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GEOCHEMISTRY OF FELDSPARS AND MUSCOVITE IN GRANITIC PEGMATITE FROM THE CAP DE CREUS FIELD, CATALONIA, SPAIN

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Abstract

The Cap de Creus pegmatite field exhibits a well-defined regional zonation of four types of granitic pegmatite: barren K-feldspar-rich pegmatites (type I), beryl–columbite-subtype pegmatites (type II), beryl–columbite–phosphate-subtype pegmatites (type III) and albite-type pegmatites (type IV). There is an increase in the albite content, relative to K-feldspar, with increasing pegmatite evolution. Type-I microcline-rich pegmatites have low contents of albite, whereas in type IV, albite is the only feldspar. The K/Rb value in K-feldspar decreases progressively from type-I to type-III pegmatites. In addition, there is a progressive enrichment in P, Rb, Cs, Y, Sr, Ga, Ce, W and Sn from type-I to type-IV pegmatites. Concentrations of Pb and Zr decrease in K-feldspar from type-I to type-III pegmatites. K-feldspar from the Cap de Creus pegmatites exhibits higher contents in Ba, Rb, Cs, Nb, Y, Sn, V and Cr than albite. Albite is richer in Sr, Zr, Cu, As, Ce and Ga. Pb, Zn, W, Co and Ni have similar affinity for K-feldspar and albite. Muscovite exhibits similar geochemical evolution to K-feldspar. These progressive trends suggest a common origin for all of the Cap de Creus pegmatites by fractionation of the same parental magma. The high P content of feldspars, up to 1.67 wt.% P_2O_5 in K-feldspar and 1.75 wt.% in albite, indicates buildup in this element from the earliest stages of pegmatite crystallization. Trace-element variation in feldspars and muscovite reflects enrichments in the melt; a higher content correlates with characteristic minerals of these elements. In particular, the phosphorus content of albite is proposed as a useful mineralogical guide to niobium–tantalum-rich oxide mineralization.

Keywords: K-feldspar, albite, muscovite, X-ray fluorescence, evolution, granitic pegmatite, Cap de Creus, Catalonia, Spain.

Sommaire

La suite de pegmatites granitiques du Cap de Creus démontre bien une zonation régionale dans la disbtribution de quatre types de pegmatite: pegmatites stériles enrichies en feldspath potassique (type I), pegmatites du sous-type à béryl–columbite (type II), pegmatites du sous-type à béryl – columbite – phosphate (type III), et pegmatites à albite (type IV). Il y a une augmentation progressive de la teneur en albite par rapport à celle du feldspath potassique, avec le degré de fractionnement. Les pegmatites du type I, riches en microcline, ont une faible teneur en albite, tandis que dans les pegmatites de type IV, l'albite est le seul feldspath. La valeur de K/Rb du feldspath potassique diminue progressivement des pegmatites du type I à celles du type III. De plus, il y a un enrichissement progressif en P, Rb, Cs, Y, Sr, Ga, Ce, W et Sn du type I au type IV. Les concentrations de Pb et Zr diminuent dans le feldspath potassique des pegmatites du type I à celles du type III. Le feldspath potassique des pegmatites du Cap de Creus fait preuve de teneurs plus élevées en Ba, Rb, Cs, Nb, Y, Sn, V et Cr que l'albite. En revanche, l'albite est plus riche en Sr, Zr, Cu, As, Ce et Ga. Le Pb, Zn, W, Co et Ni ont un degré d'enrichissement semblable dans le feldspath potassique et l'albite. La muscovite fait preuve d'enrichissements semblables à ceux du feldspath potassique. Ces enrichissements progressifs témoigneraient d'une origine commune de toutes les venues pegmatitiques du Cap de Creus, à partir d'un même magma parental. La teneur élevée des feldspaths en P, jusqu'à 1.67% P_2O_5 (poids) dans le feldspath potassique et 1.75% dans l'albite, témoigne d'une augmentation progressive de cet élément dans le magma depuis les stades précoces de la cristallisation. Les variations en éléments-traces dans les feldspaths et la muscovite sont le reflet d'enrichissements dans le bain fondu; une concentration accrue montre une corrélation avec les minéraux caractéristiques de ces éléments. En particulier, la teneur en phosphore de l'albite serait un indicateur utile d'une minéralisation en oxydes de niobium–tantale.

(Traduit par la Rédaction)

Mots-clés: feldspath potassique, albite, muscovite, fluorescence X, évolution, pegmatite granitique, Cap de Creus, Catalogne, Espagne.

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INTRODUCTION

The compositional attributes of feldspars and micas can provide valuable information on the petrogenetic evolution of granitic pegmatites (Trueman & Černý 1982, Legg & Namateba 1982, Černý 1989, 1994). In particular, these attributes can help to determine the degree of fractionation of the magma and the relationships among different pegmatites within a pegmatite field (Shearer *et al.* 1985).Furthermore, the trace-element contents of feldspars and micas indicate economic potential of pegmatites (Smeds 1992, Trumbull 1995).

In this paper, we present a geochemical study of feldspar minerals and muscovite from representative bodies of pegmatites from the Cap de Creus pegmatite field, a typical regionally zoned suite that comprises four types of pegmatite, three of which belong to the rare-element class of Černý (1989). Our aim in this work is to assess the degree of pegmatite evolution with focus upon the feldspar minerals and muscovite.

SAMPLING AND ANALYTICAL METHODS

Samples of feldspars and muscovite were selected in the different zones from all the pegmatite types. These minerals were separated manually under a binocular microscope (an average of 20 g per sample). They were then crushed in a corundum mortar and treated with nitric, oxalic and sulfuric acids, to remove impurities such as phosphates, Fe oxides and organic matter.

Feldspars and muscovite were analyzed by X-ray fluorescence at the "Servicio de Análisis de Minerales y Rocas" of the Universidad del País Vasco, using an automated Philips PW1480 sequential spectrometer. Sixty samples of K-feldspar, 58 samples of albite and 76 samples of muscovite were analyzed.

Forty-two international reference samples (Certified Reference Materials, CRM) were used to calibrate by regression the analytical lines. Correction factors were applied for line overlaps and matrix effects. Coefficients of variation for major and minor elements are usually below 1%, with a detection limit for most trace elements on the order of 2 to 5 ppm. Concentrations of the major elements (Si, Al, Ti, Fe, Mg, Mn, Ca, K, and P) and the majority of trace elements were established using fused beads, whereas pressed powder pellets were used for Cs, Y and Nb determinations. Sample-preparation procedures were described by Yusta *et al.* (1994).

Owing to the low content of albite from microcline-rich pegmatites and to compare the composition of pairs of feldspars, some additional analyses of albite from these pegmatites were performed with an electron microprobe. Albite – K-feldspar pairs were also analyzed to establish their trace-element content. We used a CAMECA SX50 electron microprobe from the Serveis Cientifico Tècnics de la Universitat de Barcelona. The analyses were conducted at an accelerating voltage of 15 kV and an intensity of 10 nA. The following standards and lines were used: orthoclase (Si$K\alpha$, Al$K\alpha$, K$K\alpha$), rutile (Ti$K\alpha$), hematite (Fe$K\alpha$), rhodonite (Mn$K\alpha$), periclase (Mg$K\alpha$), wollastonite (Ca$K\alpha$), albite (Na$K\alpha$), and apatite (P$K\alpha$).

GEOLOGICAL SETTING

The Cap de Creus area of northeastern Spain constitutes the Hercynian basement at the eastern end of the Pyrenees (Fig. 1). The Paleozoic series consists of a 2000-m-thick pelitic sequence with minor limestone, felsic volcanic rocks and coarser sedimentary rocks. This sequence has been affected by two main phases of Hercynian deformation. Phase one produced the main foliation, and the succeeding episode of deformation induced folding that ended with the development of NW–SE-trending shear bands (Carreras & Casas 1987). A low-pressure, high-temperature regional metamorphism, which increases from a chlorite–muscovite zone in the south to cordierite–andalusite, sillimanite–muscovite and K-feldspar – sillimanite zones in the north, with isograds trending northwest, occurred between the two episodes of deformation.

