

# Geochemistry of muscovite from pegmatites of the Eastern Brazilian pegmatite province: a clue to petrogenesis and mineralization potential

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**Abstract:** Single crystals of muscovite ( $N = 143$ ) from different zones of selected granitic pegmatites belonging to two well-known gem-producing districts of the Eastern Brazilian Pegmatite Province (northern Araçuaí and western Governador Valadares in Minas Gerais) were analyzed for major, minor and trace elements. Structural formulae display considerable (Fe+Mg) variation within the octahedral site, coupled with tetrahedral Si-Al substitution (Tschermak substitution – phengite component). Tetrahedral Si varies from 3.06 to 3.21 and  $\text{Al}^{\text{VI}}$  from 1.71 to 1.90 *apfu*.

Trace elements in muscovite are significant indicators for the economic potential of pegmatites as well as for the differentiation degree and origin of the magma. Muscovite with higher Li and B contents is characteristic for gem-tourmaline-bearing pegmatites. Lower concentrations in K/Rb, Ti and Mg and higher F contents are found in muscovites from higher differentiated zones within the pegmatite. In comparison with anatectic pegmatites, relatively low K/Rb ratios of muscovite from the studied pegmatites are indicative of derivation by fractional crystallization of granite magma. Zn, Ga and Y, elements rarely analyzed in micas, tend to increase with decreasing K/Rb ratios, showing them to be good indicators of the fractionation degree of magmas.

**Key-words:** muscovite, crystal chemistry, trace elements, pegmatite, magmatic differentiation, Brazil.

## Introduction

Muscovite is a common rock-forming mineral in the terrestrial crust. In granitic pegmatites, it is the fourth major mineral after quartz, K-feldspar and plagioclase (albite variety). Although muscovite is in principle a simple, water- and potassium-containing aluminosilicate with the crystal formula  $\text{KA}1_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$ , a complex exchange of cations can occur, and a formula can hence be written as  $\text{XY}_{-2}\square_{1-0}\text{Z}_4\text{O}_{10}(\text{W})_2$ , with:

$X = \text{K}^+, \text{Na}^+$ , but also Rb, Cs, Ca, Ba and  $\text{NH}_4$   
 $Y = \text{Al}^{3+}, \text{Fe}^{3+}, \text{Fe}^{2+}, \text{Mg}^{2+}$ , but also Li,  $\text{Mn}^{2+}$ , Cr, V, ...,  $\text{Ti}^{4+}$

$Z = \text{Si}^{4+}, \text{Al}^{3+}, \text{B}, \text{Be}$  and  $\text{Fe}^{3+}$

$W = \text{OH}, \text{F}, \text{Cl}$  and S

According to Rieder *et al.* (1998) the typical compositional ranges, expressed in *apfu*, are 3.0–3.1 for  $\text{Si}^{\text{IV}}$ , 1.9–2.0 for  $\text{Al}^{\text{VI}}$  and 0.7–1.0 for K. The cation ratio  $\text{VI}\text{R}^{2+}/(\text{VI}\text{R}^{2+} + \text{VI}\text{R}^{3+})$  is lower than 0.25 with  $\text{VI}\text{Al}/(\text{VI}\text{Al} + \text{VI}\text{Fe}^{3+})$  variation of 0.5 to 1.0.

The importance of muscovite composition as a clue to internal evolution of pegmatite magma has been discussed earlier (*e.g.*, Stern, 1966; Černý, 1982a; Černý & Burt,

1984; Roda-Robles *et al.* 1999; Jolliff *et al.* 1992), but systematic analyses within a pegmatite body and among pegmatites of a pegmatite province has been little investigated so far.

The K/Rb *versus* Cs in muscovite is considered to be the best indicator of pegmatite evolution (Černý *et al.* 1985, Jolliff *et al.* 1987, Černý 1991). According to Jolliff *et al.* (1992), trace-element concentrations of muscovite from a given pegmatite are normally cluster, although several can show some intra-pegmatite scatter, with large overlaps among different pegmatites. Micas from pegmatites derived from granitic precursor by fractionation normally show gradual enrichment in Li, Rb, Cs and F, with a decrease in Mg, Ti and the K/Rb ratio from the internal to the external zones.

In order to obtain a statistically representative set of analyses, 143 mica samples from two well-known gem-producing districts, located in the Araçuaí and Governador Valadares regions in Minas Gerais State (Fig. 1) were collected. The aim of this work is to obtain the compositional variation of pegmatite muscovite and to describe compositional variations across a single pegmatite body (wall, intermediate and core zones) and within distinct pegmatite

fields. Two questions are of interest: 1) does the chemical composition of muscovite reflect details of the petrogenesis (stages of magmatic differentiation)? and 2) does it reflect a certain mineral assemblage (presence of accessory minerals, mainly gem minerals)?

## Geological setting

A large quantity and variety of gemstones, particularly aquamarine and tourmaline, is produced in the Eastern Brazilian Pegmatite Province (EBPP), which comprises an area about 800 km long and 150 km wide. The pegmatites are spread over eastern Minas Gerais, western Espírito Santo and the southern Bahia States (Fig. 1). The EBPP is characterized not only by its geographic location but also by a particular geotectonic setting in the Neoproterozoic-Cambrian orogenic Araçuaí mobile belt generated during the Brasiliano – Pan-African cycle, which consisted of a set of orogenic cycles that lasted from about 850 to 550 Ma (Pinto & Pedrosa-Soares, 2001). The majority of the pegmatites of the EBPP are related to granite intrusions into the Brasiliano mobile belt generated during the consolidation of the Gondwana supercontinent. The pegmatites related to this orogeny are considered to be residual melts derived from S-type and also from I-type granites (*e.g.* Lobato & Pedrosa-Soares, 1993; Pedrosa-Soares *et al.*, 1999; Pinto & Pedrosa-Soares, 2001). Rb/Sr and U/Pb geochronological investigation of granite bodies intruding the Araçuaí mobile belt by Siga Jr. (1986) resulted in pre-tectonic (650 Ma), syn- to late tectonic (650–550 Ma), and post-tectonic (500–450 Ma) ages in relation to the Brasiliano orogeny.

