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Co genetic Cr-bearing tourmaline and emerald in the Novo Cruzeiro pegmatite field (Minas Gerais, Brazil)

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ABSTRACT
The Novo Cruzeiro pegmatite field in Minas Gerais (Brazil), located between Téofilo Otoni and the Jequitinhonha River, belongs to the areas of the EBPP with tourmaline and emerald occurrences. The pegmatites show beside of coloured tourmalines, Cr-rich black tourmaline cogenetic with Cr-bearing emerald. General crystallization conditions of pegmatites show, based on the obtained data, important changes during the final phase of tourmaline crystallisation which caused variations in tourmaline chemistry and reflects differences in the chemistry of the participating fluids. The crystallization of tourmaline ceased and after a period of co-crystallization, emerald was the only formed mineral. The obtained information suggests a crystallization sequence from tourmaline over tourmaline+emerald to emerald. A part of B and Fe may come from the host rock, Be and B from the intrusive granites and Cr from regional mafic to ultramafic rocks like amphibolite or Bt-Qz-schist.
Keywords: emeralds, Cr-rich tourmaline, co-genetic formation, pegmatite sources

RESUMO
O campo de pegmatítica Novo Cruzeiro em Minas Gerais (MG) entre Teófilo Otoni e o Rio Jequitinhonha pertence à PPOB com importantes ocorrências de turmalina e esmeralda. Os pegmatitos contêm ao lado das turmalinas coloridas, turmalina preta e rico em Cr e cogenética com Cr-esmeralda. Condições gerais de cristalização dos pegmatitos mostram em concordância com os dados obtidos, mudanças importantes durante a fase final de cristalização de turmalinas, causando variações na sua química que refletem as variações na química dos fluidos participantes. A formação de turmalina diminuiu e após um período de co-cristalização, a esmeralda foi o único mineral que continuou se formando. As informações obtidas sugerem uma sequência de cristalização de turmalina passando por turmalina-esmeralda para esmeralda. Uma parte de B e Fe provavelmente veio da rocha encaixante, Be e parte do B também dos granitos intrusivos e Cr das rochas máficas e rochas ultramáficas regionais, como do anfibolito ou do Bt-Qu-xisto.
Palavras-chave: esmeraldas, Cr-turmalinas, formação cogenética, fontes pegmatíticas

1. INTRODUCTION

Chromdravite also known as chromium dravite is a relatively rare but well-known mineral belonging to the tourmaline super group, with the general structural formula:

$$NaMg_3(Cr^{3+}, Fe^{3+})_6Si_6O_{18}(BO_3)_3(OH)_4 \qquad (1)$$

It was first described from the Velikaya Guba uranium vanadium deposit (Karelia, Russia) where it is associated with chromian phengite, taeniolite, quartz and dolomite (RUMANTSEVA, 1983). Dravite showing high chromium content are known and mined in

Kenya (Pohl et *al.,* 1980) for their gem quality. Chromium-rich tourmaline are known also from the Swat Valley (Pakistan) (QUASIM *et al.,* 1972; ARIF *et al.,* 2010), from Ural Mountains, Russia (COSSA; ARZRUNI, 1883; RUMANTSEVA, 1983), from Krivoy Rog in Ukraine (SHANDEROVA, 1955) from South East of Spain (TORRES-RUIZ, 2003), from the copper deposit of Outokompu, Finland (ESKOLA, 1933, PELTOLA *et al*. 1968) and from Line Pit, Lowes Mine, Cecil County, Maryland, USA (FOORD *et al*. 1981). Bosi et al. (2004) published a detailed geochemical study of chromdravite from Baikal Lake region (Russia).

All these chromian tourmalines are in fact dravite terms with very low Fe^{2+} content and relatively high Cr (0.13% and Cr_2O_3 up to 9.63% in the sample from Outokompu) (PELTOLA *et al*., 1968).

The Novo Cruzeiro-Teofilo Otoni area can be subdivided in two major geological and tectonically domains, the eastern domain and the western domain (Campos *et al.,* 2004; Alkmin *et al*., 2007; PEDROSA-SOARES *et al*., 2011). The Novo Cruzeiro pegmatite swarm belongs to the western domain. The host rocks are amphibolite facies metasediments of the Macaúbas and Rio Doce groups with intrusive Brasiliano granitoids (DNPM, 1979; CPRM, 2001; VIEIRA, 2007).

a. The Macaúbas Group is locally formed by schists and gneisses of Salinas Formation and overlayed by quartzites and schists of the Capelinha Formation.

b. The Rio Doce Group is constituted by schists, gneisses and quartzites of the Concôrdia do Mucuri Formation.

The granitoids intruded during the Brasiliano event (870 – 525 Ma; ALMEIDA, 1967; ALMEIDA *et al*., 1981). The Novo Cruzeiro Granite is considered as syntectonic, whereas other formed later, like Soturno granite, Viana granodiorite, Caraí, Itaipê and Faísca leucogranites.