The intrusive rocks in the Cap de Creus area consist of pre-Hercynian porphyritic granites and two types of Hercynian granitic rocks: (1) small bodies of syntectonic leucogranites to quartz gabbros related to the northern migmatite zone (Carreras *et al.* 1975, Carreras & Druguet 1994), and (2) the late Roses and Rodes granodiorites and quartz diorite stocks, emplaced within lower-grade rocks in the south and southwest.

FOUR TYPES OF PEGMATITE

Four types of granitic pegmatites have been established according to internal structure and mineralogical criteria (Corbella 1990, Corbella & Melgarejo 1993). Detailed descriptions of their internal structure and mineralogy are reported in Corbella & Melgarejo (1993) and Alfonso *et al.* (1995), and a synthesis of these features is presented in Table 1.

The pegmatites are syntectonic, and many of them are folded or display minor structures related to folding, such as boudinage and a pinch-and-swell structure. In some pegmatites of type I, a late episode of crystal growth occurs in the necking zones associated with the boudins.

The distribution of the different types of pegmatites and the mineralogical and geochemical data suggest that all these pegmatites were formed by efficient fractionation operating from a single petrogenetic source (Corbella & Melgarejo 1993, Alfonso 1995). Moreover, in the Albera pegmatite field, 50 km to the northwest of Cap de Creus, similar types of pegmatite are zonally arranged around a leucogranite intrusion (Malló *et al.* 1995).

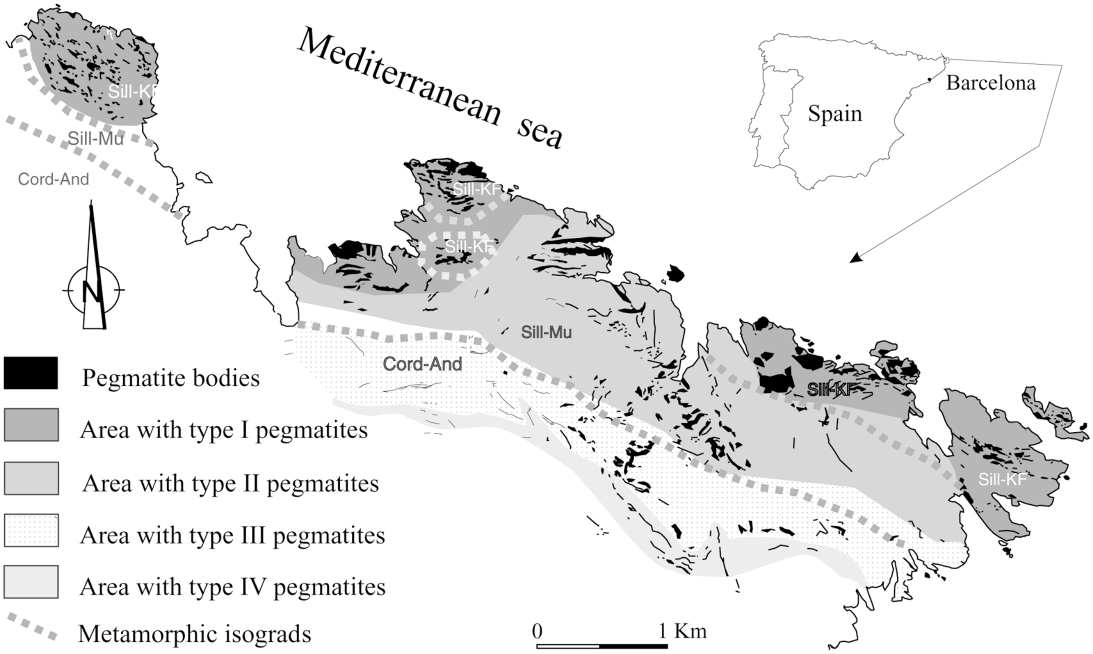


FIG. 1. Pegmatite distribution in the Cap de Creus Peninsula: 1) pegmatite bodies, 2) area with type-I barren pegmatites, 3) area with type-II transitional pegmatites, beryl–columbite subtype, 4) area with type-III beryl–columbite–phosphate pegmatites, 5) area with type-IV albite pegmatites. The metamorphic isograds are also shown. Modified from Carreras *et al.* (1975).

The most striking geochemical variations from type-I to type-IV pegmatites are: (1) increase of albitization and replacement phenomena, (2) increase in the concentrations of P and Li minerals, and (3) increase in the ratios Ta/(Ta + Nb) and Mn/(Fe + Mn) in columbite–tantalite minerals (Alfonso *et al.* 1995).

RESULTS

K-feldspar

The composition of K-feldspar from three types of pegmatite is presented in Table 2 (K-feldspar is absent from type-IV pegmatites). Systematic chemical variations among the feldspars from the different zones into a pegmatite body are absent. Hence, we will only consider the variations among the different types of pegmatite. The orthoclase component of the bulk K-feldspar ranges between Or$_{63}$ and Or$_{79}$ in type-I, Or$_{71}$ to Or$_{78}$ in type-II, and Or$_{74}$ to Or$_{98}$ in type-III pegmatites. In general, the K-feldspar from type-I and type-II pegmatites has a higher sodium content than that in type-III pegmatites (Fig. 2).

Some trace elements have long been used as indicators of the pegmatite evolution. Among them, those involving substitution in the alkali site (Rb, Cs, Sr, Ba, Pb) are the most thoroughly investigated.

The K/Rb in K-feldspar constitutes a good indicator of a pegmatite body's evolution (Černý *et al.* 1985). The K/Rb ratio ranges between 140 and 467 for type-I pegmatites (the higher values were obtained in feldspars from the late-necking zones), 88 to 264 for type-II pegmatites, and 23 to 59 for type-III pegmatites (exclusive of two samples, in which the values are 177 and 284). The content of most trace elements in K-feldspar from the Cap de Creus pegmatites correlates negatively with this ratio.

K-feldspar from Cap de Creus pegmatites exhibits a marked Rb enrichment in the progression from type-I to type-III pegmatites. The rubidium content in type-I and type-II pegmatites is low, up to 667 and 1065 ppm, respectively, but up to 3729 ppm in type-III pegmatites. Those values are typical of K-feldspar from other evolved pegmatite bodies (Černý *et al.* 1985, Neiva 1995).

The K/Rb *versus* Cs plot is commonly used as an indicator of the pegmatite evolution (Gordiyenko 1971, Černý *et al.* 1985). At Cap de Creus, cesium increases from type-I to type-III pegmatites (Fig. 3a), although it is not very abundant. Nevertheless, in type-III pegmatites, it attains a maximum of 125 ppm. Such Cs values are typical of modestly evolved pegmatites (Černý 1994). Similar Cs contents have been documented by Kontak & Martin (1997) in pegmatites from