According to Pinto & Pedrosa-Soares (2001), colored tourmaline-bearing pegmatites, *e.g.* lithium-rich pegmatites, are consolidated at intermediate crustal levels from residual melts derived from S-type granite, whereas pegmatites which are beryl-rich and poor in colored tourmaline crystallized at deeper levels from magmas derived from calc-alkaline granites. Pegmatite melts derived from the host rocks by anatexis can also be found in this province (Lobato & Pedrosa-Soares 1993).

Two pegmatite districts of the Eastern Brazilian Pegmatite Province were selected for this study: the northern Araçuaí and the western Governador Valadares districts (Fig. 1). The host rocks of the Governador Valadares pegmatites are schists, gneisses and quartzites belonging to the São Tomé Formation - Rio Doce Group, whereas those of the Araçuaí District are metasedimentary rocks of the Macaúbas Group - Salinas Formation and S-type granitoids (Fig. 1).

The Rio Doce Group comprises gneisses, a supracrustal schist sequence, sericite quartzite and intrusive granitoids (Almeida, 1981, Cunningham *et al.*, 1996; Oliveira *et al.*, 1997; Nalini *et al.*, 2000; Pedrosa-Soares & Wiedemann-Leonardos, 2000). K–Ar ages of the gneisses range from 525 to 670 Ma (Da Silva *et al.*, 1987). The rocks of this group were submitted to greenschist to amphibolite facies metamorphism, but locally sedimentary turbiditic features can still be recognized in the banded schist (Pedreira *et al.*, 1997).

The Araçuaí pegmatites are hosted by batholithic bodies of foliated biotite granitoids enclosing banded paragneisses and migmatites. According to Pedrosa-Soares *et al.* (1999) this granitic suite makes up the anatectic nucleus of the northern sector of the Araçuaí Belt, having a subalkaline to calc-alkaline, peraluminous signature. The gneissic foliation follows the regional trend of the Araçuaí Belt, indicating metamorphic recrystallization during the main deformation phase. The metamorphic ages for gneiss samples of this suite range from 655 to 591 Ma (Rb–Sr isochrons, initial Rb/Sr ratio  $\approx$  0.708; Siga Jr., 1986). Other host rocks of the Araçuaí pegmatites belong to the Macaúbas Group, a megasequence deposited on a continental passive margin in Neoproterozoic time (Pedrosa-Soares, 1995, Noce *et al.*, 1997). The Salinas Formation represents the Macaúbas Group in the internal tectonic domain of the Araçuaí Belt. Quartz-mica schist (pelitic metagraywacke) and quartzose metagraywacke predominate. Rb–Sr ages of biotite schists show that the regional metamorphism took place at about  $630 \pm 30$  Ma (Siga Jr. 1986).

Two pegmatite generations could be distinguished in the northern portion of the EBPP according to Viana *et al.* (2003). The older, 560 Ma pegmatites are related to the main stage of granitogenesis of the Brasiliano – Pan-African orogeny. They are mineralogically more complex and more evolved than the second pegmatite generation, which is dated at 500 Ma and related to the late stage of granitogenesis.

## Location and description of the samples

The Araçuaí and Governador Valadares pegmatite districts are both important gem-producing regions. In each district, three pegmatites were chosen for the chemical investigation of muscovite. The pegmatite bodies are over 20 m wide and show either a simple or complex textural and mineralogical zoning, which is almost always well defined (Table 1). According to the mineralogical assemblage, Ipê and Aci (Governador Valadares region), and Serra Cascalheira (Araçuaí region) are pegmatites of simple mineralogy, lacking Li minerals and colored tourmaline, as shown in Table 1. The barren beryl-topaz-fluorapatite-bearing Barreiro pegmatite (Araçuaí region) also lacks colored tourmaline, differing from the other mineralogically simple pegmatites because it contains F-rich minerals such as fluorapatite and topaz, and may be classified as miarolitic (Černý 1982a,b, Černý, 1992, Černý *et al.*, 1985, Černý *et al.*, 2005).

The beryl- and tourmaline-bearing Cruzeiro (Governador Valadares region) and Rocinha (Araçuaí region) pegmatites are mineralogically more complex (Table 1) and may be considered as belonging to Rare-Element class pegmatites of the LCT family (Černý 1991, 1992, Černý *et al.* 2005).

Muscovite of variable size is common in all zones, mostly as pseudo-hexagonal or fish-tail shaped books. Larger crystals are found in the intermediate zone which exceed, in some cases, diameters of 30 cm. We collected 143 muscovite samples from border, wall, intermediate and core zones for a comparative study of the internal differentiation

Table 1. Mineralogy according to the internal zoning and classification of the studied pegmatites.

