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Chemical zoning of muscovite megacrystal from the Brazilian Pegmatite Province

RÚBIA R. VIANA[1], HANNA J. EVANGELISTA[2] and WILLEM B. STERN[3]

[1]Coordenação do Programa de Pós-Graduação em Geociências, Departamento de Recursos Minerais, ICET
Universidade Federal de Mato Grosso, Av. Fernando Correa da Costa, s/n, Coxipó
Campus Universitário, 78068-900 Cuiabá, MT, Brasil
[2]Departamento de Geologia, Universidade Federal de Ouro Preto, Morro do Cruzeiro
35400-000 Ouro Preto, Minas Gerais, Brasil
[3]Mineralogisch-Petrographisches Institut, University of Basel, Basel, Switzerland

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ABSTRACT

Macroscopically homogenous muscovite plate from the Cruzeiro pegmatite, located in the Eastern Pegmatite Province in Minas Gerais, may show complex distribution patterns of some trace elements. In geochronological and petrological studies, as for example in the distinction of magmatic and post-magmatic mica, the cause of zoning could be taken into consideration. The complex chemical zoning in the studied mica plate can be best explained by growth in an evolving magma followed by alteration due to percolation of hydrothermal fluids. Enrichment of Rb towards the border is interpreted as resulting from the chemical evolution of the residual magma during crystal growth. The depletion in (^{IV}Al+^{VI}Al) as well as the increase in (Fe+Mg) and Si along a fracture could be due to the hydrothermal celadonitic substitution of muscovite. This alteration also caused depletion in the contents of Rb, Ga, Y, Nb, Sn, and Zn and residual concentration of Ti. Elements such as Ga, Y, Nb, Sn, and Zn, rarely considered in the discussion of differentiation or alteration processes in micas, have been shown to be as significant as the alkali-elements.

Key words: muscovite, chemical zoning, hydrothermal alteration, Cruzeiro pegmatite.

INTRODUCTION

Two kinds of zoning were found in micas from pegmatites from the gem-producing Eastern Brazilian Pegmatite Province (EBPP): discontinuous zoning, due to the overgrowth of muscovite on biotite as described by Viana et al. (2003), and chemical zoning of crystals of a single type of mica, investigated in the present paper.

The Cruzeiro pegmatite is located in Governador Valadares region, Minas Gerais State, Brazil (Fig. 1). This pegmatite is composed of three subvertical dikes reaching a thickness of 50 meters. The pegmatite is hosted by quartzite of the Serra da Safira Sequence (Federico et al. 1998). It shows well defined internal zonation, being composed of quartz, feldspar, muscovite, gem-tourmaline and less commonly beryl (aquamarine variety), garnet, niobotantalate, spodumene, and rarely amblygonite (Bilal et al. 2000, Cassedane et al. 1980).

Muscovite of variable size is common in all pegmatites from the EBPP, mostly as pseudo-hexagonal or fishtail shaped books. The larger crystals are found in the intermediate zone of Cruzeiro Pegmatite, in some cases exceeding 30 cm. A large muscovite plate from this zone was submitted to detailed chemical analysis in order to examine it compositional zoning and to understand the growth patterns of giant crystals. A thin fracture line crosscutting the plate from one border to the other was the only heterogeneity observed.

The chemical variation within the large muscovite plate was investigated by special analytical procedures,

Correspondence to: Profa. Dra. Rúbia Ribeiro Viana
E-mail: rubia@cpd.ufmt.br

enabling the detection of trace elements with a higher degree of accuracy than by conventional electron microprobe analysis.

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 southern Bahia States (Fig. 1). The EBPP is characterized by a particular geotectonic setting in a Neoproterozoic-Cambrian orogenic belt generated during the Brasiliano–Pan-African cycle, which consisted of a set of orogenies that lasted from about 850 to 550 Ma (e.g., Oliveira et al. 1997, Pinto and Pedrosa-Soares 2001). The majority of the pegmatites of the EBPP are related to granite intrusions into the Brasiliano mobile belts generated during the consolidation of the Gondwana supercontinent. Biotite and its muscovite overgrowth from the Ipê pegmatite, located near Governador Valadares (Fig. 1), have been dated by the K/Ar method enabling to establish a crystallization age of 575 Ma as well as a cooling rate of 3.3°C for the pegmatite (Viana et al. 2003).

