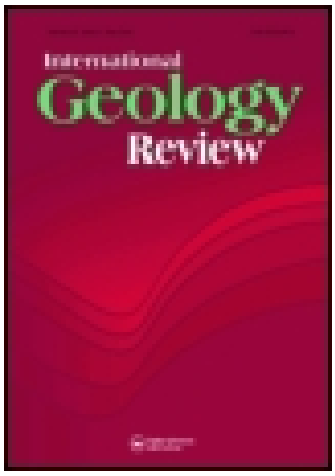
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Origin of peralkaline granites of the Jurassic Bokan Mountain complex (southeastern Alaska) hosting rare metal mineralization

Jaroslav Dostal[a] & J. Gregory Shellnutt[b]
[a] Department of Geology, Saint Mary's University, Halifax, Canada
[b] Department of Earth Sciences, National Taiwan Normal University, Taipei, Taiwan
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Origin of peralkaline granites of the Jurassic Bokan Mountain complex (southeastern Alaska) hosting rare metal mineralization

Jaroslav Dostal[a] and J. Gregory Shellnutt[b]

[a]Department of Geology, Saint Mary's University, Halifax, Canada; [b]Department of Earth Sciences, National Taiwan Normal University, Taipei, Taiwan

ABSTRACT

The Jurassic Bokan Mountain complex (BMC), composed of arfvedsonite and/or aegirine-bearing peralkaline A-type granitic rocks, is a circular body about 3 km in diameter located in southeastern Alaska. Like many other highly fractionated granitic bodies, the BMC granites were affected by late magmatic or post-magmatic processes, which, however, did not modify the contents of major elements. The granitic rocks are distinctly enriched in high-field-strength elements (HFSEs), rare earth elements (REEs), Y, Th, and U but depleted in Ba, Sr, and Eu and have high positive $\varepsilon_{Nd(T)}$ values. Unlike the variations in the major elements, Sr and Ba, which can be accounted by fractional crystallization, the abundance of REE, Y, HFSE, U, and Th (the elements which are hosted in accessory phases) were modified by F-rich hydrothermal fluids. The BMC hosts significant rare metal mineralization related to the late-stage crystallization history of the complex involving late magmatic and/or post-magmatic fluids. The mineralization includes two types: (1) a U–Th deposit which was exploited at the former Ross-Adams mine and (2) REE and Y mineralization mostly hosted in felsic dikes. Thermodynamic modelling of granites and spatially associated mafic rocks using the programme Rhyolite-MELTS implies that the granites can be derived from the mafic rocks by fractional crystallization. It is suggested that such a process (i.e. derivation of peralkaline granitic magma from the alkali or transitional basaltic magmas derived by partial melting from a lithospheric source metasomatically enriched in rare metals) can be invoked for other peralkaline granitic rocks hosting rare metal deposits.

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Introduction

Alkaline/peralkaline felsic igneous rocks have attracted a large amount of attention in the geological literature although they constitute only a relatively small volume of Earth's crust. One of the reasons for this interest is the distinct enrichment of these rocks in rare metals, particularly in rare earth elements (REEs), Y, high-field-strength elements (HFSEs), and radioactive elements (Th and U), which in some cases is of economic significance (e.g. Thompson 1988; Salvi and Williams-Jones 1990, 2005; Richardson and Birkett 1996; Cerny *et al*. 2005; Küster 2009; Verplanck *et al*. 2013; Linnen *et al*. 2014). These rocks also have distinctive mineralogical, geochemical, and petrological characteristics which are of ongoing interest (e.g. Salvi and Williams-Jones 2005; Chakhmouradian and Zaitsev 2012; Sheard *et al*. 2012; Walters *et al*. 2013; Linnen *et al*. 2014). Some, including nepheline syenites and syenites, are commonly considered to be formed by extensive low-pressure fractionation from a nephelinitic, basanitic, or basaltic parent (e.g. Platt 1996; Winter 2001). However, there is still debate on the origin of the peralkaline silica-oversaturated felsic rocks including A-type granites (e.g. Whalen *et al*. 1987; Eby 1990, 1992; Mungall and Martin 1996; Martin and De Vito 2005; Salvi and Williams-Jones 2005; Bonin 2007; Frost and Frost 2011; Chakhmouradian and Zaitsev 2012). Several petrogenetic models for A-type granites have been proposed, including: (1) crustal contamination of mantle-derived silica-undersaturated alkaline magmas (e.g. Marks and Markl 2001), (2) extensive fractional crystallization from mantle-derived mafic magmas derived by melting of enriched lithospheric mantle (e.g. Turner *et al*. 1992; Mungall and Martin 1996; Upton *et al*. 2003; Whitaker *et al*. 2008; Shellnutt *et al*. 2011b), and (3) partial melting of middle to lower crustal source rocks including underplated basalts (e.g. Eby 1992; Bonin 2007; Shellnutt and Zhou 2008; Shellnutt *et al*. 2011a; Shellnutt and Iizuka 2012). To contribute to the debate on the origin of these rocks, we investigated the Jurassic Bokan Mountain peralkaline granitic complex which hosts an important mineralization of rare metals

CONTACT Jaroslav Dostal jdostal@smu.ca
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including REE, U, and Th. The mineralization is related to the late stages of the evolution of magma. The pluton displays structural and compositional characteristics typical of anorogenic granitic complexes worldwide (e.g. Eby 1990, 1992; Clarke 1992; Salvi and Williams-Jones 2005; Bonin 2007; Frost and Frost 2011; Dostal *et al*. 2014). The purpose of this article is to present geochemical data on the BMC located near the southern tip of Prince of Wales Island (the southernmost island of Alaska, USA) in order to (1) evaluate a potential relationship between the peralkaline granites and spatially associated mafic dikes and to (2) constrain the origin of the granitic magma. We infer that the peralkaline granite pluton was derived from mantle-derived mafic magma by extensive fractional crystallization.

Geological setting

The Jurassic Bokan Mountain complex (BMC), composed of peralkaline granitic rocks, is a circular body about 3 km in diameter that intruded Palaeozoic rocks of the Alexander terrane of the North American Cordillera (Figure 1). The terrane was accreted to the western margin of North America during Mesozoic times, well after the emplacement of the BMC dated at 177 ± 1 Ma (U–Pb zircon age; Dostal *et al*. 2013). The basement in the area is composed of Neoproterozoic and Palaeozoic metasedimentary and arc-related metavolcanic rocks and associated intrusions (Gehrels and Saleeby 1987a, 1987b; Gehrels 1992; Dostal *et al*. 2013). Mesozoic rocks in this part of the terrane are relatively rare, dominantly plutonic, and include the Early Jurassic BMC, the Dora Bay pluton (DBP), and mafic and felsic dikes. The contacts of the BMC are generally steep and sharp, but the country rocks are frequently albitized. In the contact aureole of the BMC, the slate was converted to andalusite (chiastolite)-bearing hornfels, suggesting an epizonal emplacement of the intrusion into an early Palaeozoic oceanic arc complex of the Alexander terrane.