The formation of these sin- to late-tectonic intrusions of S-type affinity (G4/G5; PEDROSA-SOARES *et al*., I4/I5; BAYER *et al.,* 1986, 1987) are correlated with the mobilization of the para gneisses during the late period of the Brasiliano Event. Farther east the post-tectonic intrusive suites of Early Palaeozoic age, like Aimores Intrusive Suite, Caladão granite and Padre Paraíso charnockite, together with posterior dykes and sills, represent the latest intrusive event and are of post-tectonic age.

The first domain, Macaúbas Group s.l., shows at the level of magmatic intrusions, fractures and faults oriented toward NNE/SSW and NNW/SSE, drainage orientation, and were possibly formed by compression at the beginning of the Brasiliano intrusion, and by flux orientations during the magma intrusion and solidification. Their influence over the relief and the drainage system is noteworthy. The region is part of the Pegmatite District São José da Safira, Campo Ponte-Ladainha, together with the Campo Cruzeiro and Santa Rosa fields. These pegmatites (around 560 Ma; VIANNA *et al.,* 2003; MELLO; BILAL, 2012) are mainly > 2m thick and crosscut all rocks of the Macaúbas Group. The location of the Novo Cruzeiro Pegmatite Field can be seen in figure 1.

The interaction of the fluids issued from the final fractionation of the late- to post-tectonic intrusions with fluids mobilized from Macaúbas Group, may be responsible for the presence of beryl (*s.l.*), tourmaline-group minerals, Li-minerals like lepidolite and spodumene, columbite, tantalite, topaz and quartz in these pegmatites. When the pegmatitic bodies or veins crosscut the phlogopite schists units or are proximal to these types of rocks, they can content emeralds and Cr-rich tourmalines, that may occur both in pegmatites and in the host rocks (Figure 2).

2. EMERALD AND TOURMALINE IN THE NOVO CRUZEIRO PEGMATITES

The pegmatites of this region are mined for emerald by local miners with limited extend and investment (*garimpeiros*). In these pegmatites, emerald is often associated with black tourmaline (Figure 1).

3. MATERIALS AND METHODS

Samples were obtained during two field campaigns in the Capoeirana region and Novo Cruzeiro and Santa Rosa pegmatite fields. The selected samples of tourmalines and emeralds were cut for obtaining thin sections and a representative part was grinded down for XRD investigations and bulk mineral chemistry.

Analyses of the tourmaline-group minerals and emeralds were performed using EMPA technique at the CAMPARIS facility (Sorbonne Université Sorbonne, Paris) on a CAMECA SX 100 operated at 15 KV, 12 nA, and a counting time of 30 s.

^{57}Fe Mössbauer analyses of tourmalines were carried out at the IMMM Laboratory (Le Mans University) at room temperature (300K) and at a velocity of 4mm/s.

The SEM images were obtained using a ZEISS SUPRA 55 VP SEM with GEMINI field emission column, which allowing a spatial image resolution down to 1.0 nm ISTEP (SEM facility of Sorbonne Université -Paris VI).

X-ray elemental maps (Hyperspectral X-ray images) were produced using the BRUKER QUAD Silicon Drift Detector allowing high counting rates with following analytical conditions: voltage energy:15 kV, beam current: 40 nA.

