ALKALI-DEFICIENT ELBAITE FROM PEGMATITES OF THE SERIDÓ REGION, BORBOREMA PROVINCE, NE BRAZIL

DWIGHT RODRIGUES SOARES[1], ANA CLÁUDIA MOUSINHO FERREIRA[1,2], RILSON RODRIGUES DA SILVA[3] AND VALDEREZ PINTO FERREIRA[4]

ABSTRACT The chemical composition and structural formulae of elbaite from pegmatites of the Seridó Pegmatitic Province, northeastern Brazil, indicate important vacancies in the X structural site, with values varying from 12 up to 44%, Na being the dominant component in this site (55-85%), Ca in low amounts (<1-11%) and K in insignificant amounts (<1%). There is a linear positive correlation between vacancy in the X site and Al in the Y site, a behavior observed in tourmalines elsewhere in the world cited in the literature, and believed as due to compensation of the additional of Al in the Y-octahedral site by deficiency of cations in the X site.

Keywords: elbaite, mineral chemistry, pegmatite

INTRODUCTION The X-structural site of tourmalines can be occupied by Na, K (alkaline metals), and Ca, or can show vacancies, the total or partial occupancy usually related to the deficiency of Ca, Na and K cations. The deficiency in these alkaline cations in tourmalines are cited by several authors, usually ranging from 5% (e.g. feruvite cited by Seway *et al*. 1998) up to 74% (e.g. foitite studied by MacDonald *et al*. 1993). Vacancy of 45% in elbaite was described by Gorskaya *et al*. (1982).

In this paper, the compositional variations of elbaite from pegmatites of the Seridó pegmatitic province, NE Brazil, will be shown, and some aspects related to the alkali deficiency in this mineral will be discussed. The chemical and structural data of elbaite from Quintos (Parelhas, State of Rio Grande do Norte) and Alto Quixaba (Frei Martinho, State of Paraíba) deposits are shown, and compared with data for Gregório (Parelhas, State of Rio Grande do Norte) and São José da Batalha (Salgadinho, State of Paraíba) deposits, the two latter studied by Adusumilli *et al.* (1994) and Rossman *et al.* (1991), respectively. These deposits besides bearing excellent gem-quality elbaite, are the only ones in the region for which systematic geochemical studies have been done.

OCCURRENCE AND MINERAL ASSOCIATIONS The Seridó Pegmatitic Province is located in the Seridó foldbelt that is part of the Borborema structural province, northeastern Brazil (Fig. 1), whose major development is believed to have occurred in the Neoproterozoic during the Brasiliano orogenic cycle (e.g. Jardim de Sá 1994). The Seridó pegmatitic province is characterized by a large quantity of simple and complex, heterogeneous pegmatitic plutons, which present diverse mineralizations. Among them, aquamarine and tourmaline gems are the most famous, as well as tantalite-columbite, which has been hand-mined for over 30 years.

Tourmaline-bearing pegmatites, including some stones of exceptional gem quality, occur mainly in the Salgadinho, Junco do Seridó and Pedra Lavrada areas, State of Paraíba (e.g. Brito and Silva 1992), and in the Equador and Parelhas areas, State of Rio Grande do Norte (e.g. Diniz and Nesi 1990), although they also occur in other regions of this province. Blue elbaite tourmalines from the Seridó region have already been described in the literature, and among them can be cited Ferreira *et al.* (1990), Fritsh *et al.* (1990), Bank and Henn (1990), Henn *et al.* (1990), Rossman *et al.* (1991), Brito and Silva (1992), Adusumilli *et al.* (1993 1994), MacDonald and Hawthorne (1995), and Karfunkel and Wegner (1996).

