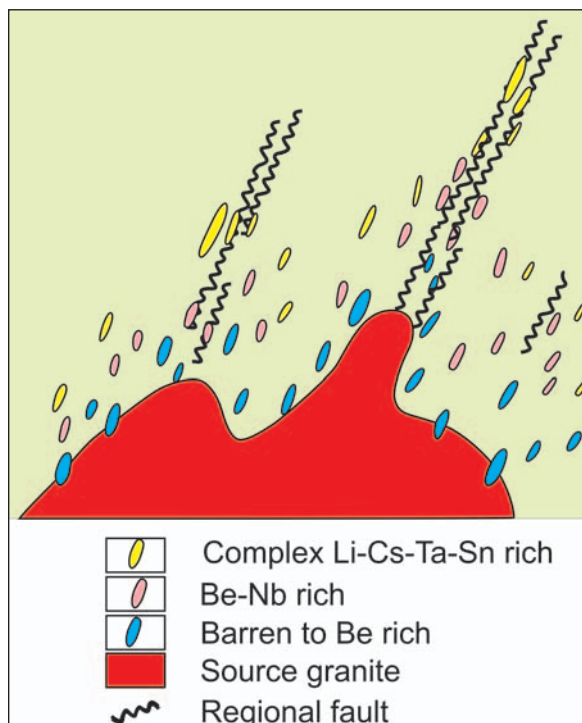


FIGURE 1 Idealized zoned pegmatite field around a source granite. The maximum distance of pegmatites from the source granite is on the order of kilometers or, at most, tens of kilometers. MODIFIED FROM ČERNÝ (1989)

Exploration and Evaluation

A useful feature of large and chemically evolved LCT pegmatites is that the wallrocks around these pegmatites are metasomatized (i.e. metamorphosed via an influx of pegmatite-derived components), and the dispersion of alkali rare elements in the metasomatic aureoles around pegmatites is used as an exploration tool. Lithium anomalies define the widest halos adjacent to pegmatites, which can be in excess of 100 m, but the dispersion of Rb and Cs is more restricted (Černý 1989; London 2008). Alkali metasomatism is a consequence of relatively high fluid–melt partition coefficients for Li, Rb, and Cs (London 2005). In fact, brine fluid inclusions from the Ehrenfriedersdorf pegmatite, Germany, contain thousands of parts per million (ppm) Li and weight percent levels of Rb and Cs; however, importantly, Ta is below the detection limit (Borisova et al. 2012). Biotite is an abundant metamorphic/metasomatic mineral in the country rocks that surround

LCT pegmatites. Because Li substitutes for Mg in biotite, and Rb and Cs for K, the formation of a biotite metamorphic/metasomatic aureole results in well-developed Li–Rb–Cs dispersion patterns around LCT pegmatites (London 2008). Primary dispersion of Be is also observed around LCT pegmatites, but this process is less well documented (Černý 1989).

Tantalum and Nb have a high ratio of charge to ionic radius, i.e. they are high-field-strength elements (HFSEs); hence their partitioning behavior is very different from that of Li, Rb, and Cs. Tantalum and Nb are highly incompatible in quartz and feldspar; thus their bulk distribution coefficients are typically very small. However, they are compatible in muscovite and partition strongly into Ti-bearing minerals, notably rutile and titanite (Linnen and Cuney 2005). The Ta content of muscovite has been used as an exploration tool, and pegmatites that contain muscovite with greater than ~80 ppm Ta are considered to have economic potential for Ta (Černý 1989). Fluid–melt partition coefficients of Ta are very low, in contrast to the alkali rare elements. Therefore, Ta is not added to the wall-rocks (Linnen and Cuney 2005); note also that most fluid inclusions lack Ta (e.g. Borisova et al. 2012).

The best-studied example of an LCT-family pegmatite is the Tanco pegmatite at Bernic Lake, Manitoba, Canada. The Tanco deposit consists of nine main mineralogical zones (Černý 2005). Different commodities are produced from different zones. Lithium production has come predominantly from the Upper Intermediate Zone, Cs production is from the Pollucite Zone, and Ta production has been primarily from the Aplitic Albite Zone and the Central Intermediate Zone. Only some of the zones are concentric, and Ta and Cs mineralization occurs in different pods within the internal structure of the pegmatite (Černý 2005; Stilling et al. 2006). Similarly, at the giant Greenbushes pegmatite, Ta mineralization occurs at a different location than the Li mineralization. Therefore, Ta and Li mineralizations may have had different controls and consequently are treated separately below.

