The Canadian Mineralogist
Vol. 40, pp. 137-151 (2002)

THE DISTRIBUTION OF RARE-EARTH ELEMENTS IN K-FELDSPAR AS AN INDICATOR OF PETROGENETIC PROCESSES IN GRANITIC PEGMATITES: EXAMPLES FROM TWO PEGMATITE FIELDS IN SOUTHERN NORWAY

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ABSTRACT

The K-feldspar from strongly zoned gadolinite-type *REE*–Nb–Ta-rich granitic pegmatites from the Evje–Iveland and Froland pegmatite fields in southern Norway was analyzed for the entire spectrum of rare-earth elements, as well as for a suite of major and trace elements, with the HR–LA–ICP–MS technique. Earlier results imply that the two pegmatite fields are nearly indistinguishable in terms of their origin and igneous evolution. An evaluation of the distribution of K, Rb, Sr and Ba in K-feldspar, elements that are strongly sensitive to igneous processes in felsic rocks, confirm this conclusion. However, the distribution of the ultra-trace elements, and particularly the *REE*, in K-feldspar is incompatible with a common origin of the two fields. The *REE* distribution in K-feldspar is buffered by the coexisting *REE*-phases. When compared to Evje–Iveland, the Froland suite of pegmatites was derived from a relatively *LREE*-deficient and *HREE*-enriched parent melt. Finally, the distribution of trace elements in feldspar implies that the Evje–Iveland pegmatites formed from progressively more differentiated melts that were emplaced in a southward-propagating system of vertical and horizontal faults and fractures.

Keywords: granitic pegmatites, K-feldspar, trace elements, rare-earth elements, HR–LA–ICP–MS data, Evje–Iveland, Froland, Norway.

SOMMAIRE

Le feldspath potassique provenant de pegmatites granitiques fortement zonées du type gadolinite, enrichies en terres rares, Nb et Ta, et faisant partie des suites d'Evje–Iveland et Froland, dans le sud de la Norvège, a été analysé pour le spectre des terres rares en entier, ainsi que pour les éléments majeurs et en traces au moyen de la technique HR–LA–ICP–MS. Selon les résultats antérieurs, les deux champs de pegmatite n'étaient pas distinguables en termes de leur origine et leur évolution magmatique. La distribution de K, Rb, Sr et Ba dans le feldspath potassique, qui dépend fortement des processus ignés dans ces roches felsiques, concorde avec cette interprétation. Toutefois, la distribution des éléments ultra-traces dans le feldspath potassique, en particulier les terres rares, n'est pas compatible avec le modèle d'une origine commune. La distribution des terres rares dans le feldspath potassique est tamponnée par des phases porteuses de terres rares. Par rapport à la suite de Evje–Iveland, la suite de pegmatites de Froland est dérivée d'un magma parental relativement appauvri en terres rares légères et enrichi en terres rares lourdes. La distribution des éléments traces dans le feldspath indique que les pegmatites d'Evje–Iveland se sont formées à partir de liquides progressivement plus différenciés qui ont été mis en place dans un système de migration vers le sud le long de failles et de fractures verticales et horizontales.

(Traduit par la Rédaction)

Mots-clés: pegmatites granitiques, feldspath potassique, éléments traces, terres rares, données HR–LA–ICP–MS, Evje–Iveland, Froland, Norvège.

INTRODUCTION

In the present study, the concentrations of major, trace and ultra-trace elements in K-feldspar are used to understand the origin and evolution of granitic pegmatite fields. Traditionally, the origin, evolution and classification of granitic pegmatites are evaluated from detailed mineralogical studies of accessory phases. Many studies have convincingly documented the success of this approach. However, the characterization of a sufficient number of bodies of granitic pegmatite in a pegmatite field is an elaborate and time-consuming procedure. Furthermore, the complex assemblages of minerals that are used in characterizing one pegmatite field

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may be difficult to apply in comparisons with other fields. Although used extensively throughout the last century in southern Norway (*e.g.*, Andersen 1926, 1931, Bjørlykke 1935, 1937, 1939, Åmli 1975, 1977), such mineralogical studies have not resolved the geochemical and the evolutionary relationships between the economically as well as mineralogically distinctive pegmatite fields of Evje–Iveland and Froland. They share many similarities and formed close to each other; however, it is not known if the pegmatite-forming melts in Evje–Iveland and Froland were derived from a single precursor, or if multiple sources were involved in their genesis. Renewed interest in economic resources of high-purity quartz and K-feldspar from granitic pegmatites also necessitates the development of efficient and unbiased methods to document the petrogenetic relationship between mineral quality and processes of formation of granitic pegmatites (Larsen *et al*. 1998, 1999, 2000).

To address these issues, I utilize major- and trace-element characterization of K-feldspar, a common phase throughout the zoned granitic pegmatites. Some studies previously reported on the chemistry and petrogenetic significance of pegmatitic K-feldspar (Černý 1982, Shearer *et al*. 1985, Černý & Meintzer 1988, Neiva 1995). However, the present study is the first to benefit from recent developments of the high-resolution laser-ablation – inductively coupled plasma –mass spectrometry (HR–LA–ICP–MS) method. This novel microbeam technique reduces the detection limit for most trace elements by orders of magnitude and facilitates the characterization of the trace-element distribution in K-feldspar, including the whole spectrum of rare-earth elements (*REE*). Using this approach to study petrogenetic processes, it is possible to characterize large pegmatite fields in terms of their origin and evolution and to reconstruct the spatial propagation of melts prior to the emplacement of granitic pegmatites.

The Granitic Pegmatites in Evje–Iveland and Froland

Granitic pegmatites in Evje–Iveland and Froland formed at the termination of the Sveconorwegian (Grenvillian) orogeny in southern Norway. Largely, the igneous activity began around 1250 Ma (*e.g.*, Pedersen 1981, Pedersen & Konnerup-Madsen 1988, Starmer 1991) and was associated with crustal extension and formation of basic and felsic igneous rocks. Large-scale emplacement of post-tectonic undeformed plutons began around 1000 Ma with massive granite, monzonite and diorite emplacement throughout the region (Fig. 1; Andersen 1997, Bingen & van Breemen 1998, Pedersen 1981, Pedersen & Konnerup-Madsen 1988). Geochronological studies of granitic pegmatites are scarce and mainly based on single-mineral Rb–Sr analyses. One recent study from the northern part of Evje–Iveland provided a well-defined isochron age of 852 ± 12 Ma (I_{Sr} = 0.7063) based on dating of K-feldspar (Stockmarr 1994). Rb–Sr dating of biotite and muscovite from two pegmatites in Telemark and the Gloserheia pegmatite in Froland (Baadsgaard *et al*. 1984) yielded overlapping ages of 893 ± 50 and 896 ± 27 Ma (Sylvester 1964), respectively, and also indicated primitive I_{Sr} values. Euxenite-(Y) U–Pb geochronology of Gloserheia gave an upper intercept age of 1060 $^{+8}/_{-6}$ Ma (Baadsgaard *et al*. 1984). Although euxenite-(Y) U/Pb dating may be regarded as more robust than the Rb/Sr method, many of the pegmatites in the Froland area intersect granites that were dated as 926 ± 8 (titanite Pb–Pb and Rb/Sr ages by Andersen 1997), hence implying that most pegmatites in Froland are younger than the Gloserheia pegmatite.

The Evje–Iveland and Froland pegmatite fields show many similarities in terms of geometry, zoning and modal distribution of major minerals, and may to a first approximation be described as one.

