Mg-RICH MICA AND TOURMALINE IN PINK-COLORED "PARELHAS QUARTZITE" INTERCALATIONS IN THE EQUADOR FORMATION, BORBOREMA PEGMATITE PROVINCE, NE-BRAZIL: RECORD OF EVAPORITIC PROTOLITH?

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ABSTRACT

Light gray, green to pink-colored banded quartzite intercalations in the Neoproterozoic Equador Formation (Seridó Group), Seridó Fold Belt, Northeast Brazil, were exploited during several decades as dimension stones under the fashion name "Parelhas Quartzite" (PQ). They can be easily sliced along intensively foliated mm-thick laminae with relatively high content of light green or pink-colored mica, in alternation with more massive, a few cm-thick bands with lower mica contents. The variable ratios between pink and light green micas in the partition laminae, define the color of the produced rock plates, used as outdoor floor or wall coating. Massive, quartzites, meta-arcoses and meta-conglomerates of the Equador Formation (EF) are the exclusive host rock of a few Li-bearing rare element class granitic pegmatites of the Borborema Pegmatite Province, mined for the prized gemstone known as "Paraíba Tourmaline". The EF hosts also nearly 10% of the Be-Li-Ta-bearing pegmatites of the same province. Pink-colored tourmaline in the form of millimeter- to decimeter-long crystals occur as prominent accessory mineral in the pink-colored PQ and are empirically described by miners and field geologists as "rubellite" and the pink-colored mica as "lepidolite". The PQ could therefore be considered as a potential source of Lithium. The tourmaline crystals occur isolated or as radial arrangements, always flattened and oriented in the foliation planes. Light green micas are dominant in the greenish-gray PQ and are always referred in the literature as common muscovite. Tourmaline with the same textural relations as the pink-colored counterpart may also occur in the gray PQ plates but in this case is always black. Electron microprobe analyses of the pink mica and tourmaline allowed establish that they are respectively Al-rich phlogopite and magnesiofoitite (this one preferentially occurring in the crystal cores) with transition to dravite and uvite (rims). Both, mica and tourmaline present insignificant lithium contents. The light green micas were identified as the Fe-Mg-rich variety of muscovite known as "phengite"). The PQ can therefore be discarded as possible source of Li for economic exploration. They can also be excluded as source for Li-enrichment in pegmatites by "in situ" anatexis of quartzite or metasomatic assimilation of ascending pegmatite forming melts. There is also no support for the hypothesis of the genesis of the pink-colored mica and tourmaline by pegmatite-related exomorphic alteration. The most probable origin of the B- and Mg-enrichment in the PQ is related to syn-sedimentary evaporitic or volcano-exhalative incursions during the deposition of the original sediments.

Keywords: Pink-colored magnesian quartzites; phlogopite and magnesiofoitite; Equador Formation; Neoproterozoic Seridó Fold Belt; Borborema Province, NE-Brazil

RESUMO

Intercalações quartzíticas com bandamento de cores variando do rosa a cinza esverdeado claro na Formação Equador, Neoproterozóica, no Grupo Seridó, Faixa Seridó, no Nordeste do Brasil foram amplamente exploradas por décadas como rochas ornamentais sob o nome comercial de "Quartzitos Parelhas" (QP). Estes quartzitos apresentam uma partição fácil ao longo de faixas milimétricas com foliação mais acentuada devido à concentração elevada de micas verde claras ou róseas, alternando com bandas centimétricas de quartzito mais maciço com menor percentagem de mica. As proporções entre as micas rosa e verde clara conferem cor às placas da rocha, usadas para revestimento em paredes e piso externos. Os quartzitos maciços, metarcósios e metaconglomerados que compõe o restante da Formação Equador constituem rochas encaixantes exclusivas de pegmatitos litiníferos lavrados para extração da famosa "Turmalina Paraíba", e também encaixam cerca de 10% dos pegmatitos mineralizados em Be-Li-Ta da Província Pegmatítica da Borborema. Turmalina rosa freqüentemente observada como proeminente mineral acessório dos QP róseos e a mica são muitas vezes empírica- e preliminarmente identificados como *"rubelita"* e lepidolita, respectivamente. Os QP poderiam neste caso ser considerados como potencial fonte de Li. A turmalina ocorre em cristais isolados ou em arranjos radiais sempre achatados e confinados aos planos de foliação. As micas esverdeadas dominantes nos QP cinza esverdeados claros são referidas na literatura como muscovita comum. Nos QP cinza-esverdeadas também pode ocorrer turmalina acessória, neste caso sempre de cor preta, com os mesmos hábitos da suposta rubelita. Análises com microsssonda eletrônica das micas e turmalina róseas revelaram tratar-se respectivamente de variedade de phlogopita rica em Al e de magnesiofoitita (núcleos dos cristais, com variação para dravita e uvita na borda), sempre sem teores significativos de Li. Já a mica macroscopicamente verde clara foi identificada como variedade de muscovita rica em Mg e Fe (ex- *"fengita"*). Os QP róseos podem, pois, ser descartados como fonte de lítio para exploração econômica direta e também como eventual fonte de lítio para geração de pegmatitos litiníferos via palingênese direta ou indireta (via geração de granitos fonte). Pode também ser descartada a origem das micas e turmalina dos QP por alteração exomórfica a partir de fusões pegmatíticas. Conclui-se outrossim que os QP são quartzitos magnesianos, com o enriquecimento de B e Mg provavelmente originado de incursões evaporíticas ou vulcano-exhalativas nos sedimentos originais da Formação Equador.