TABLE 1. MAIN CHARACTERISTICS OF THE FOUR TYPES OF PEGMATITE FROM THE CAP DE CREUS FIELD

Type of pegmatite	Main primary minerals	Characteristics of the feldspars	Primary accessory minerals	Characteristics of the micas	Late replacements
Type I Microcline-rich, barren	Qtz + Kfs + Ab + Pl	*Wall*: Kfs (dominant, non-perthitic) + Ab *Intermediate zones*: Kfs [dominant, two generations: 1) blocky, 2) graphic intergrowths with Qtz] + oligoclase + Ab (late) *Necking zones in boudin*: Kfs (blocky, perthitic) + Ab (blocky)	Sillimanite, biotite, cordierite, Tur, Grt, Ms, ixiolite and ilmenorutile	*Border*: Ms (dominant) *Wall*: Ms (dominant) + Bt *First intermediate zone*: Ms + Bt *Second intermediate zone*: Ms + Bt (both in branching crystals, up to 1 mm in length) *Necking zones in boudins*: Ms	Biotite replaced by muscovite + chlorite
Type II Beryl – columbite	Qtz + Kfs + Ab	*Wall*: Kfs dominant, non-perthitic + Ab (scarce). *Intermediate zones*: Kfs (perthitic) + Ab [two generations: 1) primary, 2) late, replacing Kfs] *Albite replacement bodies*: Ab (dominant)	Ms, Grt, Tur, sillimanite, andalusite, beryl, chrysoberyl, strüverite, gahnite, columbite, uraninite, ferberite, Ca–Mn–Mg–Fe-rich phosphates	*Wall*: Ms *Intermediate zones*: Ms (dominant) + biotite (rare) *Albite replacement bodies*: Ms	Albite, chrysoberyl, phenakite, euclase metasomatic alkali-rich phosphates, phosphates of Be
Type III Beryl – columbite – phosphate	Qtz + Kfs + Ab	*Wall*: Kfs (euhedral) replaced by Ab *Intermediate zones*: Kfs [dominant, two generations: 1) blocky, 2) graphic intergrowths with quartz] + Ab [two generations: 1) primary, 2) late, replacing Kfs) *Albite replacement bodies*: Ab (solely, saccharoidal or "cleavelandite")	Ms, Tur, beryl, columbite–tantalite, ixiolite, fergusonite, tapiolite, phosphates of Li (montebrasite and triphylite) Al–Ca–Mn–Fe phosphates	*Wall*: Ms *Intermediate zones*: Ms *Albite replacement bodies*: Ms (common) *Quartz–muscovite veins*: Ms	Albite, zircon, columbite–tantalite, tapiolite, fergusonite, strüverite, phosphates of Be, metasomatic alkali phosphates
Type IV Albite	Ab + Qtz	*Wall*: Ab *Intermediate zones*: Ab (three generations) *Late drusy veins*: Ab	Ms, beryl, gahnite, chrysoberyl, strüverite, cassiterite, columbite–tantalite, tapiolite, phosphates of Li, phosphates of Al–Ca–Mn–Fe	*Wall*: Ms *Intermediate zones*: Ms *Quartz–muscovite veins*: Ms (several generations) *Late drusy veins*: Ms	Trolleite, scorzalite, wyllieite, montebrasite, nigerite, rynersonite, microlite and other *HREE*-rich minerals

Symbols: Qtz: quartz, Kfs: K-feldspar, Ab: albite, Pl: plagioclase, Bt: biotite, Ms: muscovite, Tur, tourmaline, Grt, garnet.

Nova Scotia, Canada, and by Tindle *et al.* (2002) in beryl-type and some complex-type pegmatites of the Separation Lake area of northwestern Ontario.

Strontium exhibits the same behavior as Rb. The ratio Rb/Sr correlates negatively with K/Rb (Fig. 3b), except in some samples of type-III pegmatites, which have very high Sr content. These high values are usually encountered in non-evolved granitic pegmatites. For example, in the pegmatitic granite of Cross Lake, Manitoba, K-feldspar contains between 10 and 290 ppm of Sr (Černý *et al.* 1985), whereas in highly evolved pegmatites, the Sr contents are lower, as in most samples from the Cap de Creus pegmatites. In many other pegmatite fields, strontium in K-feldspar has a negative correlation with rubidium (Abad Ortega *et al.* 1993, Kontak & Martin 1997).

Barium does not exhibit any meaningful variation related to the evolution. Barium content is commonly used as indicator of the evolution in granites and pegmatites (Mehnert & Büsch 1981), and decreases with advancing fractionation (Černý *et al.* 1985, Roda *et al.* 1999, Morteani *et al.* 2000). Nevertheless, in the Cap de Creus pegmatites, Ba content does not seem related to the degree of evolution.

The Pb content in K-feldspar decreases with the evolution of the pegmatites (Fig. 3c). This trend is also reported in other pegmatite fields (Kontak & Martin 1997), and even a single pegmatite body (Černý & Burt 1984).

TABLE 2. COMPOSITION OF FELDSPARS FROM THE CAP DE CREUS GRANITIC PEGMATITES, CATALONIA, SPAIN

	Microcline						Albite						
	I		II		III		I	II		III		IV	
	median	range	median	range	median	range	median	median	range	median	range	median	range
	(*n* = 18)		(*n* = 9)		(*n* = 23)		(*n* = 2)	(*n* = 5)		(*n* = 16)		(*n* = 20)	
SiO_2 wt.%	65.14	[63.66–68.55]	65.71	[64.25–72.13]	64.32	[62.18–67.00]	66.93	70.30	[67.63–75.86]	68.09	[66.75–72.93]	67.44	[65.05–70.54]
Al_2O_3	19.24	[17.36–19.55]	18.88	[14.83–20.23]	19.09	[17.92–20.03]	20.51	17.54	[13.23–19.39]	18.91	[16.43–20.33]	19.12	[17.71–21.12]
TiO_2	0.01	[0.00–0.01]	0.00	[0.00–0.03]	0.00	[0.00–0.02]	0.01	0.01	[0.01–0.04]	0.01	[0.01–0.02]	0.01	[0.01–0.01]
Fe_2O_3	0.03	[0.00–0.10]	0.04	[0.01–0.09]	0.04	[0.01–0.20]	0.09	0.07	[0.04–0.10]	0.04	[0.02–0.13]	0.08	[0.03–0.14]
MgO	0.05	[0.00–0.23]	0.07	[0.01–0.13]	0.07	[0.01–0.23]	0.03	0.03	[0.01–0.18]	0.01	[0.00–0.08]	0.03	[0.00–0.07]
MnO	0.00	[0.00–0.04]	0.00	[0.00–0.01]	0.01	[0.00–0.04]	0.00	0.00	[0.00–0.01]	0.00	[0.00–0.08]	0.00	[0.00–0.04]
CaO	0.09	[0.05–0.41]	0.10	[0.03–0.38]	0.05	[0.00–1.48]	1.56	0.73	[0.07–1.54]	0.05	[0.00–1.20]	0.21	[0.00–1.50]
Na_2O	2.67	[2.07–4.66]	2.88	[2.24–4.48]	2.14	[0.00–4.53]	9.94	9.03	[5.08–11.23]	10.76	[7.61–11.35]	10.34	[8.14–11.67]
K_2O	11.13	[6.86–12.38]	11.16	[9.01–12.28]	11.02	[5.07–13.61]	0.76	0.77	[0.22–3.31]	0.65	[0.21–1.36]	0.63	[0.17–2.05]
P_2O_5	0.46	[0.21–1.09]	0.40	[0.00–0.85]	0.87	[0.13–1.67]	0.14	0.74	[0.26–1.00]	0.55	[0.20–1.39]	0.79	[0.17–1.75]
Or	71.35	[48.02–79.03]	71.29	[63.73–77.69]	73.73	[42.09–98.20]	4.41	10.11	[1.21–28.43]	4.08	[1.35–10.32]	5.13	[0.93–14.11]
Ab	27.94	[20.38–49.57]	28.04	[21.52–35.74]	25.41	0–57.14]	88.02	85.48	66.31–93.55]	95.20	87.77–98.53]	92.69	84.92–97.76]
An	0.71	[0.24–2.41]	0.67	[0.16–2.19]	0.86	[0–10.17]	7.57	4.41	[0.37–9.21]	0.72	[0–5.52]	2.18	[0–6.81]
As ppm	4	[0–6]	3	[0–4]	8	[0–14]	1	2	[0–3]	1	[0–1]	1	[0–3]
Ba	115	[25–613]	146	[0–725]	70	[4–899]	4	1	[0–2]	1	[0–61]	4	[0–91]
Ce	4	[0–28]	9	[0–45]	18	[4–36]	20	26	[22–28]	25	[1–55]	31	[10–102]
Co	0	[0–3]	1	[0–3]	2	[0–5]	1	1	[1–1]	1	[1–1]	1	[1–5]
Cr	24	[8–54]	12	[3–76]	10	[1–60]	9	22	[0–31]	5	[1–37]	3	[0–33]
Cs	11	[0–39]	12	[3–37]	41	[0–125]	0	2	[0–4]	6	[1–19]	3	[0–24]
Ga	16	[1–28]	16	[5–18]	18	[15–26]	32	25	[25–35]	27	[1–48]	24	[3–36]
Hf	3	[2–12]	5	[3–5]	4	[2–7]	5	6	[5–7]	5	[1–6]	5	[2–9]
La	1	[1–3]	8	[0–19]	6	[0–12]	10	2	[2–4]	5	[1–12]	6	[1–38]
Nb	4	[0–78]	4	[0–4]	4	[4–27]	0	0	[0–78]	0	[0–58]	0	[0–50]
Nd	5	[0–17]	6	[3–21]	6	[2–12]							
Ni	5	[1–7]	4	[4–7]	7	[5–11]	4	4	[2–12]	3	[1–4]	4	[2–11]
Pb	40	[15–121]	32	[7–39]	14	[3–132]	42	32	[23–54]	30	[17–51]	32	[16–46]
Rb	378	[217–667]	586	[44–1065]	2401	[322–3729]	6	5	[1–190]	81	[0–260]	74	[0–241]
Sn	6	[1–26]	6	[1–13]	16	[3–57]	2	1	[1–2]	1	[1–63]	6	[1–33]
Sr	24	[4–64]	50	[14–84]	57	[13–281]	56	48	[23–107]	63	[5–204]	82	[31–190]
Th	1	[0–3]	1	[0–1]	1	[0–3]	0	0	[0–0]	0	[0–1]	0	[0–10]
V	11	[1–19]	11	[8–12]	12	[0–13]	6	5	[1–14]	6	[1–10]	3	[1–15]
Y	4	[1–15]	7	[3–16]	44	[2–69]	5	5	[3–5]	5	[4–5]	5	[4–6]
Zn	2	[0–25]	2	[0–17]	8	[1–32]	8	20	[6–64]	4	[0–36]	7	[1–31]
Zr	18	[17–272]	17	[8–20]	16	[13–96]	193	338	[105–906]	122	[54–727]	148	[17–331]
K/Rb	241	[140–467]	141	[88–264]	36	[23–247]	2606	1278	[84–2282]	78	[21–1660]	74	[36–427]

