

Original paper

Geology and evolution of pegmatite-hosted U–Th \pm REE–Y–Nb mineralization, Kulyk, Eagle, and Karin Lakes region, Wollaston Domain, northern Saskatchewan, Canada: examples of the dual role of extreme fractionation and hybridization processes

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In northern Saskatchewan, granitic pegmatites intruded Early Paleoproterozoic Wollaston Group metasedimentary rocks and interfolded granitoids that unconformably overlie Late Archean gneisses, all of which have been subjected to deformation during the protracted 1.86 to 1.77 Ga Trans-Hudson Orogeny. The U–Th \pm REE–Y–Nb pegmatite intrusions and fracture-controlled U mineralization characterize the occurrences at Kulyk Lake, Eagle Lake, and Karin Lake properties in the Wollaston Domain. The pegmatites are moderately to highly evolved, ranging from mineralogically simple to complex types. These are rare-earth element class, NYF pegmatites (Nb–Y–F), and are interpreted to have formed in a late syn- to post-collisional tectonic setting.

The complex-type pegmatites are hybridized, due to metasomatic interaction with the host rocks and therefore are locally crudely zoned. Saturation of U–Th \pm REE–Y–Nb occurred at the margins (predominately the border and wall zones) of the hybridized pegmatites. Partial melts were generated at depth, and then coalesced as they intruded to higher structural levels during exhumation of this orogen. This agrees with U–Pb geochronology of these granitic pegmatites, which constrains them between peak- and late-metamorphic events of the Trans-Hudson Orogeny. The age constraints and relatively high-T partial melting conditions ($\sim 750^\circ\text{C}$) confine the pegmatite melt-forming conditions to an early deformational event (1835–1805 Ma) that was overprinted by high-T retrograde metamorphism at c. 1770 Ma. Field relationships, textures and geochemical variations provide strong evidence that the U, Th, REE \pm Y–Nb phases in the studied pegmatites were progressively enriched through extreme fractionation effects of which are evident throughout multiple pegmatite injections. In addition, volatiles and other fluxing components further enriched U, Th, REE \pm Y–Nb during complex hybridization reactions between pegmatite melt and wallrock, up to the final stages of pegmatite emplacement.

Keywords: granitic pegmatite, hybridization, fractional crystallization, assimilation, NYF-type, Wollaston Domain

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1. Introduction

In northern Saskatchewan, a majority of the rare-earth elements (REE), U, Th, Zr, and Nb mineralized showings are hosted in late-tectonic to anorogenic granitoid and pegmatite intrusions (e.g., Mawdsley 1952, 1953, 1955, 1957; Parslow and Thomas 1982; Parslow et al. 1985; Annesley and Madore 1999; Annesley et al. 2000; Madore et al. 2000; McKechnie et al. 2012a, b; 2013). These occurrences consist predominately of monazite, apatite, garnet, xenotime, zircon, allanite, uraninite–uraniothorite, thorite, and rutile. McKeough et al. (2010) have presented the field relationships between the Wollaston pegmatites and host rocks and their associated gamma-ray spectrometry data (radiometric signatures), emphasizing the U and Th contents and distribution within the late-tectonic pegmatites studied. A follow-up

article by McKeough and Lentz (2011) described the spatial and temporal relationships between the simple-type granitic and metasomatized (hybridized) pegmatites.

This paper presents the petrological, geochemical, and geochemical constraints on the mineralogically simple- to complex-type pegmatites that are mineralized with U-, Th-, REE-, Y-, and Nb-bearing phases. The overall objective of this study was to determine whether these late-tectonic pegmatites were formed via fractionation and/or *in situ* partial melting, and to refine a petrogenetic model for their evolution and their relationship with respect to the basal Wollaston sequence. The observations in this article reflect fundamental differences in how the pegmatites become enriched, and eventually saturated in U, Th, and REE at various stages from partial melting until pegmatite emplacement.

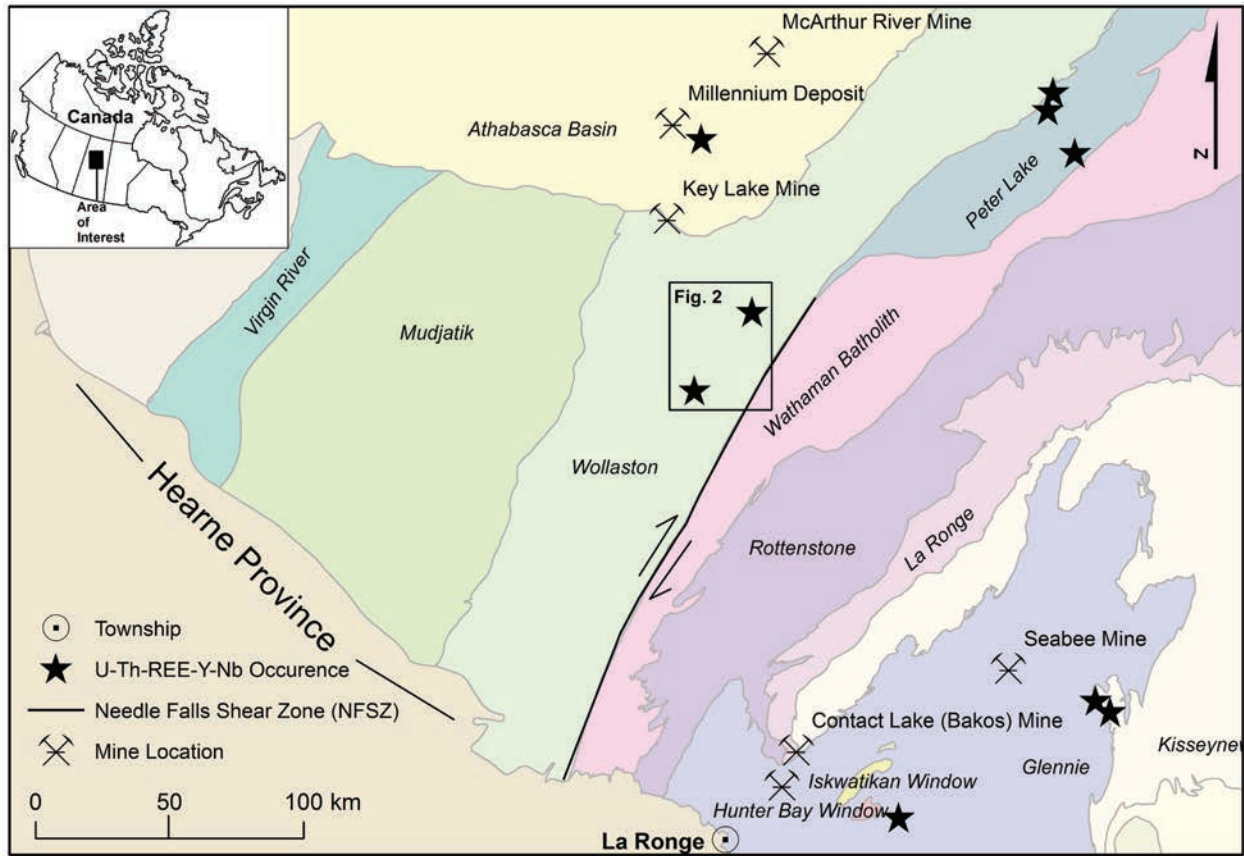


Fig. 1 Location map showing U–Th–REE–Y–Nb occurrences within each lithotectonic domain of the Hearne Province in northern Saskatchewan. The black box indicates the study area.

2. Geological setting

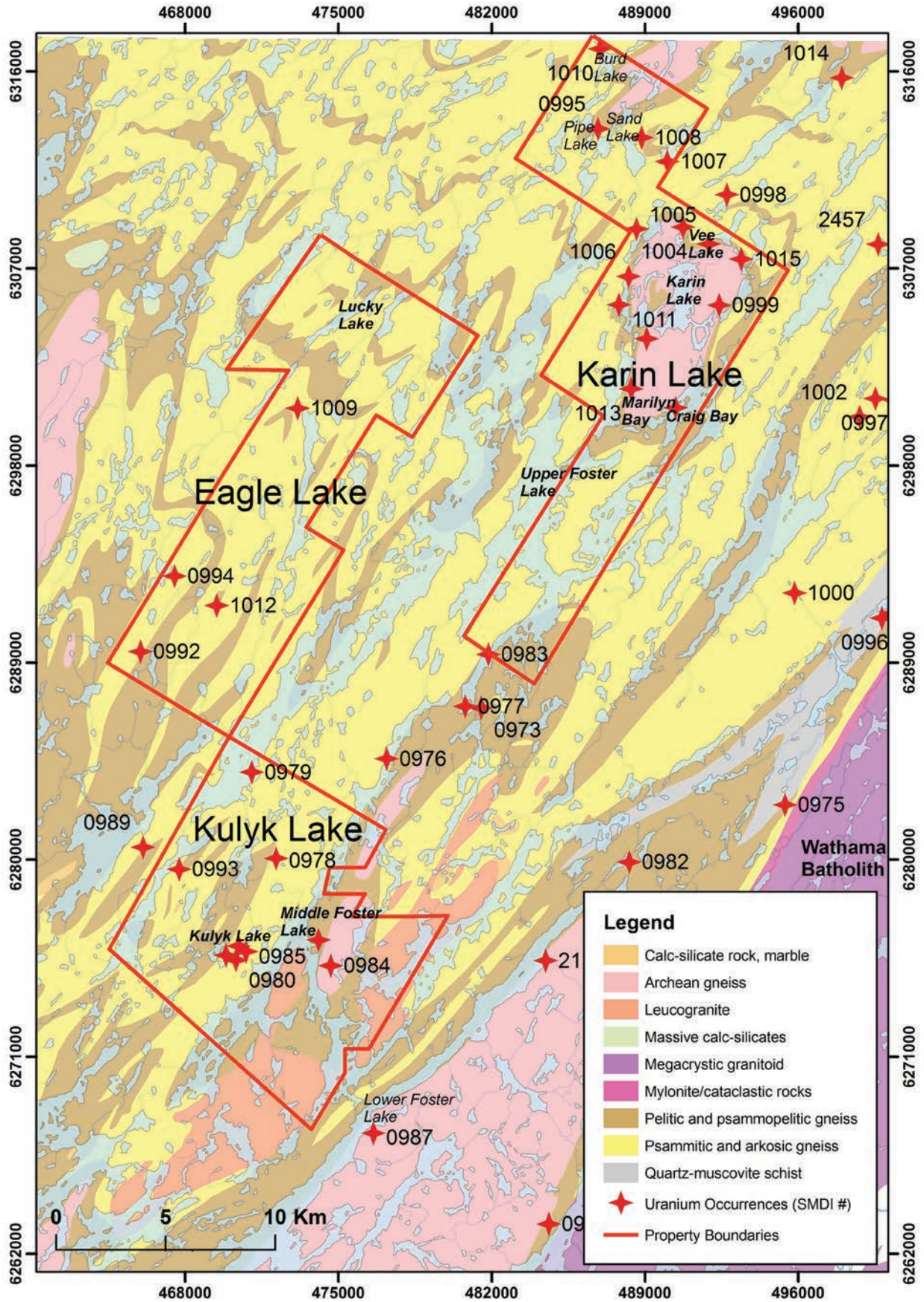
2.1. Regional geology

The Wollaston Domain formed along the southeastern margin of the Hearne Province in northern Saskatchewan, Canada (Bickford et al. 1990; Hoffman, 1990; Lewry and Collerson 1990; Fig. 1). It is comprised of a northeast-trending, tightly folded, linear belt containing Archean granitoid rocks preserved in structural domes, unconformably overlain by Paleoproterozoic metasedimentary rocks, and interfolded anatectic granitoid rocks (Ray 1977; Lewry and Sibbald 1980; Tran and Yeo 1997; Fig. 2). The supracrustal rocks, known formally as the Wollaston Group, are complexly deformed and have complicated lithotectonic relations that vary regionally (e.g., Annesley et al. 2005). Polyphase deformation was concurrent with upper amphibolite- to granulite-facies metamorphism in the region (Yeo and Delaney 2007).

Previous studies of the Trans-Hudson Orogen have used geochemical, isotopic, and geochronological data to characterize the evolution of the Paleoproterozoic sedimentary and magmatic assemblages, including the Wollaston Group and underlying basement rocks (Ray

and Wanless 1980; Bickford et al. 1990; Annesley et al. 1992, 2005; Ansdell et al. 2000; Rayner et al. 2005; Tran et al. 2008). The studies showed that the Paleoproterozoic mafic and felsic intrusive rocks record variable degrees of contamination by Archean and older Paleoproterozoic crust, underlining the complex evolutionary history of crust along the western margin of the Trans-Hudson Orogen. The Wollaston Group metasediments overlie Archean granites that range between 2650 Ma and 2500 Ma (U–Pb zircon TIMS ages; Ray and Wanless 1980; Krogh and Clark 1987; Annesley et al. 1992, 2005; Hamilton and Delaney 2000; Rayner et al. 2005). The cover sequence was described as a Paleoproterozoic syn-rift passive margin and foreland metasedimentary basin (Ansdell et al. 2000; Tran et al. 2003, 2008) that is divided into Lower and Upper subgroups, separated by a regional unconformity (Tran 2001). The lower succession includes a basal, graphitic pelitic unit overlain by, and in part interleaved with, arkose, conglomerate, quartzite, and calcareous sedimentary rocks. The upper

Fig. 2 Regional geology map of the study area in the Wollaston Domain, showing uranium occurrences as defined by the Saskatchewan Mineral Deposit Index (SMDI).



part of the succession includes conglomerates, immature arkosic rocks, and calc-silicate rocks and is thought to represent subsequent infilling of a foreland basin. Neodymium isotopic data of Tran et al. (2003) showed that the Wollaston Group detritus was likely derived from active continental margin magmatic sources, as supported by the presence of a significant proportion of 1920–1880 Ma old zircons (Tran et al. 2008). There are three main zircon populations, including: 1) older than 2400 Ma grains that represent detritus shed from the neighboring Hearne Craton; 2) ~ 2100 Ma components thought to represent reworking of syn-rift volcanic rocks, and 3) 1920–1880 Ma detritus derived from advancing volcanic arc terrains of the western Churchill Province (Ansdell et al. 2000; Hamilton and Delaney 2000; Tran 2001; Yeo and Delaney 2007; Tran et al. 2008).