SAMPLING AND EXPERIMENTAL METHODS

An 80 × 40 cm mica plate from the intermediate zone of the Cruzeiro pegmatite was selected for analysis. Mica discs were cut out on 56 points of the muscovite plate. An even, clean crystal surface of 30 mm diameter was analyzed in the Geochemical Laboratory Institute of Mineralogy and Petrography of Basel University (Switzerland) by means of X-ray Fluorescence Analysis – XFA, (SRS-3400 spectrometer of Siemens-Bruker-AXS, Germany, Specplus software) without further preparation using the analytical routine discussed by Stern (2001). No grinding process took place, and hence no contamination (W, Co) induced by sample dressing. No elements were lost in this way due to volatilization (F, Cl) as is the case with certain conventional preparation techniques such as vitrification (glass beads). The specimen thickness was measured by means of a micrometer and used for thickness correction, essential when high-energy spectral lines are used for analysis.

Since analytical reliability decreases with decreasing concentrations (Fig. 2), elements which are present mostly well above the detection limit were preferably selected for analysis in this study.

The results of the chemical analyses reported in Table I show that the main composition changes little, that is, the chemical variation of the major elements is within the analytical frame of detection. However, some trace elements vary with a factor 2 to 3, which reflects indeed the changing environment during crystal growth.

PETROLOGICAL CONSIDERATIONS TO ELEMENT DISTRIBUTION PATTERNS

The locations of the analyzed points and the distribution maps of the compositional contour lines for selected elements are presented in Figure 3. The distribution maps show very similar patterns, although the trends might be the opposite, as for example for Ti and Ga (Fig. 3). The similar distribution of the contour lines is an indication that the chemical zoning is due to the operation of some kind of geological process, i.e., it is not the result of imprecise analytical data that would certainly result in chaotic distribution patterns.

In order to evaluate the element distribution maps within the muscovite plate, the following geological processes are potential causes for the chemical variation in pegmatite minerals:

1 – The chemical evolution of residual magma during the crystallization of the pegmatite magma is responsible for an increase of Rb and other alkali-elements and a decrease of the K/Rb-ratio during fractionation (e.g. Morteani et al. 1995). Consequently, there is an enrichment of Rb and depletion of K/Rb-ratio from center to border of pegmatite minerals such as mica during growth. In melts remaining during fractionated crystallization there is an increase in Rb and decrease in Sr and Ba (Neiva et al. 1987).

2 – Zoning due to hydrothermal alteration and / or growth. In this case it is supposed that the produce enrichment in the celadonitic component that leads to increase Si and (Fe + Mg) and diminish (^{IV}Al + ^{VI}Al) when compared to magmatic muscovite (Gomes and Neiva 2000, Demster et al. 1994).