Bodies of granitic pegmatite in Evje–Iveland and Froland comprise classical "chamber" pegmatites that in most examples crystallized as subvertical or subhorizontal dikes or sills (Fig. 2C) (*e.g.*, Bjørlykke 1935). They rarely exceed 20 meters in thickness, and most pegmatites have an exposed lateral extent of less than 100 meters. Most of the pegmatites in Evje–Iveland and Froland developed in mafic host-rocks consisting of amphibolite, norite and mafic gneiss. The granitic pegmatites are extremely coarse-grained, with individual crystals varying in size from decimeters to meters. The pegmatites are strongly zoned, typically comprising a wall zone (WZ), one or several intermediate zones (IZ), and one or several core zones (CZ). Decimetric crystals of K-feldspar and plagioclase, accessory biotite, muscovite and quartz dominate WZ. In some examples, the wall zone is predominantly composed of plagioclase with minor K-feldspar, quartz and biotite. A gradual but well-defined transition is seen toward IZ. At the onset of IZ, decimeter- to meter-size single crystals of biotite grow toward the interior of the pegmatite (Fig. 2A). Macroperthitic K-felspar, plagioclase and quartz are interspersed in near-equal volumetric proportions between crystals of biotite and muscovite. In most pegmatites, and in all the pegmatites included in this study, biotite is the dominant mica. Finally, IZ grades into CZ, which normally contain rafts of K-feldspar and plagioclase (Fig. 2D).

Other than perthitic K-feldspar, plagioclase, quartz, biotite and white mica, the dominant primary magmatic phases include magnetite, spessartine, a monazite-group mineral, allanite-(Ce), xenotime, euxenite-(Y), fergusonite, gadolinite-group minerals and beryl (Andersen 1926, 1931, Bjørlykke 1935, 1937, 1939, Åmli 1975, 1977). Albite of the "cleavelandite" habit, microlite and tantalite typically formed from late hydrothermal solutions replacing primary magmatic minerals in cavities and along fractures intersecting the pegmatites. However, hydrothermal replacement-induced features are uncommon in both Evje–Iveland and Froland (Bjørlykke 1935, Fought 1993, Stockmarr 1994). Recent studies document that the late magmatic volatile phase at Evje–Iveland contained medium-salinity H_2O – CO_2 – NaCl fluids with 10–15 vol.% CO_2 during the formation of IZ, and low-salinity H_2O – CO_2 – NaCl – $MgCl_2$ – $FeCl_2$ fluids with 5–10 vol.% CO_2 during the formation of CZ (Larsen *et al.* 1998, 1999). Among the accessory minerals, a garnet dominates the outer parts of the intermediate zone, and monazite, xenotime, fergusonite and euxenite-(Y) are nearly always associated with (and probably nucleated on) blades of biotite at the transition between WZ and IZ. Together, these observations indicate that these phases formed during early and intermediate stages of crystallization, whereas allanite-(Ce), gadolinite-group minerals and beryl formed in the core zone from the last fractions of pegmatite-forming melt (*e.g.*, Bjørlykke 1935).

Sampling and Analytical Strategies

In this study, it was decided to conduct laser ablation on the $Li_2B_4O_7$-fused samples, which were already produced for major-element X-ray-fluorescence (XRF) analysis. The main benefit over solution ICP–MS is that $Li_2B_4O_7$ fusion is more efficient in decomposing the atomic framework comprising the sample and that laser ablation on the $Li_2B_4O_7$ tablets is faster than the microwave-digestion technique that otherwise is required before solution ICP–MS. Furthermore, common problems with reprecipitation of Ba- and Zr-complexes after digestion also are avoided, and the sample is not nearly as strongly diluted as in solution ICP–MS work (typically 25–50 times). However, *in situ* laser ablation on the K-feldspar separates would of course improve the detection limits considerably, since the ~7-fold dilution by $Li_2B_4O_7$ would be avoided. However, coarse-grained subsolidus exsolution-induced domains of albite in pegmatitic K-feldspar effectively prevent direct laser ablation with the relatively narrow laser beam (30–100 μm).

K-feldspar was sampled from the IZ of granitic pegmatites where biotite, potassic and sodic feldspar, and quartz all are present. This part of the pegmatite formed relatively early (*e.g.*, Bjørlykke 1935) and, contrary to the wall zone, it is relatively uncontaminated by the host-rock lithologies. Samples of K-feldspar were also gathered from the core zone, which generally is considered to be the last part of the pegmatite to have formed (*e.g.*, Jahns & Burnham 1969, London 1992). Laboratory preparations included crushing and hand picking of inclusion-free K-feldspar separates. Subsequently, the samples were fused with $Li_2B_4O_7$ and were subjected to standard XRF analysis for major and minor elements. Estimates of concentrations of ultra-trace elements by *in situ* HR–LA–ICP–MS followed the method of Ødegaard & Hamester (1997) and Ødegaard *et al.* (1998). In this method, the high precision of a Finnigan MAT ELEMENT double-focusing-sector field mass spectrometer is combined with *in situ* sampling with a Finnigan MAT–SPECTRUM UV laser. The laser operates at 5mJ, with a pulse frequency of 4 Hz, and a spot diameter of 50 μm. Sample material was acquired by automated rastering over the surface in a predefined grid. The K-feldspar was analyzed for the entire spectrum of the *REE*, Sr, Y, Ta, Nb, Hf, Zr, U and Th. Reported detection-limits are 0.003 to 0.024 ppm, and the mean uncertainty is 13.7% (Ødegaard *et al.* 1998). The results of the chemical analyses are summarized in Table 1. A complete set of analytical results may be obtained from the author.

Appearance and Composition of the K-Feldspar

K-feldspar from 14 pegmatites in Evje–Iveland has an average bulk-composition of $Or_{75}Ab_{24}An_{1}$. The composition of IZ and CZ K-feldspar is nearly indistinguishable (Table 1). The Rostadheia (Loc. 90) pegmatite comprises the most sodium-rich K-feldspar, with the stoichiometric composition $Or_{67}Ab_{32}An_{1}$, whereas Elgedalen (Loc. 93) contain the most potassium-rich K-

TABLE 1. AVERAGE COMPOSITION OF K-FELDSPAR FROM GRANITIC PEGMATITES IN SOUTHERN NORWAY

	Evje–Iveland		Froland	
	IZ	CZ	IZ	CZ
SiO_2 wt.%	65.21	65.53	65.24	65.29
Al_2O_3	18.84	18.96	18.71	18.85
CaO	0.12	0.11	0.09	0.07
Na_2O	2.60	2.60	2.18	2.12
K_2O	12.67	12.56	13.13	13.34
LOI	0.17	0.13	0.16	0.17
Sum	99.64	99.93	99.53	99.89
n	14	13	8	5
K-feldspar composition				
Or mol.%	74.7	74.8	78.7	79.6
Ab	24.4	24.5	20.6	19.8
An	0.9	0.7	0.7	0.6
XRF data				
Ba ppm	534	576	468	360
Ga	22	25	22	23
Pb	176	197	132	116
Rb	748	874	775	756
LA–HR–ICP–MS data				
Sr ppm	93.4	94.5	99.0	65.0
Y	1.343	1.479	1.764	2.841
Zr	0.527	0.525	0.360	0.241
Hf	0.053	0.064	0.070	0.045
Ta	0.151	0.138	0.164	0.111
Th	0.077	0.076	0.177	0.309
U	0.392	0.344	0.761	1.139
Nb	0.738	0.781	0.658	0.586
La	1.425	1.919	0.552	0.427
Ce	1.840	2.457	0.803	0.673
Pr	0.152	0.188	0.091	0.105
Nd	0.416	0.438	0.198	0.257
Sm	0.103	0.112	0.099	0.117
Eu	0.381	0.296	0.221	0.224
Gd	0.107	0.108	0.144	0.211
Tb	0.022	0.020	0.036	0.057
Dy	0.144	0.141	0.277	0.394
Ho	0.034	0.033	0.061	0.086
Er	0.117	0.110	0.182	0.279
Tm	0.023	0.020	0.028	0.044
Yb	0.152	0.136	0.176	0.277
Lu	0.027	0.024	0.022	0.035.