The BMC contains two major units, a dominant core, composed mainly of arfvedsonite granite, and a rim ~180 m thick, predominantly made up of aegirine granite (Figure 1). The boundary between these two types is usually transitional over about 15 m. The granites are leucocratic with about 2–10 vol.% of mafic minerals (aegirine and arfvedsonite in various proportions). The granites are mainly composed of quartz (~35–45 vol.%) and K-feldspar (microperthite and microcline) (35–50 vol.%) with albite (An < 10) as both a separate phase and a replacement of K-feldspar (5–15 vol.%). Albite pervasively replaces K-feldspar, both as a phenocryst and in the groundmass. Plagioclase with a primary magmatic composition (An ~20) is only rarely preserved. All the rock-forming minerals record a two-stage growth history (Dostal *et al*. 2014): magmatic and fluid-mediated late magmatic. The prevalent textures are primary porphyritic and late protoclastic. The porphyritic texture is defined by phenocrysts of subhedral to euhedral quartz and K-feldspar enclosed in quartzo-feldspathic groundmass with abundant albitic feldspar. The protoclastic texture shows broken or granulated phenocrysts of quartz and feldspars; this texture is particularly well developed within shear zones.

The mafic minerals were the last rock-forming minerals to crystalize from the magma. Aegirine and arfvedsonite are spatially associated with one another but in many samples only one or the other is present. Arfvedsonite ($Fe^{2+}/[Fe^{2+} + Mg] > 0.99$) is distinctly pleochroic (lilac to blue) and forms euhedral prismatic crystals typically 1–4 mm and, rarely, >1 cm in size. The crystals are interstitial and may have fretted outlines indicating continued growth into the matrix. Aegirine ($Jd_{13\text{-}19}Ae_{81\text{-}86}Di_{<1}$; $Fe/[Fe + Mg] > 0.99$) has a range of shapes from equant to prismatic or skeletal. The mafic minerals typically record a two-stage growth history. The first-stage phases are inclusion-rich cores whereas the second stage represents an inclusion-free overgrowth. Accessory minerals including zircon, monazite, xenotime, fluorite, and Fe–Ti oxides are relatively abundant. More than 20 REE-, Y, HFSE-, and U–Th-rich minerals are known from the BMC (e.g. Staatz 1978; Philpotts *et al*. 1998; Dostal *et al*. 2011, 2014). Many of these accessory minerals occur in two or more generations. They also form intergrowths, pseudomorphs, cavity fillings, and mineral rims.

The BMC hosts significant rare metal mineralization related to the late-stage crystallization history of the complex which involved hydrothermal fluids. The mineralization includes two types. The first type is confined to en echelon veins in a shear zone within the BMC and is represented by the Ross-Adams U–Th deposit (Figure 1), which was mined intermittently between 1957 and 1971. The mine produced about 77,000 tons of high-grade uranium ore that averaged about 1 wt.% U_3O_8 and nearly 3 wt.% ThO_2 (Long *et al*. 2010). The second type is the REE and Y mineralization in felsic, aplitic, and pegmatitic dikes (e.g. Dotson, I&L, Geiger, Boots, Geoduck, Cheri, Figure 1) that occur within or adjacent to the BMC (Philpotts *et al*. 1998; Long *et al*. 2010; Dostal *et al*. 2011, 2013, 2014; Hatch 2014). Steeply dipping mineralized dikes extend up to 5 km northwest and southeast from the BMC (Figure 1). The most prominent is the Dotson dike system, which consists of interwoven mm- to

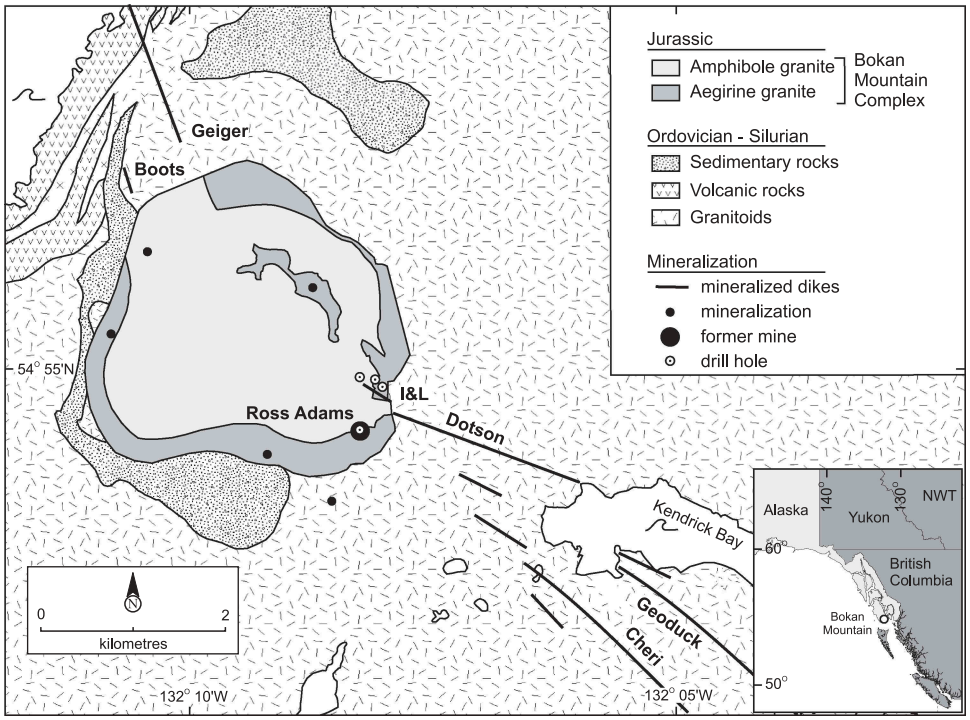


Figure 1. Geological sketch map of the Bokan Mountain intrusive complex and surrounding area showing the location of the former Ross-Adams mine, mineralized dikes, mineralized showings, and drill holes (modified after MacKevett 1963; Thompson *et al*. 1982; Gehrels 1992; Dostal *et al*. 2013). The inset shows the location of the Bokan Mountain complex.