BERYLLIUM MINERALIZATION

Pegmatites are well known for their exceptional mineral diversity, and the mineralogy of Be is a good example of this. Beryllium-bearing mineral groups include oxide, hydroxide, borate, phosphate, silicate, and aluminosilicate minerals, but production of Be from pegmatites as an industrial mineral comes almost exclusively from beryl ($\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18}$). Beryl saturation in granitic melts depends on a number of factors. In peraluminous granitic melts, the solubility of beryl can be defined as a solubility product, i.e. the product of the activities of the mineral-forming components to the power of their stoichiometric coefficients, $a^3_{\text{BeO}} \cdot a_{\text{Al}_2\text{O}_3} \cdot a^6_{\text{SiO}_2}$ (Evensen et al. 1999). In terms of the BeO concentration in the melt, beryl is less soluble in melts with high alumina content, because the solubility product depends on Al_2O_3 activity. Fluxing components, e.g. F, B, Li, and P, can increase beryl solubility, but the effects of these components are poorly understood (Evensen et al. 1999). Temperature is one of the most important parameters controlling beryl solubility. FIGURE 2 shows BeO concentrations for beryl saturation in the presence of quartz in a subaluminous (molar $\text{Al}/[\text{Na}+\text{K}] = 1$), H_2O -saturated granitic melt. This subaluminous melt composition was selected for this figure so that beryl solubility can be directly compared to the solubilities of other rare-element minerals. Extrapolation of the beryl solubility data in this figure to 600 °C indicates that several hundred parts per million Be need to be present in order for the melt to be saturated in beryl. This value is unreasonably

high, as most beryl-rich pegmatites contain no more than 250–420 ppm Be (London and Evensen 2003). However, beryl saturation could occur at as little as 50 ppm Be for a peraluminous melt at a temperature of 400 °C (London 2005).

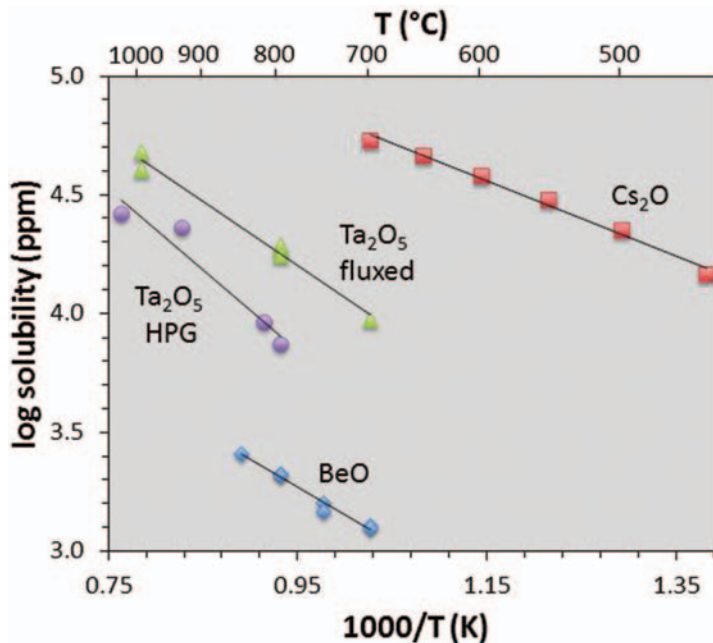


FIGURE 2 Solubility of key rare-element minerals in granitic melts. Data sources are: beryl solubility (BeO data) from Evensen et al. (1999); pollucite solubility (Cs_2O data) from London (2005); tantalite solubility (Ta_2O_5 data) – fluxed, from Bartels et al. 2010 and HPG (haplogranite), from Linnen (1998)