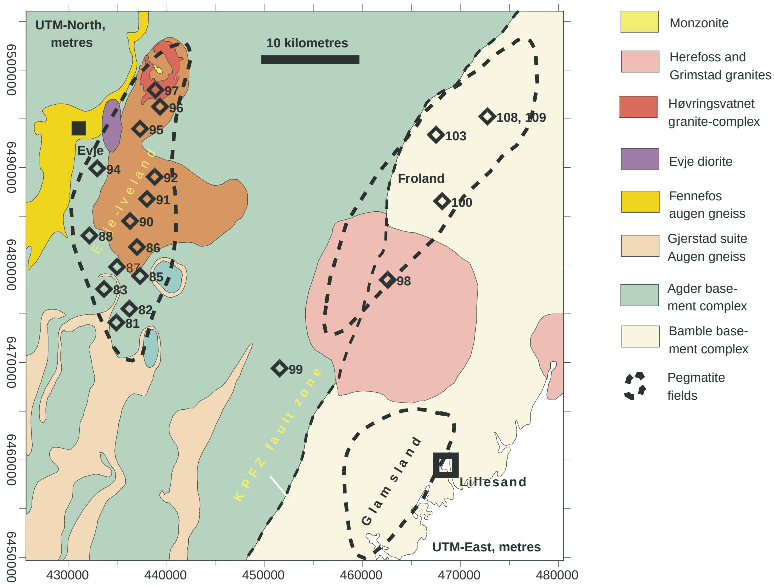


FIG. 1. Simplified geological map of the studied area, in southern Norway, based on Falkum (1982) and Padget & Maijer (1994). Samples of K-feldspar from granitic pegmatites were gathered in Evje–Iveland and Froland pegmatite fields. Two basement complexes separated by the Porsgrund–Kristiansand Fault Zone (PKFZ) dominate the area; pegmatite bodies from both complexes are included in this study. Also shown is the location of the pegmatites sampled, along with numbers referring to specific localities throughout the text.

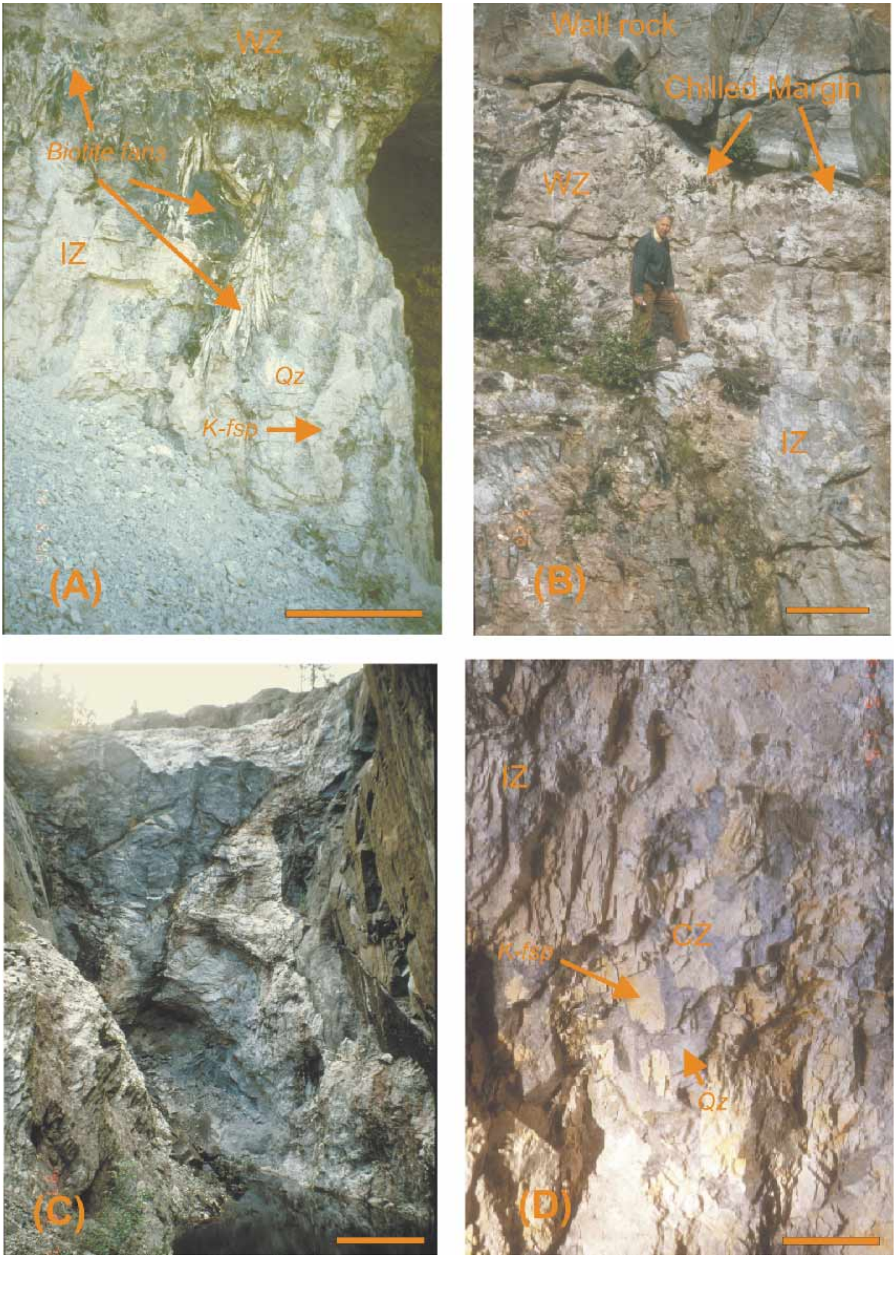
feldspar, with an average composition of $Or_{80}Ab_{19}An_1$. With an average composition of $Or_{79}Ab_{20}An_{1,}$ samples of K-feldspar in the Froland suite are markedly more potassium-rich than K-feldspar in the Evje–Iveland suite (Table 1).

Rb, Ba, Pb, Sr and Ga, in that order, are the dominant accessory elements in K-feldspar in both Evje–Iveland and Froland (Table 1). Together, these elements comprise 0.1 to 0.4 wt.% of the K-feldspar. The Evje–Iveland and Froland pegmatites show comparable patterns of distribution for these elements. The average Rb concentrations in the two fields are 748 to 874 ppm, respectively, Ba comprises 360 to 576 ppm, respectively, and Sr varies from 65 to 99 ppm (Table 1). Finally, the concentration of Pb varies from 116 to 197 ppm, and the concentration of Ga is 22 ppm on average, with insignificant variations between the two pegmatite fields. The concentration of Sr, Ba, Rb and Pb varies inconsistently from IZ to CZ.

The minor trace elements Y, Zr, Hf, Ta, Th, U, Nb and the *REE*, show the same inconsistent evolution from IZ to CZ as featured by the major elements and the trace elements mentioned above. When viewed from a regional perspective, uranium systematically attains higher concentrations in Froland compared to Evje–Iveland (Table 1). On the contrary, the concentration of the *REE*, Eu in particular, is highest in the Evje–Iveland suite (Table 1).

The chondrite-normalized distribution of *REE* in K-feldspar from Evje–Iveland and Froland generally re-

FIG. 2. Field appearance of selected granitic pegmatites. Solid bar is ~1 meter. A. Subhorizontal pegmatite dyke from Evje–Iveland, also known as the "Li" pegmatite, where dental-quality K-feldspar currently is mined. The photo show the dominant minerals and textures in the intermediate zone (IZ). Note that fan-shaped flakes of biotite extend from the boundary of the wall zone (WZ) with the IZ boundary into IZ and that quartz (Qz) and K-feldspar (K-fsp) are intergrown on a meter scale. The section is 4 meters high. B. Zoning geometry in a horizontal pegmatite dyke from Evje–Iveland featuring amphibolite wallrock, chilled margin, WZ and IZ. C. Pegmatite dyke from Evje–Iveland intersecting amphibolite host. Note that the pegmatite develops from a vertical dyke in the lower part of the section to a subhorizontal dyke in the upper part, hence illustrates the dominant fracture orientations of the area. D. Typical example of a core zone (CZ) in a pegmatite dyke from Froland. CZ is mostly composed of euhedral K-feldspar embedded in a matrix of quartz. Individual K-feldspar crystals approach 0.5 meter in diameter.