Notes: The analysed samples are from the drill holes. Details of sample locations are given in Supplementary File 1.

m-scale dikes in a zone of 50 m wide. Dikes have chilled margins, but mineralization permeates the country rocks up to 5 m.

Mafic dikes are widespread throughout the area; they are up to ~10 m thick and steeply dipping. MacKevett (1963) and Gehrels and Saleeby (1987a) recognized more than one generation of dikes. Thompson *et al*. (1982) documented that many of these dikes in an area around the former Ross-Adams mine (Figure 1) are approximately coeval with the emplacement of the pluton. These dikes are aphanitic or porphyritic; porphyritic types typically contain phenocrysts of amphibole, clinopyroxene, and/or plagioclase enclosed in an aphanitic groundmass. Some of these dikes were classified as lamprophyres (MacKevett 1963; Thompson *et al*. 1982) and may also contain biotite.

In addition to the BMC, southern Prince of Wales Island hosts another Jurassic peralkaline granitic intrusion – the DBP, a circular body ~4.5 km in diameter, which also contains rare metal mineralization (Barker 1990; Drinkwater and Calzia 1994). This intrusion which is located about 30 km north of the BMC yielded a K–Ar hornblende age of 175 ± 5 Ma (Brew 1996) and a U–Pb zircon age of 182.2 ± 5.5 Ma (Bala *et al*. 2014). Both the BMC and DBP were emplaced during regional but localized rifting event (Dostal *et al*. 2013).

Analytical methods

Samples were collected in a wider vicinity of the former Ross-Adams mine (Figure 1) from drill cores. The sample locations are given online (Supplementary File 1; see http://dx.doi.org/10.1080/00206814.2015.1052995 for supplementary files). Major and trace elements of whole rocks were determined using lithium tetraborate fusion at the Activation Laboratories, Ancaster, Ontario, Canada. The analyses of major elements were done by inductively coupled plasma-optical emission spectrometry, whereas trace element analyses were performed using a Perkin Elmer Optima 3000 inductively coupled plasma mass spectrometer. Based on replicate analyses, the precision is generally between 2% and 10% for trace elements and 3–5% for major elements (Supplementary Files 2a and b).

Two lamprophyres were selected for Nd isotopic analyses (Supplementary File 3). Samarium and Nd abundance and Nd isotopic ratios were determined by isotope dilution mass spectrometry in the laboratory of the Department of Earth Sciences at the Memorial University of Newfoundland (St John's, Newfoundland, Canada). The precision of concentrations of Nd and Sm is ±1%. The procedure for the Sm–Nd isotopic analyses was described by Kerr *et al*. (1995). Replicate analyses of JNdi-1 standard yielded $^{143}Nd/^{144}Nd$ = 0.512137 ± 1

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(n = 112). The ε_{Nd} values were calculated assuming t = 177 Ma. Nd model ages (Supplementary File 3) were calculated based on De Paolo's (1988) depleted mantle model (T_{DM1}), and on a depleted mantle model (T_{DM2}) separated from the chondrite-uniform reservoir (CHUR) at 4.55 Ga, with linear evolution and a present-day epsilon value of +10.

Geochemistry

On the mesonormative quartz–alkali feldspar–plagioclase (QAP) diagram (Figure 2) of LeMaitre *et al.* (1989), the Bokan rocks fall into the field of granite, well away from an average of 383 North American granitoid rocks (Chayes 1985) and the average composition of the upper continental crust (Taylor and McLennan 1985). The granitic rocks of the BMC are highly siliceous with SiO_2 content ranging from 72 to 77 wt.% (Supplementary File 2a). They have low abundance of Al_2O_3, CaO, and MgO but high contents of alkalis and FeO^{tot} as well as the high FeO^{tot}/MgO ratio. The rocks have molar $Al_2O_3/(Na_2O + K_2O)$ ratios <1 (Supplementary File 2a) indicative of their peralkaline composition and consistent with the presence of alkali mafic minerals (aegirine and arfvedsonite) in the mode. On de la Roche *et al.* (1980)'s multicationic diagram (Figure 3), the rocks plot in the field of alkaline granites.

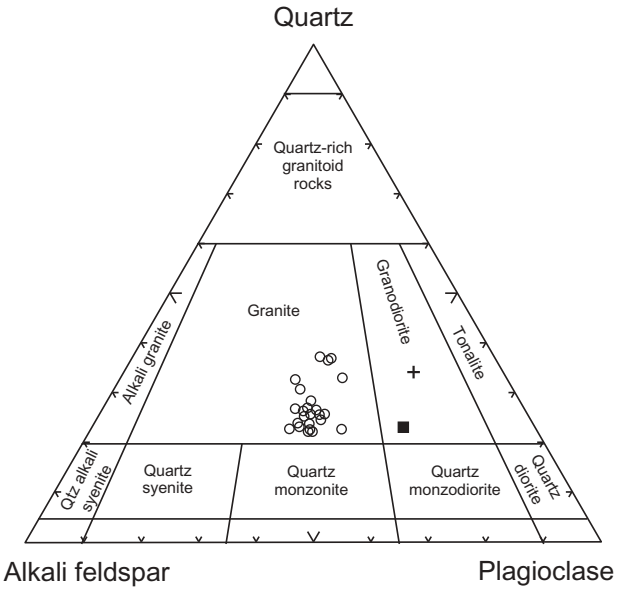


Figure 2. Mesonormative quartz–alkali feldspar–plagioclase (QAP) ternary classification diagram used to characterize the modal composition of the plutonic rocks (Le Maitre *et al.* 1989). The plot illustrates the composition of the BMC granitic rocks (circles). The average of 363 North American granitoid rocks (cross; Chayes 1985) and the average composition (solid square) of the upper continental crust (Taylor and McLennan 1985) are shown for comparison.