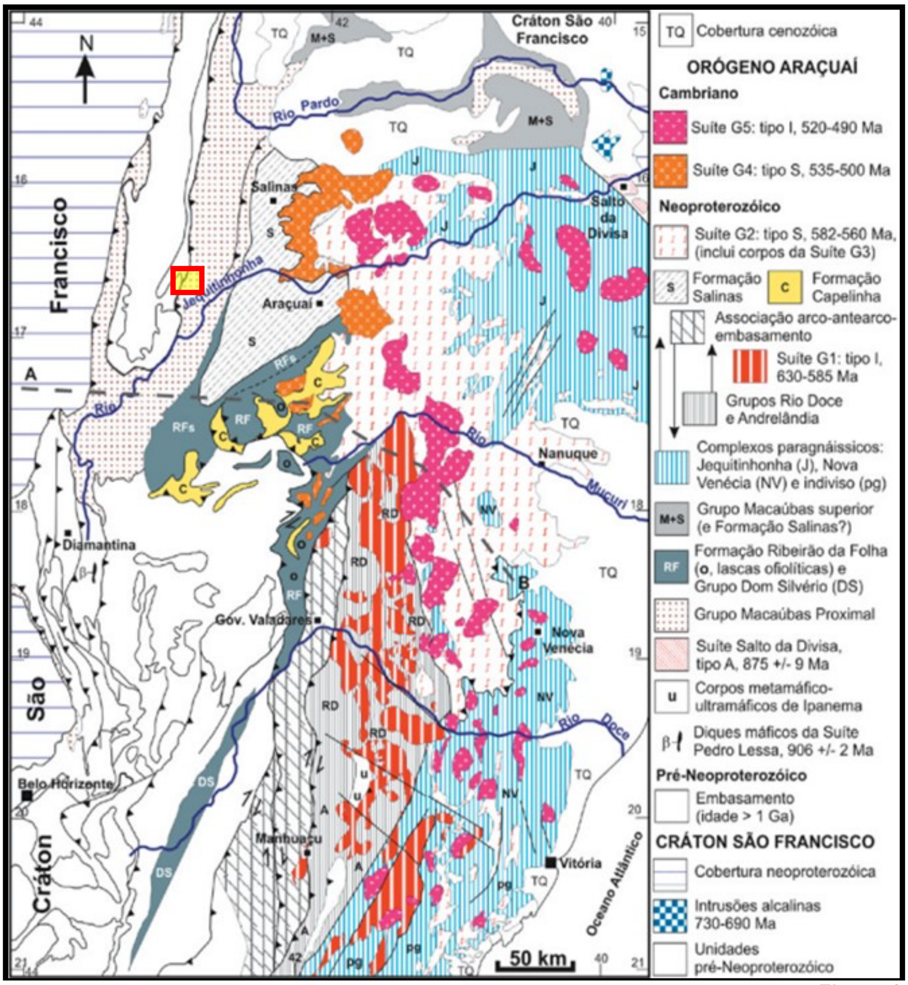

Figure 1
Geological Map of South-Eastern Brazil covering the EBPP. The location of the investigated Pegmatite Field is indicated (PEDROSA-SOARES *et al*., 2007; modified).

4. RESULTS

4.1. ANALYSES OF EMERALDS

The Cr_2O_3 content in emerald samples from Novo Cruzeiro is low as compared to the content in the emeralds from Capoeirana (table 1) with means values varying from 470 to 880 ppm, far below the Capoeirana samples (from 950 to 4190 ppm). For the Capoeirana samples it is to note that the Cr_2O_3 content in emerald is very variable even in the same crystal.

Emerald and tourmaline are imbricated together (Figure 1) so that it can be difficult to know first if they are cogenetic or not; in polished sections tourmaline appears, however, to have formed first.

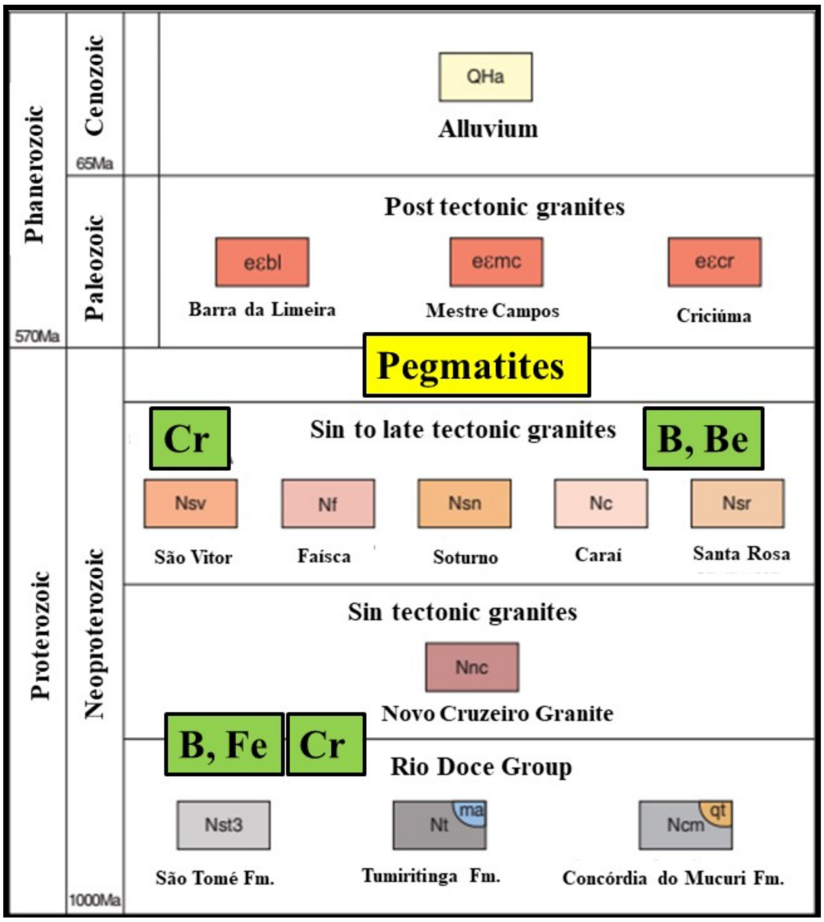

Figure 2
Stratigraphy of the investigated Field with indication of pegmatite formation and possible sources (PAES, 1997; modified).