WZ
Biotite fans
IZ
Qz
K-fsp
(A)
Wall rock
Chilled Margin
WZ
IZ
(B)
(C)
IZ
CZ
K-fsp
Qz
(D)

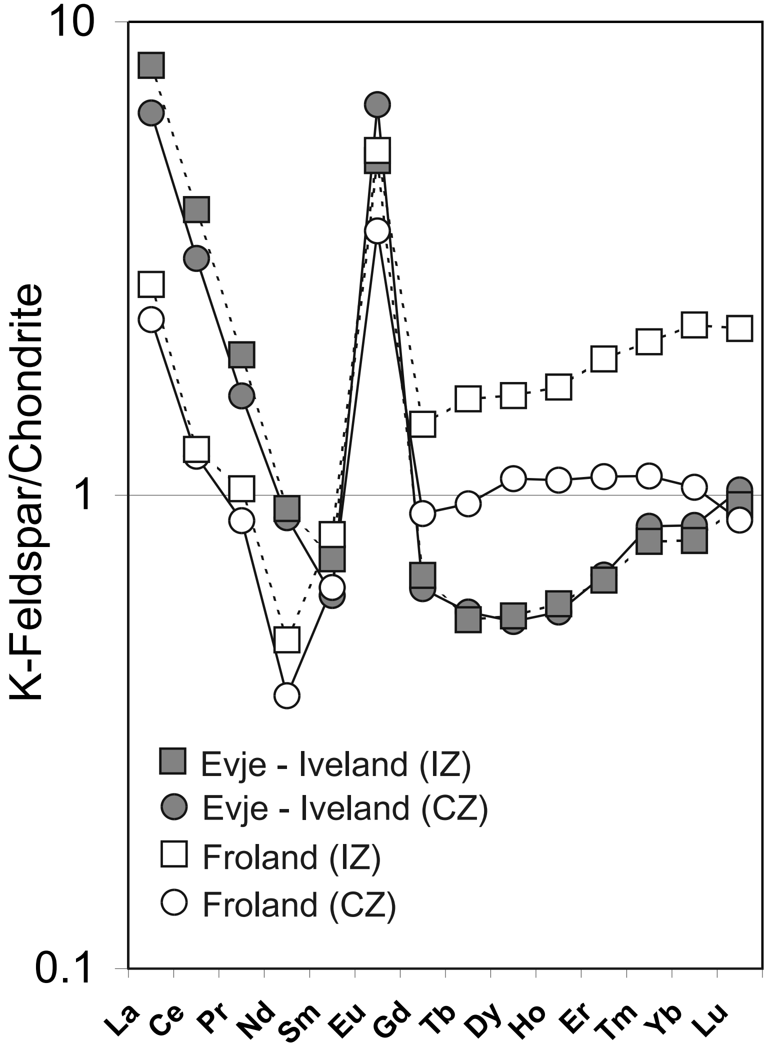


FIG. 3. Distribution of rare-earth elements (*REE*) in K-feldspar from Evje–Iveland and Froland pegmatite fields. The spectra represent the average of all IZ and CZ K-feldspar, respectively, from the two fields. Note that the *REE* spectra from the two fields are different.

sembles patterns reported in the literature for alkali feldspar that formed from a rhyolitic liquid (Nash & Crecraft 1985, Stix & Gorton 1990). Characteristically, the *REE* patterns define an asymmetrical trough broken by a moderate to strong Eu peak (Fig. 3). Accordingly, the light rare-earth elements (*LREE*) follow a steeply falling slope, whereas the middle and heavy rare-earth elements (*MREE*, *HREE*) from Gd to Tm follow an increasing slope. From Tm to Lu, the distribution pattern either continues to increase, levels out to horizontal or proceeds along a weakly falling slope. Overall, the concentration of *REE* is close to chondritic values, with a total of 4–6 ppm. In Evje–Iveland, the concentration of *LREE* is at or slightly above chondritic levels, Eu defines a strong positive anomaly, and the *HREE* fall at or below chondritic values. In Froland, the *LREE* concentrations are conspicuously lower than at Evje–Iveland and fall at or below chondritic values. In particular, La and Ce yield low concentrations, and the Eu anomaly is not as high as in Evje–Iveland. On the contrary, K-feldspar in the Froland suite is enriched in the *HREE* compared to Evje–Iveland, having concentrations at or above chondritic levels (Fig. 3). Otherwise, the Froland suite of pegmatites exhibits stronger variation in the *REE* pattern among pegmatite localities. Contrary to Evje–Iveland, the *REE* distribution, in going from the intermediate to the core zones, varies considerably and randomly (Fig. 3).

Discussion

Behavior of trace elements in igneous alkali feldspar

The distribution of trace elements in K-feldspar reflects the origin and evolution of the coexisting pegmatite-forming melts (*e.g.*, Heier & Taylor 1959, Trueman & Černý 1982, Černý *et al*. 1985, Shearer *et al*. 1985, 1992, Černý & Meintzer 1988, Abad Ortega *et al*. 1993, Neiva 1995, Icenhower & London 1996). In granitic pegmatites, the K, Sr, Rb, Ba, and Pb distributions in K-feldspar (*i.e.*, K_D) are functions of the melt composition, and depend on the extent of fractional crystallization and of the degree of differentiation of the parental magma before emplacement of the pegmatite-forming melts. Accordingly, Sr and Ba partition in favor of the primary K-rich feldspar, whereas Rb and Pb partition into the melt during differentiation of felsic igneous rocks. Sr is the most compatible, is followed by Ba, and finally Rb as the least compatible element in K-feldspar (*e.g.*, Icenhower & London 1996). Within a wide range of felsic melt compositions, the K_D of Sr is virtually independent of the albite and orthoclase concentration of the feldspar, whereas the K_D for Ba and Rb at least partially depend on the proportion of potassium.

The *REE* distribution in K-feldspar is intimately related to melt composition, rheology and temperature (Mahood & Hildreth 1983, Nash & Crecraft 1985, Stix & Gorton 1990). Although the partitioning of the *REE* into the primary alkali feldspar is not fully understood, it appears that the K_D values generally decrease from *LREE* to *HREE* (Stix & Gorton 1990), except for Eu^{2+}, which is quite easily accommodated at the alkali-site in the K-feldspar and typically induces a positive anomaly. It is also known that the total concentration of *HREE* increases with differentiation from intermediate to felsic melt compositions, whereas the concentration of the *LREE* decreases. To partially explain this pattern, Mahood & Hildreth (1983) suggested that the strictly short-range order of highly polymerized felsic melts leaves relatively small vacancies, which primarily accommodate the *HREE*, whereas the *LREE*, being too large for these vacancies, partition into the alkali feldspar. Furthermore, it is implied that the larger *LREE* compete more efficiently than the *HREE* for the K^+ and Na^+ sites in K-feldspar (Stix & Gorton 1990). Relatively Ca-rich sanidine contains even higher concentrations of *LREE* and lower concentrations of *HREE* than end-member K-feldspar; this pattern implies that the site holding the Ca *also* accommodates relatively large proportions of *LREE* in the feldspar structure (Nash & Crecraft 1985, Stix & Gorton 1990). Accordingly, the *LREE* and the *HREE* affinities in feldspar are decoupled and follow the opposite paths, *i.e.*, negative and positive trajectories, respectively, during differentiation from intermediate to felsic melt compositions. Furthermore, as it will be demonstrated below, the distribution of the *REE* in K-feldspar in pegmatite-forming environments is also buffered by various *REE*-bearing phases forming throughout the solidification of a pegmatite.