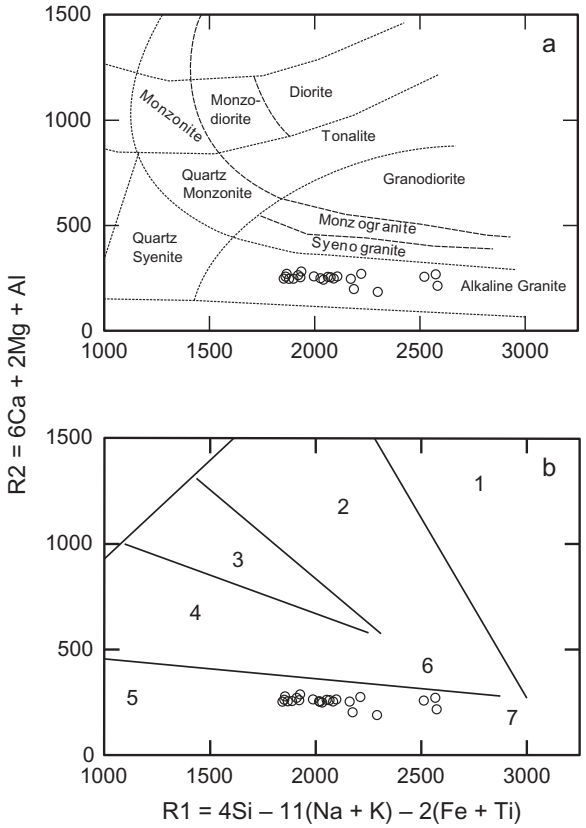


Figure 3. Multicationic classification plot of de La Roche *et al.* (1980) for plutonic rocks of BMC (circles) showing (a) the compositional/classification fields of common plutonic rocks and (b) petrogenetic/geodynamic fields for granitoid rocks (Batchelor and Bowden 1985): 1, mantle fractionates; 2, pre-plate collision; 3, post-collision uplift; 4, late-orogenic; 5, anorogenic; 6, syn-collision; 7, post-orogenic.

In general, the major element composition of the BMC granites is similar to other peralkaline A-type granites (e.g. Frost *et al.* 2001; Frost and Frost 2011). Their fluorine content is high and variable (200–3200 ppm; Thompson *et al.* 1980, 1982; Cuney and Kyser 2008) and is reflected in the widespread occurrence of fluorite, a common late-stage phase. These fluorine concentrations are probably minimum values due to the tendency of shallow-seated granitic melts to *exsolve* an aqueous halogen-rich fluid. Thompson *et al.* (1982) showed that the normative compositions of the granitic rocks plot close to the thermal minima of the normative albite–quartz–orthoclase diagrams.

Like typical peralkaline granites (e.g. Whalen *et al.* 1987; Eby 1990, 1992; Linnen and Cuney 2005; Salvi and Williams-Jones 2005; Bonin 2007; Chakhmouradian and Zaitsev 2012; Sheard *et al.* 2012), the Bokan rocks exhibit distinct enrichment in HFSE, REE, Y, Rb, Th, and U, but depletion in Ti, Ba, Sr, and Eu on N-type mid-ocean ridge basalt (N-MORB)-normalized plots (Figure 4). The Bokan granites have high but fluctuating abundances of REE,

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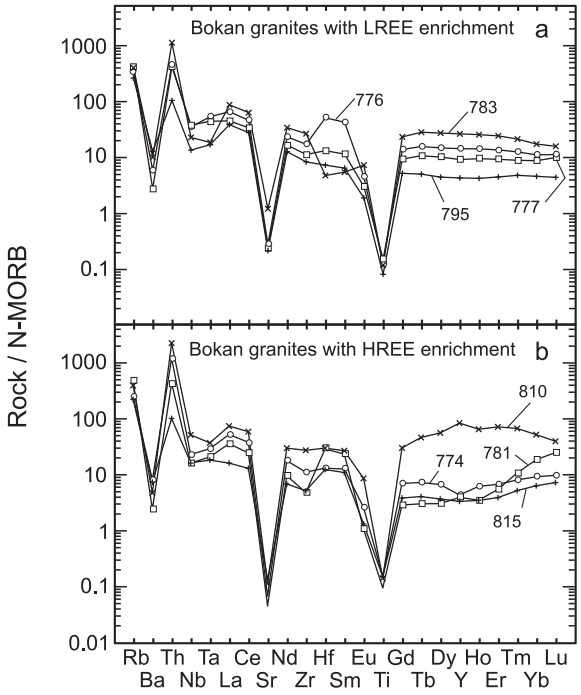


Figure 4. N-type MORB-normalized incompatible trace element patterns of the granitic rocks of the BMC. (a) Granites with a light REE enrichment and relatively flat heavy REE pattern; (b) granites with HREE enrichment. Normalizing values are after Sun and McDonough (1989).

particularly of heavy REE (HREE). The REE patterns of the granites from the Ross-Adams area, typical of the whole pluton (Dostal *et al*. 2014), are highly variable with $(La/Yb)_n$ ranging from 0.84 to 7 and $(Gd/Yb)_n$ in the range of ~0.15–1.5. This is also reflected in the shape of the REE patterns where the HREE segment shows both negative and positive slopes (Figure 5). All the patterns have a distinct negative Eu anomaly which does not correlate with the variations of Ba and Sr (Figures 4 and 6(b)). The shape of the chondrite-normalized pattern of the average REE mineralization of the Bokan complex (Dotson zone; Hatch 2014; Figure 5(c)) is similar to those of many BMC granites although the ore has higher absolute concentrations (Figures 5 and 6). The pattern of the BMC mineralization resembles those of other REE ores hosted in peralkaline granites (Figure 5(c)), including the Strange Lake (Quebec-Labrador, Canada; Miller 1986; Salvi and Williams-Jones 1990, 2005; Kerr 2011; Kerr and Rafuse 2012) and Khaldzan-Buregtey (western Mongolia; Kovalenko *et al*. 1995), suggesting they all have a similar origin.

The mafic and intermediate dikes, which seem to be roughly contemporaneous with the Bokan intrusion, have SiO_2 ranging from ~47% to 57 wt.% (Supplementary File 2b). The rocks correspond to subalkaline basalts