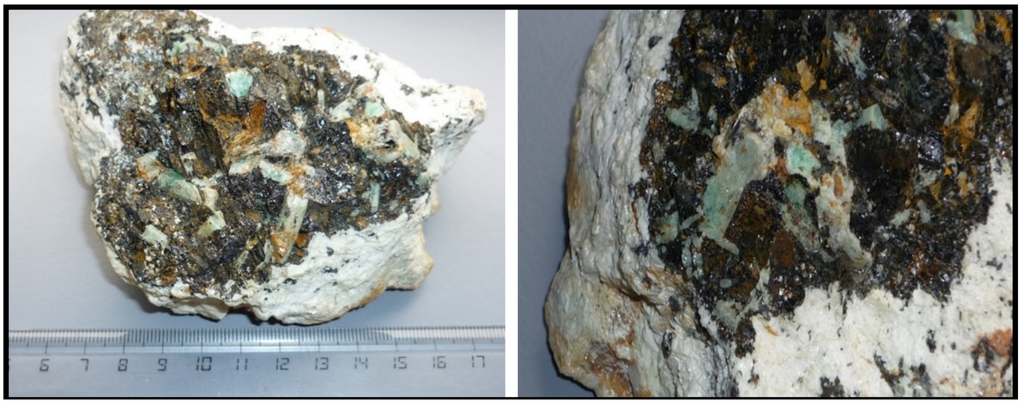

Figure 3
Left: Sample from the Novo Cruzeiro emerald *garimpo* showing the association of black tourmaline and green emerald in quartz (scale in cm). **Right:** Part of the sample showing associated cogenetic tourmaline and emerald.

4.2. ANALYSES OF TOURMALINE

Representative analyses of tourmaline-group minerals are given in Table 2. The spectra of figure 5 is different from those shown in Castaneda et al. (2003) for schorlites and blue tourmalines, but similar to the spectra of the green specimen, and to those interpreted by Pieczka, Kraczka and Zabinsky (1998) for Fe-poor tourmalines.

Scorzelli, Baggio-Saitovitch and Danon (1976) investigated Fe-rich and Fe-poor tourmalines from Minas Gerais, observing differences in the Fe-transfer visible in the Mössbauer spectra.

Figure 5 indicate less transfer than Fe-rich schorlite and similar behavior as for the green to blue-green local specimen, with different Fe-Fe site interaction than the blue Li-rich tourmalines (PIECZKA, KRACZKA AND ZABINSKY, 1998).

The data in the Table show the changes in tourmaline chemistry during its crystallization (see figure 5 and 6). The decrease in Fe and the increase in Cr from centre to rim are obvious. The Cr content of the tourmaline diminishes as the co-crystallisation of emerald begins. This can be observed comparing the crossline analyses of the core (NCTRV1 and 2) and rim (TR02B).

Table 1 - Emerald analyses from the investigated region**. NC**: Samples from Novo Cruzeiro field; **CP**: Emeralds from Capoeirana Mine. **RdF**: Samples from Ribeirão de Fogo. **NA**: not analyzed.

	NC 0709	**NC 0209**	**NC 0109**	**CP**	**CP011**	**RdF010**
Number of analyses	21	25	15	17	56	40
SiO_2	65.469	65.042	65.181	63.532	65.274	65.769
Al_2O_3	10.038	16.711	16.770	15.450	15.626	16.344
TiO_2	0.012	0.006	0.011	0.011	0.017	0.009
Cr_2O_3	0.088	0.065	0.047	0.419	0.095	0.026
V_2O_5	0.015	0.019	NA	0.029	0.045	NA
FeO	0.356	0.289	0.238	1.130	0.536	0.448
MnO	0.013	0.008	0.006	0.019	0.019	0.045
MgO	1.379	1.040	0.815	3.479	1.904	1.598
NiO	NA	NA	NA	NA	0.025	0.015
ZnO	NA	NA	NA	NA	0.069	NA
Nb_2O_5	NA	0.19	NA	NA	0.063	NA
Na_2O	1,210	0.617	0.786	1.036	1.263	1.043
CaO	0.011	0.21	0.021	0.038	0.053	0.028
K_2O	NA	NA	BDL	0.021	0.059	0.064
Rb_2O	NA	NA	BDL	NA	NA	NA
Cs_2O	NA	NA	0.059	NA	NA	NA
F	0.029	0.024	NA	NA	0.071	NA
Total	84.620	83.873	83.980	85.163	85.133	85.345