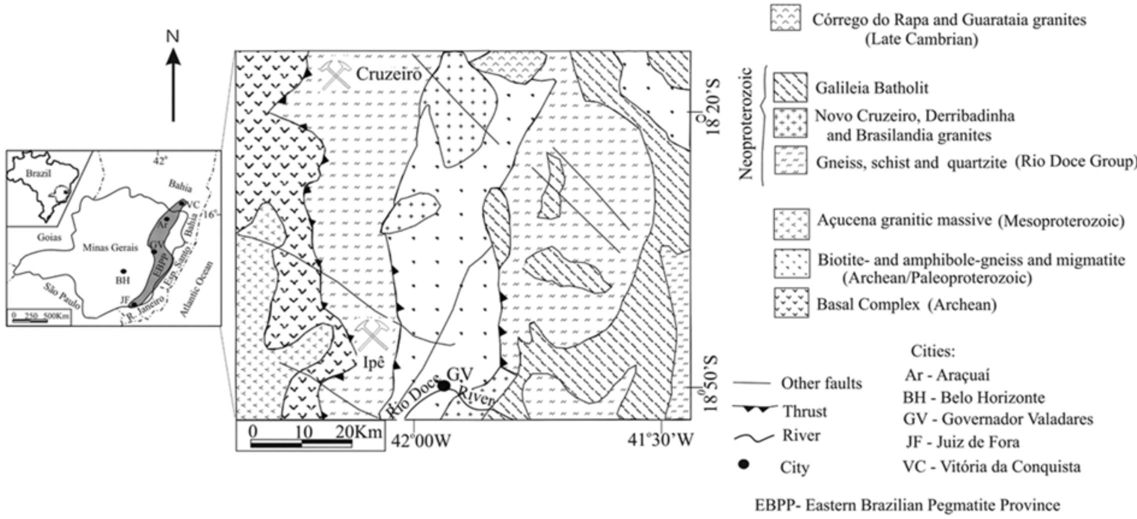

Fig. 1 – Simplified geological map of the Araçuaí and Governador Valadares regions with location of the Cruzeiro pegmatite. Outline of the Eastern Brazilian Pegmatite Province (EBPP) is also shown (modified after Oliveira et al. 1997 and Pedrosa-Soares and Wiedemann-Leonardos 2000).

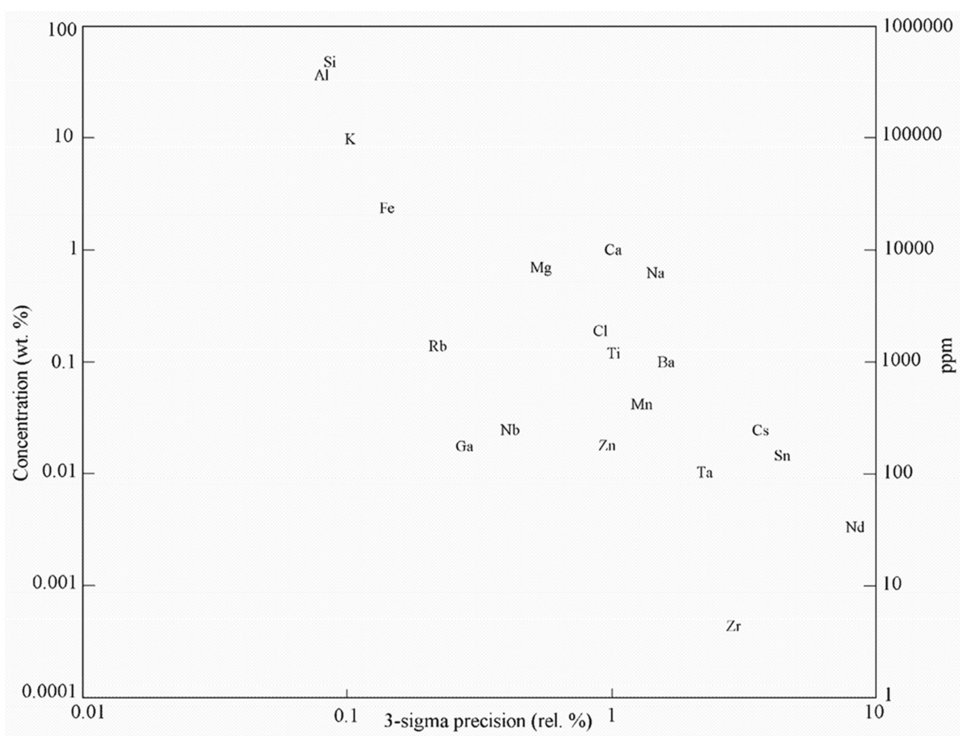

Fig. 2 – Estimated analytical error for analyzed elements at 99% confidence level.