Region	Pegmatite	Border zone	Wall zone	Intermediate zone	Core	Pockets	Host rocks/ metamorphic conditions	Pegmatite type
Aci		Quartz, muscovite, black tourmaline	Quartz - K-feldspar intergrowths, muscovite, albite	Quartz - K-feldspar, muscovite, quartz, beryl	Milky quartz	Lepidolite, albite, muscovite,	Schist and gneiss rock	Gem Simple
		Quartz, muscovite, black tourmaline, garnet	Quartz - K-feldspar intergrowths, albite, muscovite	Quartz, K-feldspar, muscovite, lepidolite, pink and green tourmaline, beryl, cassiterite, niobotantalite,	Milky quartz, muscovite	Lepidolite, albite, pink and green tourmaline, muscovite, altered beryl,	Schist and quartzite 4.5–5.0 kbar 530–650 °C ( <i>e.g.</i> , Talarico & Pereira 1997)	Gem tourmaline- bearing Complex
GV	Cruzeiro	Quartz, muscovite biotite intergrowth, garnet, black tourmaline,	Quartz - K-feldspar Intergrowths, muscovite, albite	Quartz, K-feldspar muscovite, beryl, columbite/tantalite	Hyaline/milky/ pink quartz	Quartz, muscovite, albite beryl, amethyst	Schist 4.5–5.0 kbar 530–650 °C ( <i>e.g.</i> , Talarico and Pereira 1997)	Beryl-bearing Simple
Rocinha		Quartz, muscovite, green tourmaline, albite	Quartz - K-feldspar intergrowths, muscovite, albite	K-feldspar, muscovite, quartz, green/ pink/ bi- colored tourmaline, morganite, altered aquamarine, albite	Milky quartz, pink tourmaline, lepidolite, muscovite	Quartz, albite, lepidolite, bi- colored and green tourmaline	Schist 2.5–5.5 kbar 500–740 °C ( <i>e.g.</i> , Costa 1989)	Gem tourmaline- bearing Complex
		Quartz, black tourmaline, garnet, biotite	Quartz - K-feldspar intergrowths, muscovite, albite	K-feldspar, quartz, muscovite, beryl, albite	Hyaline/milky/ pink quartz	Quartz, muscovite, altered amethyst	Granite-gneiss	Beryl-bearing Simple
Ar	Serra Casalheira	Quartz, black tourmaline, garnet, biotite	Quartz - K-feldspar intergrowths, muscovite, albite	Quartz - K-feldspar, muscovite, beryl, fluorapatite, topaz, albite	Hyaline and milky quartz	Quartz, K-feldspar partially replaced by muscovite, beryl, albite	Granite	Beryl-topaz- fluorapatite- bearing Simple
	Barreiro	Quartz, black muscovite, biotite, hematite	Quartz - K-feldspar intergrowths, muscovite, hematite	Quartz - K-feldspar, muscovite, beryl, fluorapatite, topaz, albite	Hyaline and milky quartz	Quartz, K-feldspar partially replaced by muscovite, beryl, albite	Granite	Beryl-topaz- fluorapatite- bearing Simple