I, II, III and IV: pegmatite type, *n*: number of samples.

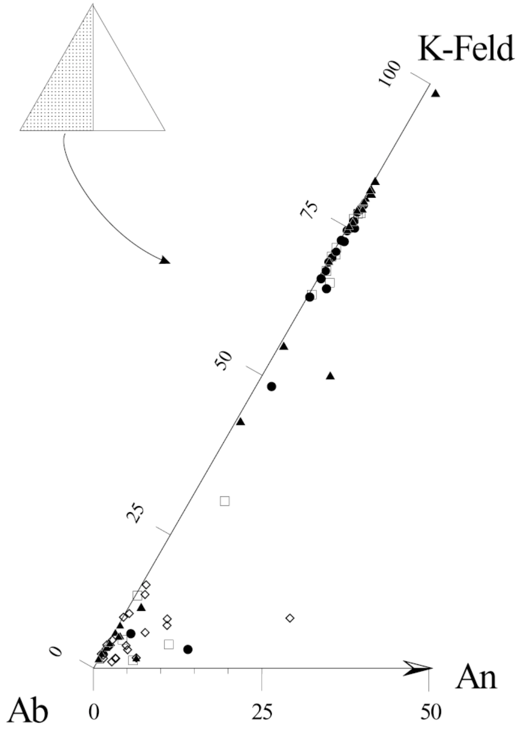


FIG. 2. Or–Ab–An diagram showing the composition of the feldspars from the Cap de Creus pegmatites. Open diamonds: type- I pegmatites, open squares: type-II pegmatites, and triangles: type-III pegmatites. Circles: ?

The role of other elements, such as zirconium, rare-earth elements, zinc and tin, is not clear. Zirconium occurs in small amounts; it is unclear if this element occur in the feldspar structure or as small inclusions of zircon.

The Y content of K-feldspar shows a good correlation with the Rb content. Yttrium increases from type-I to type-III pegmatites (Fig. 3d). This increase of yttrium is accompanied with the occurrence of Y-rich minerals, as aeschynite-(Y) and fergusonite-(Y) (Alfonso 1995). Cerium and lanthanum also increase with the pegmatite evolution, in a way similar to Rb.

The zinc content increases with the degree of evolution of the pegmatite. The tin content of K-feldspar from these pegmatites is uniform, although the richest samples belong to the latest stages of the pegmatite crystallization, where cassiterite and other Sn-rich minerals are formed (Alfonso 1995, Alfonso *et al.* 1995), and therefore contamination by inclusions of these minerals could be suspected.

Other possible substitutions in the K-feldspar involve the *T* sites: phosphorus and gallium, and probably arsenic. Phosphorus is incorporated in feldspars in tetrahedral coordination by substitution for Si ($AlPSi_{-2}$: London *et al.* 1990). K-feldspar from Cap de Creus pegmatites has a high P content, up to 1.09 wt.% P_2O_5 in type-I pegmatite. In type II, it is generally ≤0.85 wt.%, and in type III, from 0.13 to 1.67 wt.% P_2O_5. Though somewhat erratic, the phosphorus content increases with pegmatite evolution (Fig. 3e). These are among the highest contents in feldspars documented from any known pegmatite field. As a comparison, K-feldspar of beryl–columbite pegmatites from Yellowknife contains up to 0.65 wt.% of P_2O_5 and up to 0.70 wt.% of P_2O_5 in beryl–columbite–phosphate pegmatites; K-feldspar from the Cross Lake (Manitoba) granitic pegmatites reaches up to 1.2% (London *et al.* 1990).

The amount of gallium exhibits a negative correlation with K/Rb (Fig. 3f). A similar correlation has been observed in other pegmatite fields (Černý *et al.* 1985). The amount of arsenic correlates positively with rubidium.

Plagioclase

The plagioclase present in the four types of pegmatites is albite, except for one sample of oligoclase (An_{15}) in type-I pegmatites. Usually, the anorthite component of the plagioclase decreases with the degree of evolution of the pegmatite.

The chemical composition of plagioclase from the Cap de Creus pegmatite in Tables 2 and 3 reveals that P_2O_5 increases with the degree of evolution (up to 0.15 wt.% in type-I pegmatites, up to 1.00 wt.% in type-II,

TABLE 3. COMPOSITION OF PLAGIOCLASE AND MICROCLINE FROM TYPE-I PEGMATITES, CAP DE CREUS, CATALONIA, SPAIN

	Plagioclase				Microcline		
Zone	Wall		Intermediate		Wall	Intermediate	
Sample	11-3a	11-3b	12-38a	12-38b	11-3	12-38c	12-38d
SiO_2 wt.%	68.41	67.83	64.38	67.11	62.99	61.99	61.51
Al_2O_3	21.53	20.22	20.10	19.66	19.26	19.48	19.32
TiO_2	-	-	-	0.01	0.10	-	-
Fe_2O_3	0.03	-	0.05	-	0.05	0.11	0.01
MnO	-	-	-	-	0.10	0.06	0.01
CaO	1.25	0.14	0.12	0.06	-	-	-
Na_2O	8.05	10.83	10.23	11.20	1.06	3.19	1.50
K_2O	0.13	0.54	2.27	0.13	16.16	13.57	15.97
P_2O_5	0.12	0.36	1.13	0.20	-	1.47	1.44
Total	99.52	99.92	98.28	98.37	99.72	99.87	99.76
	atoms per formula unit (on the basis of 8 atoms of oxygen)						
Si *apfu*	2.968	2.963	2.890	2.976	2.936	2.859	2.863
Al	1.101	1.041	1.063	1.027	1.058	1.059	1.060
Ti	-	-	-	-	0.004	-	-
Fe^{3+}	0.001	-	0.002	-	0.002	0.004	-
Mn^{2+}	-	-	-	-	0.004	0.002	-
Ca	0.058	0.007	0.006	0.003	-	-	-
Na	0.677	0.917	0.890	0.963	0.096	0.2385	0.135
K	0.007	0.030	0.130	0.007	0.961	0.798	0.948
P	0.004	0.013	0.043	0.008		0.057	0.057
Or	0.94	3.15	12.67	0.72	90.92	76.99	87.54
Ab	91.24	96.12	86.74	98.97	9.08	23.01	12.46
An	7.82	0.73	0.59	0.31	-	-	-