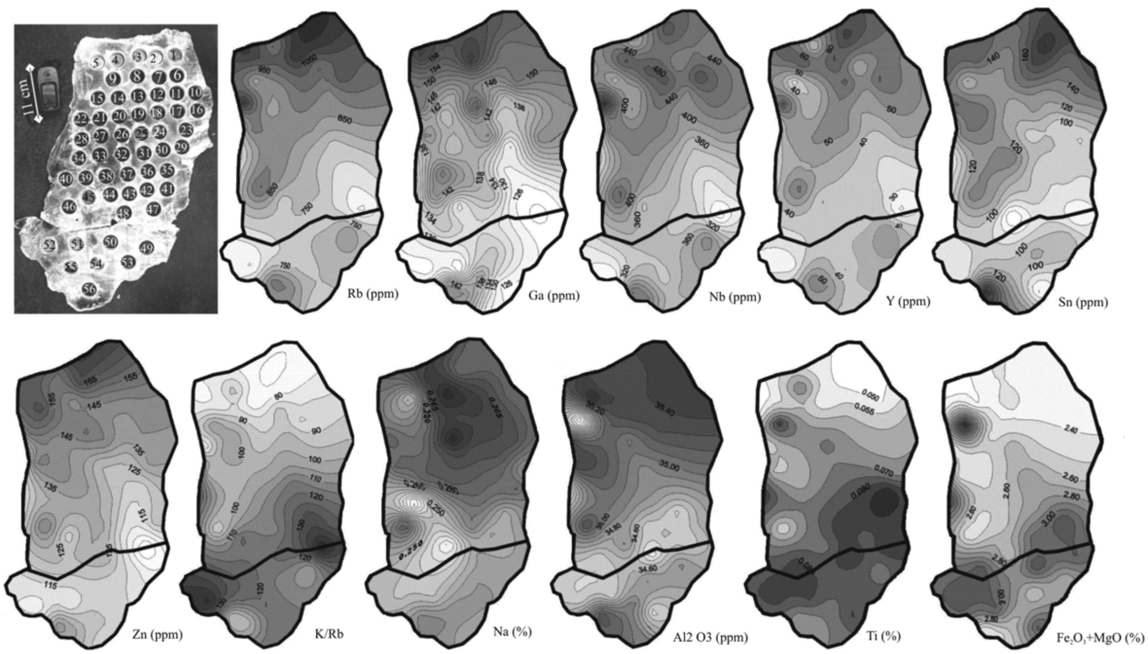

Fig. 3 – Sketch of studied muscovite plate with location of the analyzed points and distribution maps of the compositional contour lines for selected elements.

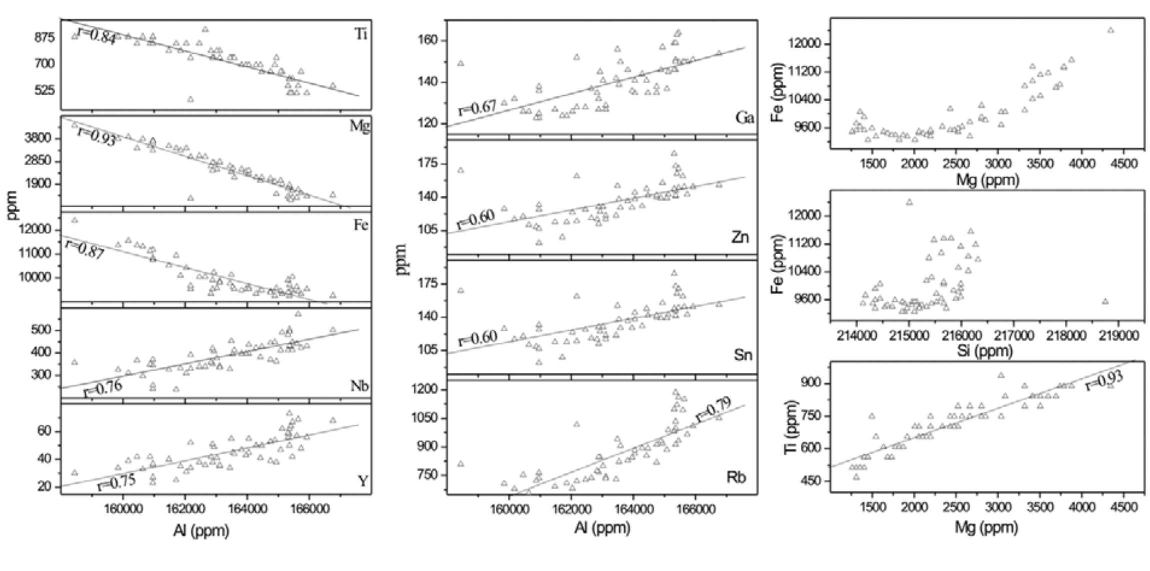

Fig. 4 – Selected bivariant diagrams showing the trend compositional of the muscovite megacrystal studied. *Solid line* is the best linear fit to the experimental data (n=56).