Palavras Chave: Quartzitos magnesianos róseos; flogopita e magnesiofoitita; Formação Equador; Faixa Seridó, Neoproterozóico; Província Borborema; NE do Brasil.

INTRODUCTION

The Borborema Pegmatite Province (BPP) in northeastern Brazil was known since World War I, when pegmatites were exploited for white mica used mainly as electrical insulator. At the end of World War II the BPP became famous as one of the world's most important Ta-ore producers and for the production of beautiful specimens of beryl and "exotic" tantalum ore minerals, being referred to as type locality for many of them. More recently, the province became important also as resource of raw materials for the Brazilian ceramics industry (feldspars, quartz and kaolin from pegmatites) and ornamental dimension stones (pegmatites, granites and quartzites). Its most famous

product nowadays is the turquoise blue copper- bearing "Paraíba Tourmaline", exclusively found in five amongst 100 of the quartzite-hosted Be-Li-Ta-mineralized pegmatites (Beurlen *et al*. 2011). Light greenish-gray- and pink-colored banded intercalations in the normally massive pegmatite hosting quartzites of the Equador Formation (EF), are themselves exploited as ornamental dimension stones known as "Parelhas Quartzites" or "Equador Quartzites" (hereafter referred as PQ*). The exploited quartzites contain strongly oriented light gray-green- and pale pink-colored micas. Millimeter thick laminae with enhanced mica contents (up to 90 modal %) interlayered with mica poorer and more massive quartzites, are responsible for a very easy partition into cm thick plates. A higher or lower proportion of pink-colored mica defines the color of the plates. Another characteristic feature of the PQ is the presence of pink- to light reddish brown-colored tourmaline in the foliation planes of the pink quartzite bands, and black tourmaline in the light gray to green quartzite bands. Sometimes tourmaline crystals are found in radial acicular to long prismatic aggregates flattened parallel to the foliation planes. The pink-colored tourmaline and micas of these quartzites, are often identified respectively as "rubellite" (pink variety of elbaite) and lepidolite by miners, "garimpeiros" and geologists, probably induced by the similarity with the lepidolite and "rubellite" frequently found in many of the quartzite- hosted pegmatites. The correct identification of the mica and tourmaline in the PQ, as well as its possible relation with the "Paraíba Tourmaline" bearing pegmatites, and the hypothesis of enhanced Li-contents in the PQ-hosted pink-colored micas and its consequently potential as Li-ore, are subject of this investigation.

GEOLOGICAL SETTING

The quarries of the focused banded ornamental "Parelhas Quartzites" are part of the Equador Formation formed mainly by massive quartzites, meta-arkoses and meta-conglomerates. The quartzites of the Equador Formation are usually referred as common muscovite quartzites in most basic geological publications (e.g. Johnston Jr 1945, Roy *et al.* 1964, Ebert 1970, Da Silva and Dantas 1989, Da Silva 1993, and many others). Only Roy *et al.* (1964) gave emphasis to the presence of color banding and suggest that the pink colored bands are due to alteration of iron-bearing accessory minerals in the rock or inclusions in the micas. The Equador Formation underlays garnet-cordierite-sillimanite-biotite schists and paragneisses of the Seridó Formation, and overlays calcic-silicate-rich gneisses that contain intercalations of marble, skarn, quartzite, rare banded iron formation and amphibolite, comprising the Jucurutu Formation. The Jucurutu, Equador and Seridó formations together, in this order from base to top, form the Neoproterozoic (Van Schmus *et al.* 2003) supra-crustal sequence of the Seridó Group, in the NNE oriented Seridó Fold Belt (SFB).