Distribution of Sr, Rb, Ba, K, Ga and Pb

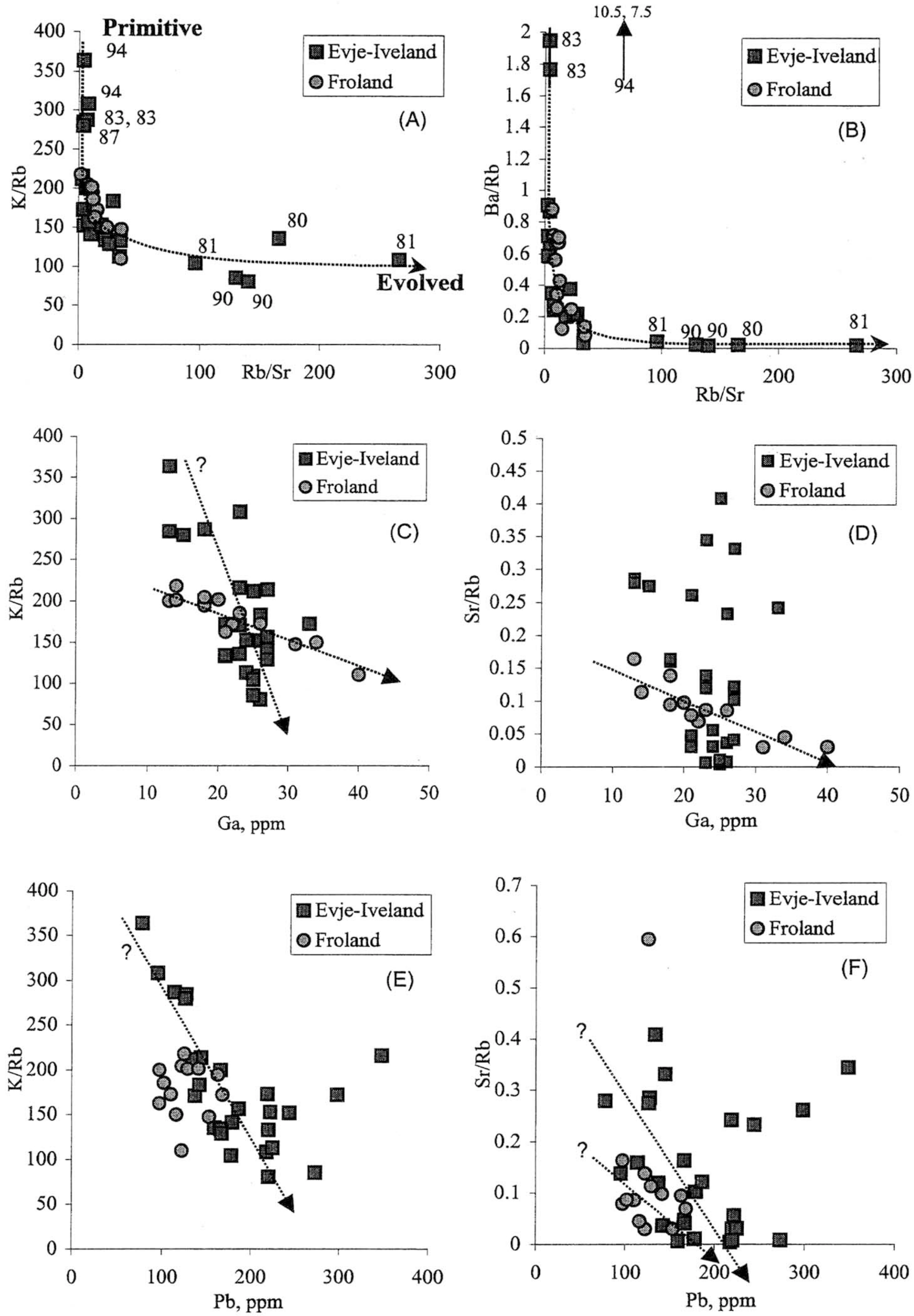
Granitic pegmatites in the Evje–Iveland and Froland fields are evaluated in terms of the ratio K/Rb, Rb/Sr and Ba/Rb, which are particularly sensitive to igneous differentiation.

In Evje–Iveland, the concentrations of Sr decrease from 248 to 4 ppm as concentrations of Rb increase from 344 to 1468 ppm. In the Froland pegmatite field, Sr decreases from 363 to 34 ppm, whereas Rb increases from 610 to 1168 ppm. Therefore, the pegmatites follow distinctive differentiation trends during evolution the pegmatite-forming melts (Fig. 4A). The most primitive pegmatites, having the highest K/Rb values, are found at localities 83 and 94 from Evje–Iveland (Fig. 4A), and the most evolved pegmatites, having the highest Rb/Sr values, are found at localities 81 and 90, also from Evje–Iveland (Fig. 4A).

In the present study, I also use the Ba/Rb and the Rb/Sr values (Fig. 4B) to evaluate the degree of evolution of the granitic pegmatites. These ratios follow the same trends as in Figure 4A, but are more discriminating in defining the degree of fractionation of the granitic pegmatites. Granitic pegmatites from both fields follows well-defined trajectories, in fact even better defined than in Figure 4A, and localities 83 and 94, and 81 and 90, respectively, again define the most primitive and most evolved pegmatites, respectively. However, notice that the trajectories are overlapping, hence it is not possible to distinguish the pegmatite fields on the basis of these elements.

Ga is incompatible in K-feldspar and follows a distinctively falling slope when compared to the Sr/Rb and K/Rb values for the Froland pegmatites (Figs. 4C, D). On the contrary, the Evje–Iveland pegmatites maintain nearly constant concentrations of Ga as differentiation proceeds. Gallium in the Evje–Iveland pegmatites probably is not controlled by K-feldspar fractionation but is buffered by other phases. The concentration of Pb increases in both fields along poorly defined trajectories (Figs. 4E, F).

K/Rb values for K-feldspar in the Evje–Iveland and Froland suites remain relatively primitive compared to K-feldspar from granitic pegmatite fields in general (Fig. 5). Pegmatites from Black Hills, South Dakota, Shatford Lake, Manitoba and Cerro de la Sal, Spain also


Primitive
Evolved
Evje-Iveland
Froland
(A)
K/Rb
Rb/Sr
(B)
Ba/Rb
10.5, 7.5
(C)
Ga, ppm
(D)
Sr/Rb
(E)
Pb, ppm
(F)

contain primitive compositions in terms of their K/Rb values, whereas well-known examples from Manitoba (Tanco, Greer Lake, Red Cross Lake) and Portugal contain much more evolved compositions. Primitive granitic pegmatites, such as at Black Hills and Shatford Lake, are also similar to the Evje–Iveland pegmatites in that they are classified as *REE* pegmatites with gadolinite, fergusonite and euxenite-(Y) as important accessory phases (Abad-Ortega *et al*. 1993, Shearer *et al*. 1992, Černý 1991).

Rare-earth elements

To a first approximation, the *REE* distribution in K-feldspar may be evaluated as a function of igneous differentiation. For example, the Rb/Sr value and Eu efficiently discriminate the pegmatites into more or less evolved types (Fig. 6). In agreement with Figures 4A–B, localities 83 and 94 comprises the most primitive pegmatites, whereas the most evolved are found at localities 90 and 81.