TABLE I
Chemical analyzed of large muscovite from Cruzeiro Pegmatite (oxides in % Wt and ions in ppm).

	1	2	3	4	5	6	7	8	9	10	11	12	13	14
SiO_2	45.85	45.81	45.87	46.00	45.85	45.85	45.87	45.84	45.89	45.80	46.01	45.97	45.98	46.04
Al_2O_3	35.36	35.37	35.42	35.49	35.45	35.28	35.39	35.39	35.37	35.38	35.67	35.37	35.32	35.31
Fe_2O_3	2.12	2.08	2.06	2.04	2.00	2.05	2.15	2.03	2.01	2.03	1.98	1.98	2.00	2.02
MnO	0.05	0.05	0.04	0.04	0.04	0.04	0.05	0.04	0.04	0.05	0.04	0.04	0.04	0.03
MgO	0.30	0.28	0.29	0.30	0.33	0.32	0.29	0.35	0.38	0.27	0.31	0.39	0.41	0.44
CaO	0.01	0.00	0.00	0.00	0.00	0.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na_2O	0.81	0.75	0.79	0.76	0.74	0.81	0.85	0.82	0.80	0.74	0.66	0.81	0.80	0.80
K_2O	10.45	10.48	10.52	10.46	10.48	10.39	10.25	10.49	10.44	10.45	10.17	10.44	10.45	10.45
TiO_2	0.12	0.11	0.11	0.11	0.14	0.16	0.11	0.13	0.13	0.11	0.12	0.13	0.14	0.14
P_2O_5	0.01	0.02	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.01	0.02	0.01	0.01	0.01
Sum	95.08	94.95	95.11	95.21	95.04	94.96	94.98	95.10	95.07	94.84	94.98	95.14	95.15	95.24
H_2O*	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5
F	0	4400	0	0	2800	3000	2800	0	2400	4000	3100	0	0	0
Cl	900	200	1600	600	500	0	0	1100	0	200	100	900	400	500
Ba	920	0	0	0	182	1182	545	607	263	1232	524	449	612	0
Cs	17	19	7	12	13	19	118	35	16	18	16	13	10	19
Nb	413	497	442	431	423	363	449	413	506	429	502	494	492	412
Rb	1098	1186	1094	1010	966	918	1161	933	1033	1120	1051	997	988	885
Sn	149	149	192	151	181	154	132	173	141	157	157	127	149	124
Zn	186	173	162	151	143	155	170	150	141	165	153	142	143	139
Ga	159	159	150	151	150	157	164	150	151	163	154	150	145	137
Pb	38	41	72	34	30	0	0	42	40	25	0	33	35	67
Sb	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Sr	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Y	56	73	57	56	48	38	67	42	62	64	68	58	62	47
K/Rb	79	73	80	86	90	94	73	93	84	77	80	87	88	98

Hydrothermal muscovite is also supposed to be poorer in Nb and Ta (Neiva 1987).

3 – Coalescence of growing crystals by synneusis (Roycroft 1991), which should generate independent zoning in each of the attached crystals surrounded by late stage zones joining the various parts (Shelley 1993). Corrosion followed by later growth of normal euhedral faces is also possible (Roycroft 1989, 1991).

RESULTS AND DISCUSSION

The following discussion about the chemical variation within the studied mica plate takes into account the above mentioned possible causes of zoning in pegmatite minerals.

Analyzing the selected bivariant diagrams (Fig. 4) it is possible to observe that there are a clear negative correlation between Ti, Mg and Fe and positive correlation between Nb, Rb, Sn, Zn, Ga, Y and Mn, both versus the Al content. In Figure 4, is also shown a positive correlation of Ti content in function of the Mg content. On the other hand, the Fe content versus both Mg and Si present two different trends, negative for lower and positive for higher Mg and Si contents. This tendency can be related to presence of Fe^{2+} and Fe^{3+} in the samples. No correlation was observed with K content, meaning that K/Rb ratio is controlled only by Rb.