** Footnote: the best known fashion name of the quatzites exploited as ornamental stones is "Equador Quartzite". We are however going to use "Parelhas Quartzite" (PQ) along this publication to avoid confusion with the formal designation of the Equador Formation (EF), which also includes massive quartzites, meta-arkoses and meta-conglomerates, also used frequently in this publication.*

The SFB is located between the Paleoproterozoic São José do Campestre Massif (with some remnants of Archean basement) in the east and the Rio Piranhas Massif in the west, altogether part of the "Northern Domain" (ND) of the Borborema Tectonic Province in NE-Brazil (Van Schmus *et al.* 2003, Dantas *et al.* 2013). Phanerozoic coastal basins (semi-graben) form the northern and eastern limits of the ND. The southern limit of the ND is set by the Patos Lineament, an E -W dextral transcurrent shear zone of transcontinental scale (Fig. 1). In the western part of the SFB a predominance of the Jucurutu Formation is observed together with, not always well distinguishable, reworked paleoproterozoic basement rocks (gneisses and migmatites) and some small Archean relics (Dantas *et al.* 2013). The eastern part of the SFB is characterized by the dominance of the Seridó and Equador formations and corresponds approximately to the area of occurrence of the Borborema Pegmatite Province (BPP), with a longitudinal NNE extension of about 150km and a transversal extension of up to 75km. The eastern limit of the SFB is set by a major transpressional lineament (Picuí-João Câmara Shear Zone), while the western limit is not well defined. Nearly 80% of the ore-mineral bearing pegmatites are hosted by the schists and gneisses of the Seridó Formation, about 10% by the Equador Formation and other 10% by gneisses of the Jucurutu Formation, basement gneisses or several types of variable aged granites (from Paleoproterozoic to Cambro-Ordovician).

The most important quarries of PQ are concentrated in a small area between 2 and 4km SSW from the town of Parelhas. The ornamental quartzites distinguish from the more massive quartzites and meta-arkoses of the Equador Formation by a higher content of white to light green- or pink-colored micas, responsible for a very easy partition along the strong developed foliation, which is parallel to the some centimeter-thick compositional banding of the rock. This banding and partition is most common near Parelhas, where it was exploited for decades as dimension stones. Other smaller quarries occur at Ouro Branco and Junco in the State of Paraíba, and at Equador in the State of Rio Grande do Norte.

The quarries near Parelhas occur at the western limb of a brachiantiformal fold, which extends along more than 100km NNE between the towns of São Tomé-RN (State of Rio Grande do Norte) in the north and Junco (State of Paraíba) in the south, and forms a prominent mountain range ("Serra das Queimadas" or "Umburanas"), standing out 500 m over a pediplane formed by the schists and gneisses (of the Seridó Formation), which by its turn crop out at about 500m over sea level at both sinformal limbs.

PARELHAS QUARTZITES: MODE OF OCCURRENCE AND PETROGRAPHY

Mode of occurrence

The most conspicuous feature of the PQ is a typical banding characterized by the color variation from pink- to light reddish brown-colored plates contrasting with more frequent light green or gray-green-colored ones. This banding, is a few centimeter thick and indicates a strong modal variation of the main components (quartz and mica) and also the type of mica (pink- or pale green-colored). The high concentration (50 to 90 modal %) of strongly oriented micas along millimeter thick "*laminae"*, is responsible for the good partition, essential for the manual production of plates with more or less regular shapes and thickness. "White" micas are dominant in the partition "*laminae"* of the light greenish-gray plates, distinguished by the absence or very low contents of pink-colored micas. Pink-

colored plates are characterized by the presence of high proportions but not always dominant pinkish mica.