Variably foliated granites, leucogranites, amphibolites, and granitic pegmatites intruded the Wollaston Group rocks and form planar lenses concordant to the regional fabric or discordant to semi-concordant dyke swarms. The LA ICP-MS U–Pb dating of monazite from a foliation-parallel pegmatite at Kulyk Lake yielded an age of 1830 ± 5 Ma (McFarlane and McKeough 2013). Using the same method, monazite grains from the Kulyk Lake monazite–apatite–Fe–Ti–oxide showing (Tab. 1) were dated at *c.* 1830 Ma and 1770 Ma (McFarlane and McKeough 2013). Furthermore, U–Pb LA ICP-MS dating of monazite from the Yellow Lake pegmatite yielded a concordant age of 1774 ± 3 Ma (McKeough 2013). Using the same methods, one point in a monazite core gave a concordant age of ~1815 Ma, but aside from this anomaly the average ($n = 8$) was 1770 ± 8 Ma with 95% confidence and probability of 0.97. The *c.* 1770 Ma date was interpreted as timing a retrograde metamorphic event followed by exhumation and rapid cooling at the terminal stages in this orogeny (e.g., Schneider et al. 2007). The U–Pb ages constrain the emplacement of the pegmatite and mineralization to the well-documented magmatism (or hydrothermal–magmatic events) in the latter stages of Trans-Hudson Orogeny (THO) deformation at *c.* 1850–1760 Ma. The timing of pegmatite emplacement in the THO has previously been supported by U–Pb zircon and monazite ages from undeformed pegmatites in the Wollaston fold belt (Chiarenzelli 1989; Bickford et al. 1990, 2005; Chiarenzelli et al. 1998; Annesley et al. 2005; Schneider et al. 2007; McKechnie et al. 2012a, b).

Evidence demonstrating melt generation in the middle crust is apparent in the Key Lake and McArthur River mine areas, where dilatant zones were created during thrusting, strike-slip faulting, and/or extensional faulting that have focused crustal melts and partitioned strain (Annesley et al. 2005). The tectonized contact between the Archean gneiss domes and the Lower Wollaston supracrustal rocks was the locus for magmatism, asso-

Tab. 1 New and historical U–Th–REE showings, defined in the Saskatchewan Mineral Deposit Index (SMDI) that were investigated as part of a regional pegmatite study completed in the Wollaston Domain (McKeough et al. 2010)

Showing Name	Commodity	SMDI	Property	Radiometrics (max cnts/s)	Pegmatite Type	Nature of Pegmatite Contact	Mineralogy	UTM mE	UTM mN
Eldorado-N	U	0980	Kulyk Lake	15,000	Simple	Sharp; magnetic psammopelitic gneiss	Kfs + Qtz + Bt	470082	6274795
Eldorado-S	U	0980	Kulyk Lake	55,000	Hybridized	Gradational; calc-silicate gneiss	Kfs + Qtz + Bt + Ca-Amp + Ca-Px	470329	6275394
Fanta (aka Kulyk Lake Occurrence)	U–Th–REE	0985	Kulyk Lake	23,000	Simple	Sharp; calc-silicate gneiss	Kfs + Qtz + /-Bt	470510	6275070
Tara Occurrence	U	0993	Kulyk Lake	3,400	Simple	Sharp; semi-pelitic gneiss	Kfs + Qtz + Bt	467730	6279358
Red October-L19	U	N/A	Eagle Lake	3,000	Simple	Sharp; semi-pelitic gneiss	Kfs + Qtz + Bt + Ilm	474290	6304415
Red October TR-1	U–REE	N/A	Eagle Lake	35,000	Hybridized	Gradational; calc-silicate gneiss and/or diorite	Kfs + Qtz + Bt + Ilm + Mt + Ca-Amp + Ca-Py	474260	6304642
Red October TR-2	U–REE	N/A	Eagle Lake	18,000	Hybridized	Gradational; diorite	Kfs + Qtz + Bt + Ilm + Mt + Amp	474234	6304562
Craig Bay Occurrence	U–REE–Mo	1001	Karin Lake	10,000	Simple	Sharp; graphitic semi-pelitic gneiss	Kfs + Qtz + Bt + Grt	490410	6300638
Yellow Lake Occurrence	U–Th–REE	0999	Karin Lake	3,000	Simple	Sharp; graphitic semi-pelitic gneiss	Kfs + Qtz + Bt	492284	6305341
Rona Showing	U	0983	Karin Lake	7,000	Simple	Sharp; semi-pelitic gneiss	Ab + Qtz + Bt	481874	6289775
Pegmatite Lake Occurrence	U–REE–Mo	1004	Karin Lake	3,000	Simple	Sharp; semi-pelitic gneiss	Kfs + Qtz + Bt	490601	6308796
Marilyn Bay	U–REE	N/A	Karin Lake	9,500	Simple	Sharp; arkosic gneiss	Kfs + Qtz + Bt	489168	6298439

ciated with deeper level anatexis during the main syn-metamorphic deformation events at the thermal peak of the Trans-Hudson Orogeny in this region. Although the Archean–Paleoproterozoic boundary here is an angular unconformity, it probably represents a parautochthonous to allochthonous contact zone that focused late-tectonic leucogranites, and underwent multiple episodes of reactivation.

2.2. Local geology and mineralization

The area of interest has been investigated for uranium by the government of Saskatchewan, as well as exploration and mining companies since the 1950's (Mawdsley 1957). It hosts historical U–Th ± REE showings, as described in the Saskatchewan Mineral Database Index (Tab. 1). The areas surrounding Kulyk and Eagle lakes are underlain by psammitic to semi-pelitic and calc-silicate metasediments. The metasedimentary supracrustal rocks at Eagle Lake were intruded by diorite, which is host to an exposed uraniferous pegmatite dyke swarm. However, the Wollaston Group metasedimentary rocks are most typically the host to these leucogranites, pegmatites and aplites. These pegmatites are syn- to post-deformational and form north-, east-, and east–north-east-trending sheets and lenses in the predominantly supracrustal rocks. It appears that the pegmatites are associated with steep north to north–northeast-trending structures. It can be challenging to distinguish localized anatectic or migmatitic phases from granitic intrusions, particularly at the contacts between the diorite and pelitic gneisses; this is a consequence of high metamorphic grades in the Upper Foster Lake region of northern Saskatchewan. The U, Th, and REE mineralization is visibly disseminated in the pegmatites, and forming veins that appear to be primarily fracture-controlled within folded and faulted metasediments locally along the contacts between the host rocks and pegmatites. North-trending fractures that cross-cut the pegmatite and the host rocks are uraniferous, which gives an indication of localized U–Th remobilization. The U–Th–REE mineralization is strongly associated with pegmatite–diorite exocontacts at Eagle Lake and pegmatite–calc-silicate gneiss contacts at Kulyk Lake.

The Karin Lake area partly lies within the Upper Foster Lake region (Fig. 2), which was mapped by Tran and Yeo (1997) at 1:20 000 scale. Historical mapping in the area of interest coincides with the contact between the Karin Lake Formation (overlying) and the Archean basement rocks (Yeo and Delaney 2007). The contact is an unconformity that is highly strained; hence the supracrustal rocks are parautochthonous, at least locally (Tran and Yeo 1997; Tran et al. 1998; Yeo and Delaney 2007). The pegmatites are host to U–Th mineralization,

and commonly associated with Mo ± REE mineralization. This is especially evident at the Rona showing (Tab. 1), where plagioclase-rich pegmatite dykes at or near margins with host garnet- and biotite-bearing gneisses, migmatite, and/or granitized metasediments are generally accompanied by finely disseminated molybdenite. The contact (and mineralized) zones are invariably fractured with localized breccia and overprinted by a moderate to strong hematite, chlorite, and clay alteration. The alteration occurs as fracture-fillings through to pervasive in highly altered zones, especially areas that are graphite-rich or proximal to graphite-rich lithologies. The presence of graphitic and calc-silicate-bearing units that are often mineralized has been noted at several uranium deposits within the Athabasca Basin (Jefferson et al. 2007).

3. Methodology

3.1. Mapping strategy and sampling methods

The entire study area encompasses a region approximately 30 × 20 km. Local- and regional-scale outcrop mapping was completed in order to determine spatial and temporal relationships between each of the pegmatite groups. A report of the various rock types encountered, as well as their associated spectrometry data, was presented in McKeough et al. (2010). Of the thirty-nine granitic pegmatites sampled from the region, twenty were K-feldspar-rich, seven plagioclase-rich, and twelve mineralogically complex pegmatites or “hybridized” types that were sampled only from Kulyk and Eagle lake areas. Seven weakly to moderately radioactive K-feldspar-rich and plagioclase-rich granites were also sampled, in attempts to define a petrogenetic relationship between them and the pegmatites.

3.2. Analytical techniques

Bulk pegmatite samples, collected from both the simple-type and hybridized pegmatites, were crushed and split at the ACME Analytical Laboratory, Vancouver, BC. Major-oxide with rare-earth and refractory elements were determined using a lithium metaborate fusion and nitric acid digestion of a >0.5 g sample prior to ICP-MS analyses. A separate >0.5 g sample was split and digested in an Aqua Regia, and analysed by ICP-MS in order to identify other precious and base metals.

Accessory minerals, especially U- and Th-bearing phases, were identified by a JEOL JSM 6400 Digital Scanning Electron Microscope (SEM) using Energy Dispersive Spectroscopy (EDS). Images were acquired using backscattered electron (BSE) imaging at the University of New Brunswick (UNB) Microanalysis and Microana-

lytical Facility. The operating conditions of the SEM-EDS were 15 kV for the accelerating voltage, 1.5 nA for the probe current, and 60 s for the spectra collection time.

4. Description of the pegmatites

Granitic pegmatites and aplites cut all of the Wollaston Group units within the region, but appear to be more common in the basal Wollaston metasediments. Limited granite outcrops were observed in close proximity to any of the pegmatite groups, especially the rare hybridized pegmatites. However, there are some outcrops of granite at Kulyk Lake, in close proximity to simple-type granitic pegmatites sampled near the Kulyk Lake and Eldorado showings (Tab. 1). They are of typical K-feldspar-quartz-biotite composition, predominately massive, and may include xenoliths of adjacent metasedimentary rock (McKeough et al. 2010). At Craig Bay (Karin Lake region; Fig. 2), it is common to observe pegmatitic segregations that have developed locally in weakly foliated granites (McKeough et al. 2010).

Steep pegmatites intruded near primary fold structures and major faults and fractures that intersect the metasediments, sometimes exploiting foliation-parallel trends that are nearly coincident with the local east-, east-northeast-, and northeast-striking gneissosity; however, the mineralized pegmatites at Kulyk and Eagle lakes are discordant to the local gneissosity. The pegmatites are between 0.1 m and 10 m wide, and vary in grain size from coarse grained to pegmatitic. It is common to see fine-grained dykes (aplite) cross-cutting pegmatite dykes at Kulyk and Karin lakes. The pegmatites display a weak to moderate igneous foliation. Compositional variations range from K-feldspar-quartz-biotite-rich, simple-type granitic pegmatites to plagioclase-quartz-muscovite-garnet-rich granitic pegmatites (Tab.1). At Kulyk and Eagle lakes plagioclase-rich pegmatite and aplites intruded parallel to the fabrics (concordant) within the diorite, calc-silicate paragneiss, and psammopelitic gneiss. At Karin Lake, plagioclase-rich pegmatites occur as dykes and lenses primarily within a grey granite suite, as well as in basal graphitic paragneisses. Pegmatites at Kulyk and Eagle lakes exhibit a more complex mineralogy, due to interaction with the adjacent host rocks. These hybridized or “hybrid” pegmatites (Rimsaite 1980; 1983a, b, 1984, 1985; Lentz 1996) form both zoned and unzoned types, with a ~ 5–10 cm contact hybrid zone, between the pegmatite and wallrock. The internal structure of the simple-type pegmatites vs. hybrid pegmatites varies texturally (Tab. 1). Overall, the simple-type pegmatites have sharp and fine-grained margins that coarsen toward the central portions of the pegmatite body. Simple-type pegmatites are rarely zoned; however some of them ex-

hibit a graphic K-feldspar-Qtz-rich intermediate zone, followed inward by a coarse-grained quartz core. The hybridized pegmatites are also zoned and unzoned. The contact between the hybridized pegmatites and the host rock is gradational and consists of medium- to coarse-grained ferromagnesian minerals, such as actinolite, diopside, biotite, and magnetite, similar to those found in the adjacent host rock. The hybridized contact (border zone) is followed inward by a medium- to coarse-grained heterogeneous intermediate zone, and a coarse-grained K-feldspar-Qtz-rich core.