LITHIUM MINERALIZATION

Economic lithium mineralization in pegmatites is dominated by spodumene ($\text{LiAlSi}_2\text{O}_6$), but other important ore minerals are petalite ($\text{LiAlSi}_4\text{O}_{10}$), lepidolite group minerals ($\text{K}[\text{Li},\text{Al}]_3[\text{Si},\text{Al}]_4\text{O}_{10}[\text{F},\text{OH}]_2$), which also can constitute Rb ore, and amblygonite–montebrasite ($\text{LiAlPO}_4[\text{F},\text{OH}]$). Few studies have determined the concentrations required for Li-mineral saturation in granitic melts, but at 100–200 MPa (H_2O saturation) and ~700 °C, petalite is a liquidus phase at approximately 2 wt% Li_2O in the melt (London 2005). The lithium aluminosilicate phase diagram (Fig. 3) has made a considerable impact on our understanding of the crystallization conditions of rare-element pegmatites. Initially, this diagram was used to interpret the pressure of pegmatite crystallization assuming equilibrium conditions; e.g. at 600 °C spodumene crystallizes at high pressure (greater than ~350 MPa), whereas petalite crystallizes at relatively low pressure. Numerical modeling indicates that where pegmatite dikes cool rapidly crystallization may occur at temperatures well below the liquidus (Webber et al. 1999; London 2008). Consequently, certain spodumene-bearing pegmatites are now interpreted as having crystallized at highly undercooled conditions, rather than at higher pressure.

CESIUM MINERALIZATION

The cesium content of beryl can reach weight percent levels, and thousands of parts per million Cs can be present in micas and K-feldspar in highly evolved LCT pegmatites (Černý 1989). However, the principal Cs ore mineral is pollucite [$\text{Cs}(\text{Si}_2\text{Al})\text{O}_6 \cdot n\text{H}_2\text{O}$], which can contain in excess of 30 wt% Cs_2O . To our knowledge, the only deposit where pollucite is mined is the Tanco pegmatite, although it also occurs in many pegmatites worldwide,

notably at Bikita. About 5 wt% Cs₂O is required for pollucite saturation in H₂O-saturated granite melt at 200 MPa and ~680 °C (London 2005). Starting with the bulk continental crust composition of 2 ppm Cs, it seems impossible that any melt could ever undergo such extensive fractional crystallization. However, like beryl, pollucite solubility is strongly dependent on temperature (Fig. 2), and at ~390°C, a granitic melt can be saturated with pollucite at a Cs concentration of ~4700 ppm (London 2005).

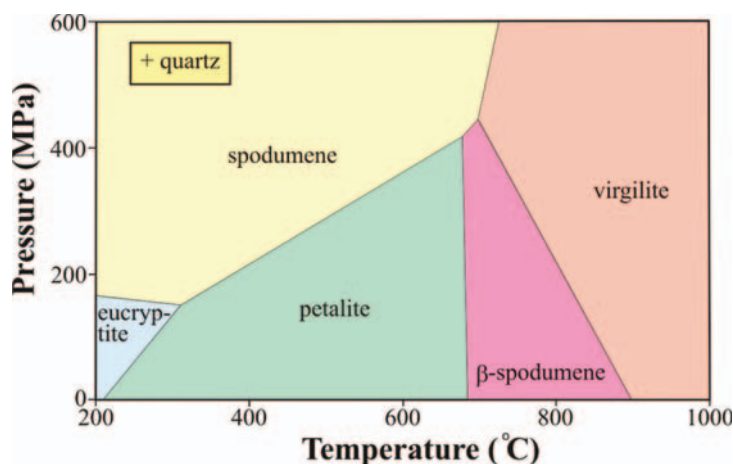


FIGURE 3 Quartz-saturated phase relationships of Li aluminosilicate minerals. MODIFIED FROM LONDON (2005)