A second preeminent feature of these quartzites is the occurrence of tourmaline as accessory mineral, in the form of two different textural types: a) as acicular prismatic crystals up to 10cm long with erratic lineation or forming radial aggregates flattened within the mica rich foliation planes; b) in fractures crosscutting the foliation and banding as stratabound aggregates in the form of centripetal "unidirectional solidification textures" *(UST)* (radial fan-like or comb textures, Fig. 2).

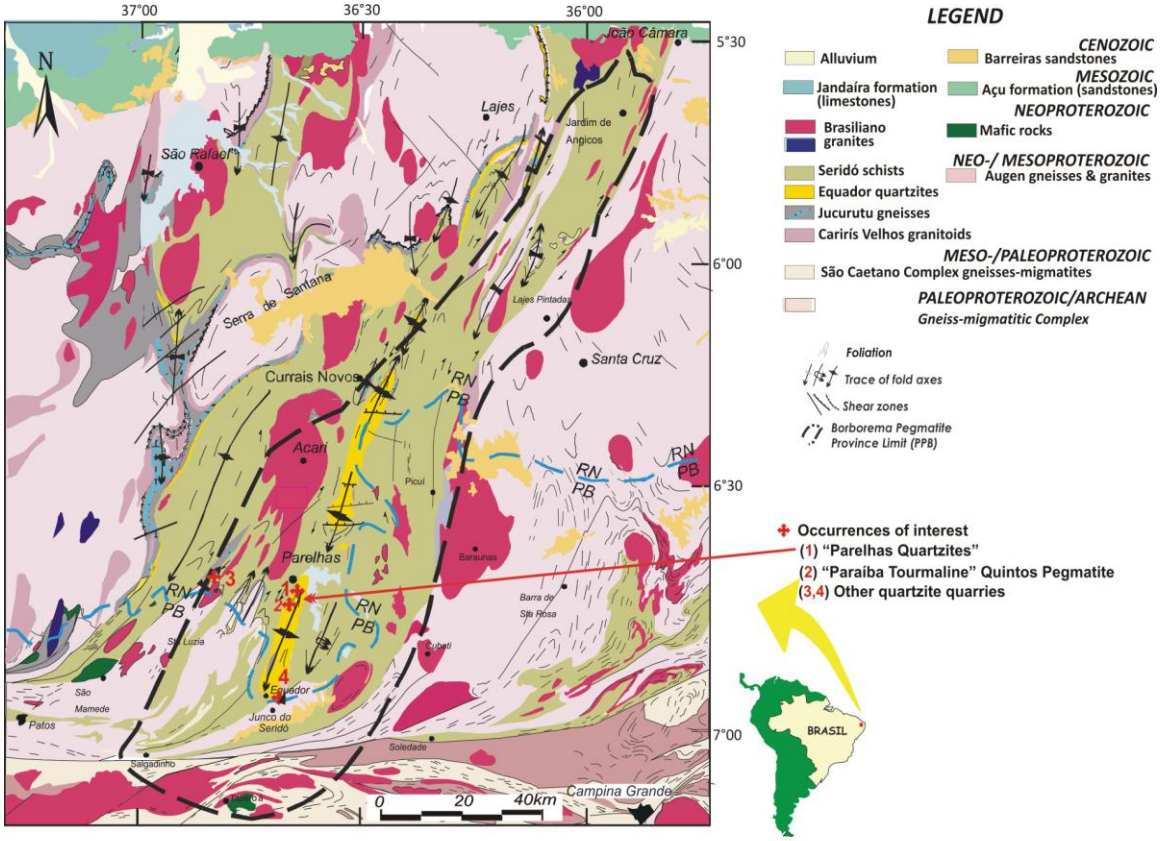

Figure 1- Simplified Geology of the Seridó Fold Belt, in the northern domain of the Borborema Tectonic Province, based on the geological maps of the State of Rio Grande do Norte (BRASIL 1998) and State of Paraíba, (BRASIL 2002). The black dotted line is the limit of the Borborema Pegmatite Province. Occurrences of interest and Parelhas Quartzite quarries (1 to 4) are shown.