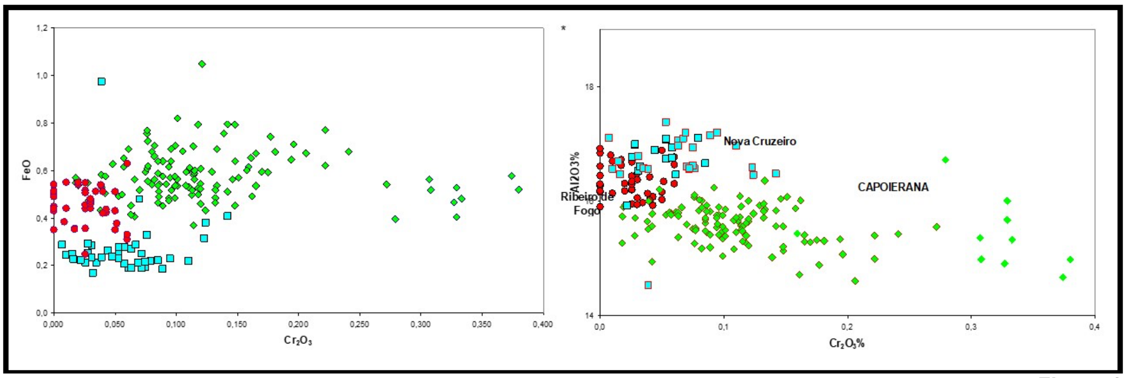

Figure 4

Each deposit has its particular chemical characteristics but variations in the Cr_2O_3 content are more important in Capoeirana quarry. **Red circles**: Ribeirão de Fogo deposit. **Blue squares**: Novo Cruzeiro deposit. **Green diamonds**: Capoeirana deposit.

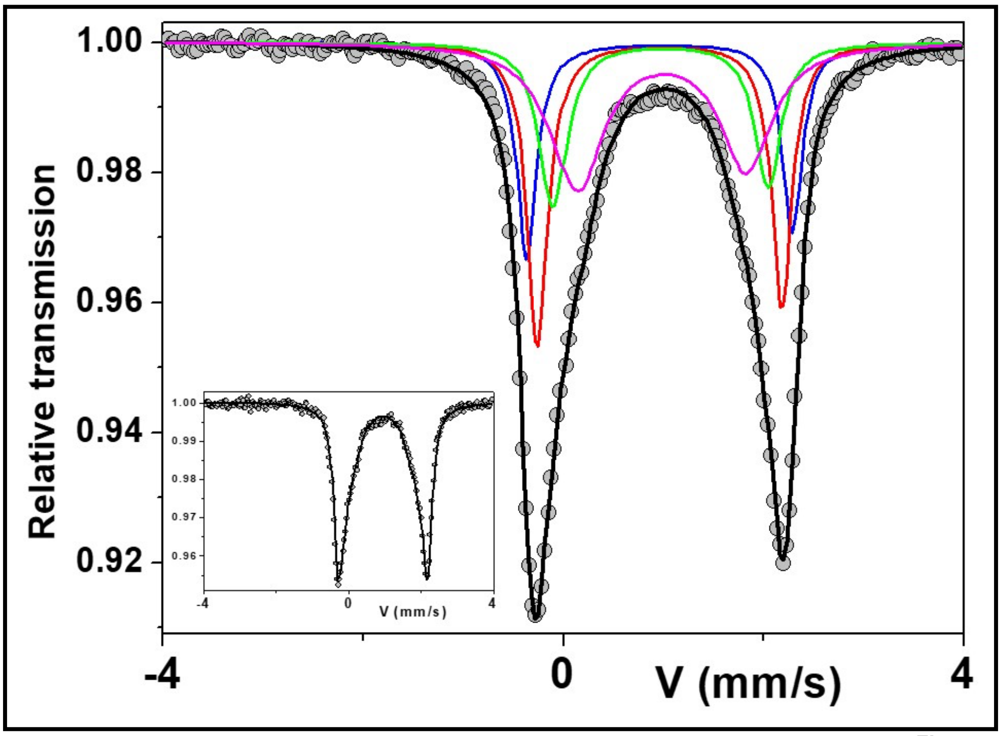

Figure 5
Fitted Mössbauer spectrum of tourmaline from the Novo Cruzeiro Pegmatite field. The small figure shows the lines of a typical tourmaline spectra.

Table 2 - Analyses of tourmalines from the Novo Cruzeiro Pegmatite field. Data represent average values from profiles on selected crystals. NC: Novo Cruzeiro Pegmatites; NA: Not analyzed.