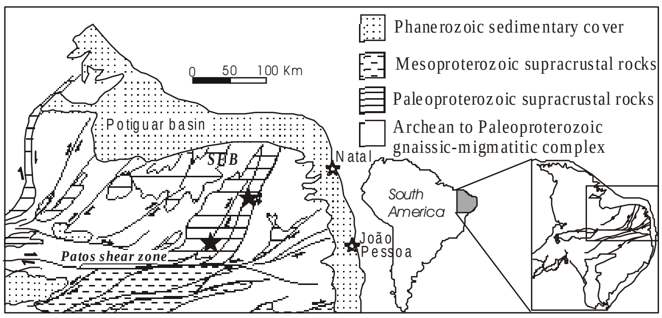

Figure 1 – Location of the studied elbaite-bearing pegmatites (stars) in the Seridó foldbelt (SFB), Borborema Province, Northeastern Brazil.

In this province, some pegmatites are intrusive in quartzite and metaconglomerate of Equador Formation, forming a trend that is roughly coincident with NNE-trending shear zones, which characterize the Seridó fold belt. Some pegmatites, however, intrude biotite schists of the Seridó Formation. Most pegmatites in the region is zoned, some of them presenting internal tubular bodies, which show typical substitution enrichment. In some places substitution is so advanced that the pegmatite zoning is masked, like in the Quintos pegmatite (Soares 1998).

Elbaite occurs mainly in these replacement bodies as accessory mineral, associated with quartz, cleavelandite and lepidolite. Some typical mineral associations of elbaite-bearing pegmatites from the Seridó province are listed in the Table 1.

Table 1 - Mineral associations of elbaite-bearing permatites from the Seridó pegmatitie province, northeastern Brazil.

Pegmatite	Mineral association	Reference
Quintos (Parelhas, State of Rio Grande do Norte)	Cleavelandite, quartz, lepidolite, kaolin	Soares 1998
Alto Quixaba (Frei Martinho, State of Paraíba)	Cleavelandite, pink mica, quartz, manganotantalite	Ferreira 1998
Gregório (Parelhas, State of Rio Grande do Norte)	Cleavelandite, lepidolite, quartz	Adusumilli *et al.* 1994
S. José da Batalha (Salgadinho, State of Paraíba)	Quartz, lepidolite, feldspar	Rossman *et al*. 1991

ANALYTICAL PROCEDURES A total of 70 electron microprobe chemical analyses of elbaite from the Alto Quixaba and Quintos pegmatites were performed. Tourmalines from the Alto Quixaba pegmatite were analyzed at the University Pierre et Marie Curie (Paris VI), France, using an electron microprobe CAMECA model SX-50, in the following conditions: 15kV, 10nA, 10mm diameter electronic beam, and 20 s peak counting time. Boron was measured using 10kV and peak counting time of 5 seconds. Li and minor elements were analyzed using ICP-AES at the School of Mines Saint-Etienne, France. Samples from the Quintos pegmatite were analyzed at the University of Brasília, Brazil, using an electron microprobe CAMECA model CAMEVAX SX50 in the following conditions:15 kV e 20 nA.

Table 2 shows significant chemical analyses of elbaite, together with their structural formulae. Cations per formula unit were calculated based on normalization to six silicons. The analytical results of the table are average values, as most studied tourmalines are zoned. Table 3 shows chemical analyses of elbaite from two other pegmatites from the Seridó province, studied by Adusumilli *et al*. (1994) and Rossman *et al.* (1991). The structural formulae listed in the Table were calculated using the chemical analyses provided by Adusumilli *et al.* (op cit.) and Rossman *et al.* (1991) on the basis of 31(O,OH).

1 - Pós-Graduação em Geociências, Centro de Tecnologia e Geociências, UFPE, Cidade Universitária, 50740-530, Recife, PE, Brasil;
2 - Centro Gemológico do Nordeste, Depto. de Mineração e Geologia, UFPB - CEP: 58109-170 Campina Grande, PB, Brasil
3 - Dept. de Engenharia de Minas, UFPE, Cidade Universitária, 50740-530, Recife, PE, Brasil *(in memoriam)*
4 - NEG-LABISE, Depto. de Geologia, UFPE, Cidade Universitária, 50732-970, Recife, PE, Brasil

Table 2 Representative chemical compositions of elbaites from the Gregório pegmatite (Adusumilli et al. samples 1 and 2) and Alto Quixaba pegmatite(Ferreira 1998; samples 3 through 7). Structural formulae for samples 1 and 2 were calculated on the basis of 31 (O, OH); for samples 3 through 7, according to the procedures described by Henry & Guidotti (1985), on the basis of 29 oxygens.