Near the contact with pegmatite dikes the content of tourmaline and micas of the PQ and the foliation are gradually reduced until complete disappearance, forming a several centimeter or decimeter thick contact zone of bleached granular quartzite, almost devoid of mica and foliation in abrupt, nearly planar contact with the dike. This field relation indicates that, both, tourmaline and micas (pink and green) within the quartzites are not formed by ionic or metasomatic diffusion or external fluids originated from the pegmatites as suggested as alternative origin, named "exomorphic effects" or "exomorphic halos" by Da Silva (1991, 1993). It suggests, at least

in the observed cases (considering the possibility of more than one pegmatite generation), that the micas and tourmaline in the PQ are older and were destabilized (and assimilated ?) by the pegmatite (Fig. 3). Instead, if there is a relation of the colored minerals in the quartzites with the pegmatites, it would be that the heat of the intruding pegmatite melt destabilized the colored minerals of the quartzite and assimilated elements like Li and B of these minerals (by diffusion or fluid release) from the wall-rocks. This could be in partial agreement of a palingenetic origin of pegmatites or by static recrystallization of quartzites (Ebert 1969, 1970), depending on the relative age of the melt intrusion and the assimilation of wall rocks (which should be almost simultaneous in the case of the supposed palingenetic process). In the observed case, however, the melt intruded clearly much later than the metamorphic peak of the wall-rock, under brittle deformation conditions (as indicated by the abrupt and nearly planar contact between the dike and the quartzite).

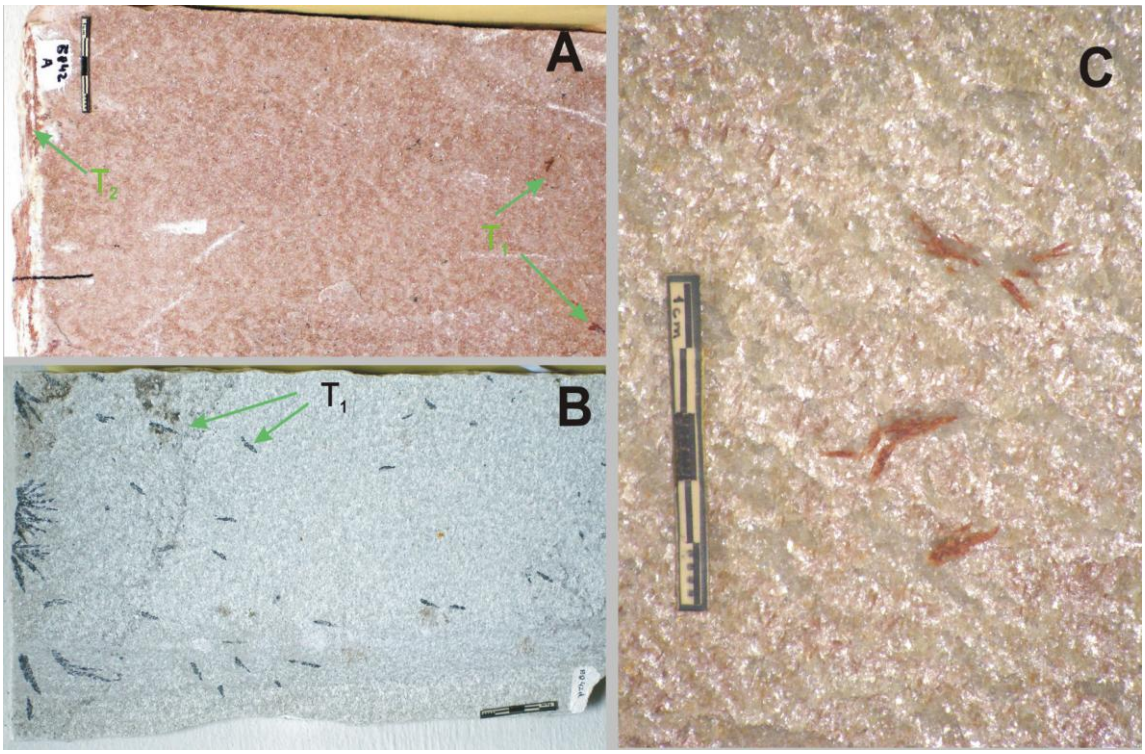

Figure 2 Plates of the color-banded Parelhas Quartzites, respectively with dominance of either pink-colored mica or tourmaline (A and C) or pale gray-green-colored mica and black tourmaline (B). The porphyroblastic tourmaline crystals or aggregates are flattened within the foliation laminae (C, detail of A) and are in contrast with a later generation of comb- or fan-like aggregates of idiomorphic tourmaline found in quartz-filled fractures, as shown at the left edges of the plates A and B.