The pattern becomes considerably more complex when the entire *REE* dataset is considered (Figs. 7A, B). In Figure 7, the *REE* patterns for all the studied localities are sorted in terms of increasing Rb/Sr values (direction of arrow in figure), hence arranged with progressively more evolved pegmatites appearing toward the bottom of Figures 7A and 7B. The positive Eu anomaly is seen to diminish as Rb/Sr increases. In agreement with Figure 6, this pattern simply reflects the decreasing anorthite component of K-feldspar with differentiation. Furthermore, the *MREE* and, to a lesser extent, the *HREE* increase in concentration as the pegmatites become more evolved. Some *MREE* are actually enriched by an order of magnitude if one compares the most primitive with the most evolved pegmatites (Fig. 7A). Overall, the *LREE* distribution remains constant. The combined effect of decreasing the Eu anomaly and increasing the *MREE* and the *HREE* concentrations is to flatten the *REE* pattern. As a result, the characteristic trough shape, a feature of the most primitive K-feldspar, evolves toward an almost flat chondritic pattern (bottom, Fig. 7A). Samples from the intermediate and core zones in Evje–Iveland cannot be distinguished in terms of their *REE* distribution in K-feldspar.

In the Froland pegmatite field, the positive Eu anomaly also decreases with increasing evolution. Contrary to the Evje–Iveland pegmatite field, however, the total concentration of the *LREE* increases along with differentiation of the relevant melts (*i.e.*, increasing Rb/

FIG. 4. Distribution of various elements in K-feldspar, which are informative about the evolution of the granitic pegmatite as the pegmatite-forming liquids become more evolved. Numbers in Figures 4A, B refer to pegmatite localities that either represent the most primitive (83, 87, 94) or the most evolved pegmatites (80, 81, 90), respectively. See text for further explanation.

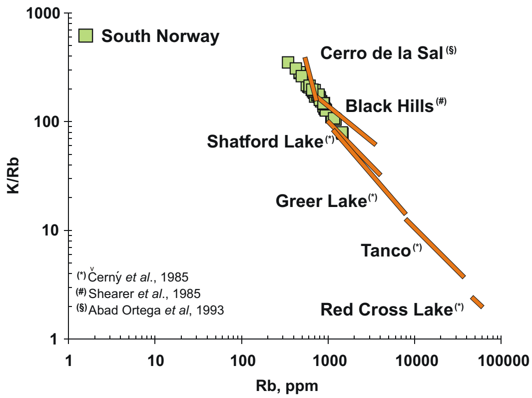


FIG. 5. K/Rb *versus* Rb distribution in K-feldspar from southern Norway compared with the composition of K-feldspar at well-studied localities from around the world. Compared to these localities, the granitic pegmatites in southern Norway contain very primitive compositions. Data extracted from Černý *et al*. (1985), Shearer *et al*. (1985), Abad-Ortega *et al*. (1993) and the present study.

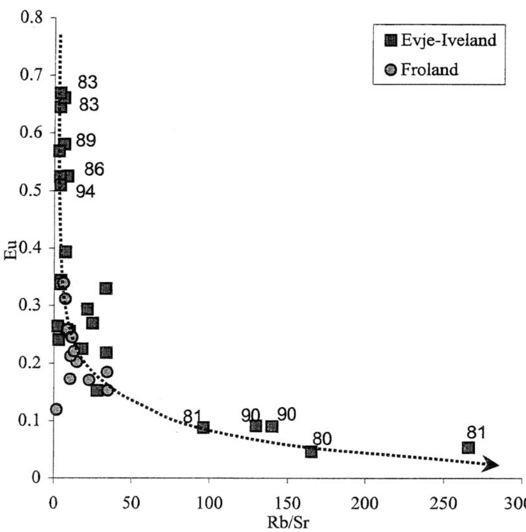
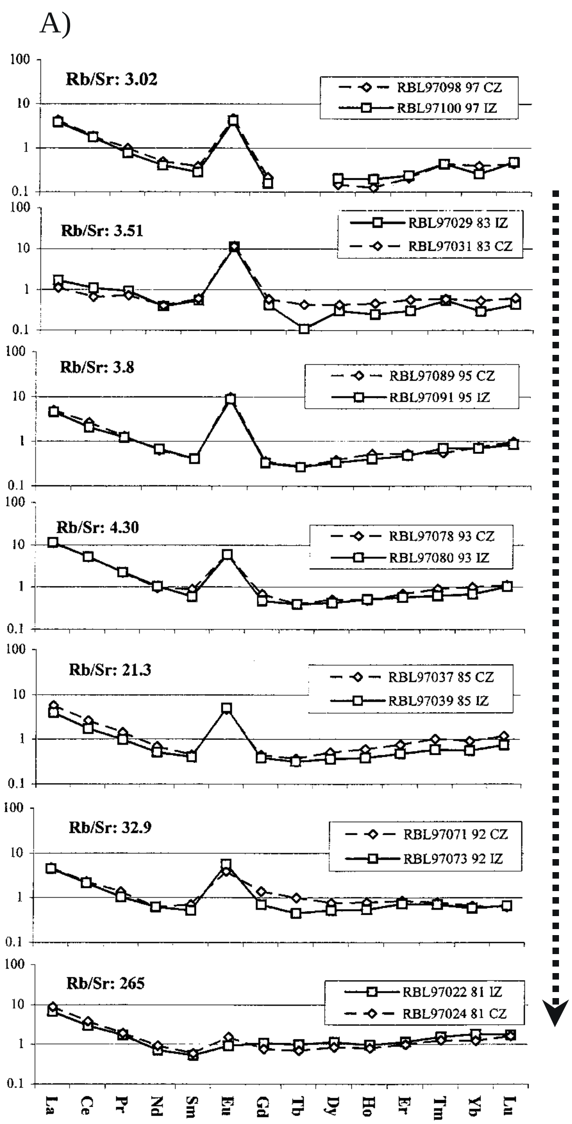


FIG. 6. Evolution of the Eu concentration in K-feldspar as a function of igneous differentiation. The concentration of Eu decreases along a steep slope as differentiation proceeds and the Rb/Sr value, therefore, increases. Numbers refer to the most primitive and the most evolved pegmatites in Evje–Iveland (see Fig. 4 and text).

Sr values), whereas the concentration of the *HREE* maintains chondritic values (Fig. 7B).

In conclusion, the *REE* distribution in K-feldspar from granitic pegmatite follows a well-defined evolutionary path during igneous differentiation. The systematic increase in the concentrations of the *HREE* in K-feldspar at Evje–Iveland follows the pattern defined in earlier studies (Mahood & Hildreth 1983, Nash & Crecraft 1985, Stix & Gorton 1990). However, the *LREE* concentration of K-feldspar in the Evje–Iveland suite and the entire *REE* distribution at Froland do not evolve as expected. On the contrary, the *LREE* in Froland increase in concentration with fractionation, whereas the *HREE* remain rather constant.

Accordingly, the K_D values for the $REE_{\text{K-feldspar}}/REE_{\text{melt}}$ equilibrium in granitic pegmatites from Evje–Iveland and Froland cannot only be functions of melt composition, melt rheology and temperature, as Mahood & Hildreth (1983), Nash & Crecraft (1985), Stix & Gorton (1990) implied. The importance of accessory *REE*-rich minerals in these pegmatite fields implies that the evolution of *REE* patterns in K-feldspar, largely, is buffered by the formation of these phases.



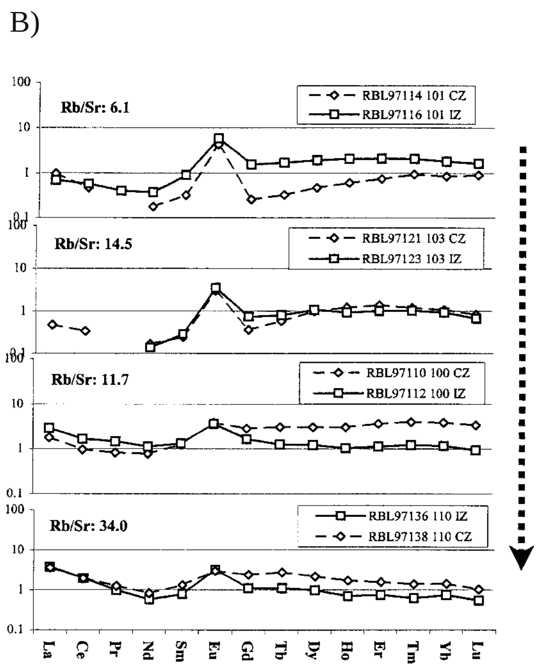


FIG. 7. Evolution of the *REE* distribution in K-feldspar as differentiation proceeds and the Rb/Sr value (shown in upper left corner of figures) increases. Solid lines for K-feldspar in IZ, and broken line for that in CZ. A. Evje–Iveland. B. Froland. Broken arrow indicate the direction of melt evolution. See text for detailed interpretation.