Color Oxides	Green (01)	Blue (02)	Blue (03)	Violet Blue (04)	Green (05)	*Bottle* Green (06)	Greenish Blue (07)
SiO_2	38.08	37.80	37.92	37.32	37.52	37.07	37.63
Al_2O_3	41.47	39.78	38.08	38.34	38.06	38.70	38.30
Na_2O	1.981	2.25	2.64	2.58	2.72	2.64	2.78
CaO	0.044	0.308	0.29	0.24	0.17	0.13	0.16
K_2O	0.017	0.02	0.01	0.01	0.014	0.03	0.00
MgO	0.004	0.495	0.01	0.02	0.004	0.13	0.002
MnO	0.67	2.05	2.87	2.45	2.09	1.73	1.69
FeO	2.225	1.328	2.97	3.87	2.58	5.04	2.82
TiO2	0.007	0.073	0.05	0.05	0.01	0.12	0.08
CuO	0.01	0.488	0.02	0.00	0.01	0.01	0.00
Cr_2O_3	0.014	0.006	---	---	---	---	---
ZnO	0.06	0.796	0.45	0.42	2.87	0.23	2.98
PbO	0.14	0.22	---	---	---	---	---
Bi_2O_3	0.02	0.118	---	---	---	---	---
Li_2O	1.50	1.56	1.737	1.810	1.586	1.665	1.665
V_2O_3	0.003	0.012	---	---	---	---	---
B_2O_3	9.80	9.00	10.97	10.80	10.86	10.73	10.75
H_2O	4.19	4.17	---	---	---	---	---
Cl	0.01	0.007	---	---	---	---	---
Total	100.23	100.47	98.02	97.91	97.49	98.225	98.865
Si	5.96	5.96	5.98	5.89	5.93	5.86	5.94
Al_Y	1.63	1.36	1.06	1.01	1.02	1.06	1.06
Al_Z	6.00	6.00	6.00	6.00	6.00	6.00	6.00
Al_T	0.04	0.04	0.02	0.11	0.07	0.14	0.06
Na	0.60	0.68	0.81	0.79	0.833	0.807	0.85
Ca	0.07	0.05	0.05	0.04	0.03	0.022	0.027
K	0.002	0.04	0.002	0.002	0.003	0.003	0.00
Mg	0.0009	0.114	0.0047	0.009	0.0019	0.06	0.00
Mn	0.09	0.27	0.38	0.32	0.28	0.23	0.22
Fe	0.29	0.17	0.39	0.51	0.34	0.66	0.37
Ti	0.0001	0.008	0.0057	0.0057	0.00114	0.014	0.009
Cu	0.0009	0.06	0.0024	0.00	0.00114	0.0011	0.00
Zn	0.006	0.08	0.05	0.047	0.33	0.026	0.34
Pb	0.006	0.00	---	---	---	---	---
Bi	0.00	0.004	---	---	---	---	---
Li	0.95	0.99	1.04	1.15	1.01	1.058	1.058
V	0.00	0.00	---	---	---	---	---
B	2.70	2.60	2.99	2.95	2.964	2.93	2.93
OH	4.35	4.40	4.00	4.00	4.00	4.00	4.00
Cl	0.003	0.00	---	---	---	---	---
	0.328	0.266	0.138	0.168	0.134	0.168	0.123

Table 3 Chemical composition of elbaites from the São José da Batalha pegmatite studied by Henn et al. *(1990, sample 1) and Rossman* et al. *(1991; samples 2 through 6), and from Quintos pegmatite studied by Soares (1998, samples 7 and 8). Structural formulae calculate in this work on the basis 31 (O, OH). * from Rossman* et al. *(1991)*