Distribution patterns of indicator elements such as Rb and K/Rb in the studied megacrystal is unrelated to crystal center-border geometry (Fig. 3). Rb shows a gradual increase towards the upper border and a left to right-oriented low crosscutting the lower half of the crystal, which is more or less coincident with the fracture line. Elements showing similar trends to Rb are Ga, Y, Nb, Sn, Zn, Na, and (^{IV}Al+^{VI}Al). K/Rb ratio, on the other hand, shows an opposite trend, which is also found for Ti and (Fe+Mg) and less pronounced also for Si. Other elements, such as Ba, K, Pb, F, not shown in

TABLE I (continuation)

	15	16	17	18	19	20	21	22	23	24	25	26	27	28
SiO_2	46.00	46.79	45.99	46.13	46.06	46.12	46.02	46.08	45.90	46.06	46.10	46.08	46.17	46.13
Al_2O_3	35.24	34.69	33.89	35.17	34.97	35.09	35.07	35.04	35.43	35.24	35.09	34.85	34.89	34.79
Fe_2O_3	2.02	2.04	2.65	2.03	2.17	2.06	2.04	2.03	2.02	2.01	2.04	2.19	2.11	2.15
MnO	0.03	0.04	0.04	0.04	0.04	0.03	0.03	0.03	0.04	0.03	0.03	0.03	0.03	0.03
MgO	0.45	0.28	0.93	0.44	0.52	0.50	0.52	0.54	0.36	0.46	0.53	0.60	0.60	0.65
CaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na_2O	0.79	0.74	0.71	0.75	0.85	0.79	0.78	0.70	0.77	0.74	0.78	0.78	0.76	0.76
K_2O	10.44	10.28	10.38	10.48	10.31	10.51	10.45	10.52	10.47	10.45	10.50	10.44	10.47	10.44
TiO_2	0.14	0.10	0.19	0.15	0.16	0.15	0.15	0.15	0.12	0.14	0.15	0.16	0.17	0.20
P_2O_5	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01
Sum	95.12	94.98	94.79	95.19	95.08	95.27	95.07	95.10	95.12	95.14	95.23	95.16	95.21	95.17
H_2O*	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5
F	0	3800	6700	0	2600	0	0	2600	0	2300	0	0	0	0
Cl	500	200	1200	300	200	300	1400	300	1000	100	600	600	700	0
Ba	1060	254	289	732	201	0	929	0	116	0	0	490	350	1217
Cs	11	15	14	15	9	8	12	12	23	10	13	29	14	13
Nb	445	392	357	382	454	398	398	397	573	371	436	409	342	340
Rb	912	1016	809	853	941	864	843	828	1151	817	883	846	743	736
Sn	127	162	105	127	114	122	92	89	154	149	116	92	105	92
Zn	140	162	168	149	152	145	129	136	151	142	138	125	121	118
Ga	138	152	149	141	156	135	136	146	150	135	141	145	127	137
Pb	0	55	50	47	39	29	0	49	69	91	46	47	29	0
Sb	0	0	0	0	0	0	166	186	0	0	0	215	0	214
Sr	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Y	53	52	30	41	55	45	43	47	69	39	55	49	35	36
K/Rb	95	84	106	102	91	101	103	105	76	106	99	102	117	118

Figure 3, have erratic distribution patterns, while Ta and Sr, which are elements considered of petrological importance, present contents lower than the limit of detection.

Considering the mentioned variation patterns, growth during evolution of residual magma could not be the only cause of zoning, because chemical variation of diagnostic elements such as Rb, which should gradually increase from center to border, shows distribution patterns unrelated to crystal geometric contour. However, the much higher Rb-contents found along the upper border (Fig. 3) suggest that the influence of growth during magmatic differentiation cannot be excluded.

Coalescence of several crystals by synneusis is improbable because the various areas corresponding to the old crystals that should present similar concentric distribution patterns cannot be recognized. The observed lower contents in Rb, Ga, Y, Nb, Ta, Sn, Zn, Na, and (^{IV}Al+^{VI}Al) as well as the higher values of K/Rb, Ti, Si, and (Fe+Mg) along a more or less E-W oriented stripe in the southern half of the plate seem to be related to the fracture line (see Fig. 3).