	NC TRV1 Cross-line	NC TRV2 Cross-line	NC-01 Rim	NC-02 Rim	TR02B Rim
Distance between 2	30µ	30µ	unknown	unknown	30µ
Number of analyses	63	29	18	15	10
SiO_2	36.355	36.376	36.656	36.702	36.646
Al_2O_3	32.905	32.780	32.099	32.717	32.955
TiO_2	0.880	0.835	0.279	0.269	0.373
Cr_2O_3	0.136	0.111	0.263	0.334	0.065
V_2O_5	0.020	0.005	NA	0.022	0.012
FeO	5.771	5.644	4.293	4.237	4.524
MnO	0..037	0.050	0.045	0.076	0.059
MgO	6.451	6.528	7.352	7.934	7.412
CaO	0.454	0.432	0.434	0.374	0.333
Na_2O	2.034	2.040	2.326	2.300	2.183
K_2O	0.039	0.040	0.53	NA	0.048
Cs_2O	NA	NA	0.018	NA	NA
F	0.185	0.222	NA	NA	0.362
Total	85.267	85.053	83.848	85.450	84.963

4.3. ^{57}Fe MÖSSBAUER SPECTROSCOPY

The EMPA and Mössbauer data allow the calculation of the structural formula of the core tourmaline as in (2) and of the rim as in (3). The formulas show a significant compositional change and evolution from core to rim with significant variations of Mn, Fe, Cr and F contents. Structural vacancies are indicated by a square.

$$(Fe^{2+}_{0.150}Cr_{0.019}Al_{5.831})(Al_{0.003}Si_{5.997})O_{18}(BO_3)_3(OH)_3(OH)_{0.849}O_{0.055}F_{0.096} \quad (2)$$

$$\square_{0.250}Na_{0.681}Ca_{0.072}1K_{0.007})(Mg_{1.628}Mn_{0.013}Fe^{2+}_{0.787}Ti_{0.024}V_{0.005}Cr_{0.014}Al_{0.318})(Al_{6.00})$$
$$(Al_{0.037}Si_{5.963})O_{18}(BO_3)_3(OH)_3(OH)_{0.815}O_{0.046}F_{0.139} \quad (3)$$

Table 3 - Values of the isomer shift (δ), of the quadrupole splitting (Δ) and the site attributions and percentage for the NC tourmaline.

	δ(mm/s) ± 0.02	Δ δ(mm/s) ± 0.02	Attribution	%
NC	1.09	2.62	Fe^{2+} in Y1	19
	1.09	2.41	Fe^{2+} in Y2	28
	1.10	2.13	Fe^{2+} in Y3	19
	1.10	1.68	Fe^{2+} in Z	34
NC Magic Angle	1.08	2.49	Fe^{2+} in Y1	44
	1.09	2.25	Fe^{2+} in Y2	15
	1.06	1.98	Fe^{2+} in Y3	7
	1.06	1.63	Fe^{2+} in Z	34

5. CONCLUSIONS

The obtained results permit the drawing of conclusions about the genesis of the Novo Cruzeiro deposit. In some sections it is possible to see the initial contact between the tourmaline formed during a first phase and the later crystallized rim. The rim of the first formed tourmaline shows also important compositional variations

This zonation can be seen by mapping of samples for different elements using a scanning Electron Microscope.

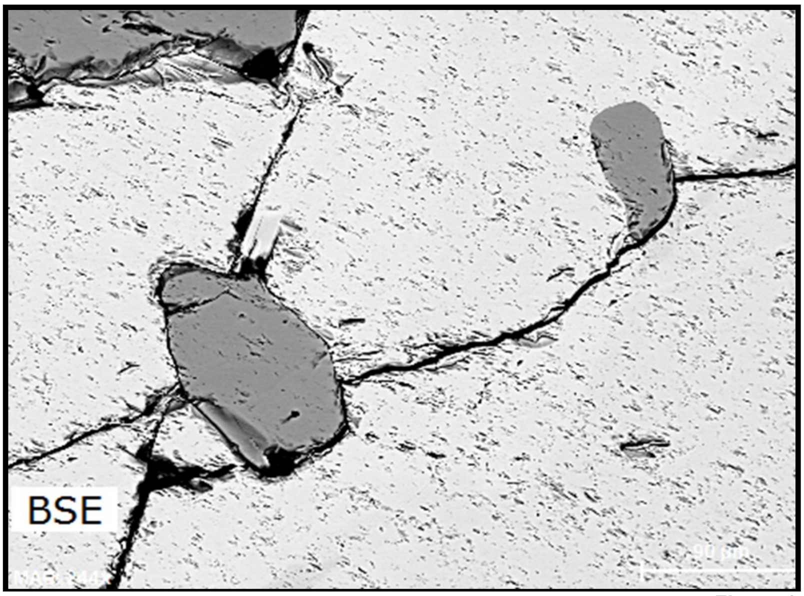

Figure 6
Back-scattered electron mapping of the rim of a tourmaline crystal with included tiny emerald crystals (darker colour).

Back-scattered electron images from the rims of some tourmaline crystals, near the outer contact of the pegmatitic body, show included microcrystals of emerald (from 40 up to 100 µm in length) with their c* axis oriented following the direction of the tourmaline growth.