Figure 3 Outcrop relations of pegmatites with the PQ: the pink and green bands of the quartzite, at the left, are "bleached" close to the contact with a pegmatite dike at the right.

Petrography

As described in the previous section, the main feature of the pink-colored bands of the PQ is the significant presence of macroscopically pink-colored mica. Under the microscope this mica (hereafter be cited as "pink mica") has intensive pleochroism from light orange brownish color (parallel to foliation, Nω) to colorless (normal to foliation, Nε). This optical property contrasts with the dominant mica in the gray- to green-colored quartzite bands, which have only macroscopically light green micas, and are colorless and not pleochroic in thin sections under the microscope (hereafter cited as "white mica", designation used in the literature for species of the muscovite-celadonite series, margarite and paragonite and could be expanded to include polylithionite). Only in 250 microns thick sections, some grains of the "white mica" seem to be very weakly pleochroic from colorless to very pale pink (Fig. 4).

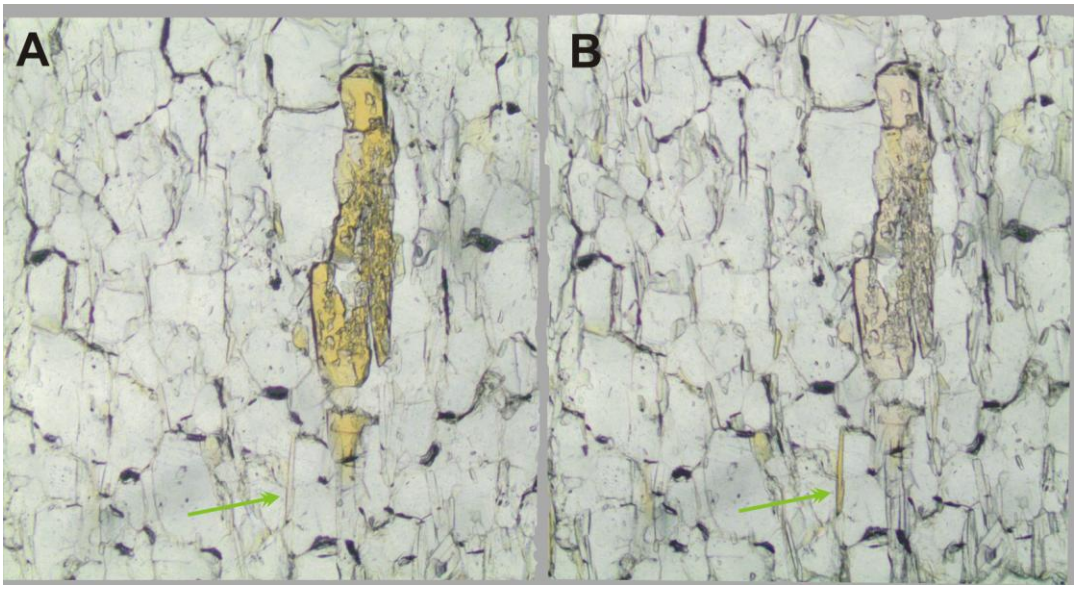

Figure 4 Photomicrography of a polished 250 micrometer thick section (transmitted plane-polarized light) illustrating the strong pleochroism of a small poikiloblastic tourmaline crystal (see the small rounded quartz inclusions in the core) from very light pink (B at the right,- horizontal position) to light brownish-orange (A, vertical position) and of "pink mica" (green arrow at the center bottom of the photomicrography) from brownish orange(B) to nearly colorless (A). Mica and tourmaline are well oriented along the foliation. The poikiloblastic nature of the tourmaline (with a stronger-colored magnesiofoitite core rimmed by dravite and uvite) is documented by the small rounded inclusions of incompletely assimilated quartz. The width of the photos is 2.5mm.

Both pink and "white" micas occur together in green- and pink-colored bands of the quartzite in variable proportions defining the color of the bands. The pink and white micas occur in textural equilibrium with each other, (grain size 0.3 mm long in average, but sometimes reaching up to 1.0 mm in length) with simple planar contacts and strong orientation with more than 90% of the crystal plates oriented with less than 10 degrees of deviation from the average foliation, parallel to the compositional banding. Pink and "white" micas therefore seem to be synchronic each other and syn-deformational. More massive, up to few centimeter thick quartzite bands with less than 20 vol. % of micas, alternate with millimeter- to sub millimeter-thick, strongly foliated high micaceous *laminae* with up to 90 vol. % content of micas. Besides the two micas, the main component of the massive bands of the rock is quartz, which characterizes the texture with grains usually elongated along the foliation with around 0.3mm in length. Feldspar was not observed (at least not with the typical feldspar twinning).