4.1. Mineralogy of the pegmatites

The mineral assemblages in individual types of pegmatites are shown in Tab 2. The most common of the mineralogically simple-type granitic pegmatites are K-feldspar-Qtz-rich types with lesser amounts of micas. The mineralogy from the center to margin of the simple-type pegmatites is relatively consistent, with K-feldspar >> plagioclase, quartz, and biotite. They are inequigranular to equigranular, variably magnetic, and display typical pegmatite granophyric and graphic textures in quartz and feldspars. To a lesser extent, myrmekites are visible adjacent to pegmatite-wallrock contact zones. Orthoclase is more common than microcline. Plagioclase feldspars are albite that generally forms very fine (<2 mm) perthitic intergrowths with orthoclase. The feldspars have moderately sericitized cores and unaltered rims. The K-feldspar-rich granitic pegmatites locally include up to 5 % fine-grained, earthy hematite replacement of feldspars. Biotite is mostly interstitial between quartz, K-feldspar, and plagioclase and forms aggregates. Chlorite and hematite partially replace biotite flakes. The U-Th-REE accessory phases are predominately microscopic (<2 mm) thorite, monazite, apatite, and zircon, with rare allanite and xenotime. They are associated with biotite in simple-type granitic pegmatites, but are also noted as inclusions in quartz-rich zones.

The second most common variety of the simple-type granitic pegmatites is plagioclase-quartz-rich. Plagioclase feldspar is almost always albite, and K-feldspar, when present, is identified as microcline, and locally interstitial to quartz. Both myrmekite and, less common, granophyre texture are present. Biotite occurs as medium- to coarse-grained radiating booklets, with finer grained quartz and feldspar between the layers. Muscovite is fine-grained and interstitial to K-feldspar. Both micas are associated with hematite, chlorite, and epidote alteration. At Kulyk and Eagle lakes, the plagioclase-rich granitic pegmatites are sparse and rarely contain garnets, but at Karin Lake, they show a higher abundance of aluminous minerals, such as muscovite, garnet, cordierite and sillimanite. Uranium-thorium-REE accessory phases are

Tab. 2 Petrological and mineralogical descriptions at selected U–Th–REE showings visited, emphasizing differences between simple-type pegmatites and hybridized pegmatites in the Wollaston Domain

Location	Simple pegmatites		Comments	Hybridized pegmatites		Comments
	Rock-forming minerals	Accessory minerals		Rock-forming minerals	Accessory minerals	
Kulyk Lake Eldorado N (simple) Eldorado S (hybridized)	75 % K-feldspar 20 % quartz <5 % albite	tr. ilmenite tr. titanite tr. monazite tr. apatite	Accessory phases disseminated throughout pegmatite.	45 % K-feldspar 10 % quartz 15 % albite <10 % biotite <10 % diopside <10 % actinolite	0.5 % titanite 0.1 % monazite Tr. uraninite Tr. zircon	Accessory phases associated with biotite, diopside, and actinolite in pegmatite border zone.
	65 % K-feldspar 20 % albite 15 % quartz 10 % biotite	5 % ilmenite 0.2 % titanite tr. uraninite	Ilmenite and titanite associated with biotite; uraninite is disseminated throughout pegmatite.	50 % K-feldspar 20 % quartz 10 % biotite <10 % actinolite <5 % diopside	<5 % magnetite 0.1 % titanite Tr. rutile Tr. monazite Tr. apatite Tr. zircon	Albite altered to sericite and/or scapolite; accessory phases associated within pegmatite border zone.
	50 % K-feldspar 20 % quartz 15 % albite 5 % biotite 5 % muscovite tr. sillimanite	<5 % garnet 0.1 % molybdenite 0.1 % monazite tr. pyrite	–	60 % albite 20 % quartz <10 % biotite 5 % K-feldspar <5 % actinolite <5 % diopside	<5 % magnetite 0.1 % titanite/rutile Tr. xenotime Tr. monazite Tr. apatite Tr. zircon	Accessory phases associated with biotite in metasomatic front along pegmatite–diorite contact.
Eagle Lake Red October TR-2	60 % K-feldspar 15 % quartz <5 % albite 5 % biotite 2 % calcite	5 % ilmenite 3 % monazite 5 % apatite tr. zircon	–	–	–	Albite partially altered to sericite; accessory phases associated with selective hematite and chlorite along fractures.
	55 % albite 20 % quartz 15 % biotite <10 % muscovite	0.5 % molybdenite tr. uraninite tr. monazite tr. zircon	–	–	–	There are two different zones within the sample; the host rock consists of K-feldspar, quartz, albite, calcite and biotite; the other zone consists of cumulate-textured apatite and monazite
	55 % K-feldspar 20 % quartz 10 % albite <10 % biotite	<5 % ilmenite tr. apatite tr. monazite tr. zircon	–	–	–	Accessory phases are strongly associated within biotite segregations.
Marilyn Bay	–	–	–	–	–	The accessory phases are disseminated throughout the pegmatite.

predominately monazite and xenotime, with lesser apatite and zircon. Co-existing microscopic monazite and xenotime are associated with biotite- and/or garnet-rich segregations and/or occur as inclusions in feldspars.

At Kulyk and Eagle lakes, pegmatites that are interpreted as products of hybridization consist of fine- to coarse-grained K-feldspar, albite, and quartz, with high concentrations of biotite, actinolite, diopside, magnetite, ilmenite, and titanite. The ferromagnesian and Fe–Ti-oxide phases together make up over 30 vol. % of the pegmatite hybrid border zone. The border zone is followed inward by an intermediate zone that consists of plagioclase >> K-feldspar, quartz, and variable amounts of fine- to medium-grained biotite, actinolite, diopside, magnetite, ilmenite, and titanite. The central portion of the hybridized pegmatites is composed of medium- to coarse-grained quartz and K-feldspar. Coexisting ilmenite, magnetite, and titanite are commonly associated with biotite, closer to the pegmatite–wallrock contact, and less abundant in pegmatite cores. Albite is the primary plagioclase feldspar within the intermediate zone, and is partially altered to sericite and/or scapolite. Radioactive minerals uraninite, monazite, xenotime, apatite, and zircon are present at higher concentrations in the hybrid border zone, are less abundant in the intermediate zone, and rare in the hybridized pegmatite cores.

5. Geochemistry of the pegmatites

The whole-rock major- and trace-element compositions for pegmatite groups are presented in Tab. 3. Selected variation diagrams illustrate the chemical compositions of the simple-type and hybridized pegmatites (Fig. 3). The simple-type pegmatites vary from subaluminous to peraluminous. Their Alumina Saturation Index ($ASI = Al_2O_3 / (CaO + Na_2O + K_2O)$) ranges from 1.06 to 2.36. The complexly altered hybridized pegmatites range between metaluminous and peraluminous, having ASI values that are between 0.42 and 1.72. All pegmatites in the region vary from magnesian to ferroan as indicated by the $FeO / [FeO + MgO]$ vs. SiO_2 plot (Fig. 4; Frost et al. 2001; Frost and Frost 2008). This is attributed to the variable amounts of biotite, tremolite, actinolite, cordierite vs. ilmenite and magnetite with various degrees of hematization. The simple-type K-feldspar–quartz-rich and plagioclase–quartz-rich granitic pegmatite varieties fall within the calcic to alkalic series in the $Na_2O + K_2O - CaO$ vs. SiO_2 plot (Fig. 5; Frost et al. 2001). The hybridized pegmatites from Kulyk and Eagle lakes are restricted to alkali-calcic and alkalic fields.

All simple-type granitic and hybridized pegmatites contain oxides, phosphates, and silicates that carry Nb > Ta, Y, REE, U, Th, and Zr, and as a result are classified

as NYF pegmatites (see Černý 1991). There is a moderate correlation between U and Th in each of the pegmatite groups (Fig. 6a). The simple-type pegmatites range between low Th/U (Eagle Lake) and high Th/U (Karin Lake). There is a tendency to decrease in this ratio with increasing SiO_2 between the hybridized and simple-type pegmatites (Fig. 6b). The hybridized pegmatites show the strongest correlation between U and ΣREE and Th and ΣREE (Fig. 6c–d). There are granitic pegmatites, particularly those from the Karin Lake region, that show a stronger link between Th and total REE, because of high concentrations of monazite compared to other U–Th–REE-bearing phases. The chondrite-normalized REE patterns (Fig. 7) for both the simple-type (granitic) and hybridized pegmatites show similar features, but distinct REE profiles. All pegmatite groups are LREE-enriched, mostly with moderately negative Eu anomalies: Kulyk Lake La_N / Lu_N of 0.53–158.56 and Eu / Eu^* of 0.30–2.66; Eagle Lake La_N / Lu_N of 0.61–21.06 and Eu / Eu^* of 0.21–1.11; Karin Lake La_N / Lu_N of 0.16–61.80 and Eu / Eu^* of 0.16–1.95.

The pegmatites, aplites, and granites are characterized by high SiO_2 , $Na_2O + K_2O$, Fe/Mg, F, Zr, Nb, Ga, REE, and Y, representative of A-type granite geochemistry (e.g., Whalen et al. 1987; Eby 1990). Crustally derived A-type granite signatures are characteristic of REE-enriched NYF-pegmatites (Eby 1990; Černý and Ercit 2005; Martin and Devito 2005; Martin 2006). Furthermore, the Y–Nb–Ce ternary diagram (Eby 1992) discriminates between mantle-derived and crustally-derived A-type granites (Fig. 8); the hybridized pegmatites fall completely within the field of fractionated, crustally derived A_2 granites, as do nearly all the simple-type pegmatites.

5.1. Hybridization and fractionation indicators

A ‘hybridization index’ of $CaO + MgO + FeO_t$ is useful in quantifying deviations from leucogranitic melt compositions (see Lentz 1996). A statistically strong correlation between the hybridization index and U, Th, ΣREE , Nb, and Zr concentrations in the pegmatites at Kulyk and Eagle lakes was observed by McKeough and Lentz (2011; Fig. 9). Uranium, Th, REE, Y, and Nb show a slightly stronger correlation to the hybridization components, confirming the empirical observation of high radioactivity along the margins of the hybrid pegmatites. The P_2O_5 increases to a slightly lesser degree and was most likely incorporated with other mineralizing elements enriched autometasomatically with magmatic–hydrothermal (orthomagmatic) fluids.

The pegmatites are weakly to moderately evolved, as demonstrated by Rb–Ba–Sr trends that vary between signatures of a normal and a moderately fractionated

Tab. 3 Whole-rock major- and trace-element geochemical data for selected simple-type and hybridized pegmatites in the Kulyk, Eagle, and Karin lakes region of the Wollaston Domain