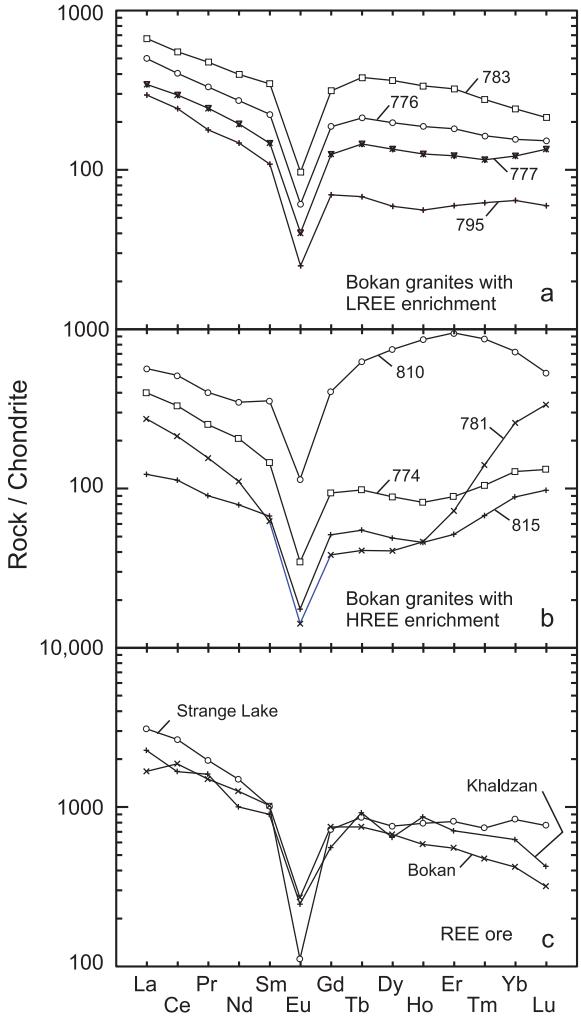


Figure 5. Chondrite-normalized REE abundances in the granitic rocks of the BMC. (a) Granites with a light REE enrichment and relatively flat HREE pattern; (b) granites enriched in HREE; (c) REE mineralization/ore in peralkaline granites: Strange Lake (Quebec-Labrador, Canada; Hatch 2014), Khaldzan-Buregtey (western Mongolia; Kovalenko *et al*. 1995), and Bokan (Hatch 2014). Normalizing values are after Sun and McDonough (1989).

and andesites; they include the lamprophyres of Thompson *et al*. (1982). The dikes have highly variable abundances of rare metal elements including HFSE and REE with the abundances of La and Yb between 9 and 18 ppm and between 1.9 and 20 ppm, respectively (Supplementary File 2b). The $(La/Yb)_n$ and $(Gd/Yb)_n$ ratios range from 0.5 to 5 and from 1 to 1.6, respectively.

Age-corrected (177 Ma) ε_{Nd} values for the granitic rocks of the Bokan complex are high and positive but highly variable (+2.7–+8.6; Philpotts *et al*. 1998; Dostal *et al*. 2014). The ε_{Nd} values of the mafic dikes (~+6.5; Supplementary File 3) are also high and positive, within the range of the granites (Figure 7).

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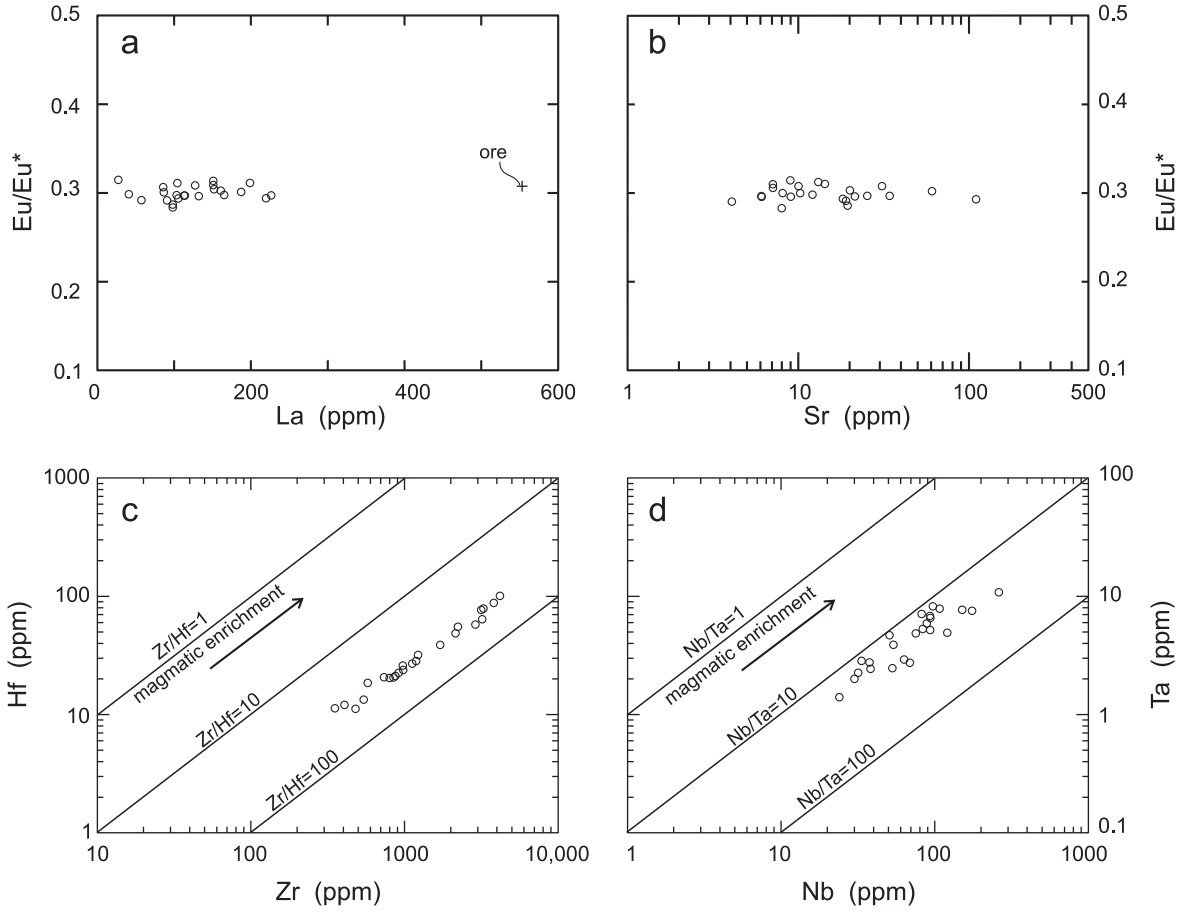


Figure 6. Variations of (a) La (ppm) *versus* (Eu/Eu*), (b) Sr (ppm) *versus* (Eu/Eu*), (c) Zr *versus* Hf, and (d) Nb *versus* Ta (ppm) in granitic rocks of the BMC (circles). Cross in (a) denotes the average ore of the BMC (Hatch 2014). A vector of magmatic enrichment is shown.

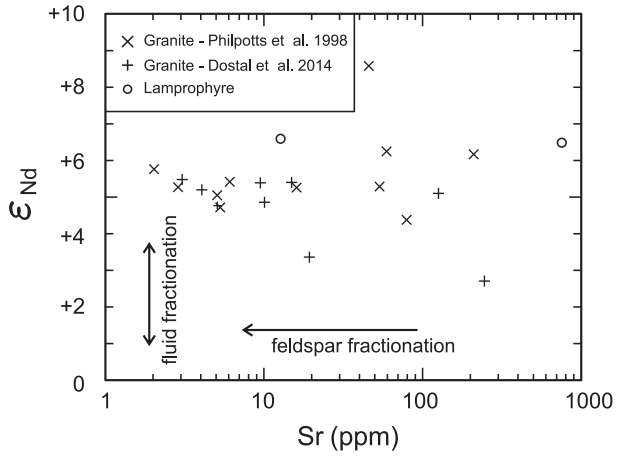


Figure 7. $\varepsilon_{Nd(T)}$ *versus* Sr (ppm) plot for the granites and lamprophyre dikes of the Bokan Mountain complex. Isotopic data for the lamprophyres are given in Supplementary File 3, whereas the data for the granites are from Philpotts *et al.* (1998) and Dostal *et al.* (2014). Vectors for fluid and feldspar fractionation are shown.