Color Oxides	Green (01)	Bluish green (02)	Light yellowish green(03)	Light blue (04)	Light purplish blue (05)	Light blue (06)	Neon blue (07)	Red (08)
SiO_2	37.12	36.53	37.27	36.97	37.11	37.06	38.26	38.21
Al_2O_3	40.20	38.58	39.04	38.95	39.66	38.47	43.78	44.25
Na_2O	2.20	2.49	2.27	2.26	2.16	2.35	1.846	1.815
CaO	0.37	0.05	0.46	0.55	0.62	0.47	0.078	0.078
K_2O	0.03	0.02	0.03	0.02	0.02	0.02	0.041	0.007
MgO	0.01	0.00	0.54	0.00	0.00	0.00	0.02	0.0004
MnO	1.20	1.48	1.47	2.30	1.32	2.55	0.32	0.466
FeO	0.02	0.07	0.22	0.00	0.00	0.00	0.195	0.006
TiO_2	---	0.06	0.10	0.01	0.00	0.01	0.016	0.022
CuO	2.14	1.76	0.37	0.72	0.62	0.74	0.67	0.23
Cr_2O_3	---	0.00	0.00	0.00	0.00	0.00	0.0008	0.013
ZnO	---	0.25	0.08	0.01	0.00	0.01	---	---
PbO	---	0.01	0.00	0.01	0.00	0.02	---	---
Bi_2O_3	0.54	0.01	0.39	0.11	0.15	0.08	---	---
Li_2O	1.52	1.62	1.62	1.62	1.62	1.62	1.62*	1.62*
V_2O_3	---	0.01	0.01	0.00	0.00	0.00	---	---
B_2O_3	10.71	10.94	10.94	10.94	10.94	10.94	10.94*	10.94*
H_2O	3.06	3.13	3.13	3.13	3.13	3.13	2.232	2.24
Cl	---	0.01	0.01	0.00	0.00	0.01	---	---
F	0.91	---	---	---	---	---	---	---
Total	100.03	97.02	97.95	97.60	97.35	97.48	100.11	99.88
Si	5.9	5.752	6.05	6.026	6.055	6.054	6.06	6.04
Al_Y	1.53	1.492	1.472	1.485	1.544	1.410	2.16	2.24
Al_Z	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00
Al_T	0.1	0.248	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.68	0.822	0.714	0.715	0.682	0.744	0.566	0.55
Ca	0.06	0.009	0.08	0.096	0.108	0.082	0.01	0.013
K	0.01	0.004	0.006	0.004	0.004	0.004	0.007	0.001
Mg	0.002	0.00	0.131	0.00	0.00	0.00	0.0005	0.00
Mn	0.16	0.214	0.165	0.317	0.182	0.352	0.04	0.06
Fe	0.003	0.01	0.03	0.00	0.00	0.00	0.026	0.0007
Ti	---	0.007	0.012	0.001	0.00	0.001	0.002	0.003
Cu	0.26	0.023	0.045	0.088	0.076	0.091	0.076	0.03
Cr	---	0.00	0.00	0.00	0.00	0.00	0.00	0.0015
Zn	---	0.032	0.01	0.001	0.00	0.001	---	---
Pb	---	0.001	0.00	0.001	0.00	0.001	---	---
Bi	0.02	0.002	0.016	0.004	0.006	0.004	---	---
Li	0.97	1.109	1.057	1.062	1.063	1.064	1.028	1.026
V	---	0.002	0.002	0.00	0.00	0.00	---	---
B	2.94	3.229	3.065	3.078	3.081	3.085	2.99	2.98
OH	3.24	3.553	3.389	3.403	3.406	3.411	2.36	2.354
Cl	---	0.003	0.003	0.00	0.00	0.003	---	---
F	0.46	---	---	---	---	---	---	---
	0.25	0.165	0.2	0.185	0.206	0.170	0.417	0.436

RESULTS Total Fe determined by microprobe analysis is reported as FeO. There is not a good correlation between Fe and Li values (Fig. 2), suggesting the presence of both ferrous and ferric Fe. The green color of some of the studied tourmalines may indicate the presence of Fe^{3+} in combination with Fe^{2+} (Jollif *et al.* 1986), although it may result also from Fe^{2+}-Ti^{4+} charge-transfer processes (Faye *et al.* 1974, in Jollif *et al.* 1986).