GV: Governador Valadares AR: Araçuaí

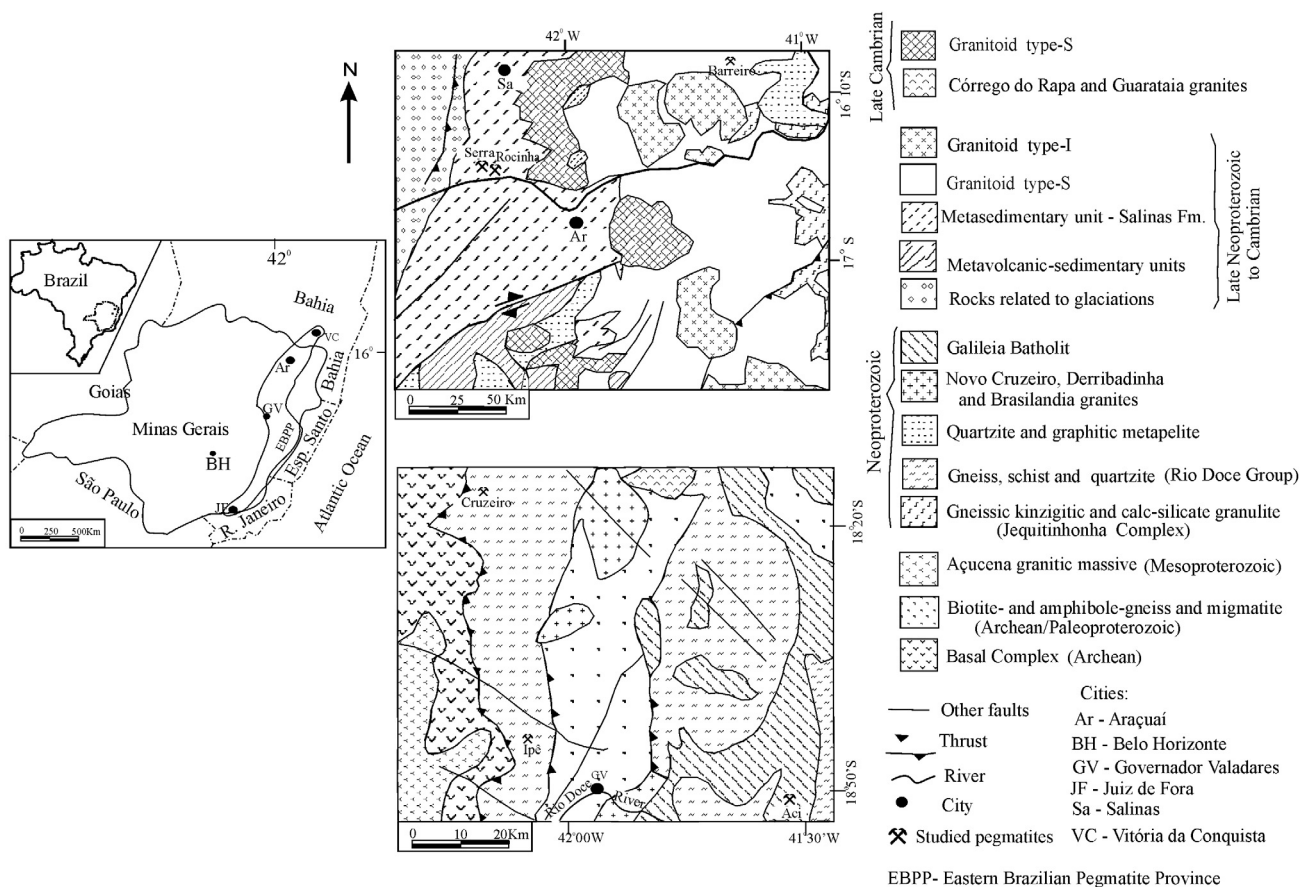


Fig. 1. Simplified geological map of the Araçaí and Governador Valadares regions with location of the studied pegmatites. Outline of the Eastern Brazilian Pegmatite Province (EBPP) is also shown (modified after Oliveira *et al.*, 1997 and Pedrosa-Soares and Wiedemann-Leonardos, 2000).

within these bodies, and to be able to compare pegmatites of different mineralogy.

### Analytical techniques

Chemical analyses were performed at the University of Basel, Switzerland, using the techniques described below.

High-power X-ray fluorescence analysis (XRF, 4 kW excitation voltage) was employed in the present study because of its good sensitivity not only for the main constituents of muscovites, but also for many important elements such as Ti, V, Cr, Ga, Zn, Y and Nb, as well as elements with high atomic numbers above 45 (Sn, Ba, Cs, La and Ce) and elements with low atomic numbers, below 11 (B and F).

Inductively-coupled plasma optical emission (ICP-OE, Spectro-flame, Spectro, Germany) was applied in selected cases for Li, Be and B analysis, and combustion analysis — CA (RT-412 of Leco, USA) was used for the determination of the water contents following the technique described by Petrova *et al.* (2001).

Chemical analyses by XRF were performed on 30 mm wide and 3 to 30 mm thick discs, which were cut from

larger flakes. For small samples XRF analyses were performed on glass beads made of 300 mg of finely powdered mica calcinated at 1000 °C with 4700 mg dried  $\text{Li}_2\text{B}_4\text{O}_7$  (Merck). For some samples both glass beads and discs were analyzed: the results from both techniques were identical within the frame of counting statistics. For CA analyses 5 to 10 mg powdered mica and for ICP, 50 mg powdered ignited mica were used.

ICP analysis needs complete dissolution, which is possible by first fusing the powdered mica with  $\text{Li}_2\text{B}_4\text{O}_7$  and subsequent attack by  $\text{HNO}_3$  or  $\text{H}_2\text{SO}_4$ , or by acid digestion with HF, depending on the elements to be analyzed.

Since analytical reliability decreases with decreasing concentration (Fig. 2), trace elements which are present mostly well above the detection limit were preferably selected for analysis, namely, Ga, Zn, Y and Nb.

### Chemical composition of muscovite: results and discussion

Due to its crystal structure, muscovite may incorporate several trace elements, many of which are potential indicators of the fractionation degree of evolving pegmatite

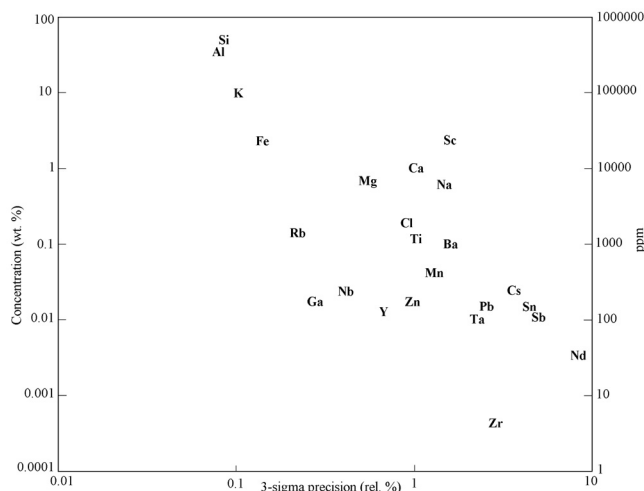


Fig. 2. Estimated analytical error for analyzed elements at 99% confidence level.

melt  $\pm$  fluid. The indicator elements reported in the literature (*e.g.* Černý, 1982a,b; Černý, 1992; Černý & Burt, 1984; Černý *et al.*, 1985) are mobile elements such as Na, Ba and Cs which occur preferably in the X-site, while others, such as Ti, V, Cr, Ga, Zn, Y, Nb, Sn, La and Ce are merely incorporated in the Y-site or, to a lesser extent, possibly in the Z-site.

Major, minor and trace constituents of muscovite from the studied pegmatites are listed in Tables 2 and 3. As it is not feasible to report the results of all analyses, only the average chemical compositions of the muscovite of each pegmatite zone are listed. When the major elements are considered, the composition does not show significant variations (Table 2). Subordinate elements show larger variation, *e.g.*, total Fe as FeO, which ranges from 0.73 up to 2.28 wt%, Na<sub>2</sub>O from 0.55 to 0.80 wt%, MgO from 0.13 to 1.07 wt% and MnO from nearly 0.03 to 0.16 wt%.