The granitic pegmatites in Evje–Iveland and Froland have been thoroughly studied in terms of assemblages of their *REE* minerals in 108 pegmatite localities in Evje–Iveland and 57 localities in the Froland suite (Bjørlykke 1935, 1937, 1939). Bjørlykke concluded that the most common *REE* minerals that fractionate *LREE* are monazite and allanite-(Ce) (Table 2), whereas the dominant *HREE*-enriched minerals, in decreasing abundance, are euxenite-(Y), xenotime, fergusonite and gadolinite (Table 2). From textural studies, he concluded that monazite and xenotime formed during crystallization of the wall and intermediate zone of the granitic pegmatites, whereas allanite-(Ce) and gadolinite formed late (IZ to CZ) (Table 2). Fergusonite and euxenite-(Y), in that order, formed after crystallization of xenotime, but before gadolinite (Bjørlykke 1937). Similarly, the first pegmatite-forming melts that were extracted from the source area gave higher proportions of early-forming *REE* phases whereas the late pegmatite-forming melts gave higher proportions of late-forming *REE* phases (Fig. 8).

Given this association of *REE* minerals, the concentration of *REE* in the melt was buffered by various *REE*-phases throughout solidification of the pegmatite. During initial crystallization, the *LREE* may partition into monazite, and the *HREE*, into xenotime and fergusonite. At more advanced stages of crystallization, the *LREE* may partition into allanite-(Ce), whereas the *HREE* first would partition into euxenite-(Y), then gadolinite (Table 2, Fig. 8).

In Froland, early-forming *LREE* phases (*i.e.*, monazite) are rare (Bjørlykke 1939), and the pegmatite-forming melts in most cases were undersaturated with respect to an *LREE* carrier phase. It was not until final crystallization that the melts became saturated with the *LREE* carrier allanite-(Ce) (Fig. 8).

This *REE*-mineral paragenesis is supported by the *REE* distribution in K-feldspar from Froland (Fig. 7B). In the most primitive pegmatites, the *LREE* concentration of K-feldspar is below the detection limit (Fig. 7B), whereas the most evolved pegmatites feature *LREE* concentrations comparable with those at Evje–Iveland.

Origin of the magmas in Evje–Iveland and Froland pegmatite fields

Previous studies imply that the Evje–Iveland and Froland pegmatite fields largely evolved simultaneously; they both contain primitive granitic pegmatites that are strongly zoned, comprise comparable assemblages of minerals, are rich in *REE*, Nb and Ta, and were derived from primitive parental magmas with low I_{Sr} (Bjørlykke 1935, 1937, 1939, Pedersen 1981, Starmer 1991, Pedersen & Konnerup-Madsen 1988, Stockmarr 1994, Sylvester 1998). The present study confirms these conclusions in that the evolution of the K-feldspar in these granitic pegmatites, in terms of the Rb, Sr, Ba and K distributions, follows overlapping trajectories from primitive to more evolved compositions; in discrimination diagrams, they plot together with primitive pegmatites from elsewhere in the world (Fig. 5). If the pegmatite-forming melts were derived from compositionally comparable source-areas, the distribution of all the trace elements in K-feldspar should also coincide. However, Ga, the *REE* distribution in K-feldspar, and the evolution of these elements with differentiation are fundamentally different in the two fields. Hence a common source appears unlikely. Apparently, the Evje–Iveland melts continuously maintained saturation with regard to both *LREE* and *HREE* minerals, whereas the Froland melts were undersaturated with regard to *LREE*-minerals until final crystallization. The Froland suite also has significantly higher *HREE* concentrations throughout their formation, compared to the Evje–Iveland suite. Most likely the parental liquids to the Froland suite of pegmatites were extracted from a relatively *LREE*-deficient source.

Apparent propagation of melt at Evje–Iveland

The Sr/Rb, Ba/Rb and K/Rb values may be used to distinguish between more or less differentiated granitic pegmatites within a pegmatite field; these values also may be applied in tracing the direction of melt propagation and the apparent source of the liquids (Trueman & Černý 1982, Shearer *et al.* 1992). The Evje–Iveland pegmatites show an overall decrease in the K/Rb and Sr/Rb values toward the south and east, with the highest values obtained in the far north and the lowest appearing ~30 km farther south (Fig. 9A). However, halfway between these two extremities, the generally declining trend is partially reversed to values approaching the values in the northern extremity of the profile. The two maxima are clearly defined on the contoured maps

TABLE 2. DISTRIBUTION (%) OF EARLY- AND LATE-FORMING *REE*-PHASES IN EVJE–IVELAND AND FROLAND SUITES, SOUTH NORWAY

Mineral group	Stage	REE	Classification	E–I RA	E–I *f*	F RA	F *f*
Monazite	early	*LREE*	(Ce,La,Nd)-phosphate	52	56	12	7
Allanite	late	*LREE*	(Ce,La,Nd)-silicate	17	19	51	30
Xenotime	early	*HREE*	(*HREE*)-phosphate	26	28	8	5
Fergusonite	early-medium	*HREE*	(*HREE*)-niobate	19	20	10	6
Euxenite	medium	*REE*	(Y,Er,La,Ce,)-niobate	50	53	22	13
Gadolinite	late	*HREE*	(*HREE*)-silicate	20	21	7	4

Early phase / late phase	Category	E–I %	F %
Monazite / allanite	*LREE* minerals	75	16
(Xenotime + fergusonite) / (Euxenite + gadolinite)	*HREE* minerals	39	39
Xenotime / gadolinite	*HREE* minerals	57	56

E–I: Evje–Iveland (108 localities); F: Froland (57 localities). RA: relative abundance, in %; *f*: frequency, in %.

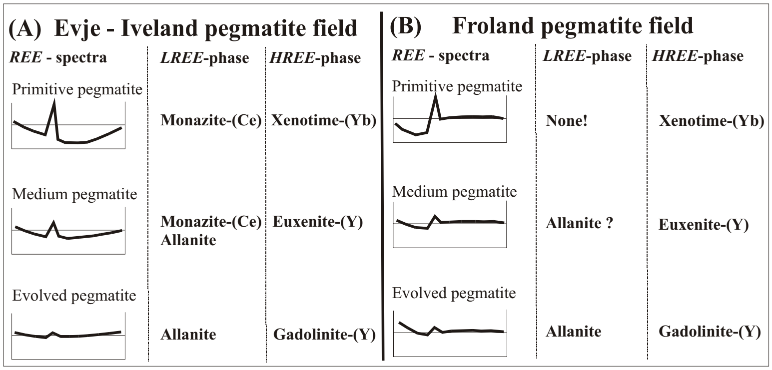


FIG. 8. Model of the evolution of the *REE* distribution in K-feldspar from Evje–Iveland and Froland pegmatite fields, respectively. The basic assumption is that the distribution of *REE* in K-feldspar is partially or fully buffered by the formation of *REE*-minerals that either consume *LREE* or *HREE* from the pegmatite-forming melt. In both Evje–Iveland and Froland, it is implied that pegmatite-forming melts were derived from a reservoir of parent magma, from progressively more evolved residual melts generated during terminal crystallization of a parental magma. Here, this igneous evolution is symbolized with three principal *REE* spectra representing primitive, medium and evolved *REE* compositions of the pegmatitic K-feldspar. A. In Evje–Iveland, the distribution of *REE* in K-feldspar is buffered by the formation of *REE*-bearing minerals throughout the differentiation history of granitic pegmatites. B. In Froland, primitive granitic melts were mostly undersaturated with respect to a *LREE*-bearing mineral. Only after some extent of differentiation and evolution of the granitic melts did an *LREE*-bearing phase, here allanite-(Ce), would begin to form. The *HREE* distribution in K-feldspar is apparently buffered by various *HREE*-bearing minerals throughout the crystallization of primitive, medium and evolved pegmatites, respectively.