All studied crystals are elbaite, data plotting next to the schorlite-elbaite line (Fig. 3), as nearly complete solid solution exists between these two end-members (Foit and Rosenberg 1977); samples from the Quintos pegmatite are almost pure elbaite. Individual crystals are normally zoned, those from the Quintos pegmatite presenting higher concentrations of Mg, Mn and Fe, and lower Na and K in the core relative to the rims, indicative of crystal fractionation as the main magmatic differentiation process.

The elbaite crystals have rather high Fe contents (1.3-5wt%) and very low Mg (0-0.5wt%) resulting in high Fe/(Fe+Mg) ratios, which vary from 0.58 to 1. Samples from the Gregório and São José da Batalha pegmatites have low Fe and Mg contents. Fe and Mn tend to decrease, and Zn to increase, with increasing crystallization, as observed when using (Fe+Mg)/(Fe+Mg+Mn+Zn) as differentiation index (Fig. 4), a behavior typical of crystal fractionation processes. Very low Mg in most samples do not allow to visualize good correlation trends, except for samples from the Alto Quixaba pegmatite, for which negative correlation is observed, with curved trends, typical of fractional crystallization processes.

Vacancy and Alkali Deficiency Most studied elbaite crystals are Ca-poor (0.08-0.33%) and Al-rich (37-39%). In most cases Al in the Y structural site corresponds to over 50% total of the site. There are vacancies in the X structural site of all analyzed samples, sometimes with values as high as 0.44 (Quintos pegmatite). Na is the dominant component in this site, occupying from about 55% up to 85% of the total, contrasting with Ca and K, which occupy from ≈1to11% and < 1%, respectively.

Vacancy (□) in the X site and percentage of Al in the Y structural site of all samples have a good positive linear correlation (Fig. 5A), indicating that increasing of Al in the Y site corresponds to an increasing in the vacancy in the X site. This behavior was also observed by Gorskaya *et al.* (1982) in elbaite from USSR, who believe that additional amounts of Al in the Y-octahedral site are compensated by deficiency of cations in the X site. They also found that some elbaite crystals show vacancies of 0.45 and that percentage of Al in the Y structural site represents up to 74% of the total, i.e., there is a relationship between alkaline cation deficiency and Al content in the Y site. MacDonald *et al.* (1993) suggested that vacancy can be dominant in the X site of tourmaline, showing an example of foitite in which vacancy represents up to 75% of the total.

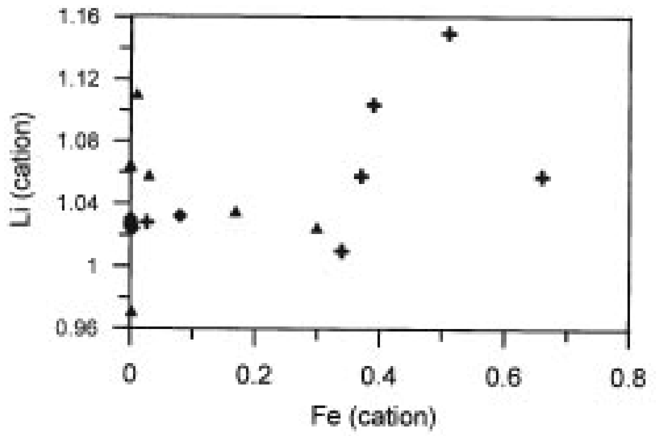

Figure 2 - Fe vs. Li (cations) plot of elbaite from the Seridó province, NE Brazil. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991).