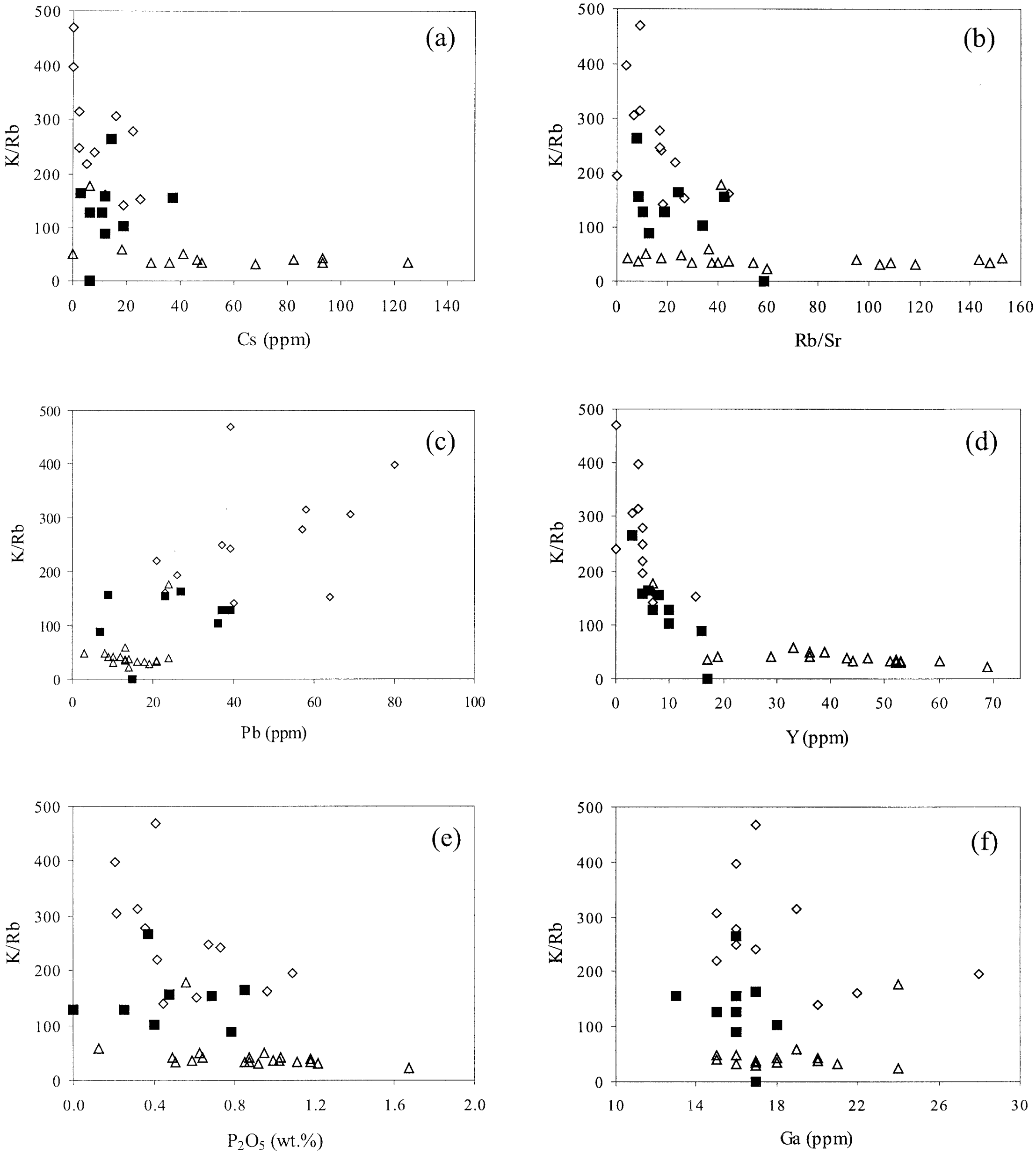


FIG. 3. K/Rb *versus* a) Cs, b) Sr, c) Pb, d) Y, e) P_2O_5, and f) Ga in K-feldspar from the Cap de Creus pegmatites. Open diamonds: type-I pegmatites, filled squares: type-II pegmatites. and open triangles: type-III pegmatites.

1.39 wt.% in type-III and up to 1.75 wt.% in type-IV pegmatites). These values are significantly higher than those in the literature (Kontak *et al.* 1996). In type-I pegmatites, apatite is rare, and the only phosphate present. The P_2O_5 – CaO plot indicates that these components evolve in an opposite sense (London *et al.* 1990). In the presence of Ca and P, apatite is formed preferentially. In the presence of only one of these components, the other, Ca or P, goes to form part of the feldspars.

Plagioclase contains only small amounts of the trace elements mentioned above, presumably because of the small size of the Na^+ (Černý 1994). In general, there is no direct relation between the trace-element content of plagioclase and the type of pegmatite. Nevertheless, the content of Rb, Sr and Ba is higher in type-III and -IV pegmatites relative to type-I and -II pegmatites. The Rb content in albite is up to 42 ppm in type-I pegmatites and up to 241 ppm in type-IV, and Sr is between 23 and 107 ppm in type-I and type-II pegmatites, and up to 204 ppm in type-III pegmatites. The barium content is similar in all the pegmatite types, but the highest values are found in type-IV pegmatites (up to 91 ppm).

Other elements, such as Cs (up to 24 ppm), Ga (1–48 ppm) and Ce (1–102 ppm), do not exhibit a marked variation with pegmatite type. Zirconium is mostly below 190 ppm, and exceptionally can reach 400 ppm. These peaks may well be related to inclusions of zircon.

The contents of yttrium and tin are practically negligible in albite, in spite of the fact that in many cases the albite is associated with cassiterite and *REE*-bearing minerals. Therefore, the higher yttrium and tin contents in K-feldspar coexisting with albite suggest that these elements can be accommodated in the K-feldspar structure.

Muscovite

The chemical composition of the muscovite from the Cap de Creus pegmatites is given in Table 4. The interlayer-cation (I-site) occupancy is not complete, with values between 1.57 and 2.00, median of 1.80,