Discussion

Petrogenesis

The BMC granites are peralkaline within-plate A-type granitoids (Figure 8). Their major element composition is characteristic of post-orogenic to anorogenic rocks (Figure 3(b)). Like many highly fractionated granitic bodies, these granites were affected by the magmatic or post-magmatic processes. However, the contents of major elements in most granitic rocks of the pluton did not significantly change during these secondary processes although primary minerals could have been partially recrystallized (Dostal *et al.* 2014). This is supported by (1) the similarities of the major element composition of the Bokan pluton and other peralkaline granitic intrusions, (2) variation trends of major elements when plotted on the multielement diagram (Figure 3) where the rocks plot into the alkaline granite field, and (3) the position of the Bokan granites on the normative

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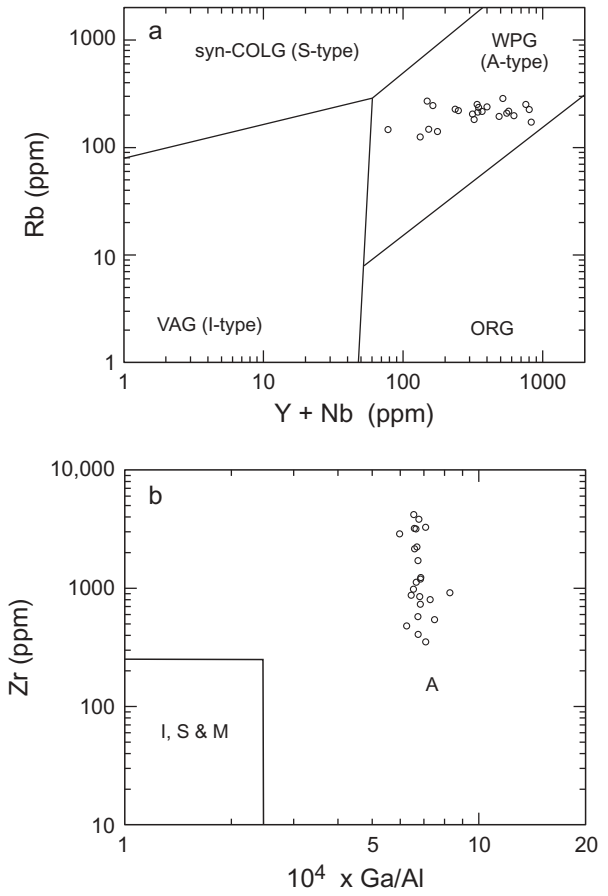


Figure 8. (a) (Y + Nb) *versus* Rb (ppm) geotectonic discrimination diagram of Pearce *et al*. (1984) for the BMC granites (circles). Fields: syn-COLG (S-type granite), syncollisional granite; VAG (I-type granite), volcanic arc granite; WPG (A-type granite), within-plate granite; ORG, ocean-ridge granite. (b) 10,000*Ga/Al *versus* Zr (ppm) discrimination diagram of Whalen *et al*. (1987) showing that the BMC granites can be classified as A-type granites. Fields: IS&M field indicates the distribution of I-, S-, and M-type granites while the A field is for the A-type granites.

albite–quartz–orthoclase plots (Thompson *et al*. 1982), where they plot close to ternary minima. However, HFSE, REE, Y, Th, and U, which are typically hosted in accessory phases, cannot be readily explained by fractional crystallization using published mineral-melt partition coefficient values for these elements in highly silicic rocks. The distinct enrichment and large variations of the REE, HFSE, Th, and U in the highly evolved granitic rocks with relatively uniform major element composition can be attributed to the interaction of melts with the coexisting fluids (e.g. Keppler and Wyllie 1990; Charoy and Raimbault 1994; Monecke *et al*. 2002; Dostal *et al*. 2004; Salvi and Williams-Jones 2005; Agangi *et al*. 2010). In the Bokan granites, this is also consistent with the textural features and compositions of the accessory minerals.

Highly positive ε_{Nd} values of the granitic rocks of the BMC (Figure 7) indicate that the rocks were derived from a reservoir with a history of light rare earth element depletion, such as depleted upper mantle, and were not affected by contamination by ancient continental crusts. However, Dostal *et al*. (2014) suggested that the large variations of ε_{Nd} values in the relatively homogeneous complex is evidence that during the late-stage evolution of the complex, F-rich fluids remobilized REE, leading to a modification of the age-corrected Nd isotopic values. The spread of ε_{Nd} values does not correlate with various petrogenetic parameters, including the concentration of Sr (Figure 7), which are related to feldspar fractionation, but it is consistent with the remobilization of REE.

The fluid phases associated with highly evolved granitic systems can be rich in halogens (F and Cl; London *et al*. 1988; London 1992; Thomas *et al*. 2005; Dolejs and Baker 2007), which have a great effect on the complexing and mobility of rare metals such as HFSE, REE, and Y and play an important role in natural systems (e.g. Manning 1981; Dostal and Chatterjee 1995, 2000, 2010; Haas *et al*. 1995; Williams-Jones *et al*. 2000; Webster *et al*. 2004; Salvi and Williams-Jones 2005; Schönenberger *et al*. 2008; Agangi *et al*. 2010). The widespread occurrence of fluorite in the BMC indicates that the fluids had an F-rich composition. It has been generally assumed that the REEs are transported as fluoride complexes (e.g. Smith and Henderson 2000; Williams-Jones *et al*. 2000) although Migdisov and Williams-Jones (2014) recently questioned this process.

Dostal *et al*. (2014) argued that the hydrothermal fluids are related to the last stages of the evolution of the magma. Such an origin is also consistent with the correlations of Zr with Hf and Nb with Ta (Figure 6), which, as suggested by the texture and nature of the occurrences of accessory minerals, are mainly controlled by fluids. The absence of significant negative Nb and Ta anomalies relative to N-MORB (Figure 4) is probably also due to the late fluid enrichment. The variation of Sr does not correlate with Eu (0.7–10 ppm) and Eu/Eu* (Figure 6), which were likely modified by fluids. The significant variations of rare metals in the mafic rocks (Supplementary File 2b) are also assumed to be related to the fluid modifications. However, like in the granitic rocks, the major elements of the mafic rocks were not noticeably modified by fluids.