The most preeminent accessory mineral in these quartzites is tourmaline in the form of acicular to long prismatic crystals (usually as poikilitic to -porphyroblastic crystals with a few mm in length, but sometimes reaching up to 10 cm along the c axis). The tourmaline crystals are strongly flattened in sections across the banding and foliation, and do not show the typical

trigonal prismatic habit (different speed of crystal growth of the prism surfaces?). In the pink-colored bands of the PQ the tourmaline is macroscopically light orange to reddish brown. Under the microscope this tourmaline shows a medium strong birefringence, negative uniaxial interference figures, and the typical extinction parallel to the C axis. The birefringence is approximately 0.030 ($N\omega$ = 1.648 and $N\varepsilon$ = 1.618, with pleochroism respectively from light orange brown (parallel to foliation) to colorless or very light yellowish pink) as shown in Fig. 4. These birefringence and refraction index values are close to those reported by Hawthorne *et al.* (1999), with ω = 1.650 and ε = 1.624), but overlap the data of other species of the group (e.g. schorl-dravite series and elbaite). In light green bands the tourmaline is usually black under naked eye (supposedly iron rich dravite). Other, very rare accessory minerals are apatite, sphene, zircon and ilmenite in very small grains (usually less than 0.05 mm). The poikiloblastic intra-foliation flattened habit of the individual tourmaline crystals (without linear orientation) and its aggregates indicate syn- to late-, probably peak-metamorphic crystal growth, related to ductile deformation. The tourmaline associated with quartz veins crosscutting the foliation and banding, probably represents another generation, related to retrograde metamorphism and brittle deformation conditions. The tourmaline of this second generation is formed by lateral segregation as indicated by the stratabound distribution: light brownish orange tourmaline in fractures cutting pink-colored quartzite bands, and black tourmaline in gray-green bands.

MINERAL CHEMISTRY

Preliminary approaches by wet chemical analyses of a few samples of bulk rock and mineral concentrates, followed by powder X-ray diffractometry of mineral concentrates and, in addition, observation of optical properties of both mineral groups (micas and tourmalines) during petrographical examination, did not allow an inequivocal conclusion if mica and tourmaline species in the quartzites were Li-bearing or not. Thus, during the systematic electron microprobe analyses (EMPA) of tourmalines from pegmatites (Soares *et al.* 2008, Beurlen *et al.* 2011), two pink-colored tourmaline crystals and six mica plates of a pink-colored band of the PQ were also analyzed, aiming to obtain a correct mineral identification.

Methodological aspects

The two tourmaline crystals were analyzed using EMPA along cross sections from core to rim (totalizing 13 data-points) and six mica plates, with two points each, totalizing respectively six points for the light orange-brownish to pink micas and also six points for three "white mica" plates. The preliminary results were reported during the 2010 IMA meeting in Budapest (Beurlen *et al.* 2010). The results were very surprising and will therefore be discussed here, separately from the previous studies on pegmatite hosted tourmalines.

The EMPA were performed in a Jeol Hyperprobe, model JXA-8500F, using the standards and working conditions as described in detail by Beurlen *et al.* (2011).

The analytical results in weight percent were recalculated using the *.xls worksheets for "tourmaline", "biotite" and "muscovite" developed by Tindle *et al.* (2002a), available for open access at the web address: *http://www.open.ac.uk/earth-research/tindle/*.

The recalculated data in apfu (atoms per formula units) are summarized in Table 1 (average formulae) and Appendix I (complete results). Li_2O and H_2O in mica were calculated according to Tindle and Webb (1990), and Li calculation in tourmaline according to Selway and Xiong (2002) and Tindle *et al.* (2002b). These worksheets for all three mineral groups ("tourmaline", "biotite" and "muscovite") allow two modes of formula calculation*[1]: either considering the possible presence of not analyzed Li (=Li*), or presuming the absence of Li. In this context it may be opportune to be pointed out for those not familiar with mineral chemistry analyses, that the EMPA equipments do not allow to analyze Li. All "results" reported here for Li, B, H_2O, are calculated considering ideal formula compositions as supposed by the worksheets by Tindle *et al.* (2002a). The use of the SIMS facility at the GFZ-Potsdam was programmed to obtain analyses of Li and B (and their isotopes), but was out of work during our stay at this Institute.)