(Fig. 9A), although the maxima to the north are the highest. Largely, these trends imply that the pegmatite-forming melts propagated toward the south and southeast (Fig. 9B**)** from an origin somewhere in the north or northwest. This distribution of K/Rb and Sr/Rb values imply that the pegmatite-forming melts followed a major conduit along the western edge of the pegmatite field and, further south, branched out eastward (Fig. 9B). It is noteworthy that the main conduit coincides with the lithological boundary between the *Gjerstad suite augen gneiss* and the *Agder basement complex* (Fig. 1). Using the field data from Bjørlykke (1935), the reversal in the chemical gradients (Fig. 9A) coincides with an abrupt change in the strike of the pegmatite dykes from generally N–S to generally E–W (Fig. 9B).

Many other pegmatite fields around the world also feature the most primitive pegmatites in a proximal position with respect to the presumed source-area for the pegmatite-forming melts. In the Black Hills area of South Dakota, for example, the most primitive pegmatites occur in a proximal position with respect to the parent granitic intrusion (*e.g.*, Shearer *et al*. 1992). In the Birse Lake pegmatite group, in Manitoba, the melt-propagation pattern is also characterized by smaller conduits branching off from a single major conduit (Trueman & Černý 1982).

CONCLUSIONS

1. LA–ICP–MS analysis of K-feldspar samples from pegmatites for major, trace and ultra-trace elements (including *REE*) rapidly provides a wealth of unbiased chemical information. This information allows petrogenetic interpretations of the origin, evolution and distinction of granitic pegmatite fields.

2. Compared to the Evje–Iveland pegmatite field, K-feldspar from the Froland pegmatites is more K- and U-rich, whereas it is depleted in Ba, Pb and *LREE*. The chondrite-normalized *REE* spectra consistently follow a decreasing slope from La to Nd, show a strong positive Eu-anomaly, and feature a flat or slightly increasing *HREE* spectrum. Froland may also be distinguished from Evje–Iveland in that the decreasing slope is broken at Nd rather than at Sm and in having much lower concentrations of *LREE* and higher concentrations of *HREE*.

3. The distribution of the *REE* in K-feldspar reflects that in the coexisting pegmatite-forming melts. Partitioning of *REE* into K-feldspar is buffered by fractional crystallization of *REE*-phases. The Froland pegmatites remained undersaturated with respect to *LREE* phases until allanite-(Ce) formed during the terminal stage of crystallization of the pegmatites.

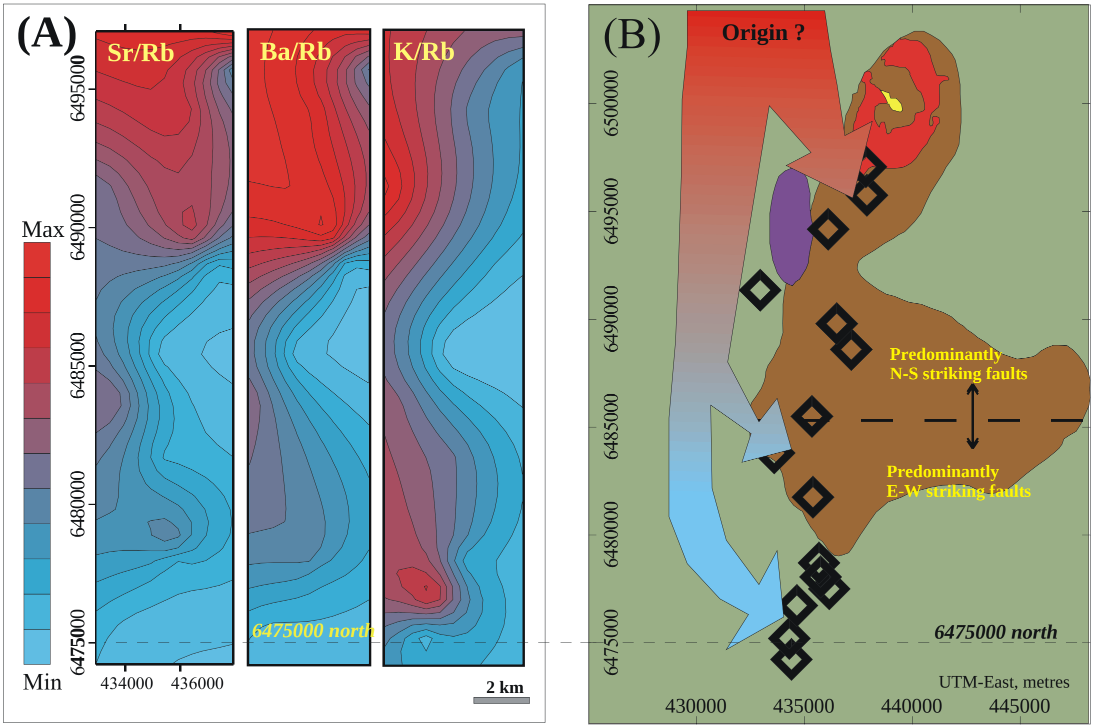


FIG. 9. Spatial representation of primitive and progressively more evolved granitic pegmatites in the Evje–Iveland area and an apparent direction of propagation of the pegmatite-forming melts. A. Distribution patterns of key indicators that are strongly sensitive to igneous differentiation are shown on these contoured maps. The Sr/Rb, Ba/Rb and K/Rb values decrease toward the south and east, implying that progressively more evolved granitic melts are emplaced in that direction. The highest values are confined to the northern part of the area. B. Apparent direction of propagation of the pegmatite-forming melts estimated from the Sr/Rb, Ba/Rb and K/Rb values of K-feldspar. See text for detailed explanations and interpretations.

4. In both pegmatite fields, the concentration of Sr, Ba and Eu decreases as differentiation proceeds, whereas the concentration of Rb, Pb and Ga increases. For Sr, Ba, Rb and Eu, the evolutionary paths followed by the pegmatites coincide, whereas for Ga the trends are distinctively different for the two fields. These features, together with conspicuous differences in the *REE* and U distributions, indicate that pegmatite-forming melts were derived from different sources. Judging from the *REE* spectra alone, the Froland pegmatite suite seems to have been derived from a relatively *HREE*-rich and *LREE*-poor source.

5. The Sr/Rb, Ba/Rb and the K/Rb values decrease towards south and east in the Evje–Iveland field. Accordingly, granitic pegmatites in that suite were derived from parent melts in the north. Progressively more differentiated pegmatite-forming melts propagated toward the south and east along a system of vertical and subhorizontal faults and fractures. The main conduit of the pegmatite- forming melts coincides with a lithological boundary, whereas minor conduits, branching off toward the east, primarily are controlled by E–W-striking vertical faults.

ACKNOWLEDGEMENTS

The author is grateful to North Cape Minerals, Lillesand, which partially funded the present study. Discussions with S. Pedersen and accessibility to unpublished REE-data (University of Copenhagen) are much appreciated. B. Bingen (NGU) commented an earlier draft of this article, and S. Dundas and B. Flem (NGU) carried out the HR–LA–ICP–MS analyses. Review by D.J. Kontak and R.F. Martin considerably improved an earlier draft of this article. Comments from an anonymous reviewer are acknowledged.

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Received August 20, 2000, revised manuscript accepted February 13, 2002.