The structural formulae (Table 2) for the average chemical compositions of Table 2 were calculated on the basis of 12 anions (O, OH, F and Cl) as suggested by Rieder *et al.* (1998). Tetrahedral Si varies from 3.05 to 3.21 *apfu*. By substituting Al<sup>3+</sup> by Fe<sup>2+</sup>, Mg<sup>2+</sup> and Mn<sup>2+</sup> in the octahedral site (Fig. 3), a compensating substitution of Al<sup>3+</sup> by Si<sup>4+</sup> takes place in the tetrahedral position (Tschermak substitution). The sum (Fe+Mg) in the octahedral site displays some variation, tending to decrease from external to internal zones as shown in the Ipê pegmatite, where (Fe+Mg) varies from 0.117 in the border to 0.054 in the central zone. Al<sup>IV</sup> varies from 0.78 to 0.94, Al<sup>VI</sup> from 1.71 to 1.90, K from 0.88 to 0.96 and Li from 0.02 to 0.21 *apfu*. All other elements occur in small quantities, with the exception of <sup>VI</sup>Fe, which reaches up to 0.12 and 0.14 *apfu* in the samples of the intermediate zone from the Serra and Barreiro pegmatites, respectively. These results show that there is an excess of Si and an Al deficiency in muscovite samples from all studied pegmatites. Figure 3 shows that the octahedral substitutions are more extensive in muscovites from the Araçuaí pegmatites compared with those from Gover-

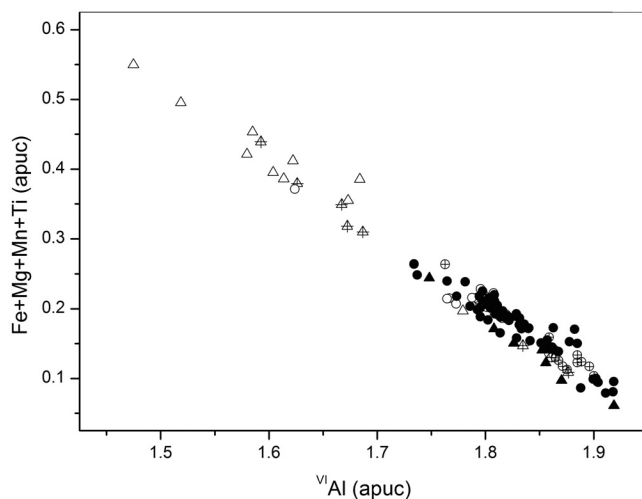


Fig. 3. Octahedral substitutions in muscovite from the Araçuaí region (open triangle: Barreiro; filled triangle: Rocinha; crossed triangle: Serra Cascalheira) and the Governador Valadares region (open circle: Cruzeiro; filled circle: Ipê; crossed circle: Aci).

nador Valadares, with the exception of the Rocinha pegmatite.

Comparative evaluation of the chemistry of muscovite from the different pegmatites and from distinct zones within each pegmatite intends to verify if the chemical composition of muscovite can be an indicator of mineralization potential concerning gem minerals, of differentiation degree and of genesis.

From the studied pegmatites, the most differentiated Cruzeiro and Rocinha pegmatites have the greatest economic potential since they bear colored gem-tourmaline. The highest Li contents in muscovite belong to Cruzeiro and Rocinha pegmatites with 0.63 and 0.80 weight%, respectively (Table 2), which also contain lepidolite. In the other pegmatites, where gem-tourmaline is missing, Li contents vary between 0.07 and 0.35 weight%. Thus, higher Li contents in muscovite may be indicative of the presence of gem-tourmaline.

Barreiro, the only pegmatite where tourmaline is absent in all zones, has muscovite with the lowest B contents ( $\leq 4$  ppm, Table 4), and highest Nb, Y and F contents, and can be considered as belonging to the NYF family (Černý 1991, 1992, Černý *et al.* 2005).

In the other pegmatites, where tourmaline can be found at least in the border zone, B varies from 103ppm in Rocinha to 746ppm in Cruzeiro. These data show that B deficiency in magmas, inhibiting crystallization of tourmaline, is also repassed into muscovite. Therefore, the amount of B in muscovite may indicate the presence or absence of tourmaline in unknown pegmatites.

In terms of internal zoning, the complex Cruzeiro pegmatite presents the lowest B contents in muscovite from the border and intermediate zones (360 and 489 ppm, respectively), which are tourmaline-bearing. Higher values are found in the wall (746 ppm) and core zones (591 ppm), where no tourmaline is found. The mentioned

Table 2. Mean chemical composition (weight wt%) and structural formulae of muscovite from selected pegmatites of the EBPP.