The abundances of REE, Y, HFSE, U, and Th as well as Nd isotopic values of the pluton and dikes were modified by hydrothermal halogen-rich fluids, and thus they cannot be used to evaluate the potential petrogenetic relationship between the granites and mafic dikes. However, the major elements mainly hosted by

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rock-forming minerals appear to escape modification and their limited variations can be accounted for by feldspar-dominated fractional crystallization. This process is also supported by distinct negative anomalies of Sr and Ba on the N-MORB-normalized plots (Figure 4), while the depletion of Ti is probably a result of the fractionation of Fe–Ti oxides or titanite.

Thermodynamic modelling parameters and results

In order to evaluate the petrogenetic relationship between the granites and mafic magmas, we have applied the thermodynamically modelled major element composition of granitic rocks assuming that associated mafic dikes (Figure 1, Supplementary Files 1 and 2a, b) are parental. The thermodynamic modelling of mafic and felsic rocks can be performed using the programme Rhyolite-MELTS (Gualda *et al*. 2012). The programme is calibrated to rocks within the SiO_2–TiO_2–Al_2O_3–Fe_2O_3–Cr_2O_3–FeO–MnO–MgO–CaO–Na_2O–K_2O–P_2O_5–H_2O compositional range. The software is specifically optimized for silicic systems and enables the user to adjust parameters such as relative oxidation state (fO_2), pressure (bars), and water (wt.%) content of the composition that is being modelled in order to constrain the possible magmatic conditions and processes (i.e. fractional crystallization or partial melting) of the system being modelled.

Fractional crystallization modelling was carried out for the Bokan granitic intrusion using a starting composition equal to the syn-plutonic lamprophyric dikes that are found in the area. We selected a starting composition that is approximately equal to the average composition of the lamprophyres (Supplementary File 2b). The initial pressure used for the modelling is 0.1 GPa (i.e. 1 kbar) which is based on the likely depth of emplacement of the granitic rocks (MacKevett 1963). The relative oxidation state we selected is within the range (i.e. fayalite-magnetite-quartz (FMQ) buffer ± 2) of the mantle (Frost and McCammon 2008) and initial water content is typical of lamprophyric rocks (Righter and Carmichael 1996).

The model used sample 828 as the starting composition with a water content of 3 wt.% and a relative oxidation state equal to the FMQ buffer (i.e. FMQ 0). The results (solid line) are shown in Figure 9 and indicate that the starting composition, under the magmatic conditions outlined in Supplementary File 4, can produce granitic compositions that are reported from the Bokan system (Thompson *et al*. 1982). The model calculates that plagioclase (An_{85}) will crystallize first at a temperature of 1080°C followed by clinopyroxene ($Wo_{48}En_{39}Fs_{13}$) at a temperature of 1045°C, Ti-rich spinel (i.e. titanomagnetite) at 1005°C, and orthopyroxene (Mg# = 7) at 995°C. Clinopyroxene ($Wo_{34}En_{20}Fs_{46}$) stops crystallizing after the temperature drops below 995°C, whereas all other minerals (e.g. plagioclase = An_{75}, orthopyroxene, spinel) continue to crystallize until the residual liquid composition is similar to the Bokan granites at ~870°C which represents ~35% of the starting material (i.e. ~65% has crystallized). The mineral proportions and compositions are outlined in Supplementary File 5 and the complete modelling results are available online (Supplementary File 6).

The calculations show that there is good agreement between the modelled liquid evolution curves and the bulk composition of the granitic rocks from Bokan. However, the CaO content and the P_2O_5 content are consistently too high in the models. The likely explanation for the discrepancy between the model curves and the data is related to phosphate mineral (i.e. apatite, xenotime, monazite) and fluorite fractionation. The effect of F-rich fluids is well documented in the Bokan system (e.g. Dostal *et al*. 2014) and would likely be a factor also in alkali enrichment in the magma and the crystallization of alkali amphibole and pyroxene (Bailey 1977; Giordano *et al*. 2004; Giehl *et al*. 2014). From the models, it is clear that apatite/xenotime/monazite does not fractionate; therefore, we adjust the models to take into consideration the role of apatite crystallization. Since the bulk P_2O_5 content of the granitic rocks is rarely above 0.3 wt.%, we maintain the concentration between 0.2 and 0.3 wt.% and remove the corresponding amount of CaO to compensate. As shown in Figure 9(b), the adjusted CaO (dashed line) content of the modelled curves start to intersect the bulk composition of the Bokan granites. It is also possible that fluorite crystallization affected the system which would further suppress the CaO curve downwards and within the known composition range of the Bokan granitic rocks. Therefore, we conclude that the Bokan granitic rocks are likely derived by fractional crystallization of a mafic parental magma which resembles the lamprophyric dikes.

Conclusions

Our calculations are consistent with a model suggesting that magma with a composition similar to the syn-plutonic lamprophyric dikes from BMC can produce a residual liquid which resembles the observed compositions of the granitic rocks of the complex under geologically reasonable conditions. The exact parental magma composition is unknown; however, the models place some constraints on the possible initial composition and magmatic conditions. This conclusion helps to elucidate one