TANTALUM MINERALIZATION

There are two dominant styles of tantalum mineralization (Černý 2005; Van Lichtervelde et al. 2007). Magmatic Ta oxides are disseminated or occur in layers in aplite (commonly albite rich; Fig. 4). Tantalite solubility in granitic melts depends strongly on temperature (Fig. 2), on the ratio of alkalis to aluminum in the melt (Linnen and Cuney 2005), and on the abundance of fluxing components in the melt (Bartels et al. 2010). Grades in most Ta deposits are a few hundred parts per million, but the Tanco pegmatite is exceptional, with grades in excess of 1000 ppm Ta (Stilling et al. 2006). The solubility data in Figure 2 indicate either that tantalite-(Mn) (MnTa₂O₆) crystallizes at low temperature (<500°C) or that Ta concentrations in highly evolved melts are much higher than a few hundred parts per million. Low temperatures are plausible because of the high concentrations of fluxing components, and it is possible that the loss of these components and/or alkalis (chemical quenching; London 2008) may trigger tantalite crystallization. Other experiments show that columbite-(Mn) (MnNb₂O₆) has a lower solubility than tantalite-(Mn) (Linnen and Cuney 2005). Consequently, the decrease of Nb/Ta observed at scales ranging from individual columbite group crystals to zoning in pegmatite fields can be explained by fractional crystallization of columbite-group minerals.

The second style of Ta mineralization is associated with late replacement units, which produce rocks composed mostly of albite (Černý 2005; Kontak 2006) or muscovite (Van Lichtervelde et al. 2007). Figure 5 shows replacement of K-feldspar in the Tanco pegmatite by albite veins and by muscovite and associated Ta oxides. Van Lichtervelde et al. (2007) proposed that this metasomatism involved highly fluxed melts rather than hydrothermal fluids, based on the high solubility of Ta in highly fluxed silicate melts, the low

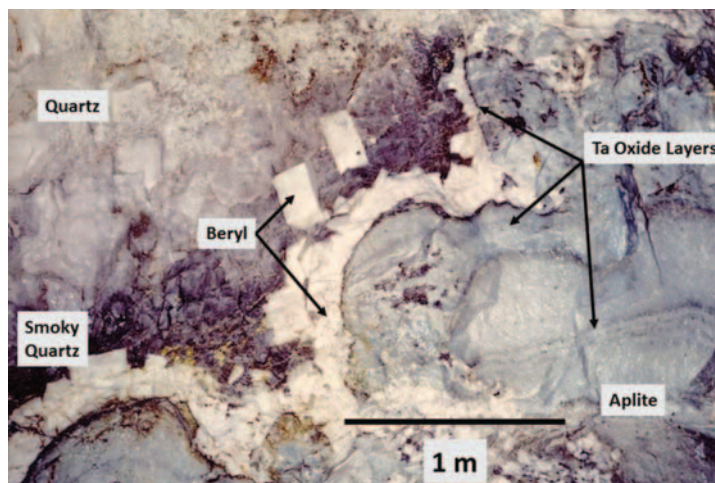


FIGURE 4 Aplitite-hosted Ta mineralization at the Tanco pegmatite. Aplite (bottom) with layers of dark Ta–Nb oxide minerals, separated from quartz by a layer of Li–Cs-rich beryl. Smoky quartz at the contact with beryl is due to the presence of U-bearing microlite. ORIGINAL PHOTO BY A. C. TURNOCK, MODIFIED FROM ČERNÝ (2005)

solubility of Ta in aqueous fluids, and the textures and chemistry of Ta oxide assemblages. Highly fluxed melts are capable of transporting weight percent levels of Ta (Bartels et al. 2010). Such melts possess low viscosities (London 2008) and thus are highly mobile. They may originate as boundary layers that can transport Ta and other HFSEs via a zone refinement process (London and Morgan 2012 this issue). Alternatively, highly fluxed melts may be related to immiscibility (Thomas et al. 2006), but Ta distribution does not appear to be controlled by immiscibility (Borisova et al. 2012). Regardless of the origin of the highly fluxed melts, Ta and other HFSEs are highly soluble in these melts. It is unlikely that temperature (i.e. undercooling; see London and Morgan 2012) is the sole control of mineralization. Chemical quenching, in which the fluxing components are dissipated or removed by crystallization, diffusion, or loss to a vapor phase, may trigger tantalite crystallization. Textural evidence for this process lies in the intimate intergrowth of HFSE minerals (tantalite, zircon, and apatite; Van Lichtervelde et al. 2007).