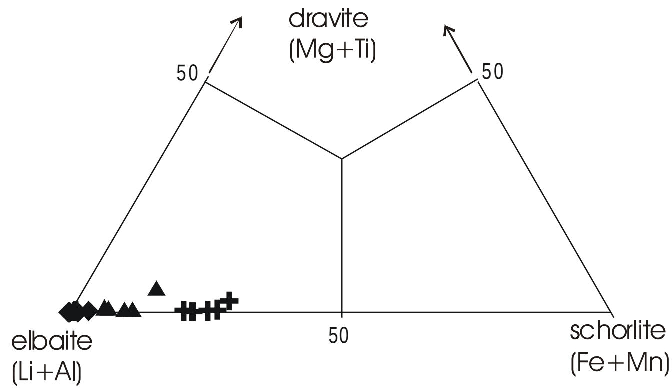

Figure 3 - Classification of the studied tourmaline in the (Mg+Ti)-(Li+Al)-(Fe+Mn) triangular plot. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991).

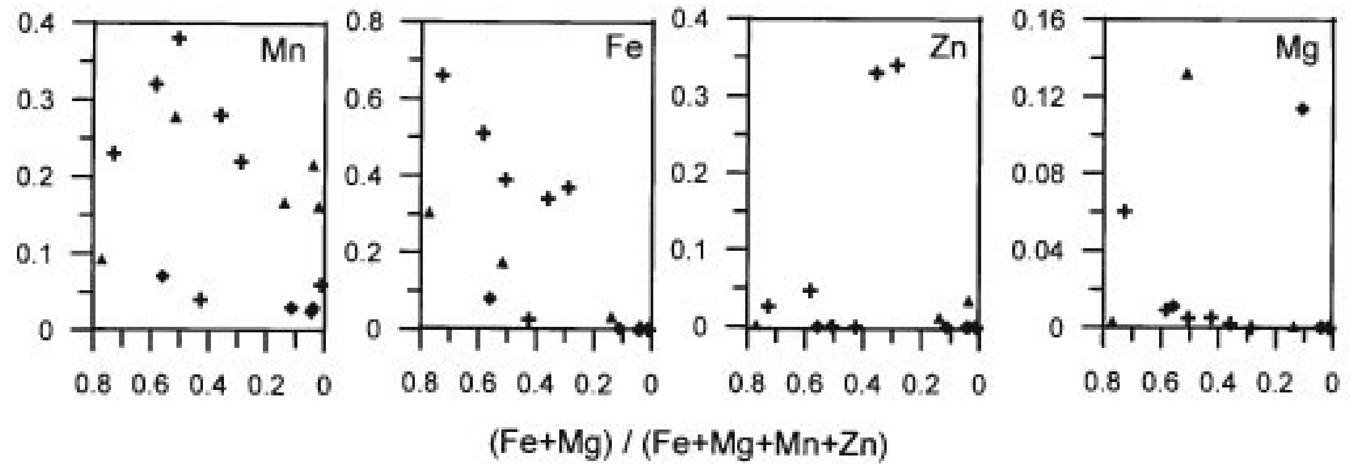

Figure 4 - Fe, Mg, Zn, and Mn vs. [(Fe+Mg/(Fe+Mg+Mn+Zn)] plot of elbaite from the Seridó province, NE Brazil. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991).