Sample	MMELR 008	MMELR 015	MMELR 003	MMELR 007	MMELR 012	MMELR 010	MMELR 009	MMKJR 008	MMKJR 010	MMKJR 017	MMKJR 022	MMKJR 014	MMKJR 015	MMKJR 028
Type	Simple	Simple	Simple	Hybrid	Hybrid	Hybrid	Hybrid	Simple	Simple	Simple	Simple	Hybrid	Hybrid	Hybrid
SiO ₂	72.0	74.5	73.5	65.4	68.3	51.3	51.2	71.5	68.9	73.0	73.1	65.0	64.1	67.9
TiO ₂	0.10	0.09	0.35	0.30	1.16	2.60	1.72	0.07	0.06	0.04	0.02	0.10	0.01	0.44
Al ₂ O ₃	14.4	13.9	11.1	15.7	14.2	14.2	15.3	14.9	16.9	16.7	14.9	14.6	16.4	15.1
Fe ₂ O ₃	1.03	1.03	3.10	2.40	6.23	10.86	10.47	0.76	0.72	0.52	0.52	5.22	6.69	5.76
MnO	0.02	0.02	0.01	0.05	0.03	0.18	0.16	0.01	0.02	0.01	0.01	0.01	0.01	0.01
Cr ₂ O ₃	0.003	0.003	0.002	0.003	0.003	0.002	0.014	0.003	0.002	0.003	0.003	0.003	0.002	0.006
MgO	0.50	0.22	1.13	1.55	1.05	4.60	5.77	0.12	0.22	0.23	0.05	3.48	3.05	0.80
CaO	2.42	1.53	1.30	2.68	1.09	7.93	8.05	0.97	1.00	0.83	0.34	0.85	0.69	1.37
Na ₂ O	3.85	4.35	4.78	6.57	3.08	4.25	4.32	2.69	3.54	3.17	4.19	9.09	6.99	6.57
K ₂ O	4.37	3.71	4.38	5.18	4.57	2.67	2.39	8.27	7.95	5.40	6.08	1.72	1.54	2.05
P ₂ O ₅	0.08	0.07	0.04	0.17	0.09	1.23	0.33	0.60	0.55	0.17	0.03	0.03	0.03	0.11
LOI	0.4	0.6	0.6	0.3	0.6	0.8	0.8	0.6	1.2	0.8	0.6	0.8	0.7	0.7
SUM	99.17	100.02	100.29	100.30	100.40	100.62	100.52	100.49	101.06	100.87	99.84	100.90	100.21	100.82
Hybrid Index	4.05	2.87	5.88	6.93	9.53	25.99	26.01	1.92	2.00	1.62	0.93	9.65	10.44	8.37
ASI	1.35	1.45	1.06	1.09	1.62	0.96	1.04	1.25	1.35	1.78	1.40	1.25	1.78	1.51
Ba	3389	1301	2453	2736	3989	1678	757	753	924	630	491	127	186	667
Be	3	4	1	2	2	8	4	1	1	4	2	10	8	3
Cs	8.2	1.7	4.0	4.2	7.0	3.3	3.6	1.8	1.2	1.3	0.5	2.1	2.5	3.2
Hf	0.8	1.0	57.2	1.5	2.0	6.1	3.4	0.8	0.3	0.2	1.2	7.2	6.9	3.2
Rb	122	130	161	135	226	50.3	44.6	233	296	220	146	30.7	25.2	65.4
Nb	2.6	6.1	6.8	13.0	5.6	131.0	19.6	9.4	3.3	1.2	0.1	30.2	3.0	11.6
Sr	369	137	125	303	231	286	518	85.4	109	98.8	69.4	56.7	60.2	124
Ta	0.2	0.8	0.7	1.3	0.5	23.1	1.3	1.4	0.4	0.1	0.1	8.2	0.4	0.9
Zr	20	34.1	1831	39.6	64.8	195	107	23.7	7.3	7.9	36.5	129	119	122
Y	32	14.1	15.3	53.3	36.8	608	86.4	49.9	56.9	20.8	3.9	24.2	26.6	14
K/Cs	0.5	2.2	1.1	1.2	0.7	0.8	0.7	4.6	6.6	4.2	13.5	0.8	0.6	0.6
K/Rb	0.04	0.03	0.03	0.04	0.02	0.05	0.05	0.04	0.03	0.02	0.04	0.06	0.06	0.03
Rb/Sr	0.33	0.95	1.29	0.45	0.98	0.18	0.09	2.73	2.72	2.23	2.10	0.54	0.42	0.53
Rb/Ba	0.04	0.10	0.07	0.05	0.06	0.03	0.06	0.31	0.32	0.35	0.30	0.24	0.14	0.10
Nb/Ta	13.0	7.6	9.7	10.0	11.2	5.7	15.1	6.7	8.3	12.0	1.0	3.7	7.5	12.9
Y/Nb	12.31	2.31	2.25	4.10	6.57	4.64	4.41	5.31	17.24	17.33	39.00	0.80	8.87	1.21
Th	119	9	11	43.4	111	35.2	6.6	27	2.1	3.8	0.8	51.9	163	38.7
U	450	15.7	44.7	100	151	469	316	19.8	11.2	8.2	1.1	44.6	12.5	22
Th/U	0.3	0.6	0.2	0.4	0.7	0.1	0.0	1.4	0.2	0.5	0.7	1.2	13.0	1.8
La	13.7	6.0	7.0	19.7	75.2	100.0	43.5	44.9	15.2	9.1	3.3	3.9	5.0	142.0
Ce	35.9	14.0	15.8	47.6	162.0	312.0	109.0	99.4	36.6	23.4	6.5	15.3	13.2	291.0
Pr	4.36	1.37	1.73	6.32	14.40	47.50	15.50	11.47	4.88	2.72	0.64	1.56	1.67	30.00
Nd	21.0	4.7	6.6	26.5	50.0	222.0	67.5	42.5	19.8	10.8	2.1	7.2	6.9	102.0
Sm	5.99	1.28	1.24	6.89	7.99	68.50	14.50	8.33	6.07	2.30	0.58	1.93	2.21	11.30
Eu	1.05	0.50	0.45	1.36	1.23	6.74	2.70	1.08	1.11	0.66	0.32	0.27	0.36	1.39
Gd	5.67	1.55	1.27	7.55	6.35	79.70	13.60	7.47	7.25	2.49	0.49	2.06	2.68	5.33
Tb	1.22	0.34	0.24	1.52	1.29	16.30	2.46	1.18	1.25	0.40	0.08	0.42	0.39	0.66
Dy	8.23	2.26	1.78	9.96	8.53	108.00	15.30	7.73	9.07	3.04	0.57	3.62	2.83	2.83
Ho	1.62	0.49	0.53	2.18	1.74	23.50	3.08	1.65	1.94	0.66	0.13	0.78	0.63	0.48
Er	4.97	1.71	2.40	6.67	5.25	73.10	9.84	5.33	6.16	2.29	0.39	2.92	1.93	1.35
Tm	0.74	0.28	0.60	1.04	0.81	10.90	1.67	0.88	1.00	0.36	0.06	0.63	0.32	0.20
Yb	4.26	1.89	5.72	6.60	5.11	68.74	10.73	5.78	5.97	2.52	0.46	4.95	2.10	1.10
Lu	0.51	0.26	1.19	0.86	0.60	8.13	1.42	0.81	0.84	0.37	0.08	0.77	0.33	0.17
ΣREE	109.2	36.6	46.6	144.8	340.5	1145.1	310.8	238.5	117.1	61.1	15.7	46.3	40.6	589.8

The 'hybrid index' refers to the CaO + MgO + FeO (wt. %) in each pegmatite.

The 'ASI' is the Alumina Saturation Index ($ASI = Al_2O_3 / (CaO + Na_2O + K_2O)$ in mol. %).

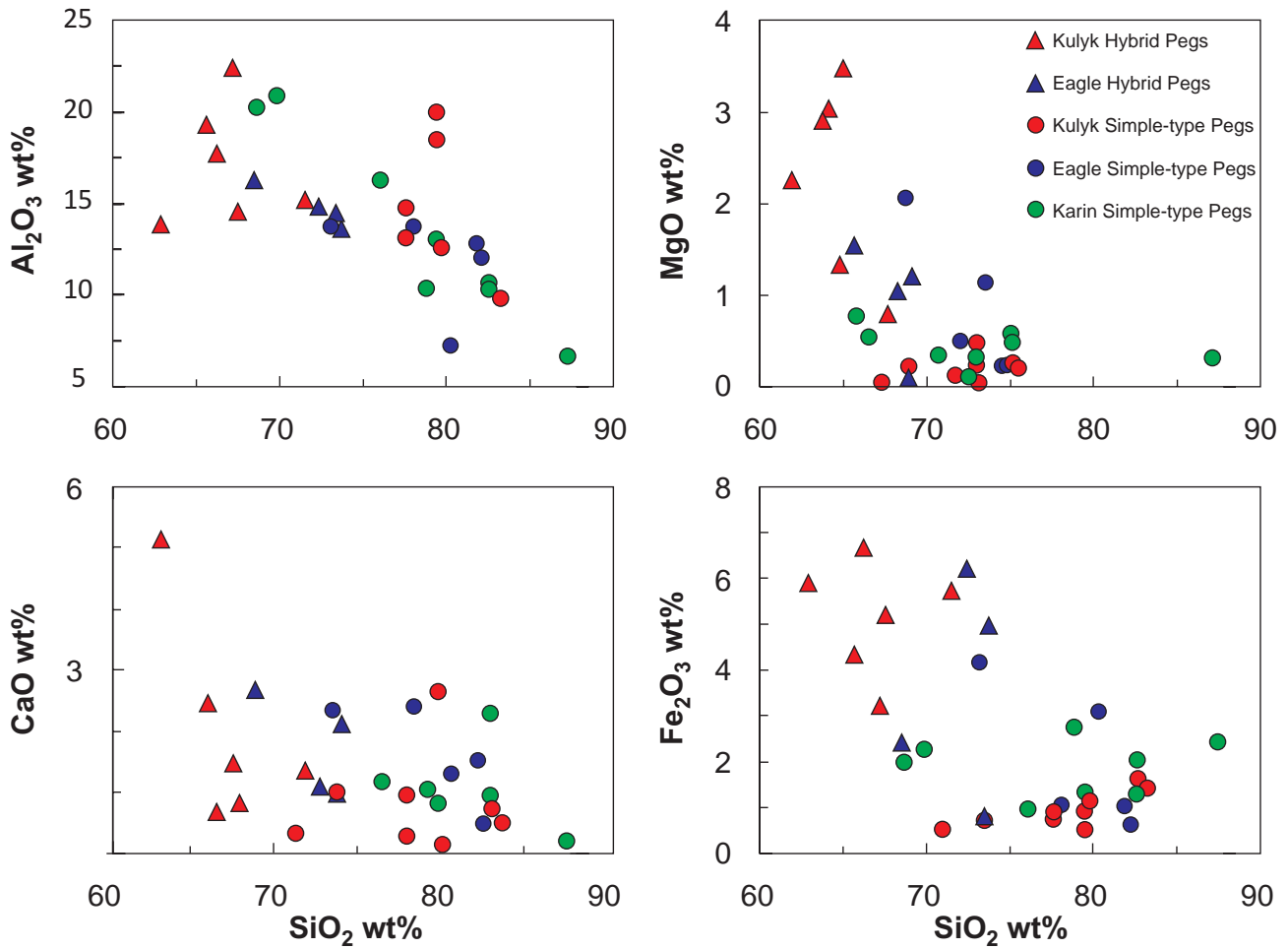


Fig. 3 Whole-rock variations of Al_2O_3 , MgO , CaO , and Fe_2O_3 (wt. %) vs. SiO_2 (wt. %) of the Wollaston pegmatites. The data represent compositions from pegmatites in the Kulyk, Eagle, and Karin lakes region discussed in this study.

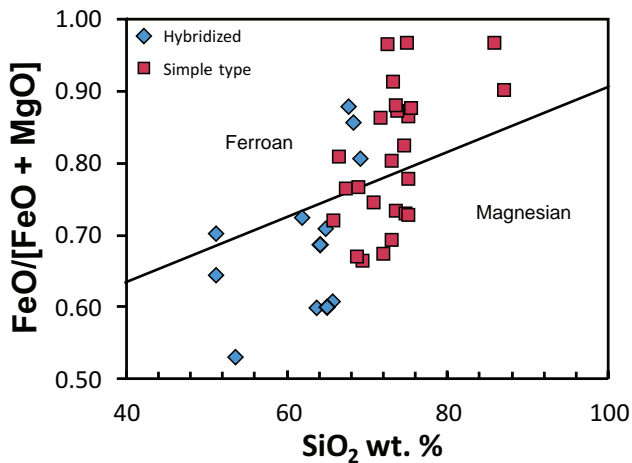


Fig. 4 The $\text{FeO}/(\text{FeO} + \text{MgO})$ versus SiO_2 diagram showing the boundary between ferroan and magnesian intrusions of Frost et al. (2001).

granitic pegmatite (Fig. 10a–b). The granites sampled in close proximity to some of the pegmatites at Kulyk Lake and Karin Lake are plotted for comparison, in order to further emphasize the distinct differentiation

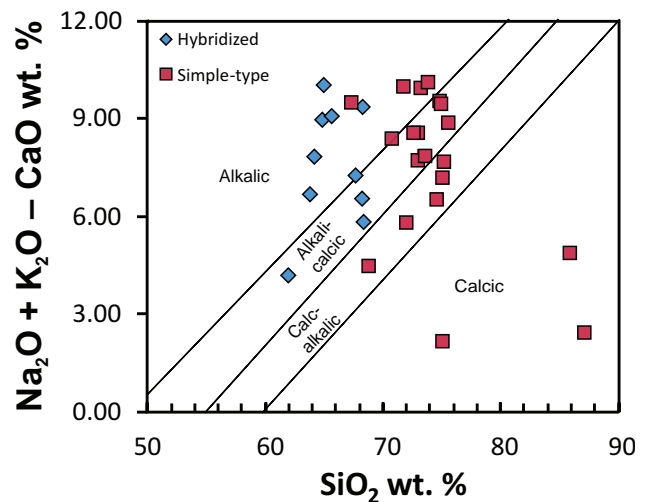


Fig. 5 The $\text{Na}_2\text{O} + \text{K}_2\text{O} - \text{CaO}$ vs. SiO_2 diagram (Frost et al. 2001) showing the approximate range for the alkalic, alkali-calcic, calc-alkalic, and calcic series.

styles of hybridized pegmatite relative to “normal” granitic signatures. The majority of the simple-type

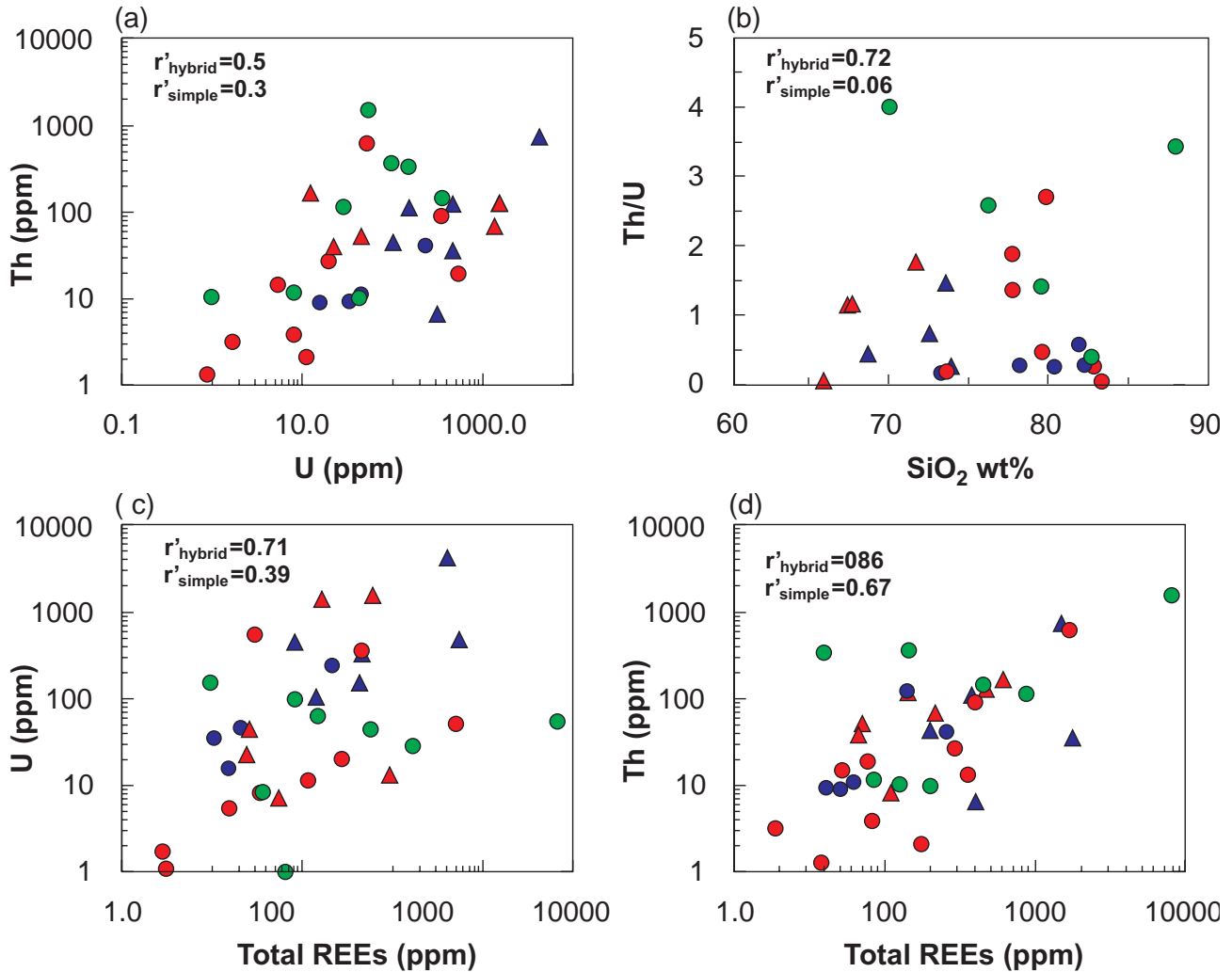


Fig. 6a – Correlation between U and Th for the simple-type pegmatites ($n = 25$) and the hybridized pegmatites ($n = 13$) from all of the study regions. **b** – Correlation SiO₂ vs. Th/U, reflecting the saturation of the radioactive phases in the melt (with hybridization); the simple-type granitic pegmatites strongly decrease in Th/U with increasing SiO₂, the hybridized pegmatites overall have low SiO₂ and Th/U ratios of 1.6. **c** – Total REE vs. U. **d** – Total REE vs. Th, further emphasizing the strong correlation between the hybridized pegmatites.