	Aci			Ipê			Cruzeiro									
	IZ[5]	$\sigma$	CZ[9]	$\sigma$	WZ[2]	$\sigma$	IZ[36]	$\sigma$	BZ[7]	$\sigma$	WZ[6]	$\sigma$	IZ[45]	$\sigma$	CZ[7]	$\sigma$
SiO <sub>2</sub>	46.18	0.31	46.02	0.58	46.46	0.47	45.97	0.17	46.52	0.47	46.22	0.16	46.26	0.45	45.67	0.41
Al <sub>2</sub> O <sub>3</sub>	35.27	0.69	35.17	0.95	33.75	0.91	34.49	0.09	34.35	0.84	34.13	0.24	34.54	0.87	35.76	0.50
FeOt	0.85	0.10	0.98	0.73	1.10	0.28	1.12	0.35	1.01	0.13	1.10	0.15	0.94	0.33	0.73	0.29
MnO	0.03	0.00	0.04	0.02	0.04	0.00	0.04	0.01	0.04	0.01	0.04	0.00	0.06	0.06	0.16	0.08
MgO	0.33	0.19	0.25	0.15	0.82	0.11	0.72	0.15	0.60	0.11	0.79	0.13	0.50	0.24	0.13	0.15
CaO	0.01	0.01	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.01	0.01	0.02
Na <sub>2</sub> O	0.71	0.10	0.78	0.05	0.70	0.01	0.74	0.07	0.71	0.03	0.71	0.05	0.73	0.07	0.71	0.05
K <sub>2</sub> O	10.55	0.05	10.44	0.10	10.76	0.13	10.49	0.14	10.43	0.21	10.45	0.05	10.56	0.11	10.39	0.11
Li <sub>2</sub> O	0.28 (2)	0.08	*		0.20 (1)		0.19 (14)	0.02	0.49 (4)	0.08	0.57 (4)	0.08	0.49 (27)	0.13	0.63 (2)	0.26
TiO <sub>2</sub>	0.10	0.07	0.11	0.10	0.19	0.11	0.17	0.09	0.15	0.04	0.18	0.01	0.13	0.04	0.07	0.03
Cs <sub>2</sub> O	0.01	0.01	0.01	0.02	0.00	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.01	0.02	0.02	0.02
Rb <sub>2</sub> O	0.14	0.06	0.18	0.04	0.12	0.01	0.22	0.11	0.22	0.05	0.17	0.01	0.26	0.14	0.38	0.14
P <sub>2</sub> O <sub>5</sub>	0.04	0.01	0.04	0.01	0.01	0.00	0.01	0.00	0.01	0.00	0.01	0.00	0.01	0.00	0.02	0.00
H <sub>2</sub> O	4.68	0.32	4.73	0.28	4.69	0.28	4.50	0.00	4.62	0.20	4.50	0.00	4.66	0.24	4.50	0.00
F	*		0.54	0.96	*		0.04	0.12	0.14	0.17	0.47	0.17	0.28	0.22	0.93	0.16
Cl	0.03		0.01		0.01		0.04		0.01		0.06		0.03		0.03	
Subtotal	99.21		99.30		98.86		98.75		99.30		99.40		99.46		100.14	
O=F, Cl	0.006		0.229		0.002		0.026		0.062		0.033		0.079		0.125	
Total	99.204		99.071		98.858		98.724		99.238		99.367		99.381		100.015	
K/Rb	149	72	125	35	188	37	114	70	105	43	124	10	97	20	99	2

Note: Number of samples in brackets, except for Li<sub>2</sub>O (in parentheses after content). \*: not analyzed; nd: below detection limits;  $\sigma$ : standard deviation for 95% confidence; BZ: Border Zone; WZ: Wall Zone; IZ: Intermediate Zone; CZ: Core Zone.

Table 2. Continued.

	Rocinha				Serra		Barreiro			
	IZ [4]	$\sigma$	CZ[3]	$\sigma$	IZ [8]	$\sigma$	WZ[6]	$\sigma$	IZ[5]	$\sigma$
SiO <sub>2</sub>	46.89	0.88	45.90	0.33	46.49	0.26	46.63	0.14	45.37	0.20
Al <sub>2</sub> O <sub>3</sub>	33.50	0.18	34.31	0.04	30.66	0.76	31.10	1.94	31.21	0.64
FeOt	1.06	0.22	1.00	0.39	2.11	0.28	1.98	1.09	2.28	0.33
MnO	0.16	0.03	0.12	0.03	0.08	0.02	0.11	0.03	0.11	0.03
MgO	0.18	0.02	0.12	0.02	0.92	0.35	0.82	0.68	1.07	0.43
CaO	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.00	0.00	0.01
Na <sub>2</sub> O	0.80	0.01	0.79	0.09	0.55	0.08	0.58	0.15	0.67	0.08
K <sub>2</sub> O	10.54	0.18	10.56	0.01	10.86	0.09	10.78	0.25	10.11	0.14
Li <sub>2</sub> O	0.59 (1)		0.80(2)	0.22	0.07 (1)		0.35 (1)		0.25(1)	
TiO <sub>2</sub>	0.19	0.12	0.10	0.02	0.70	0.23	0.35	0.30	0.51	0.09
Cs <sub>2</sub> O	0.06	0.00	0.11	0.08	0.10	0.04	0.00	0.00	0.01	0.01
Rb <sub>2</sub> O	0.37	0.02	0.41	0.12	0.23	0.06	0.32	0.11	0.40	0.03
P <sub>2</sub> O <sub>5</sub>	0.03	0.00	0.03	0.01	0.02	0.03	0.02	0.01	0.03	0.00
H <sub>2</sub> O	5.09	0.11	5.56	0.68	4.83	1.93	4.99	0.21	4.50	0.00
F	*		*		*		*		1.60	0.38
Cl	nd		nd		nd		0.01		0.02	
Subtotal	99.46		99.82		97.63		98.05		98.14	
O=F, Cl	nd		nd		nd		0.002		0.677	
Total	99.460		99.820		97.630		98.048		97.463	
K/Rb	60	13	44	10	126	52	73	29	51	4