The impoverishment in (^{IV}Al + ^{VI}Al) and the increase in (Fe+Mg) and Si near to the fracture region could be due to the celadonitic substitution of muscovite as discussed by Demster (1992) and Gomes and Neiva (2000):

$$\underset{(muscovite)}{KAl_2Si_3AlO_{10}(OH)_2} + Fe^{3+} + Si^{4+} =$$

$$\underset{(celadonite)}{KFe^{3+}(Mg, Fe^{2+})Si_4O^{10}(OH)_2} + 3Al^{3+}$$

This substitution is considered to be characteristic of hydrothermal alteration/growth (Demster et al. 1994, Gomes and Neiva 2000). In the studied case alteration could have been caused by hydrothermal fluids circulating along the fracture, implying in a deformational event of late Brasiliano age, following the crystallization of the pegmatite. Elements that were also depleted during the alteration include Rb, Ga, Y, Nb, Sn, Zn, Na.

TABLE I (continuation)

	29	30	31	32	33	34	35	36	37	38	39	40	41	42
SiO_2	46.12	46.03	45.97	46.18	46.20	46.09	46.23	46.24	45.96	45.93	46.07	46.19	46.23	46.13
Al_2O_3	34.59	35.37	35.18	34.98	34.75	34.36	34.66	34.26	34.99	34.87	34.43	34.54	34.43	34.62
Fe_2O_3	2.34	2.01	2.03	2.06	2.15	2.42	2.23	2.47	2.04	2.05	2.31	2.25	2.32	2.16
MnO	0.03	0.04	0.04	0.03	0.03	0.03	0.03	0.03	0.05	0.03	0.03	0.03	0.03	0.03
MgO	0.73	0.40	0.47	0.55	0.66	0.81	0.73	0.83	0.47	0.54	0.71	0.75	0.80	0.71
CaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na_2O	0.69	0.76	0.76	0.76	0.75	0.68	0.72	0.68	0.57	0.68	0.73	0.67	0.69	0.68
K_2O	10.56	10.46	10.48	10.49	10.50	10.57	10.51	10.38	10.68	10.62	10.40	10.57	10.56	10.47
TiO_2	0.18	0.13	0.14	0.16	0.18	0.19	0.18	0.19	0.16	0.17	0.19	0.17	0.19	0.17
P_2O_5	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Sum	95.26	95.21	95.08	95.23	95.24	95.16	95.30	95.09	94.92	94.90	94.89	95.19	95.26	94.99
H_2O*	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5
F	0	0	2500	0	0	0	0	2900	5600	4400	4100	0	0	3400
Cl	700	600	500	100	300	0	100	100	0	0	0	200	0	300
Ba	426	110	0	243	346	1599	0	481	0	927	1672	832	670	738
Cs	1	13	11	116	13	10	12	10	92	61	12	24	19	13
Nb	238	432	428	395	339	297	310	311	412	377	345	327	241	333
Rb	590	912	892	822	726	658	681	678	906	800	763	691	577	709
Sn	78	138	127	141	89	92	87	78	130	111	100	84	68	81
Zn	98	147	142	138	127	111	124	117	129	130	132	114	92	114
Ga	124	146	135	139	128	126	126	132	150	141	138	127	123	124
Pb	61	40	50	55	46	34	62	33	32	0	44	40	38	33
Sb	0	0	0	0	216	0	183	187	0	0	0	216	208	194
Sr	16	0	0	0	14	0	0	18	33	28	0	0	0	0
Y	25	50	50	45	38	33	31	39	44	41	36	40	23	35
K/Rb	149	95	98	106	120	133	128	127	98	110	113	127	152	123

Ti-contents, on the other hand, may have been increased due to (i) residual concentration because of the immobile character of Ti or (ii) reaction/alteration of other phases, e.g. rutile needles included in muscovite.

The similarity of the trends of Ga, Y, Nb, Sn, and Zn to the trend of Rb, which is considered as being an important petrological indicator, demonstrates that in case of the studied mica these elements show the same geochemical behavior as Rb. Therefore elements such as Ga, that shows significant variation, can also be valuable indicators for discrimination of different mica generations in rocks.