granitic pegmatites follow the normal granite trend. Ratios of K/Rb vs. Cs (see Černý et al. 1985) are good proxies to evaluate the K–Rb and K–Cs substitution in potassium feldspar and micas within the bulk samples (Fig. 10c–d). The granites differ from the granitic pegmatites by having significantly lower Rb, but comparable Cs contents. The hybridized pegmatites follow a similar fractionation pathway on the K₂O/Rb vs. Rb and K₂O vs. Cs plots (Fig. 10c–d). The trends are very steep, increasing sharply in Rb and Cs, on separate fractionation trajectories than the simple-type pegmatites and granites. The granites and simple-type pegmatites clearly lie on a shallower K₂O/Rb vs. Rb path, but are parallel to the hybrid pegmatite K₂O vs. Cs path (Fig. 10c–d). The hybridized pegmatites and simple-type granitic pegmatites and granites are grouped separately, as illustrated by K/Cs vs. REE and K/Cs vs. U + Th diagrams (Fig. 11a–b). The figures show that

low K/Cs ratios (K/Cs < 1) are attributed to hybridized pegmatites, which are consistent with higher total REE and U + Th, although there is some overlap between the mineralized simple-type granitic pegmatites.

Variations in Nb/Ta and Zr/Hf also infer a moderate to high degree of fractionation in the Wollaston NYF-type pegmatite melts (Fig. 12). The Nb/Ta ratios for Kulyk, Eagle and Karin Lake pegmatites range between 1.0 and 28.5. The average Nb/Ta for hybridized pegmatites (Kulyk and Eagle lakes) is 9.8, and 11.1 for the simple granitic pegmatites. The Zr/Hf ratios for the all studied pegmatites range from 16.4 to 39.6. There is no significant difference in Zr/Hf ratios between the hybridized and simple-type pegmatites. The pegmatites at Karin Lake differ from all of the pegmatite groups; there is an increase in Hf, with relatively constant Zr/Hf (Fig. 12c–d), and increasing Hf and Zr with increasing Th/U.

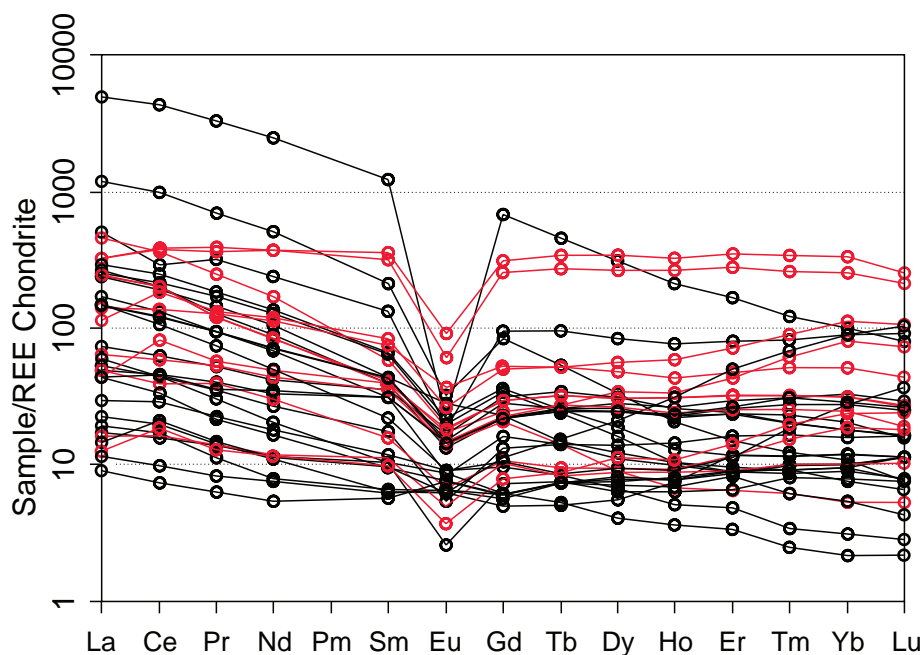


Fig. 7 Chondrite-normalized REE diagram (average chondrite values after Boynton 1984) for the simple-type pegmatites (black) and hybridized pegmatites (red).

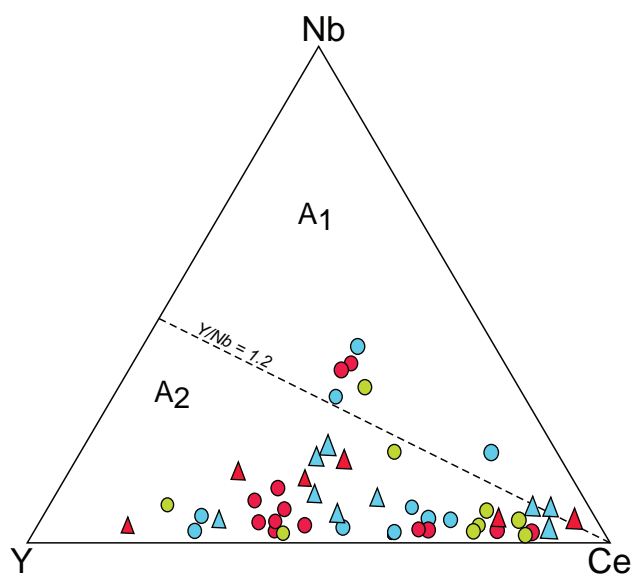


Fig. 8 Y–Nb–Ce ternary diagram (Eby 1992) for Wollaston pegmatites in the Kulyk, Eagle, and Karin lakes regions, discriminating between mantle-derived (A_1) and crustally-derived (A_2) A-type granites.

6. Temperature evaluation

Temperature estimations are based on oxide data from EDAX analyses of coexisting magnetite and ilmenite (Tab. 3; McKeough 2013). The used program ILMAT (LePage 2003) calculates temperature and oxygen fugacity using empirical methods of Powell and Powell (1977), Spencer and Lindsley (1981), and Andersen and Lindsley (1985). The temperature for ilmenite–magnetite pairs from a weakly hybrid pegmatite at Eagle Lake (MMELR010) and a simple-type granitic pegmatite from Kulyk Lake (MMKJR008) is 605–789 °C. The oxygen fugacity obtained for both pegmatites ranges between a $\log f(\text{O}_2)$ of –11.5 and –20 (Tab. 4). The broad range in temperature estimates, specifically the lower (underestimated) temperature range, is due to a lower Ti-in-magnetite composition used in this study compared to the higher Ti-in-magnetite used for the calibration of the magnetite–ilmenite geothermometer. The temperature estimates of pegmatites from Eagle and Kulyk lakes overlap temperature estimates of titaniferous magnetite and ilmenite from NYF pegmatites at Fraser Lakes, Wollaston Domain (20 km northeast of the study area), which average 750 °C (McKechnie et al. 2012a, b).

Tab. 4 Average temperature and oxygen fugacity of selected pegmatites from the Kulyk and Eagle lakes regions of the Wollaston Domain

Methods	Average T °C	Range T °C	Average $f(\text{O}_2)$	Range $f(\text{O}_2)$
Powell and Powell (1977)	683	605 to 728	–16.0	–13.0 to –20.0
Spencer and Lindsley (1981)	709	644 to 770	–14.2	–12.3 to –16.8
Anderson and Lindsley (1985)	728	664 to 775	–14.2	–12.7 to –16.2
Nasir (1994)	745	662 to 789	–13.2	–11.5 to –15.8
All methods	716	605 to 789	–14.4	–11.5 to –20.0

Wollaston Domain (20 km northeast of the study area), which average 750 °C (McKechnie et al. 2012a, b).

The melting temperature of the pegmatite can also be estimated using monazite (Montel 1993) and zircon saturation models (Watson and Harrison 1983). These phase saturation thermometers assume that the

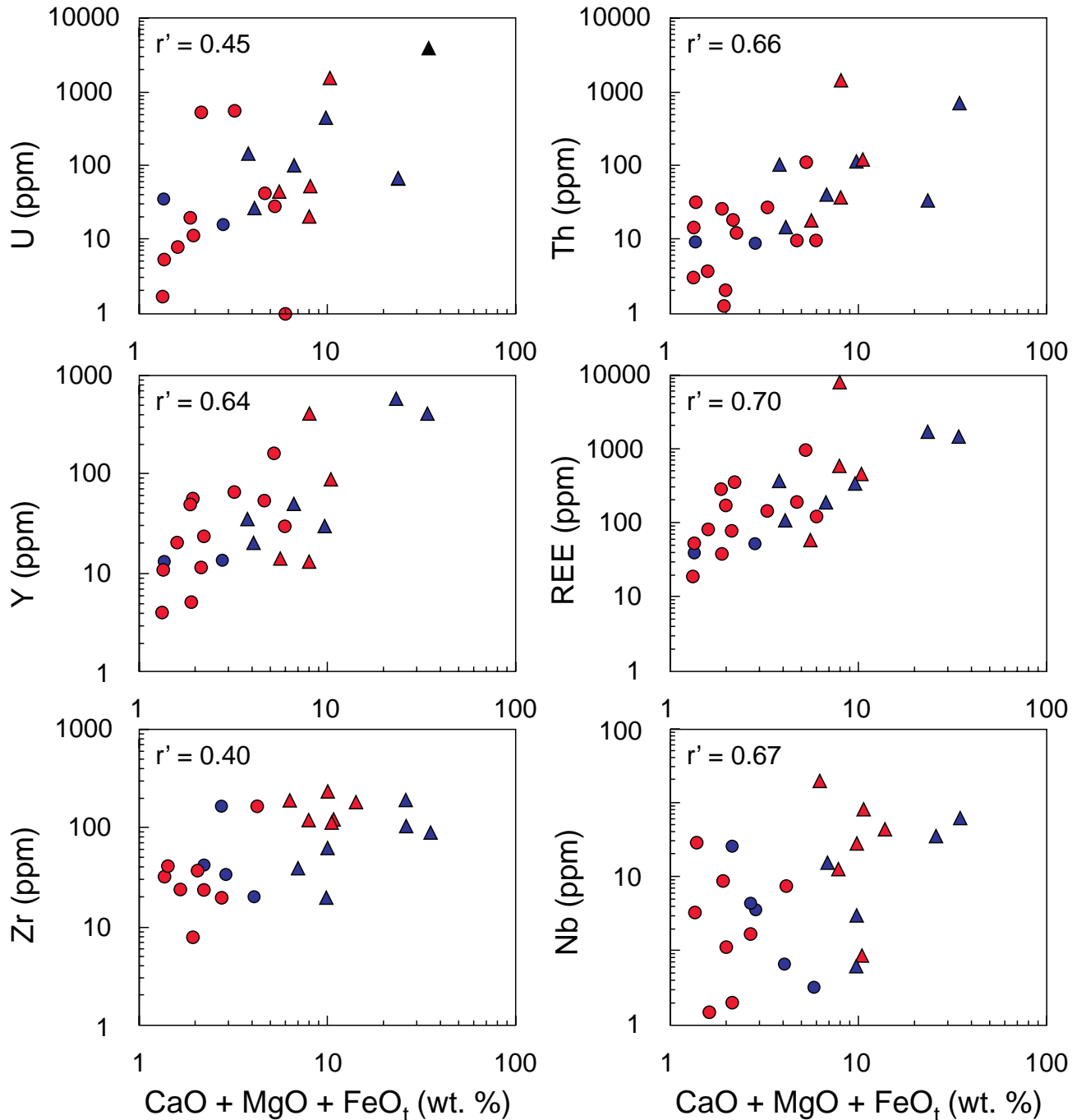


Fig. 9 A ‘hybridization index’ of $\text{CaO} + \text{MgO} + \text{FeO}_t$ is used to show comparisons between P_2O_5 , TiO_2 , U, Th, ΣREE , Nb, and Zr contents in the Wollaston pegmatites. Spearman’s rank correlation coefficients (r) are presented at a 95% confidence level with a critical number (p -value) of 0.37 for the sample size of $n = 30$ (see McKeough and Lentz 2011). Statistically, if the correlation values are above the critical number 0.37, they represent a moderate to strong relationship between hybridization effects and enrichment of the mineralizing phases.

given accessory grew from the melt and was not inherited, but also that Zr and LREE contents in the source were high enough to achieve the saturation in the melt. The decrease in Th/U (and total REE) with increasing SiO_2 (Fig. 6b) documents fractionation of monazite and zircon, implying that not all of the accessory phases are

inherited. The calculated monazite saturation temperatures exhibit a range (625–894 °C), similar to calculated zircon saturation temperatures (609–948 °C), although it is evident that inheritance accounts for the higher temperature values and only the lower temperatures reflect the likely crystallization conditions.