Mapping of the area allow to observe that the tiny emeralds crystals are located in the outer rim of the tourmaline larger crystals. This area shows a particular chemistry that differs from the general composition of the tourmaline crystals especially for Fe/Mg ratio and Cr content.

The automatic system of the Cameca electron microprobe analyser made it possible to get profiles showing variations in the composition of tourmaline crystals at the level of their rim. The increase of the MgO content and correlatively the decrease of the FeO reinforce the trend toward the dravite end-member.

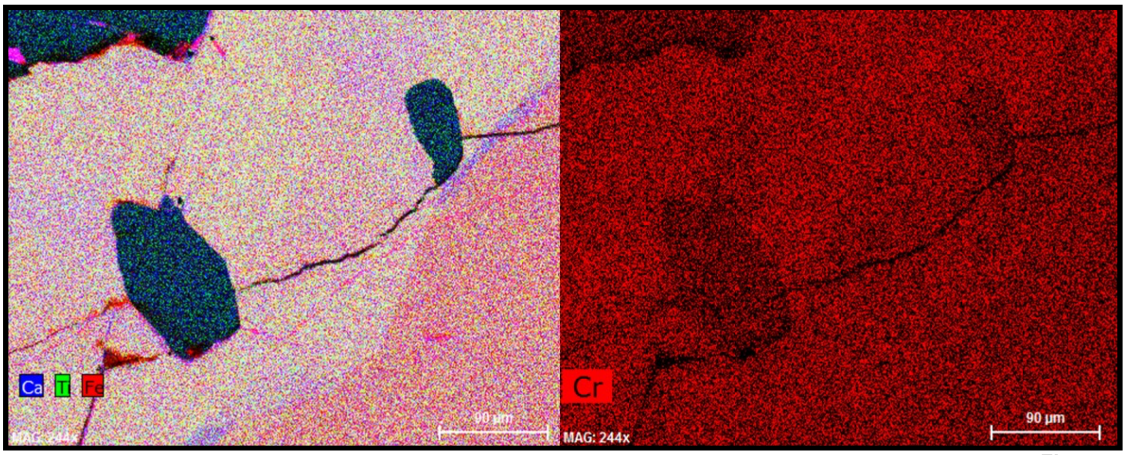

Figure 7
Microprobe mapping showing the differences in composition between the host tourmaline and the included emerald crystals (small areas in blue on the left side). Left: Ca, Ti and Fe distributions. The Fe concentration increases with tourmaline zoning from inside to the external parts. Right: Variation of the Cr content from tourmaline to emerald. The rim with the emerald inclusions is 300 up to 600 microns across.

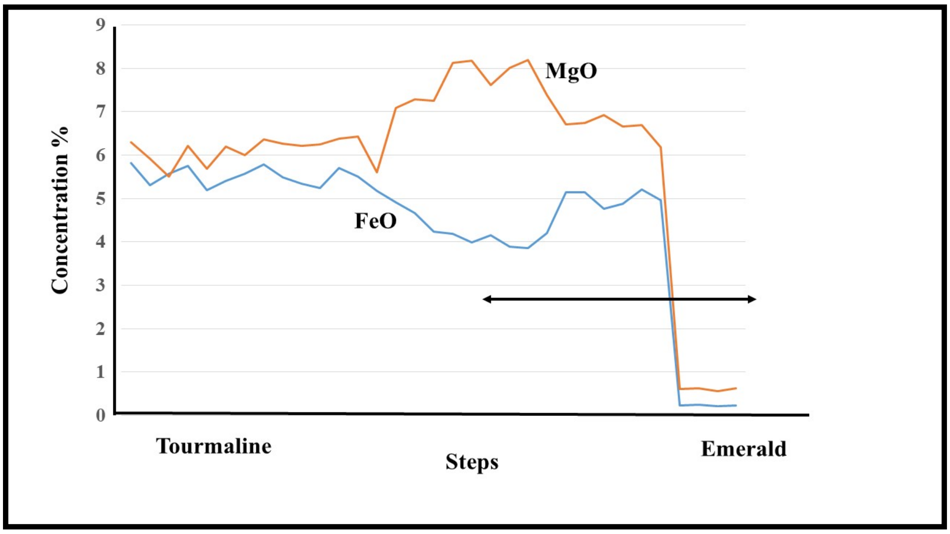

Figure 8
Diagram showing the evolution for FeO and MgO concentrations at the contact between tourmaline and included emerald crystals. The black arrow shows the extension of the rim where emerald crystals occur.