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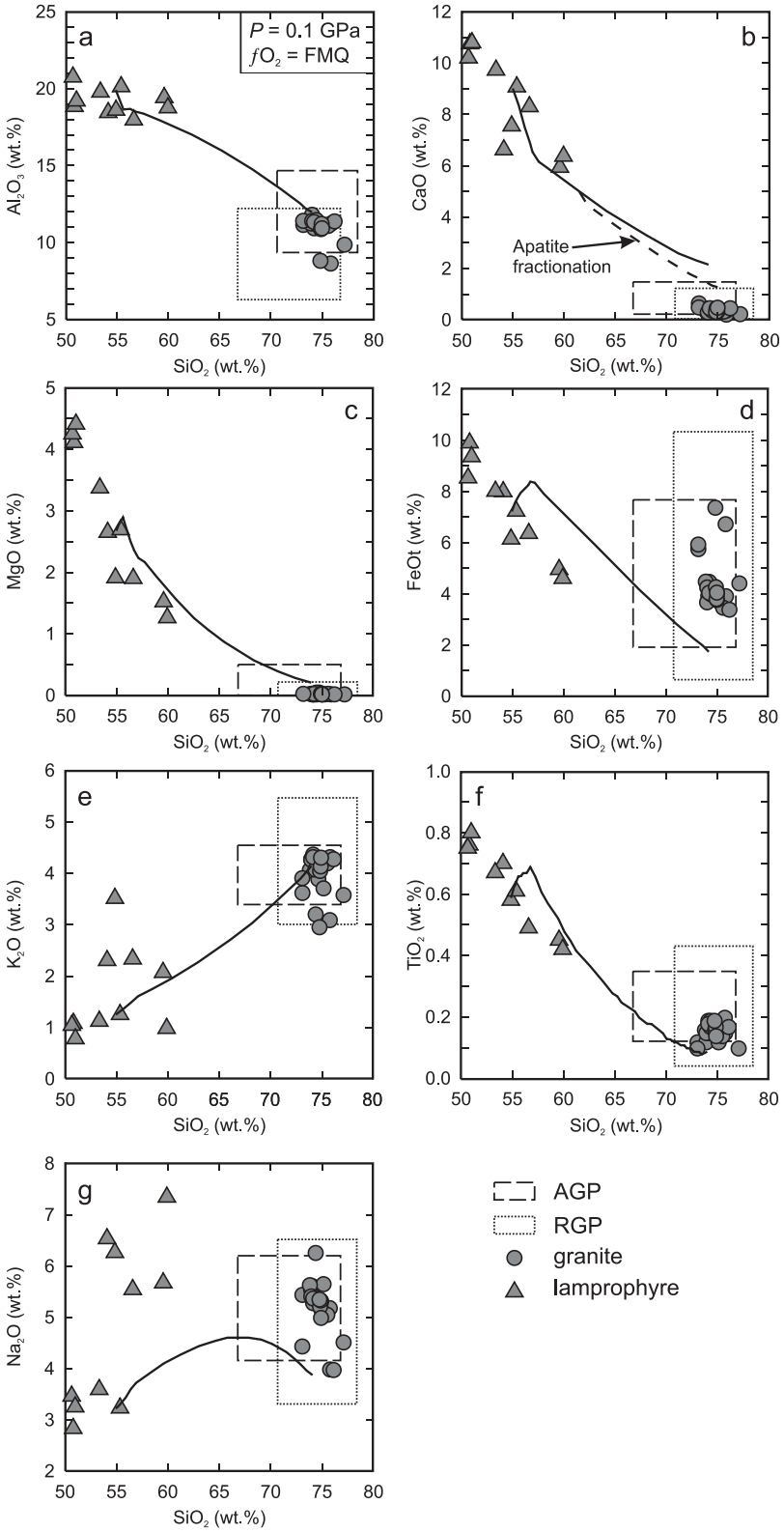


Figure 9. Results of Rhyolite-MELTS modelling showing the liquid compositional evolution curves. The solid line is the liquid evolution curve of model 828. The Ca content is adjusted to account for phosphate fractionation (see the text for details). The range of data for the aegirine granite porphyry (AGP) and riebeckite granite porphyry (RGP) from Thompson *et al*. (1982). The full modelling results are given online (Supplementary File 6).

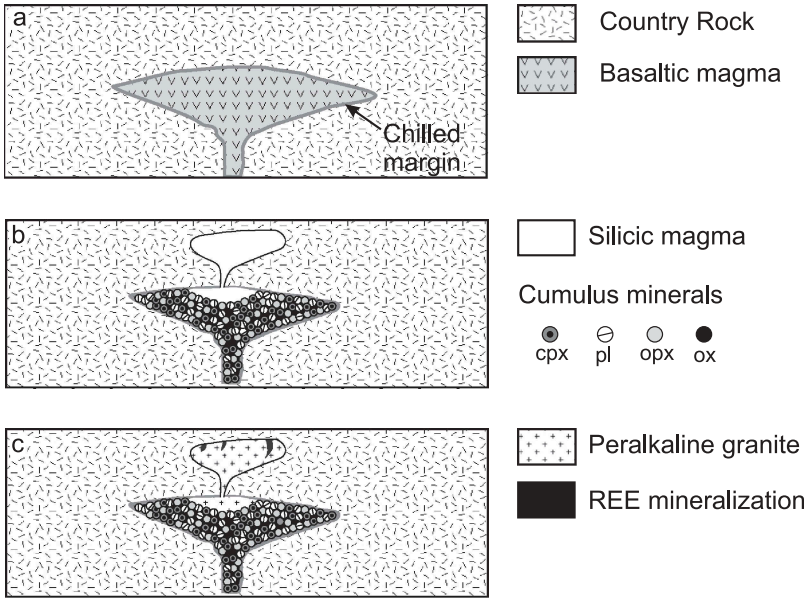


Figure 10. Origin of peralkaline granites of the Bokan Mountain complex. (a) Injection of basaltic magma into the crust; (b) fractional crystallization of parental magma to produce a residual silicic magma and a lower cumulate intrusion (cpx, clinopyroxene; pl, plagioclase; opx, orthopyroxene; ox, Fe–Ti oxide). (c) Crystallization of the residual silicic magma in the units of the Bokan complex (i.e. peralkaline granite) and the formation of magmatic REE deposits from late-stage fluids.

of the most debated stages of the origin of the rare metal deposits associated with peralkaline granitic rocks, i.e. the origin of the host rocks.

Rare metal mineralization (REE, Y, HFSE, U, and Th) of peralkaline felsic rocks is closely related to the crystallization of magma, especially to the late stages of the evolution. According to this model (e.g. Mungall and Martin 1996; Chakhmouradian and Zaitsev 2012; Dostal *et al*. 2014), the parent of the granitic rocks – the alkali or transitional basaltic magmas – was derived from a metasomatically enriched lithospheric source by a low degree of partial melting. The melts underwent fractional crystallization achieving a granitic composition (Figure 10). The fractional crystallization was protracted in part due to high contents of halogens and alkalis. The bulk of rare metals is hosted by ore minerals, which are frequently disseminated in the rocks (e.g. Richardson and Birkett 1996; Salvi and Williams-Jones 2005; Chakhmouradian and Zaitsev 2012; Dostal *et al*. 2014). Rare metal-bearing minerals started to crystallize only during the late stages of crystallization when magmas become fluid-saturated and rich in rare metals. The mineralizing fluids were dominantly magmatic, released during the last stages of the magma evolution. The fluids did not significantly modify the major element composition of the granites. Such a process (i.e. derivation of the peralkaline granites from mafic magmas) can probably be applied to other rare metal deposits associated with peralkaline granitic rocks such as the Strange Lake, Labrador/Quebec (Canada), Khaldzan-Buregtey (Mongolia), and Ghurayyah (Saudi Arabia; Drysdall and Douch 1986; Küster 2009).

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Supplemental data

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