## Structural on the basis of 12(O + F + Cl + OH)

	Ipe		Serra		Barreiro		Rocinha		Cruzeiro			Aci	
	WZ	IZ	IZ	WZ	IZ	IZ	CZ	BZ	WZ	IZ	CZ	IZ	CZ
Si	3.134	3.101	3.213	3.206	3.158	3.157	3.097	3.117	3.105	3.105	3.057	3.095	3.104
Al <sup>iv</sup>	0.866	0.899	0.787	0.794	0.842	0.843	0.903	0.883	0.895	0.895	0.943	0.905	0.896
Z	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000	4.000
Al <sup>vi</sup>	1.818	1.843	1.711	1.726	1.719	1.815	1.827	1.830	1.808	1.838	1.878	1.881	1.900
Ti	0.010	0.009	0.036	0.018	0.027	0.010	0.005	0.008	0.009	0.007	0.004	0.005	0.006
Fe	0.062	0.063	0.122	0.114	0.133	0.060	0.056	0.057	0.062	0.053	0.041	0.048	0.055
Mn	0.002	0.002	0.005	0.006	0.006	0.009	0.007	0.002	0.002	0.003	0.009	0.002	0.002
Mg	0.082	0.072	0.095	0.084	0.111	0.018	0.012	0.060	0.079	0.050	0.013	0.033	0.025
Li	0.054	0.052	0.019	0.097	0.070	0.160	0.217	0.132	0.154	0.132	0.170	0.075	*
Y	2.029	2.041	1.988	2.045	2.066	2.071	2.124	2.088	2.114	2.083	2.115	2.044	1.988
Ca	0.001	0.000	0.001	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.001	0.000
Na	0.092	0.097	0.074	0.077	0.090	0.104	0.103	0.092	0.092	0.095	0.092	0.092	0.102
K	0.926	0.903	0.957	0.945	0.898	0.905	0.909	0.891	0.895	0.904	0.887	0.902	0.898
Rb	0.005	0.010	0.010	0.014	0.018	0.016	0.018	0.009	0.007	0.011	0.016	0.006	0.008
Cs	0.000	0.000	0.003	0.000	0.000	0.002	0.003	0.000	0.000	0.000	0.001	0.000	0.000
X	1.023	1.009	1.045	1.038	1.006	1.027	1.034	0.993	0.995	1.011	0.997	1.001	1.008
OH	1.999	1.987	2.000	1.999	1.646	2.000	2.000	1.969	1.983	1.960	1.937	1.997	1.884
F	0.000	0.009		0.000	0.352	0.000	0.000	0.030	0.010	0.036	0.059	0.000	0.115
Cl	0.001	0.005		0.001	0.002	0.000	0.000	0.002	0.007	0.004	0.003	0.003	0.001
Al total	2.684	2.742	2.498	2.520	2.561	2.658	2.729	2.713	2.703	2.733	2.821	2.786	2.796
Fe+Mg	0.144	0.135	0.217	0.198	0.244	0.078	0.068	0.117	0.141	0.103	0.054	0.081	0.080
Fe/Fe+Mg	0.214	0.233	0.281	0.288	0.272	0.384	0.412	0.243	0.219	0.257	0.380	0.296	0.343

trends suggest that the amount of B in muscovite may be at least partially controlled by preferential incorporation into tourmaline. Be contents in muscovite, on the other hand, do not seem to be controlled by the crystallization of beryl, tending to be relatively uniform across the different internal zones, as shown by Cruzeiro pegmatite. In Ipe the amount of Be is even higher in the beryl-bearing intermediate zone (14 ppm) than in the beryl-free wall zone (3 ppm).

It is known that K/Rb ratios decrease during the differentiation of magmas (*e.g.* Morteani *et al.* 1995). In most

pegmatites K/Rb ratios of muscovite tend to diminish from the outer towards the inner zones (Fig. 4), thus reflecting the increasing differentiation degree of the pegmatite from border to core (Table 2).

In Fig. 5 K/Rb ratios of muscovite from each pegmatite were plotted against the trace elements Zn, Ga, Y and Nb. These elements were chosen because their contents are well above the detection limits. Rather than in clusters, the individual points for muscovite from a single pegmatite body plot along trends showing significant compositional



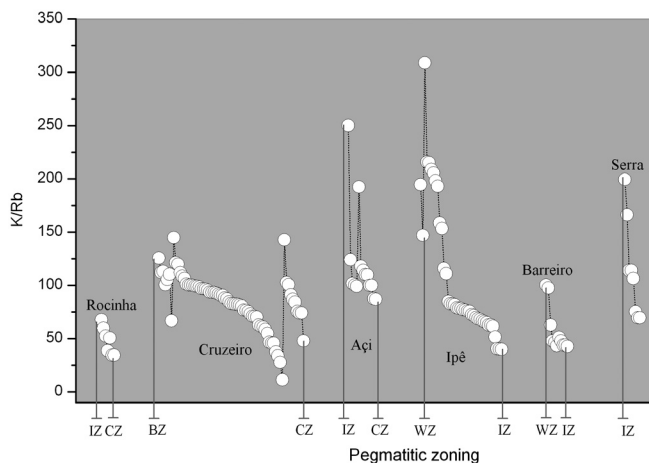


Fig. 4. Variation in K/Rb in muscovite from the outer to the inner zones of the studied pegmatites (BZ: Border zone. WZ: Wall zone; IZ: Intermediate zone; CZ: Core zone).