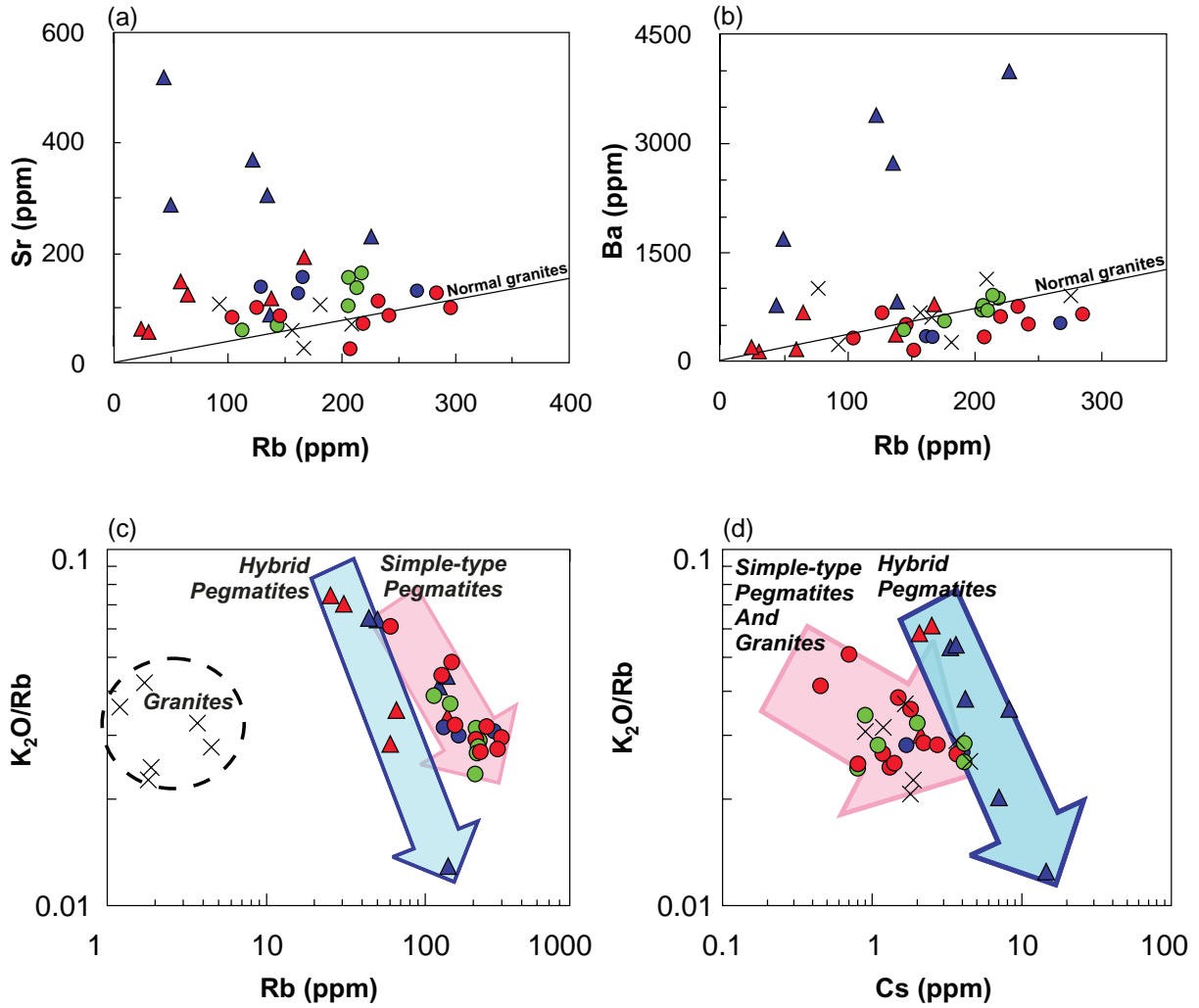


Fig. 10a – Rb vs. Sr and **b** – Rb vs. Ba plots showing variable degrees of fractionation within the Wollaston simple-type and hybridized pegmatites and granites; the trend for normal granites (El Bouseily and El Sokkary 1975) is plotted for comparison. **c** – K_2O/Rb vs. Rb and **d** – K_2O/Rb vs. Cs plots both evaluate the K–Rb and K–Cs fractionation in simple-type and hybridized pegmatites.

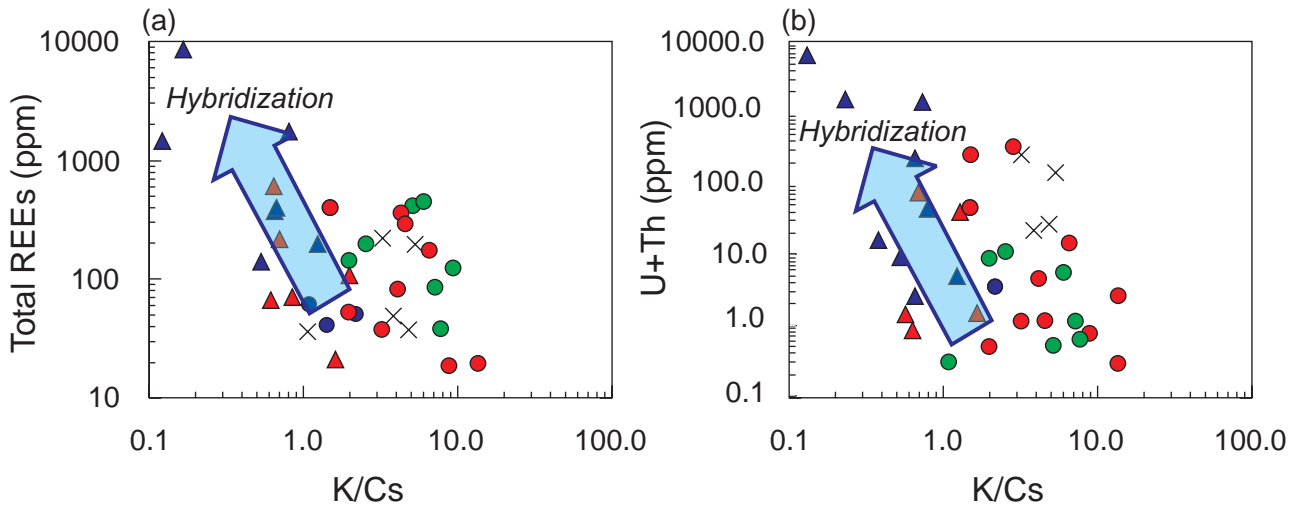


Fig. 11a – K/Cs vs. Total REE (ppm) and **b** – K/Cs vs. U + Th (ppm) plots, with arrows indicating the direction of progressive fractionation, leading to enrichment in U + Th (ppm) and REE (ppm); further evidence linking pegmatite fractionation-hybridization and the enrichment of U–Th–REE.

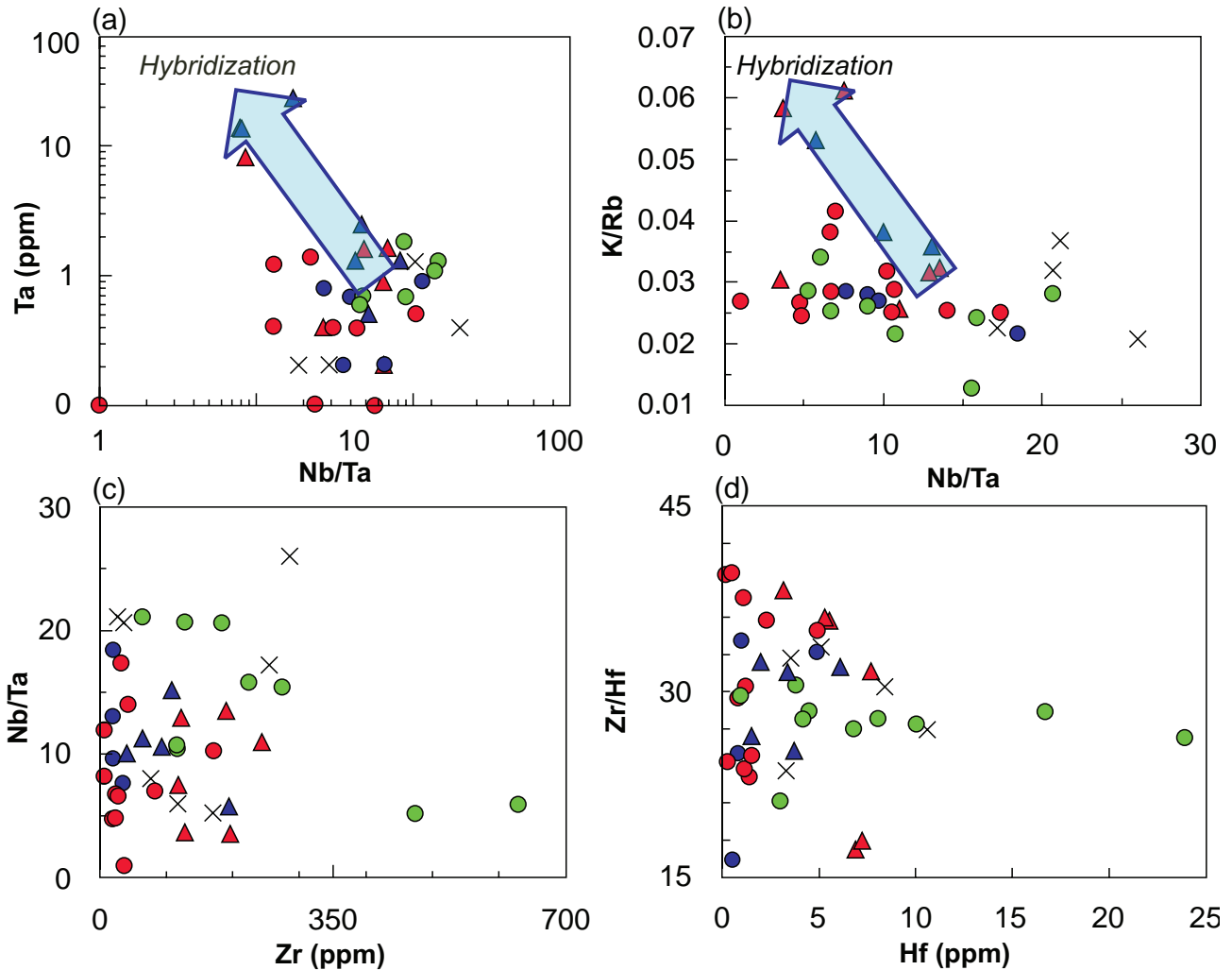


Fig. 12a – Nb/Ta vs. Ta and **b** – Nb/Ta vs. K/Rb plots show variations in Nb/Ta indicate fractionation between simple-type pegmatites vs. hybridized pegmatites; the arrows indicate the direction of a more evolved (i.e. fractionated) melt undergoing hybridization. **c** – Zr vs. Nb/Ta and **d** – Hf vs. Zr/Hf plots reflect relatively consistent Zr/Hf ratios, except in Karin Lake peraluminous pegmatites.

7. Discussion

7.1. Pressure and temperature constraints

The knowledge of pressure–temperature conditions of pegmatite formation is necessary in order to model petrogenetic evolution. A P–T path for the Wollaston pelitic gneisses has been determined based on detailed metamorphic equilibria, and defines a partial melting zone at depth and an emplacement path that is consistent with prograde near-peak metamorphic conditions of 6–9 kbar and ~750 °C (Annesley et al. 2005). The U–Pb age (*c.* 1800 Ma) and relatively high-T partial melting conditions (~750 °C) confine the origin of the pegmatite-generating melts to syn- to post-deformational events of the Trans-Hudson Orogeny.

Petrographic studies have revealed the presence of garnet and cordierite in the pegmatites (at least in the Karin

Lake region), and in similar pegmatites of the Fraser Lakes area in the Wollaston Domain (e.g., McKechnie et al. 2012a, b) indicating a high temperature of intrusion. The geochemical reactions include: biotite + sillimanite + quartz ↔ garnet + cordierite + K-feldspar + melt and biotite + sillimanite + quartz + plagioclase ↔ garnet ± cordierite ± K-feldspar + melt, and reflect partial melting that occurred under fluid-absent conditions involving biotite breakdown at moderate to high temperatures (e.g., Christiansen et al. 1983; Clemens et al. 1986; Clemens 2006; Brown and Korhonen 2009; Brown 2010). The clearly hypersolvus composition (perthitic exsolution in K-feldspar) provides additional evidence for higher T pegmatite emplacement (e.g., Parsons and Brown 1984).

The path of the segregated partial melt forming in rocks undergoing adiabatic decompression is indeed steep, and would bring about the subsolidus dehydration of the biotite to form metamorphic orthopyroxene. The

initial segregation of the fluid-absent melt would have occurred because the magma inevitably retains a certain H₂O-content at its pressure of formation (e.g., Clemens and Droop 1998). The generation of melts with H₂O activities less than total pressure is the key to their mobility and differentiation. Consequently, during adiabatic decompression the pegmatite (granitic) melts intruded and crystallized into higher structural levels in the Wollaston Domain.