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RESUMO

Um grande cristal de muscovita, macroscopicamente homogêneo, procedente do Pegmatito Cruzeiro, localizado na Província Pegmatítica Oriental, em Minas Gerais, exibe padrão de distribuição complexa para alguns elementos traços. Em estudos geocronológicos e petrológicos, como, por exemplo, na separação entre micas magmáticas e pós-magmáticas, a causa de zoneamento deve ser levada em consideração. O complexo zoneamento químico no cristal de mica estudado é melhor explicado pelo crescimento em um magma evoluído, seguido pela alteração, proveniente da percolação de fluidos hidrotermais. O enriquecimento de Rb nas bordas é interpretado como resultado da evolução química do magma residual durante o crescimento do cristal. A diminuição em ($^{IV}Al+^{VI}Al$), bem como o aumento de (Fe+Mg) e Si ao longo da fratura é explicado pela substituição hidrotermal celadonítica da muscovita. A altera-

TABLE I (continuation)

	43	44	45	46	47	48	49	50	51	52	53	54	55	56
SiO_2	45.92	46.20	46.01	46.16	46.20	46.26	46.14	46.20	46.10	46.13	46.27	45.96	46.03	46.19
Al_2O_3	35.36	34.69	34.84	34.19	34.84	34.43	34.83	34.41	34.96	34.32	34.43	35.27	35.14	34.89
Fe_2O_3	2.01	2.07	2.04	2.43	2.12	2.39	2.00	2.38	2.09	2.43	2.30	1.98	2.00	2.10
MnO	0.04	0.03	0.04	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.04	0.04	0.03
MgO	0.37	0.65	0.53	0.81	0.60	0.77	0.57	0.75	0.57	0.73	0.79	0.43	0.47	0.61
CaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na_2O	0.78	0.65	0.60	0.69	0.69	0.65	0.70	0.66	0.71	0.70	0.66	0.72	0.72	0.69
K_2O	10.41	10.59	10.78	10.41	10.59	10.50	10.50	10.59	10.61	10.66	10.49	10.47	10.51	10.54
TiO_2	0.12	0.16	0.16	0.19	0.16	0.18	0.17	0.18	0.16	0.18	0.18	0.15	0.15	0.16
P_2O_5	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01
Sum	95.02	95.05	95.01	94.92	95.24	95.22	94.96	95.22	95.24	95.20	95.16	95.02	95.06	95.22
H_2O*	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5
F	3200	3600	4700	4800	0	0	4800	0	0	0	0	2900	2800	0
Cl	600	0	0	100	500	0	0	400	100	1300	0	0	0	700
Ba	0	350	0	634	0	531	535	378	585	0	1478	344	457	237
Cs	12	76	1	77	8	11	10	31	12	8	1	41	13	11
Nb	481	330	352	326	419	371	358	350	328	367	257	449	423	336
Rb	980	720	767	707	846	735	773	722	730	752	591	931	894	733
Sn	138	114	54	89	111	97	116	114	103	65	81	197	143	122
Zn	149	115	130	128	112	127	117	109	121	120	107	139	131	117
Ga	146	128	132	130	127	136	135	123	142	126	125	146	144	129
Pb	40	0	0	41	58	0	28	34	41	39	27	0	0	37
Sb	0	0	240	224	0	0	0	0	0	143	182	0	0	0
Sr	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Y	59	34	38	34	51	37	42	42	34	42	27	54	50	36
K/Rb	88	122	117	122	104	119	113	122	121	118	147	93	98	119

* estimated value.

ção hidrotermal causou, também, a diminuição nos conteúdos de Rb, Ga, Y, Nb, Sn e Zn ao longo desta fratura, além da concentração residual de Ti. Elementos tais como, Ga, Y, Nb, Sn, e Zn, pouco considerados em discussão de diferenciação ou processos de alteração, mostraram significância tanto quanto os elementos alcalinos.

Palavras-chave: muscovita, zoneamento químico, alteração hidrotermal, pegmatito Cruzeiro.

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