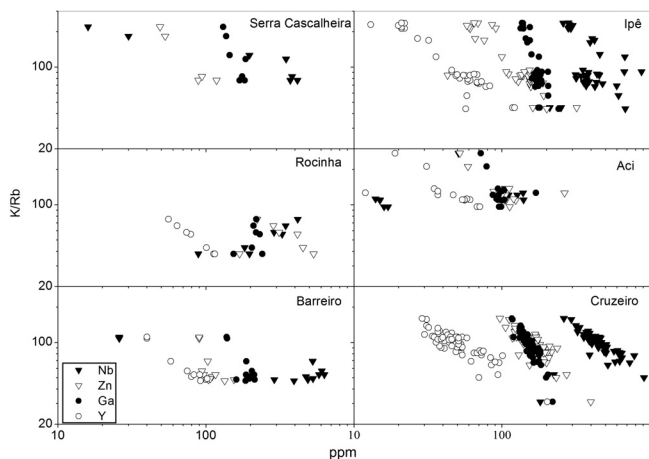


Fig. 5. K/Rb versus Ga, Y, Nb and Zn (ppm) for muscovite from the studied pegmatites.

variations that are much larger than the scatter due to analytical uncertainties. In general, the trends show negative slopes, especially visible in Cruzeiro, enabling us to conclude that Zn, Y and, less pronounced, Ga increase with decreasing K/Rb ratios (that is, with the progress of differentiation of the pegmatite magma). Niobium displays at first sight a controversial pattern (Fig. 5). In the less differentiated Serra Cascalheira and Aci pegmatites with  $K/Rb > 70$  Nb the Nb trend shows no correlation. In the more differentiated Rocinha and Barreiro pegmatites (muscovite with  $K/Rb < 70$ ), the Nb trend displays a positive slope, whereas Cruzeiro and Ipê show a negative slope. Since niobium does not only enter the muscovite structure, but also forms minerals of its own such as minerals of the columbite group as found in Ipe and Cruzeiro pegmatite, where muscovite presents high Nb contents, the niobium enrichment can be reflecting an extended fractional crystallization of original magmas.

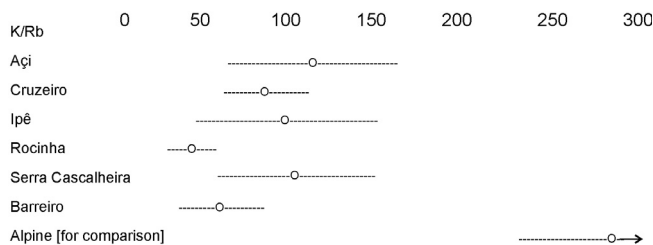


Fig. 6. K/Rb ratios ( $\pm 1\sigma$ ) for muscovite from the studied pegmatites. Muscovite from alpine anatectic pegmatites (Stern, 1966) is also shown for comparison.

Figure 6 presents the mean K/Rb values and their respective standard deviations, enabling one to visualize that muscovite from less differentiated pegmatites (Aci and Serra Cascalheira) displays higher K/Rb ratios and larger variation than muscovite from higher differentiated ones (Rocinha, Barreiro and Cruzeiro). Alpine muscovites of anatectic beryl-bearing pegmatites (Stern, 1966) have much higher K/Rb ratios when compared with the muscovites studied here (Fig. 6).

Concerning the discussion of an “anatectic” or a “magmatic” origin for pegmatites, muscovite of even the simplest pegmatites such as Aci and Serra Cascalheira has much lower K/Rb ratios (around 100, Table 2, Fig. 6) than muscovite from anatectic pegmatites as found in the Alps ( $> 250$  ppm). Thus, an origin by granite differentiation processes and not by partial melting may be postulated for the studied pegmatites.

Ti and Mg tend to diminish in muscovite from the wall zone towards the core, as shown particularly well in the Cruzeiro pegmatite, which is the only one where muscovite from all zones was analyzed. Thus, Mg and Ti contents in muscovite reflect the differentiation degree of the pegmatite. It is noticeable, however, that muscovite from the Cruzeiro border zone shows an opposite trend, with lower amounts of Ti and Mg than in the wall zone, possibly due to metasomatic interaction with the country rock. Also, Barreiro does not follow the pattern of decreasing Ti and Mg towards the pegmatite interior, since it has higher Ti and Mg in the intermediate zone than in the wall zone. This pegmatite is also quite particular concerning other chemical aspects (see below).

In the Cruzeiro pegmatite the amount of F increases from 0.14 weight% in muscovite from the border zone to 0.93 weight% from the core. Fluorine is an element with a low atomic number, tending to be concentrated in residual magmas since it is not easily accommodated in the structure of earlier minerals such as feldspars. Thus, the amount of fluorine in muscovite may indicate the relative degree of differentiation of the pegmatite. Barreiro has muscovite with the highest F content (1.60 weight%). This pegmatite is the only one possessing fluorapatite and topaz and is hosted in granite, belonging to the miarolitic-type class of pegmatites of Cerny (1982a), which are crystallized at higher levels in the crust.

In summary, geochemical data show that higher amounts of Li and B in muscovite from pegmatites may constitute

a potential indicator for the presence of gem-tourmaline. Lower K/Rb, Ti and Mg and higher F contents are characteristic of muscovite crystallized in the latest stages of pegmatite differentiation. In addition, K/Rb ratios may indicate if the pegmatite is of anatectic origin (high ratios) rather than of crystallization from an evolved granitic magma (relatively low ratios), as seems to be the case of the studied pegmatites. Zn, Ga, Y and Nb, rarely considered in the discussion of differentiation processes, tend to increase with decreasing K/Rb ratios, showing them to be useful indicators of the fractionation degree of magmas.

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