Although hydrothermal fluids may not play a critical role in the transport of Ta, they may nevertheless be important in controlling mineralization by adding to the melt the divalent cations Fe, Mn, and Ca, which are essential components of columbite–tantalite-(Fe,Mn), wodginite (MnSnTa₂O₈), and microlite ([Ca,Na]₂Ta₂O₆[O,OH,F]). Complex intergrowths of Ta oxide minerals at Tanco (Fig. 6) suggest that the low chemical activities of the divalent cations, rather than the Ta concentration, are the limiting factors for mineral growth (Van Lichtervelde et al. 2007). Tantalum minerals are also intimately intergrown with apatite and zircon, suggesting that the crystallization mechanism for different HFSE minerals may be interrelated (see also Černý et al. 2007). The role of fluids is still a matter of debate. London (1986) documents microlite as a daughter mineral in crystal-rich fluid inclusions at Tanco, and Thomas et al. (2011) suggest that immiscible fluids may have been critical for Ta–Nb transport in the Borborema pegmatite province, Brazil.

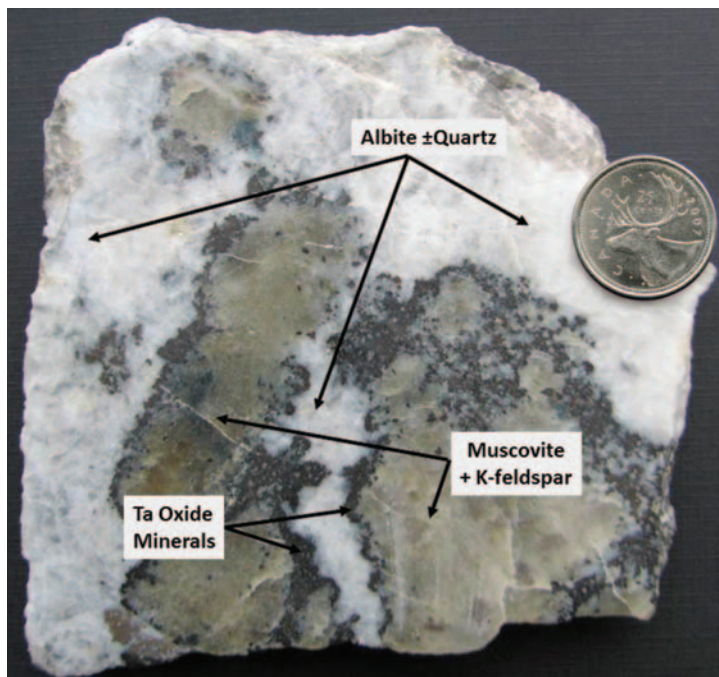


FIGURE 5 Metasomatic Ta mineralization in the Tanco pegmatite. Grey K-feldspar is in part replaced by green muscovite and veined by white albite ± pale grey quartz, with metallic black Ta oxide minerals deposited along the contact between the two assemblages. The coin is 2.4 cm in diameter. PHOTO BY R. LINNEN

OTHER STRATEGIC METALS IN PEGMATITES

Pegmatites of the NYF family are enriched in Nb, Y, and F (along with Be, REEs, Sc, Ti, Zr, Th, and U), and they are commonly derived from A-type and I-type granites (Černý and Ercit 2005). These pegmatites have generally not been exploited for this suite of rare elements, although they have been mined for other industrial minerals (e.g. feldspar). The deposit at Strange Lake, Canada, is a notable exception (Salvi and Williams-Jones 2005). The REE–Zr–Nb mineralization is hosted by a strongly altered pegmatite. There is little doubt that rare elements were concentrated by igneous processes, but the close relationship between mineralization and alteration indicates that rare elements were enriched by hydrothermal fluids (Salvi and Williams-Jones 2005). Uranium is another strategic metal that is mined from abyssal-class pegmatites. The Rössing deposit, Namibia, contains disseminated Th-bearing uraninite in leucocratic (low-iron) dikes of “alaskite” granite and pegmatite (Cuney and Kyser 2008). Grades in this deposit are very low, ~300 ppm U, but at the end of 2007, over 90,000 tonnes of U had been produced. Other past-producers of uranium from abyssal-class granite pegmatites include the Bancroft area of Ontario (see references in Cuney and Kyser 2008).