7.2. Structural controls on pegmatite emplacement

After anatexis within the thickened crust during the early deformation stages of the Trans-Hudson Orogeny, crustal melts migrated upwards along major transcurrent shear zones. The Kulyk and Eagle lakes simple-type and hybridized pegmatites formed in steep structures discordant to the S₂ foliation of the host rocks. The overall features of the pegmatites at Kulyk and Eagle lakes indicate that they were injected into dilatant regions in and around shear zones in the host rocks, propagating through low-stress domains. The steep pegmatite bodies were injected E–W adjacent to NE–SW-trending dextral faults, representing lateral expansion sub-orthogonal to the principle compression direction (e.g., Lopez-Moro et al. 2012). On the other hand, at Karin Lake a majority of the pegmatites are concordant to the metasedimentary gneisses. The host rocks to the pegmatites are characterized by tensile-strength anisotropies, such as pre-existing fractures, cleavage, schistosity, and layering. These planes of weakness would play an important part in controlling the orientation of the pegmatites, however variations in the tensile strength combined with lithostatic and directed stresses must also be considered when evaluating pegmatite intrusion (Brisbin 1986). The pegmatites would have intruded at sites where pegmatite fluid pressures overcame the lowest combined effects of normal and tensile strengths (e.g., Brisbin 1986). Steep structures were vital for the accumulation and segregation of these melts, as well as channeling fluids that facilitate geochemical reactions (i.e., hybridization).

The upward migration of the melts proceeded at faster rates during exhumation; therefore the later (most fractionated) pegmatites intruded at higher crustal levels compared to earlier melts in the Wollaston Domain.

7.3. Textural and mineralogical considerations

In the Kulyk and Karin lakes regions, it is common to see aplites cross-cutting pegmatitic dykes that are otherwise compositionally similar. The cross-cutting relationships of the texturally different aplitic/pegmatitic dykes indicate multiple felsic melt injections in the domain. Textur-

al evidence also suggests that the Kulyk Lake and Eagle Lake pegmatites were intruded into low-stress domains. At Karin Lake, the quartz in pegmatites is strained, and accompanied by fine-grained, oriented mica and/or albite. These textures support the idea that the Karin Lake pegmatites were emplaced into a shear zone that was locally reactivated prior to complete crystallization.

There are distinct mineralogical differences between the K-feldspar–quartz-rich pegmatites and plagioclase–quartz-rich pegmatites, as well as the simple-type granitic pegmatites and hybridized pegmatites. The plagioclase–quartz-rich pegmatites at Karin Lake exhibit an S-type affinity, while the K-feldspar–quartz-rich pegmatites are dominantly A-type. The plagioclase–quartz-rich pegmatites do not show any characteristics of hybridizing with adjacent metasedimentary rocks. It is the K-feldspar-rich pegmatites, specifically at Eagle and Karin lakes, that show evidence of hybridization and/or metasomatic reactions along pegmatite–wallrock contacts and locally into more internal zones of the pegmatite. The compositional differences between the individual pegmatite groups are likely due to the distinct sources, genesis and timing within the Trans-Hudson Orogeny. For example, the presence of magnetite and K-feldspar ± biotite in the simple to complex (hybridized) pegmatite indicates that more oxidizing conditions prevailed at crystallization. Without age constraints it is difficult to distinguish between the different pegmatite groups, or to assign them to specific generations of pegmatite formation. We can only conclude from cross-cutting relationships that the K-feldspar-rich are younger than the plagioclase-rich pegmatites in the region.

The gradational transition between mineralogical zones (hybrid border to the central zone) in the pegmatites, and between the hybridized pegmatites and their host rocks, indicates that there may have been further “mixing” during and after emplacement of the melt (e.g., Novák et al. 1999; Dini et al. 2004; Novák 2007). Infiltration of a volatile phase, such as H₂O or F and the separation of a supercritical fluid (SCF) from the crystallizing pegmatite may have been responsible for the formation of the “pegmatitic” textural zones, as well as the net transfer of exsolving, metalliferous SCF out from the crystallizing pegmatite (e.g., Lentz and Suzuki 2001). The textural and mineralogical differences between the simple-type and hybridized pegmatites imply that an active chemical potential gradient existed between the hybridizing contact zone and the internal portion of the injecting pegmatite melt. This chemical gradient would have enhanced the diffusion of U, Th, REE, Y, Nb, and other HFSE to the outer reaction interface (hybrid border zone). It is these reactions that account for the intrapegmatite compositional differences, and the variations between mineralized hybrid pegmatites and barren non-hybridized pegmatites in the Wollaston Domain.

7.4. Geochemical aspects of the pegmatites

In addition to McKeough and Lentz (2011), we have elaborated on the geochemical evidence for hybridization reactions, which have resulted in extreme geochemical differences between the studied Wollaston pegmatites. The presence of metasomatic reaction zones and “hybrid” (exococontact) border zones, as well as geochemical variations in these NYF-type granitic pegmatites indicate the operation of assimilation and fractional crystallization (AFC) processes. Processes such as assimilation, crystal fractionation, hybridization (before and during volatile saturation), and autometasomatic reaction all contributed to the pegmatites’ trace-element signatures. This interaction has been the focus of many studies that described the processes in which host rock interactions enriched the pegmatite margins in U, Th, and REE accessory phases (e.g., Simmons and Heinrich 1980; Rimsaite 1985; Morgan VI and London 1987; Williams, 1987; Kretz et al. 1989; Owen 1989; Shearer et al. 1992; Lentz 1996). These studies gave examples of enriched granitic pegmatites that range from unzoned to strongly zoned, and can be compositionally simple or complex. Good examples of these are the mineralogically complex pegmatites in the Grenville Province, which are interpreted as products of hybridization, based on field observations (Rimsaite 1985; Lentz 1996). Similar to the Grenville pegmatites, the whole-rock relationships between simple- and complex-type Wollaston pegmatites reflect variations between the two types of pegmatite groups. The inverse relationship between whole-rock SiO_2 and CaO, FeO, and MgO (Fig. 3) can be explained by the lower proportion of quartz (SiO_2) in the hybridized pegmatites, which are more enriched in biotite, diopside, actinolite, magnetite, titanite, and ilmenite.

The AFC processes were responsible for the varying ratios between Rb–Ba–Sr and contrasting Cs vs. K/Rb (Fig. 10) for distinctive textural and compositional evolution paths (e.g., Shearer et al. 1992). The K/Cs vs. U + Th and K/Cs vs. ΣREE diagrams illustrate similar degrees of increasing fractionation (Fig. 11). Therefore, we can assume that with increasing degrees of fractionation, the abundance of U–Th–REE increased in the pegmatites. The hybridized pegmatites are more evolved, exhibiting greater enrichment of U–Th–REE; their fractionation path was ultimately affected by the increase of volatiles to the melt. The REE profiles of all of the Wollaston NYF-type granitic pegmatites in this study are characteristic of a complex interrelationship between melt fractionation and multiple injections of magma (e.g., Simmons et al. 1987). The Wollaston pegmatite REE profiles are consistent with previously studied LREE-enriched NYF pegmatites, in having weakly to moderately negative Eu anomalies. The evolution of these REE patterns, with increasingly

negative Eu anomalies, suggests moderate plagioclase fractionation in the pegmatites, possibly modified by feldspathization of calcic plagioclase under oxidizing conditions (Fowler and Doig 1983a).

This study has shown there is geochemical evidence of internally derived mineralizing fluids, i.e., fluid fractionation, which has modified ratios, such as Nb/Ta, K/Rb and Zr/Hf (Fig. 12). These have been used to explain fractionation of other rare metal granitic pegmatites (Pan 1997; Linnen 1998; Dostal and Chatterjee 2000; Linnen and Keppler 2002), whereby all three ratios will decrease with crystal fractionation and with the introduction of fluids. Primitive-mantle melts have rather constant Nb/Ta ratios of 17.5 ± 2 (Hofmann 1986; Green 1995), a value comparable to that of chondrites (Sun and McDonough 1989). However, there is controversy on whether a typical continental crust value is within the range of the mantle-derived melts (Hofmann et al. 1986; Jochum et al. 1986) or is significantly lower (~ 11 ; Taylor and McLennan 1985; Green 1995), implying that these two elements were fractionated from each other in the continental crust (e.g., Linnen and Keppler 2002). The Wollaston pegmatites show distinct fractionation trends when comparing the Nb/Ta vs. Ta and vs. K/Rb ratios, as a result of hybridization reactions (Fig. 12a–b). There is a sharp decrease in Nb/Ta, specifically within the hybridized pegmatites. The simple-type granitic pegmatites still show a negative correlation between K/Rb and Ta, albeit as a much shallower trend. Decreasing Nb/Ta is typical of these types of NYF granitic pegmatites, and may indicate a high activity of F, promoting the fractionation of Nb and Ta (Černý et al. 1986; Černý 1991). The crystallization of accessory phases such as rutile and ilmenite, which preferentially incorporate Nb over Ta, may be responsible for the decrease of Nb/Ta ratios in residual peraluminous granitic melts (e.g., Linnen and Keppler 1997). We conclude that the lower Nb/Ta ratios in the more evolved (hybridized) pegmatites could have resulted from the crystal/liquid fractionation of Ti-rich minerals. The observed constant Zr with decreasing Nb/Ta was probably related to the abundance of restitic zircon buffering the whole-rock Zr contents. The Zr/Hf ratios for the pegmatites remain relatively constant, except for the strongly peraluminous pegmatites at Karin Lake (Fig. 12d). The latter contain abundant garnet (up to 5 vol. %), which prefers Zr over Hf, therefore producing a decrease in Zr/Hf with fractional crystallization (Linnen and Keppler 2002). The Zr/Hf vs. Hf plot for the Karin Lake pegmatites is quite typical of moderately fractionated peraluminous granites (e.g., Linnen and Keppler 2002); otherwise the Zr/Hf ratio is less than chondritic value. Hafnium and Zr concentrations should remain constant in unfractionated igneous rocks, having chondritic values between 33 and 40 (Jochum et al. 1986). Fluid metaso-

matism has been proposed to account for non-chondritic Zr/Hf values in granitic rocks (e.g., Bau 1996), but it is suggested that this ratio can reflect crystal fractionation involving accessory phases in the pegmatite as well (e.g., Pan 1997; Linnen 1998; Linnen and Keppler 2002). The decrease in Zr/Hf of the studied pegmatites is comparable to the moderately to highly fractionated granites associated with F-rich melts (Linnen and Keppler 2002).

The above geochemical data interpretations additionally support hybridization reactions between hypersolidus–subsidius pegmatite and host rock, likely aided by volatiles that effectively enhanced pegmatite crystallization. Network-forming cations (Al and Si) and U, Th, REE and other HFSE would diffuse during viscous stress relaxation in melts, which typically occurs with the addition of volatiles (e.g., Bagdassarov et al. 1993; Mungall 2002). Experimental studies of volatile-rich pegmatite melts (Bartels et al. 2011, 2012) have also shown the effectiveness of the of U, Th, and REE diffusion related to the changes in residual melt composition, either as a result of fractionation or volatiles loss. We have proposed that the separation of a SCF and infiltration of a volatile phase through crystallized portions of the pegmatite (and eventually the host rocks) would facilitate metasomatic transfer, i.e., assist in the diffusion and advection of elements, such as rare alkalis and HFSE from the melt. The SCF phase would enhance the autometasomatic reactions along the chemical potential gradient between the host-rock major elements (e.g., Ca, Fe, Mg and Ti) and the pegmatites (e.g., Si, Al, K, Na, U, Th, REE, Nb, Zr, F, and Cl). Fluid–melt partitioning would enrich the SCF in HFSE (U, Th, REE, Zr, and Nb) during diffusive exchange and/or infiltration (interaction) with the outer portions of the pegmatite undergoing hybridization. This may explain why the simple granitic pegmatites, as well as the central zones of the hybridized pegmatites, are depleted in U, Th, REE, Y, and Nb accessory phases.

7.5. U, Th, REE, Y, and Nb enrichment processes

Analogous to the study of Lentz (1991) in the southwestern Grenville province pegmatites, *in situ* gamma-ray spectrometry proved that the hybrid and metasomatized contact (border) zones correspond to the greatest enrichment in U and Th (see McKeough et al. 2010). Detailed mapping of mineralized pegmatites and petrographic analysis of the hybridized pegmatites have provided further evidence that allanite, monazite, apatite, garnet, xenotime, uraninite, and other U-, Th-, and REE-bearing minerals are concentrated within the metasomatized and hybridized contact-zone (Tab. 2), similar to other fractionated NYF-type pegmatites (e.g., Shearer et al. 1992).

Most fertile crustal lithologies contain sufficient amounts of REE, Y, Zr, and P to supply an anatectic melt in these components and allow early precipitation of accessory phases (Watt and Harley 1993). However, the evolved NYF pegmatitic melts in the Wollaston Domain were undersaturated in REE; therefore, the primary deposition of U, Th, and REE occurred later, at lower temperatures. Early crystallization of apatite, allanite, and/or titanite was favored in the metaluminous melts at Kulyk and Eagle lakes, and the crystallization of monazite and/or xenotime in peraluminous melts at Karin Lake. Previous studies on mineralized pegmatites showed coexisting U–Th–REE–Y accessory phases in an intermediate field of a slightly peraluminous hybrid pegmatite, with moderate CaO content (less than 2.5 wt. % – e.g., Dini et al. 2004). Stability of accessory phases, such as monazite, allanite, and apatite are mainly controlled by the activity of Ca and degree of peraluminosity of the melt (Dini et al. 2004). We have shown (in this study, and McKeough and Lentz 2011) that there is a distinct correlation between the CaO + MgO + FeO_i (hybridization index) and U, Th, ΣREE, Y, and Nb contents of the pegmatites, confirming that the saturation of relevant minerals in the hybrid border and wall zones was directly related to the introduction of Ca, Mg and Fe, caused by hybridization reactions. The hybridized pegmatites have higher bulk concentrations of P₂O₅ (compared to other pegmatites in the region), providing phosphorus to form xenotime, monazite and apatite. Therefore we can assume that chemical potential gradients between the melt and/or fluid from core (P₂O₅) and hybrid border (CaO) of the pegmatite favored saturation of U–Th–REE in the hybridized zone. The higher contents of allanite, uraninite, thorite, monazite, xenotime, and apatite in the hybrid border compared to pegmatite intermediate and core zones indicate high diffusivities within and between pegmatite and host rock for LREE, U, and Th.