TABLE 4. COMPOSITION OF MUSCOVITE FROM CAP DE CREUS GRANITIC PEGMATITES, CATALONIA, SPAIN

	I		II		III		IV	
	median	range	median	range	median	range	median	range
	($n = 4$)		($n = 19$)		($n = 34$)		($n = 19$)	
SiO_2	46.62	[45.33–47.81]	46.80	[42.46–51.27]	45.74	[42.55–63.96]	45.89	[43.42–47.33]
Al_2O_3	36.83	[36.04–38.97]	36.32	[33.71–37.99]	38.31	[22.46–39.96]	38.72	[36.35–40.42]
TiO_2	0.20	[0.07–0.48]	0.13	[0.03–0.36]	0.08	[0.03–0.20]	0.02	[0.01–0.17]
Fe_2O_3	1.34	[0.64–1.67]	1.83	[1.22–2.98]	0.92	[0.58–1.39]	0.86	[0.52–2.52]
MgO	0.21	[0.01–0.42]	0.35	[0.13–0.52]	0.07	[0.00–0.32]	0.09	[0.06–0.41]
MnO	0.01	[0.01–0.03]	0.02	[0.01–0.08]	0.03	[0.01–0.11]	0.02	[0.01–0.04]
CaO	0.05	[0.01–0.21]	0.06	[0.00–0.24]	0.06	[0.00–1.87]	0.08	[0.00–0.64]
Na_2O	0.82	[0.69–1.06]	0.73	[0.48–0.99]	0.85	[0.56–1.59]	1.14	[0.74–1.32]
K_2O	9.49	[9.09–10.09]	9.40	[7.02–10.13]	8.99	[5.16–9.95]	8.95	[7.56–9.88]
P_2O_5	0.11	[0.04–0.21]	0.08	[0.05–0.29]	0.11	[0.06–1.33]	0.15	[0.08–0.54]
Pg	11.82	[10.34–13.77]	10.77	[6.72–13.65]	13.77	[9.96–22.91]	15.87	[11.10–19.44]
Cel	2.30	[0.10–4.43]	3.72	[1.56–5.66]	1.07	[0–4.56]	1.47	[0.70–4.43]
Ms	85.87	[83.51–86.96]	85.51	[82.90–89.41]	85.16	[72.87–89.34]	82.66	[78.46–88.14]
As	10	[7–14]	5	[0–13]	7	[0–30]	8	[2–26]
Ba	410	[82–651]	283	[51–899]	219	[13–786]	169	[5–505]
Co	5	[0–16]	10	[0–20]	17	[1–59]	37	[9–76]
Cr	6	[3–9]	10	[1–16]	8	[0–16]	8	[1–40]
Cs	100	[44–220]	69	[3–273]	115	[9–631]	526	[18–1604]
Cu	20	[7–163]	16	[5–103]	17	[1–140]	12	[0–65]
F	1568	[1006–1632]	1924	[187–4016]	2290	[20–48887]	1059	[62–12261]
Nb	358	[304–493]	344	[217–447]	348	[229–2437]	325	[219–452]
Ni	17	[5–30]	14	[1–32]	13	[1–50]	9	[1–30]
Pb	22	[7–51]	9	[0–46]	16	[1–103]	22	[7–122]
Rb	1479	[918–4610]	1837	[1108–3192]	3066	[792–9587]	5756	[2650–13996]
Sn	231	[166–437]	274	[109–1565]	349	[86–1122]	649	[195–1358]
Sr	21	[4–57]	13	[8–61]	15	[5–188]	14	[10–41]
V	5	[2–8]	12	[1–31]	6	[0–29]	9	[1–63]
W	102	[10–132]	30	[10–174]	17	[1–109]	9	[1–22]
Y	10	[3–37]	4	[1–16]	13	[1–112]	22	[1–118]
Zn	97	[64–126]	89	[51–156]	149	[46–286]	112	[65–227]
Zr	10	[0–21]	44	[0–291]	36	[2–492]	21	[5–229]
K/Rb	54	[18–83]	43	[26–69]	23	[8–54]	13	[5–30]

major oxides in wt.%, trace elements in ppm. I, II, III and IV: pegmatite type. *n*: number of samples. Symbols: Pg: paragonite, Cel: celadonite, Ms: muscovite.

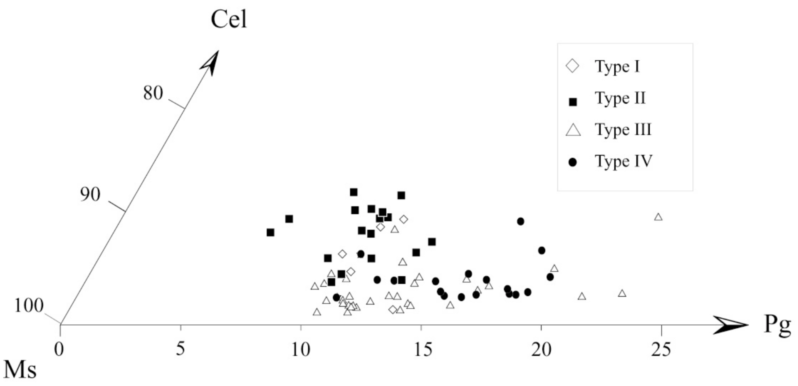


FIG. 4. Muscovite – paragonite – celadonite diagram showing the composition of muscovite from the Cap de Creus suite of pegmatites.

owing to a low K content. K_2O usually is below 9.8 wt.%, at least 1 wt.% lower than in muscovite from other pegmatite occurrences (Jolliff *et al.* 1992, Roda *et al.* 1995). The same result was reported in muscovite from the McAllister Sn–Ta-bearing pegmatite, Alabama, by Foord & Cook (1989), who attributed this content to near-surface weathering and leaching.

Muscovite from all the Cap de Creus pegmatites exhibits a relatively high proportion of the paragonite component (Fig. 4), in comparison with that from other pegmatites. Type-I and type-II pegmatites have similar paragonite contents, between 6.7 and 13.8 mol.%. Muscovite from some of type-III pegmatites also exhibits a similar range, but muscovite from the later episodes (quartz–muscovite veins) has higher values, up to 22.9%. The muscovite in type-IV pegmatites has the highest paragonite component, common values being between 15.9 and 19.4%. This tendency for sodium enrichment in muscovite is consistent with the geochemical trend in the pegmatite field, and correlates with the increase in the albite content of the pegmatites. On the other hand, these contents are higher than those expected for pegmatites (Zane & Rizzo 1999). The paragonite component of micas from Black Hills pegmatites is lower; it ranges from 2.5 to 11 mol.% (Jolliff *et al.* 1987). However, Černý & Burt (1984) also indicated similar high values in the paragonite component for the mica pegmatites from the north of Karelia.

The celadonite component of the Cap de Creus muscovite is very low, in agreement with the general composition of muscovite in granite pegmatites (Zane & Rizzo 1999). There is a decreasing trend with advancing pegmatite evolution.

Certain trace-element contents in muscovite exhibit a progressive geochemical evolution from type-I to type-IV pegmatites. The K/Rb *versus* Cs in muscovite is the best indicator of the pegmatite evolution (Černý *et al.* 1985, Jolliff *et al.* 1987, Černý 1991). In the Cap de Creus pegmatites, there is a progressive enrichment in rubidium from type-I (up to 4600 ppm) to type-IV pegmatites (up to 14000 ppm). Similarly, cesium increases, and is especially notable in type-IV pegmatites, with up to 1360 ppm (Fig. 5a).

Barium exhibits a broad range of values in these pegmatites, without a strong correlation with pegmatite type, although the less evolved pegmatites usually have higher Ba contents, as expected according to Černý & Burt (1984).

The Y content of muscovite shows a good negative correlation respect to the K/Rb (Fig. 5b). There is an increase in Y in these pegmatites with degree of evolution. This enrichment in yttrium is probably due to inclusions of Y-rich rare-earth-bearing minerals.

The tin content of muscovite has a negative correlation with K/Rb; the highest values belong to the most evolved pegmatites (Fig. 5c). In addition, type-IV pegmatites exhibit the highest tin contents in the quartz–muscovite veins relative to the zones formed earlier. Sn reaches up to 1350 ppm. According to Smeds (1992), the tin content of micas is an indicator of cassiterite, with the upper limit in his study being 600 ppm. In the present case, the increase in tin content in the quartz–muscovite veins is accompanied by the occurrence of higher proportions of tin-rich minerals, such as cassiterite and nigerite. This tin content of 1350 ppm is one of the highest reported in literature for muscovite from pegmatites (Jolliff *et al.* 1992, Roda *et al.* 1995, Wise 1995), and from the Panasqueira Sn–W deposits from Portugal (Neiva 1987).