RESEARCH IN A STATE OF FLUX

Strategic metals in pegmatites are closely linked to highly fluxed silicate melts. There is strong evidence that much of the rare-element mineralization is magmatic in origin, but our understanding of the roles of the different fluxing compounds, both individually and in mixtures, remains limited. Van Lichtervelde et al. (2010) proposed that there are differences in the solution behavior of each HFSE in melts: Ta has an affinity for Al in melts, whereas Zr tends to form an alkali zirconosilicate species. Fluxing components, in particular F and P, can also form Al or alkali species, depending on the bulk-melt composition. Consequently, there is a complicated competition for Al and alkalis among

both HFSEs and fluxing components (also see London 2008). Few experimental studies have been carried out for weakly alkaline granitic melts. Mineralization in the NYF family of pegmatites, and the role of fluorine in that process, remains poorly understood.

The summary above advocates magmatic as well as magmatic–metasomatic styles of mineralization, but there is little doubt that hydrothermal metasomatism also occurs in rare-element pegmatites, particularly in NYF pegmatites such as the Strange Lake deposit. The precise role that hydrothermal fluids play in concentrating strategic metals has yet to be resolved. However, geological relationships suggest that F, B, and P may be important for rare-element transport in aqueous fluids, but only a few experiments have tested this hypothesis. The role of F, B, and P in the transport of rare elements in hydrothermal fluids remains poorly understood, and the different effects that these elements have in peralkaline, metaluminous, and peraluminous systems similarly are not well constrained. On one hand, more experiments are needed to explain different features of strategic-metal mineralization in pegmatites. On the other hand, pegmatites are natural laboratories that record the different processes and provide critical constraints on the behavior of this group of metals in the Earth’s crust.

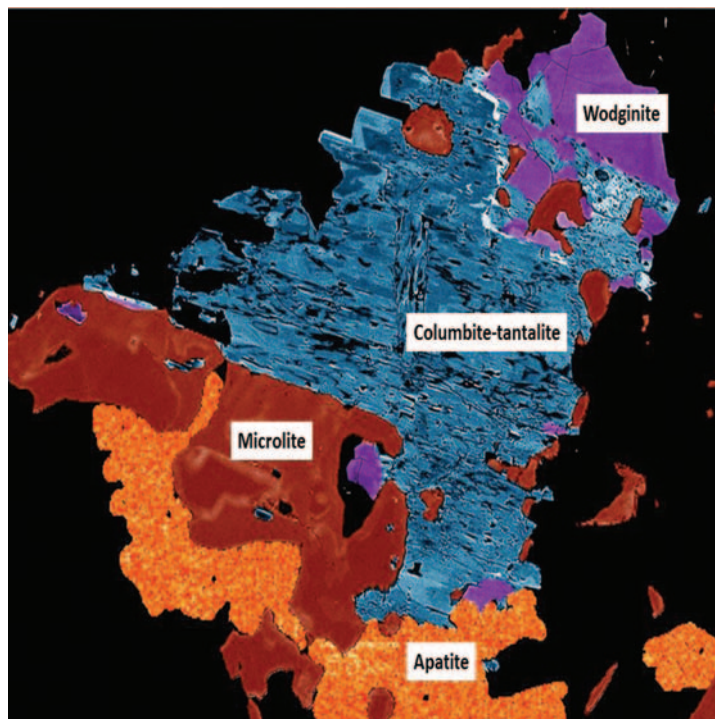


FIGURE 6 Backscattered electron false-color image of complex Ta-mineral associations from metasomatic-style mineralization in the Tanco pegmatite. Orange represents a combination of Ca + P (apatite), brown is Na + Ca + Ta (microlite), blue is Fe + Mn + Ta + Nb (columbite–tantalite), and purple represents Sn + Ta (wodginite). Field of view is 500 μm across. MODIFIED AFTER VAN LICHTERVELDE ET AL. (2007)

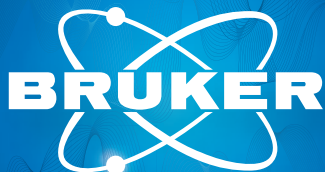
ACKNOWLEDGMENTS

Karen Ferreira was instrumental in helping to put this manuscript together, and her efforts are greatly appreciated. Many colleagues and students have contributed ideas over the years, but in particular we thank David London and Dan Kontak for numerous discussions and also for the opportunity to present our ideas in *Elements*. We are also grateful to three anonymous reviewers for their comments.

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