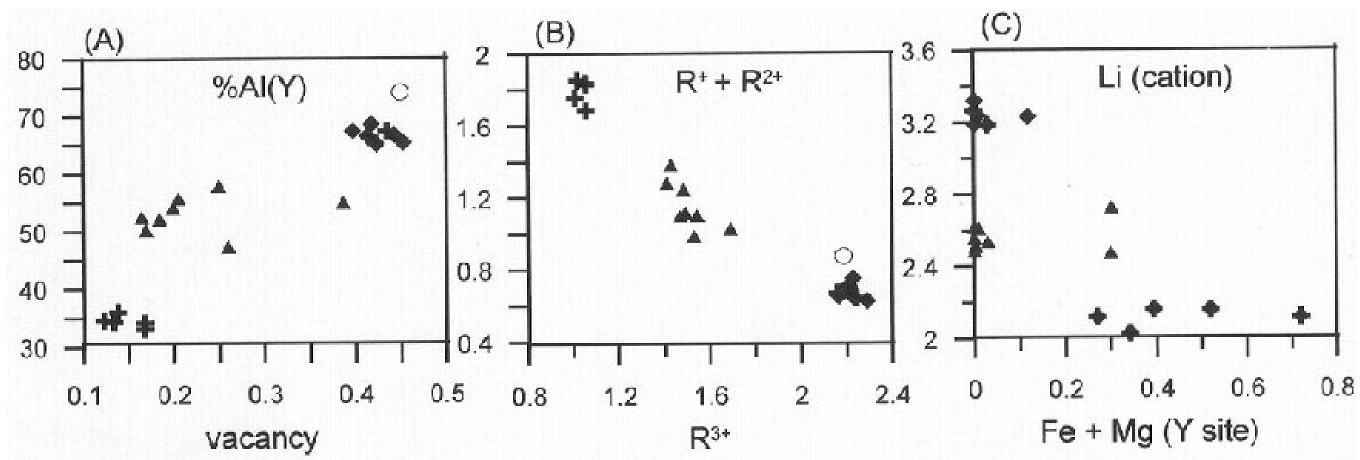

Figure – 5 – (A) - vacancy in the X-site vs. % Al in the Y-site plot for elbaite from the Seridó province, NE Brazil. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991). Highly alkali-deficient elbaite from USSR (open circle; Gorskaya et al. 1982) is plotted for comparison. (B) - $R^{3+} = (Al^{3+})_Y$ vs. $R^+ = (Na^+ + 2Ca^{2+} + K^+) + R^{2+} = (Fe^{2+} + Mg^{2+} + Mn^{2+} + Zn^{2+})$ diagram for elbaite from the Seridó province, NE Brazil. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991). Highly alkali-deficient elbaite from USSR (open circle; Gorskaya et al. 1982) is plotted for comparison. (C) - (Li+Al) vs. (Fe+Mg) in the Y site (cations) plot for elbaite from the Seridó province, NE Brazil. crosses = Alto Quixaba pegmatite; diamond = Quinto pegmatite; triangle = pegmatites studied by Adusumilli et al. (1994) and Rossman et al. (1991).

The alkali deficiency according to Foit and Rosenberg (1977) can be estimated by the relationship $(R^+ + R^{2+})/R^{3+}$, where $R^+ = (Na^+ + 2Ca^{2+} + K^+)_X$, $R^{2+} = (Fe^{2+} + Mg^{2+} + Mn^{2+} + Zn^{2+})_Y$ and R^{3+} $(Al^{3+})_Y$; the most common substitutions are either the alkali defect or desidroxylization types (OH)- + $R^{2+} = R^{3+} + O^{2-}$. The studied elbaite crystals show negative correlation (R^{3+}) x $(R^+ + R^{2+})$ (Fig. 5B), i.e. increasing in the Al content in the Y site is followed by decreasing in the alkali contents in the X site and R^{2+}, suggesting moderate substitutions. On the other hand (Li + Al) correlates negatively with (Fe + Mg) in the Y site (Fig. 5C), suggesting a major substitution in this site.

CONCLUDING REMARKS The chemistry and structural formulae of gem-quality elbaite from the Seridó pegmatitic province indicate:

presence of both ferric-and ferrous-Fe, with rather high concentrations (up to 5wt%, contrasting with very low Mg (up to 0.5wt%), although low Fe and Mg contents can also be found.

crystal fractionation as the main differentiation process is suggested by crystal zoning and compositional variation trends, using (Fe + Mg) as differentiation index;

the X structural site is incomplete in all samples studied, with vacancy values varying from 12 to 44%;

Na is the dominant (55-85%) cation in the X site, in which Ca varies from <1 up to 11% and K is usually < 1%.

There is a positive linear correlation between vacancy and % Al in the Y site, a tendency observed elsewhere in the world by different authors.

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