Tungsten shows a positive correlation with K/Rb (Fig. 6a), with muscovite in type-I pegmatites being the richest in W (10–132 ppm), whereas that in type-IV pegmatites is the poorest (up to 22 ppm). The titanium content also shows a positive correlation with the K/Rb (Fig. 6b). Zinc does not exhibit a strong correlation with K/Rb, although the highest values belong to the most evolved pegmatites.

According to the data of Corbella (1990), the lithium contents of muscovite are invariably lower than 300 ppm, and the variation among the different types of pegmatite is not significant. Fluorine content has no direct relation with the pegmatite type, although the highest contents are in type-III pegmatites. Lithium and fluorine can be incorporated in three different mineral associations in pegmatites: anhydrous Al-silicates, phosphates and Li- and F-rich micas. The incorporation in one or the other will depend on the absolute and relative activities of PO_4^{3-}, F, H^+, Na^+ and K^+ (Černý *et al.* 1985). In the Cap de Creus pegmatites, the PO_4^{3-} activity is very high, as demonstrated by the high content of phosphate minerals (Corbella & Melgarejo 1993, Alfonso *et al.* 1996) and the high content in feldspars. In such specialized pegmatites, lithium is incorporated

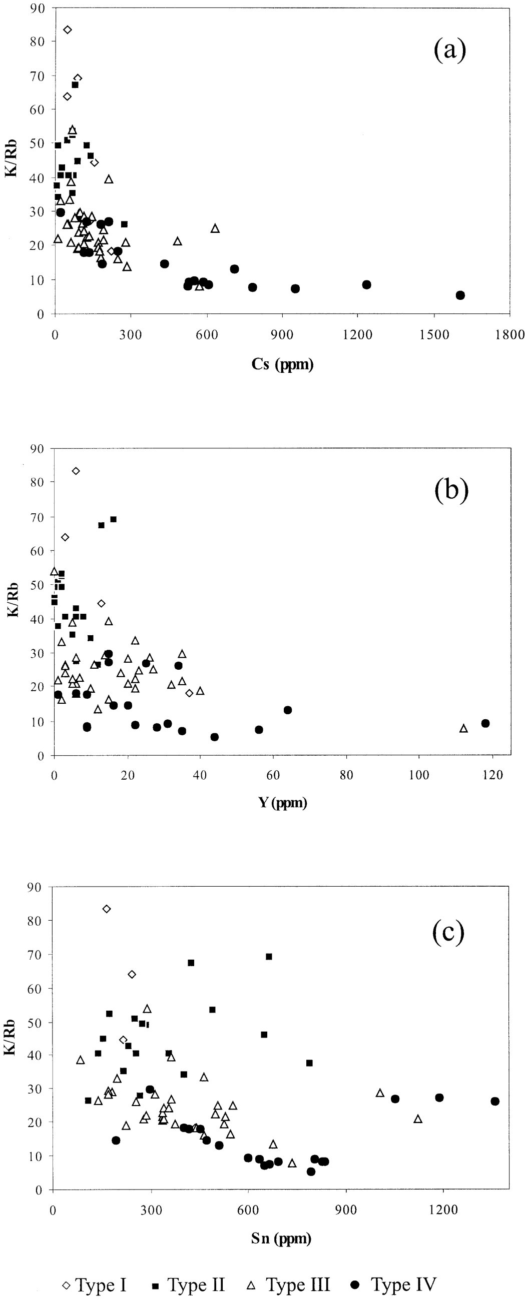


FIG. 5. K/Rb *versus* a) Cs, b) Y, and c) Sn in muscovite from the Cap de Creus pegmatites.

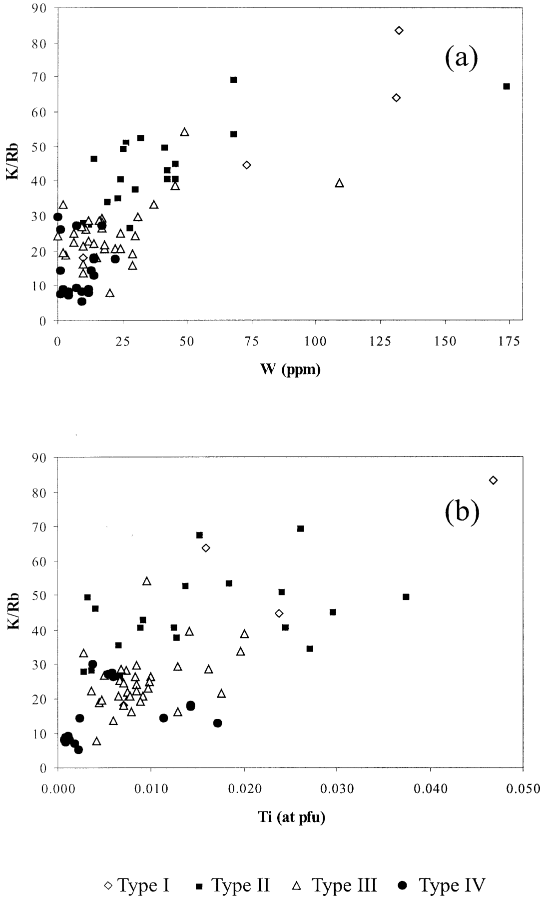


FIG. 6. K/Rb *versus* a) W, and b) Ti in muscovite from the Cap de Creus pegmatites.

preferentially in phosphates (mainly of the amblygonite–montebrasite series) relative to lithium aluminosilicates such as spodumene and petalite.

Niobium content shows no correlation with K/Rb; values are higher than data reported in other examples of muscovite, as in lithian muscovite from spodumene-bearing pegmatites (Wise 1995).

Fractionation of Components

K-feldspar – albite

K-feldspar from the Cap de Creus pegmatites exhibits higher contents of Ba, Rb, Cs, Y, Sn, Nb, V and Cr than albite. Albite is richer in Sr, Cu, As, Ce and Ga (Table 2, Fig. 7). Other elements, usually present in lower concentrations, have a comparable affinity for K-feldspar and albite: Hf, Pb, Zn, W, Co and Ni. These results are similar to those of Neiva (1995) in the granitic aplites and pegmatites from Alijó–Sanfins, Portugal, except for Sr, Pb and Ga.

Phosphorus contents are similar in K-feldspar and in albite, contrary to the results of London *et al.* (1990). Nevertheless, where they coexist (Table 3), the amount of phosphorus generally is slightly higher in K-feldspar than in albite.

K-feldspar – muscovite

Figure 8 shows the evolution of the content of the elements in muscovite and K-feldspar that best represent the geochemical evolution among the different types of pegmatite. These trace elements are more abundant in the structure of muscovite than in K-feldspar. Whereas the rubidium content in the K-feldspar of the Cap de Creus pegmatites has a maximum of 3729 ppm, that in muscovite is much higher, up to 9587 ppm in type-III pegmatites and 13,996 ppm in type-IV pegmatites. Similarly, cesium, in the same type of pegmatite, is about five times higher in muscovite than in K-feldspar. This trend is also followed by Y, Co, Cu, Nb, Zn and Sn. Similar results have been reported by many authors, *e.g.*, Černý & Lenton (1995) in rare-element pegmatites of the complex type, petalite subtype in the Bernic Lake pegmatite group, Cat Lake – Winnipeg River field, Manitoba, and Morteani *et al.* (1995), in granitic pegmatites from the Achala granitic complex, Argentina.

Strontium is one of the few elements that exhibits a greater affinity for feldspar than for muscovite.