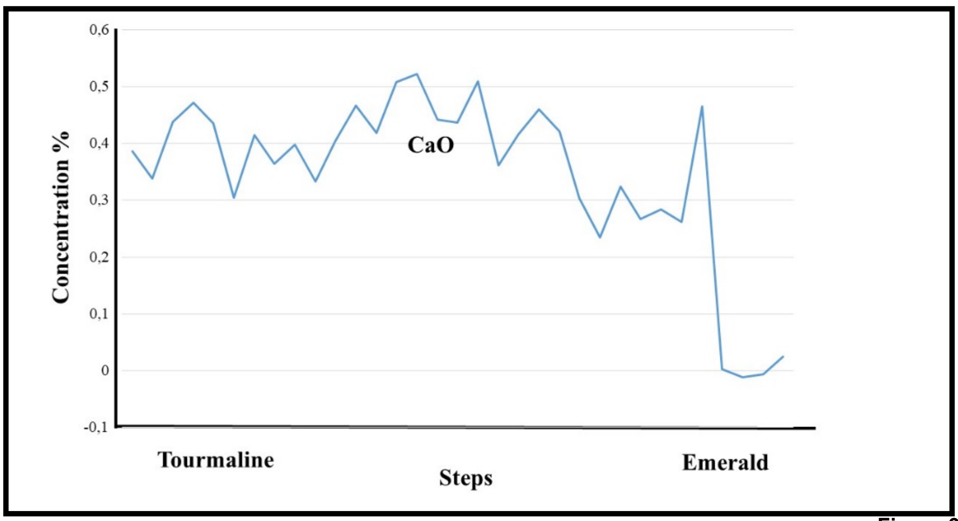

Figure 9
Diagram showing a similar distribution of CaO as for FeO near the contact of tourmaline with emerald. At the very contact between tourmaline and emerald, a wider variation can be observed.

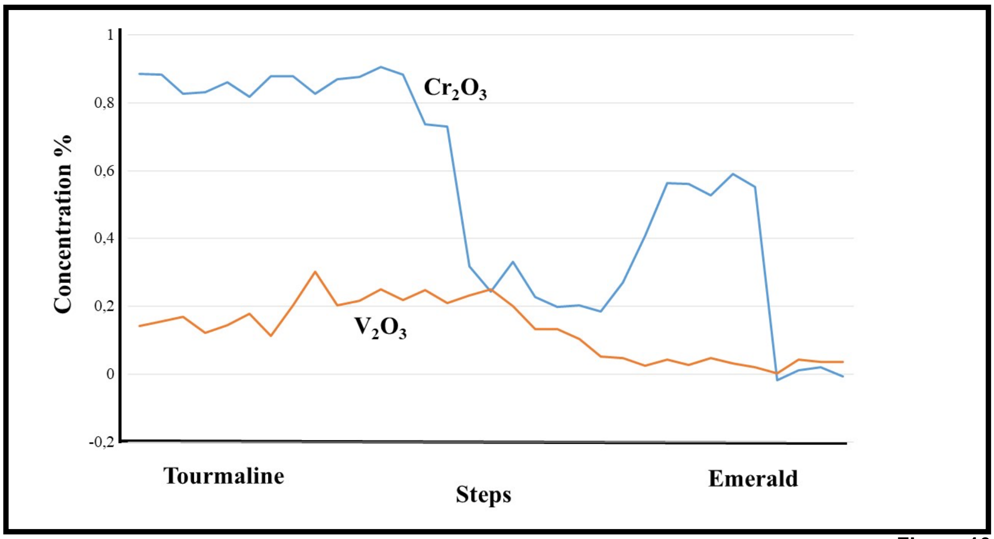

Figure 10
The evolution of the Cr_2O_3 content (blue line) is similar to those of FeO and CaO but differs from that of V_2O_3 (brown line). The V concentration in emerald is very low, what is typical for the samples originating from this region.

In the Capoeirana region, another emerald deposit in the southern part of the same field at Minas Gerais, chromium spinel (picotite), chromium-rich pargasite and chromium-rich phlogopite are abundant in the host rocks. The EMPA analyses of these minerals showed a mean content of 43.4% Cr_2O_3 for picotite. Other minerals of the host rocks of the Capoeirana deposit show mean values for Cr_2O_3 between 950 ppm and 1.75% in pargasite and between 0.95% and 1.4% in phlogopite. These metamorphic rocks can be a good source for fluids involved in the crystallisation of the emerald of Capoeirana deposit. In Novo Cruzeiro, ortho-amphibolites are also present in the metamorphic host rocks but no outcrop was found in the neighbourhood of pegmatite.

The end of the Cr-tourmaline crystallisation, the changes observed in the rim of some tourmaline crystals, the lower content in chromium in emerald, the formation of emerald inclusions in the rim of the tourmaline crystals are in favour of a change in the fluids chemistry that could possibly be reported to a late phase of granite intrusion.

Further research is in progress to identify exactly the sources of the chromium and the link between the crystallization pathway, the change of fluids and the evolution of the late phases of the Brasiliano orogeny.

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