7.6. The effect of fluxes on melt enrichment

Research by London (1987), London (2008), Simmons and Webber (2008), Nabelek et al. (2010), Van Lichtenvelde et al. (2010), and Linnen (2012) have shown that U-, Th-, Nb-, Y- and REE-minerals are closely linked to highly fluxed silicate melts. Fluxes, including F and P, are important for the transport and deposition of these metals, depending on the melt composition. However, the role of fluorine in NYF-type pegmatites is still poorly understood (Linnen 2012).

There is relatively high F-content (> 1 wt. %) in metasomatic biotites from pegmatites sampled at Kulyk Lake and Eagle Lake (McKeough 2013). Fractionation in the Wollaston pegmatites was likely enhanced by F

(and P), in addition to providing a possible mechanism for saturation of the U-, Th-, and REE-bearing phases. Even at low concentrations, fluorine may influence fractionation of REE and the transition elements (e.g., Mahood and Hildreth 1983). At high concentrations, the two elements reduce liquidus and solidus temperatures, decrease viscosity, and increase the solubility of H₂O in the melt (London et al. 1993; Dingwell et al. 1996; London 1997; 2009). The incorporation and increase of fluorine concentration in these particular NYF-type melts could have led to the formation of F–REE complexes, lowering the activity of the REE, and therefore reducing REE crystal–melt partition coefficients (e.g., Ponander and Brown 1989).

8. A proposed model for the origin of the Wollaston pegmatites

A petrogenetic model for the Wollaston pegmatites must include key observations, such as high U, Th, Nb, Y, and REE contents, evidence for hybridization, crustal contamination, and absence of compositionally similar, granitic plutons in the area. We have established that the age constraints, textures and relatively high-T partial melting conditions (~750°C) confine the pegmatite melt-forming processes to late deformational events (c. 1830–1805 Ma) followed by retrograde metamorphism (c. 1770 Ma). Two models are proposed for their origin: (1) extreme fractionation of syn- to late-tectonic granites (e.g., Černý 1990; Lentz 1991, 1992, 1996; Bowden et al. 1995; Kinnaird and Nex 2007), or (2) *in situ* partial melting of U-rich metasedimentary rocks (e.g., Cuney 1980, 1982; Annesley et al. 2005).

The geological, geochemical, and geochronological evidence provided for the pegmatites implies anhydrous partial melting of a refractory crust coupled with extreme fractionation. In addition, these pegmatites are texturally and compositionally similar to numerous U–Th–REE–Mo occurrences in pegmatites of anatectic origin and related metasomatic rocks of the Grenville Orogen (Fowler and Doig 1983b; Lentz 1991, 1996 and references therein). The Grenville pegmatites are often associated with a facies containing at least 50 % mafic minerals at, or near the contact, predominantly pyroxene, with inclusions of uraninite, uranophane, magnetite and allanite (Rimsaite 1985). The Bancroft district (Grenville Orogen) U–Th–REE deposits are complex and suggested to have formed through pegmatite melt injection, accompanied by AFC processes, followed by metasomatic replacement and a magmatic–hydrothermal vein stage (Lentz 1996). Analogous to the Grenville pegmatites, the proposed fractionation model for the Wollaston pegmatites described in this study involves adiabatic decompressional partial

melting of chemically favourable crustal rocks. The geochemical evidence presented in this paper confirms that the partial melts of Wollaston granulite-facies crustal rocks show characteristics of both A-type and S-type (hybrid), H₂O-undersaturated granites. The observed enrichment in U and other incompatible elements in the Wollaston pegmatites can be explained by the low degree of partial melting. Robb (2005) showed that in order to achieve concentrations of around 300 ppm U in the leucogranitic partial melts, the degree of partial melting would have to be very low (i.e. <5 %), or the protolith had to be significantly enriched (>> 10 ppm U). The Wollaston pegmatites average 133 ppm U (simple-type) and 667 ppm U (hybrid pegmatites). They represent generations of leucogranites derived from a fertile protolith, whereby enrichment of the trace elements Rb, U, Th, REE, Zr, Y and Nb was formed by small degrees of partial melting (e.g., Rossing leucogranites – Nex et al. 2001). Further enrichment of these elements proceeded during fractional crystallization (± assimilation) and increased exponentially as crystallization proceeded, i.e., the fraction of melt decreased (Robb 2005). After an advanced degree of solidification, the U, Th, REE, Zr, Y and Nb content of the residual magma was sufficiently high and promoted thorite, allanite, xenotime, monazite, apatite, rutile, garnet, and zircon crystallization. If the pegmatites were generated from a suite of uranium-bearing metapelites, then it is anticipated that U, Th, REE, and volatiles would be depleted during the early stages of partial melting (e.g., Kinnaird and Nex 2007), however there are uraniferous pegmatites dated over long time intervals (multiple pegmatite injections), throughout the Trans-Hudson Orogeny.

A proposed multistage genetic model for the studied pegmatites is presented in Fig. 13. Stage (1) represents a basal heating event at pressures >12 kbar (lower crust) to 6–9 kbar (middle crust) during (D₂?) thrusting and folding (1830–1825 Ma), immediately followed by decompression to ~3 kbar (1820–1810 Ma) at near-peak temperatures of ~750 °C (Annesley et al. 2005). Stage (2) involved a high P–T partial melting event between metamorphic peak and pegmatite emplacement. Adiabatic decompression, accompanied with shearing and faulting, led to melt generation and its relatively rapid emplacement. At this stage, the main melting zone would progress with strain-induced melt segregation and the pegmatites intruded at sites where pegmatite fluid pressures overcame the effects of normal and tensile strengths. Increased basal heating with the associated deformation and the introduction of volatiles, assisted the intrusion of fluid rich, highly mobile NYF-type melts. The third stage (3) approached peak thermal conditions in the Wollaston Domain, whereby the partial melting front migrated upwards in the crust. Consequently the

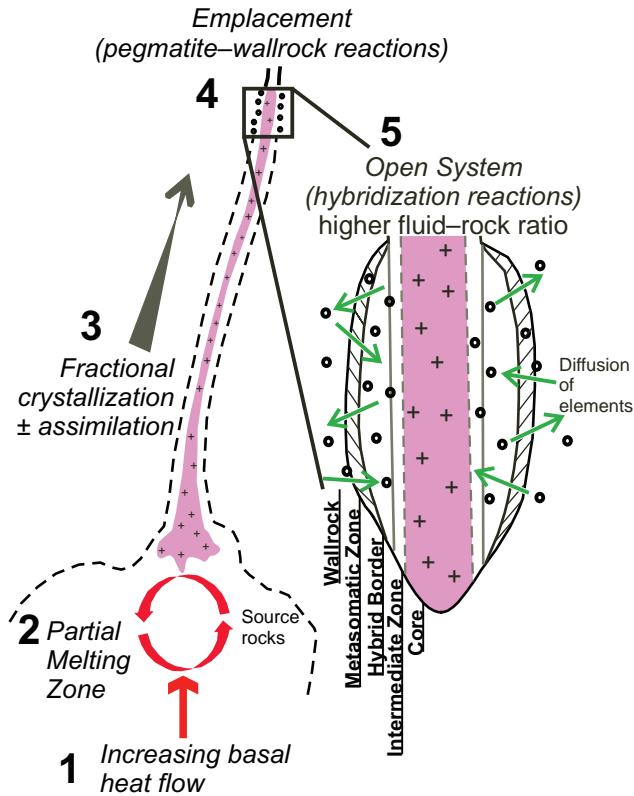


Fig. 13 A five-stage model for the evolution of the pegmatites in the Kulyk, Eagle, and Karin lakes region of the Wollaston Domain, northern Saskatchewan.

melt was transferred through conduits reaching shallow crustal levels before solidification. Melt coalescence and magma ascent was accompanied by fractional crystallization during this stage, therefore the youngest late-tectonic pegmatites at the present exposure level should be the most fractionated. The pegmatite evolved in part in an open-system environment in which AFC processes (during ascent) resulted in an exchange of accessory phases between continuous multiple pegmatite injections and the wallrock. Stage (4) was emplacement of the pegmatite, as it reached the point of crystallization. Undoubtedly the chemical reactions at stages (3) and (4) overlapped; field relationships, mineralogical, and textural evidence suggest that the hybridization and/or metasomatic reactions occurred both during ascent and emplacement. These processes took place while the pegmatite was still partially molten, but also volatile saturated, because of exocontact metasomatism is also evident. The pegmatite continued to evolve into the last stage (5), through sub-solidus alteration, i.e. infiltration of a volatile phase through crystallized portions of the pegmatite (and eventually the host rocks). It would facilitate the metasomatic transfer, i.e., assisted in the diffusion and advection of elements, such as rare alkalis and HFSE from the melt. Progressive changes in the residual melt compositions, either due

to fractionation or loss of SCF to the wall rock, would additionally influence the melt viscosity (i.e., degree of melt polymerization) and affect the diffusivities of U, Th, and REE (Bartels et al. 2011). High concentrations of F and/or P in the hydrous melt or the pegmatite-derived SCF would result in formation of complexes with HFSE and thus aid their progressive enrichment.

9. Conclusions

1. The Wollaston pegmatites association to local (steep) structures is consistent with deformation-induced rapid melting in the Trans-Hudson Orogeny (c. 1830–1805 Ma).
2. The pegmatites have originated from within the centers of crustally-derived A- or S-type (hybrid) magmas at depth. The heat loss from the plutons facilitated migration of the melt along fractures away from the main body, and thus compositionally related granites are not present at the current exposure level. These distal, i.e., the most chemically evolved, pegmatites were injected into rocks hundreds of degrees below temperatures of the pegmatite-forming melt.
3. There are two types of pegmatites in the Wollaston region. The simple-type pegmatites are K-feldspar-quartz-rich and plagioclase-quartz-rich, with minor ferromagnesian phases present. The complex-type pegmatites are composed of a more diverse suite of minerals, including abundant ferromagnesian phases, and this complex mineralogy is interpreted by interaction with the host rocks (“hybridized” pegmatites).
4. Temperature estimations reveal high-T partial melting conditions (~750 °C), confining the origin of the pegmatite-generating melts to syn- to post-deformational events of the Trans-Hudson Orogeny. Pegmatite mineralogy additionally supports partial melting that occurred under fluid-absent conditions involving biotite breakdown at moderate to high temperatures. Furthermore, petrographic studies indicate a hypersolvus composition, also supporting a high-T environment during pegmatite emplacement.
5. Textural relationships, such as the presence of crosscutting aplitic and pegmatitic dykes, and the zonation observed in hybridized pegmatites vs. simple-type pegmatites (of the same region) combined with the occurrence of pegmatite intrusions over a long time interval indicates multiple melt injections in the Wollaston Domain.
6. There are anomalous abundances of U, Th, Σ REE ± Y–Nb in the simple-type pegmatites, and, even more so, in the hybridized types. Similar to Grenville hybridized pegmatites, there is a distinct correlation of these elements with CaO + MgO + FeO_i (hybridization

index). This confirms that the saturation of minerals hosting these elements in the pegmatite hybrid border and wall zones was directly related to the mafic cations introduced by open-system reactions.

7. The contact, or border zones, of the studied pegmatites represented a dynamic environment in which hybridization processes evolved at the site of pegmatite emplacement, potentially aiding in the exchange of U, Th, ΣREE, Y, and Nb accessory phases along an evolving chemical potential gradient between pegmatite hybrid border and wall rock.
8. Field, mineralogical, and textural evidence supports the infiltration of a volatile phase and the separation of a supercritical fluid from the NYF-type Wollaston pegmatites, which resulted in the partial loss of U, Th, REE, and other HFSE to the wallrock. The postulated volatile loss effectively increased the diffusivities of the U, Th, and REE, allowing the corresponding accessories to precipitate in the hybrid border and exocontact of these pegmatites. Indeed, the simple-type granitic pegmatites, as well as the central zones of the hybridized pegmatites, are depleted in U, Th, REE ± Y – Nb accessory phases due to the same diffusion processes.
9. The geochemical data support a fractionation model of the Wollaston NYF-type pegmatites that was inevitably enhanced by added fluxes such as F (and may be P). Fluorine would form F–REE complexes, lowering the activity of the REE, and reducing REE crystal–melt partition coefficients, therefore driving U–Th–REE fractionation in the Wollaston pegmatites.

Overall, the documented case underlines the importance of open-system interactions between the moderately fractionated late-tectonic melts and their metasedimentary host rocks in origin of U–Th–REE–Y–Nb mineralized pegmatites. The metasomatic and hybridization processes have strongly affected the chemical evolution of the fractionating magmas, and were the factors critical for concentrating the U–Th–REE–Y–Nb rich accessory minerals at contacts of the hybrid pegmatites. This study has significant implications not only for similar evolved felsic igneous systems worldwide containing the same style of mineralization, but should also help to explain why other high-level fractionated pegmatite systems remain barren. The processes responsible for saturating the U–Th–REE–Y–Nb elements remain poorly understood and need further analysis.

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