In some cases, these trace-element contents can be partly justified by the later formation of muscovite with respect to K-feldspar. The latest units to be formed, such as the quartz–muscovite veins, lack K-feldspar, and are the richest in Rb, Cs and Sn. Nevertheless, this fact cannot justify all the difference in the trace-element contents between K-feldspar and muscovite. The different level of acceptance of trace elements between K-feld-

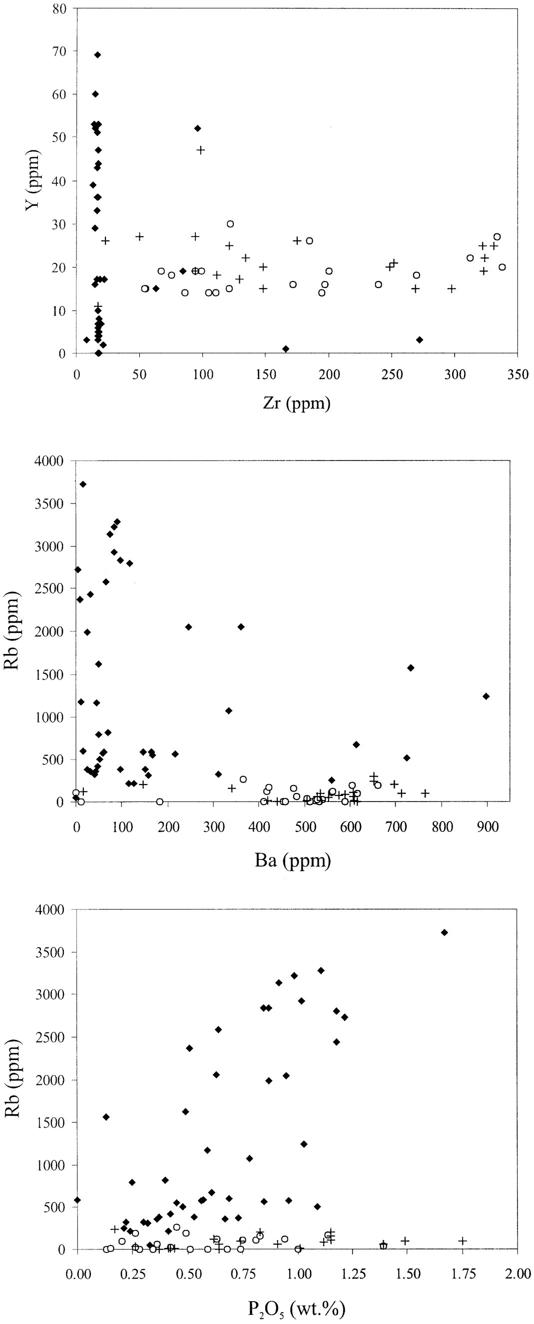
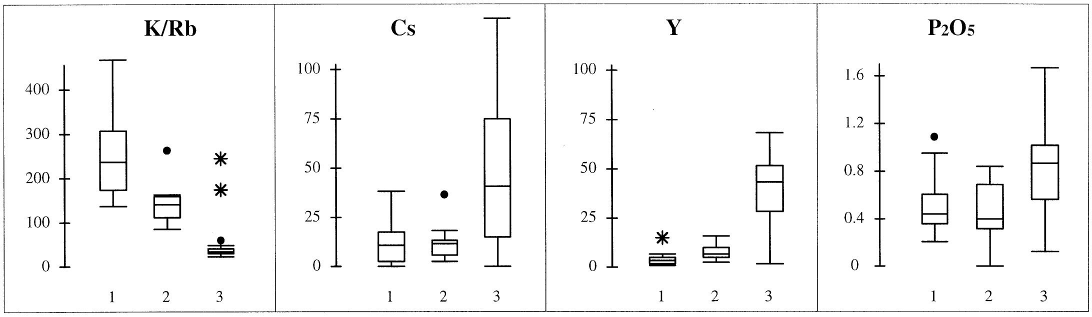


FIG. 7. Comparison of some trace-element contents between K-feldspar and albite from the Cap de Creus pegmatites. Filled diamonds: K-feldspar (from type I, II and III pegmatites), open circles: albite from type-I, -II and -III pegmatites, and crosses: albite from type-IV pegmatites.

K–feldspar



Muscovite

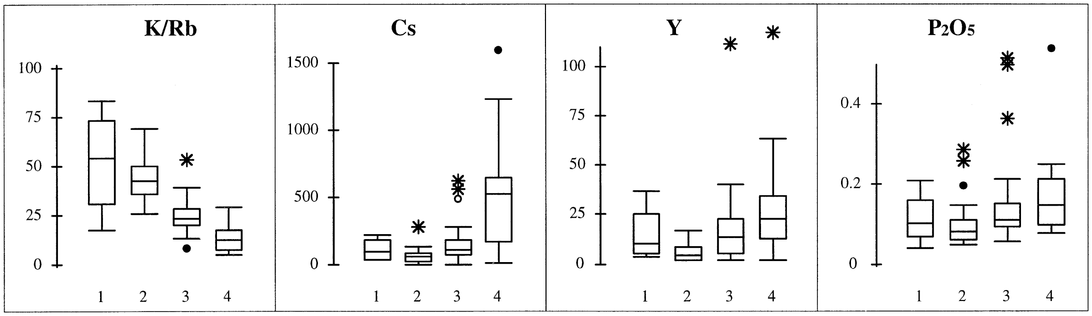


FIG. 8. Distribution of K/Rb, Cs (ppm), Y (ppm) and P_2O_5 (wt.%) in K-feldspar and muscovite from the Cap de Creus suite of pegmatites. In each plot, the central box depicts data between the 25th (Q1) and 75th (Q3) percentiles; the stars stand for extreme outliers (values $\geq$ Q3 + 3 IQR), and the dots, for mild outliers (Q3 + 1.5 IQR $\leq x_i \leq$ Q3 + 3 IQR); IQR = InterQuartile Range.

spar and muscovite may be related to the structure of these minerals. Černý *et al.* (1985) advanced this interpretation to explain the higher content in some trace elements in white mica, and especially lepidolite, compared to K-feldspar.

For some elements, such as Rb, Cs, Ba and Sr, the partition coefficients between feldspars and melt and muscovite and melt are well known (Icenhower & London 1995, 1996). The partition coefficients between muscovite and melt for Rb, Cs and Ba are higher than those between K-feldspar and melt, whereas those coefficients for Sr are higher in K-feldspar than in muscovite. These data are in accordance to the distribution of these elements observed in feldspars and muscovite from the Cap de Creus pegmatites.

CONCLUSIONS

The chemical composition of feldspars and micas is rather homogeneous throughout all the zones of a single body of pegmatite. This fact may be a consequence of subsolidus re-equilibration phenomena.

There is a progressive increase in the level of P, Rb, Cs, Y, Sr, Ga, Ce, W and Sn in K-feldspar from type-I to type-III pegmatites. Concentrations of Pb and Zr decrease in K-feldspar from type-I to type-III pegmatites. In albite, there is no correlation between the concentration of most trace elements and pegmatite type. This fact may reflect the overlap among the different generations of this mineral.

The high P content of feldspars indicates high levels of concentration of this element in the melt from the earliest stages of pegmatite crystallization. This fact agrees with the continuous crystallization of primary phosphate minerals in the pegmatite sequence. The content of the other trace elements in the feldspars reflects their buildup in the pegmatite-forming melt. A higher content is usually correlated with the occurrence of characteristic minerals of these elements. Thus, the feldspars are useful indicators for mineral exploration of commodities such as tantalum and cesium. Moreover, the high P content in albite in phosphate-enriched pegmatites is inherited from the replaced K-feldspar and can be used as a criterion for late metasomatic processes in pegmatites. Since these processes are linked with Nb–Ta enrichment, the P content in albite can be used as an additional criterion for rare-element mineral exploration.

Muscovite from the Cap de Creus pegmatites exhibits a progressive geochemical evolution from type-I to type-IV pegmatites. There is a progressive enrichment in Rb, Cs and Y from type-I to type-IV pegmatites. The highest Zn and Sn contents in muscovite belong to pegmatites of type III and type IV. The content of W has a positive correlation with the K/Rb ratio.

The paragonite component (9.3–22.0%) increases with the evolution, and correlates with the increase in the albite content of the pegmatites. The celadonite component is very low.

The progressive geochemical trend in feldspars and muscovite from the Cap de Creus pegmatites suggests a genetic linkage by fractionation of a common parental magma.

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