Results

MICAS

The EMPA data of analyzed micas are plotted in Figure 5, the mica classification diagram proposed by Tischendorf *et al.* (2007).

In the case of "white mica" in the pink-colored quartzite bands both calculation modes indicated absence of Li. The average formula in apfu (with standard deviations for all elements below 1%) for six analyses in three petrographically homogeneous "white mica" crystals is:

$$(K_{1.87},Na_{0.051})\,[Al_{3.35}(Mg_{0.39},Fe_{0.37}\,Mn_{0.01})]\,(Si_{6.30},\,Al_{1.70},\,Ti_{0.07})O_{20}(OH_{3.995},Cl_{0.005})_{4.00,}$$

This formula is very close to the ideal "phengite" formula suggested by Bailey (1984a, b):

$$K_2[Al_3(Mg,Fe)_{1.0}](Si_7,Al_{1.0})O_{20}\{OH,F)_4.$$

"Phengite" however, was discredited by IMA as valid mineral species, following Rieder *et al.* (1998), because it has an intermediate composition in a series with muscovite and celadonite as end members but with intermittent solid solution. However a considerable excess of ^{T}Si and significant $^{Y}(Mg + Fe)$ contents distinguish "phengite" from muscovite, Fe+Mg values being to low for celadonite. Therefore Rieder *et al.* (1998) suggest maintain the use of the name "phengite", to designate the discontinuous muscovite-celadonite series, the classical "phengite" being an intermediate variety now called "Mg-Fe-rich muscovite" by Tischendorf *et al.* (2007). It is not possible to know if the obtained composition also represents the "white micas" in the gray-green PQ, not yet analyzed.

The average formulae obtained for 6 data-points in three plates of "pink-colored" mica are:

$$(K_{1.87},\,Na_{0.04})\,(Mg_{4.77},\,Fe_{0.21},Al_{0.68}\,Ti_{0.09})\,(Al_{2.36},Si_{5.64})\,O_{10}\,(OH)_{4.00}\text{ (without Li)}$$

or, if the presence of Li is supposed,

$$(K_{1.83},Na_{0.04}\,)\,Li^*_{\,0.98}(Mg_{4.66},Fe_{0.20},Ti_{0.08}Al_{\,0.49})\,(Al,_{2.49}Si_{5.51})\,O_{10}\,(OH)_4$$

The average formulae above are very representative with the standard deviation for all elements below 0.03 apfu (see Table 1)

The best (almost perfect) fit for the formula of the pink-colored mica, if calculated without Li*, is with the "Al-rich phlogopite" variety, identified (amongst more than 6,700 mica analyses) by Tischendorf *et al.* (2007) as shown in Table 1. The formula obtained considering the theoretically possible presence of Li* (0.98 apfu, 1.7 wt. %) is unrealistic and can be discarded because: a) there is no fit with any of the tainiolite varieties proposed by Tischendorf *et al.* (2007), which are the only known high (Mg + Li)-bearing micas; b) there is no correlation between Li* and ^{VI}Al, as usually observed in the siderophyllite-polylithionite series; c) all varieties of tainiolitic micas identified by the dataset of Tischendorf *et al.* (2007), have high F-contents (>1.0 of 4.0apfu), not present (found below detection limit) in the studied pink micas; d) this considerable F-content is necessary to stabilize a high Mg-Li solid solution according to Monier & Robert (1986); e) the Si contents (5.64 +/- 0.02 apfu) of the pink-colored micas match well those of the Al-rich-phlogopite variety of Tischendorf *et al.* (2007), and are much lower than those of all varieties of Fe-poor tainiolitic micas (between 5.99 and 7.77 apfu, for the formula normatized to total site ^{iv} = 8.0).

The considerations a) to e) above strongly support the identification of the pink-colored micas as Al-rich phlogopite, with insignificant Li-contents according to Tischendorf *et al.* (2007) below 0.02 wt % or 0.01 apfu. This is assumed as real, while direct analytical results for Li by LA-ICP or other methods are under way.

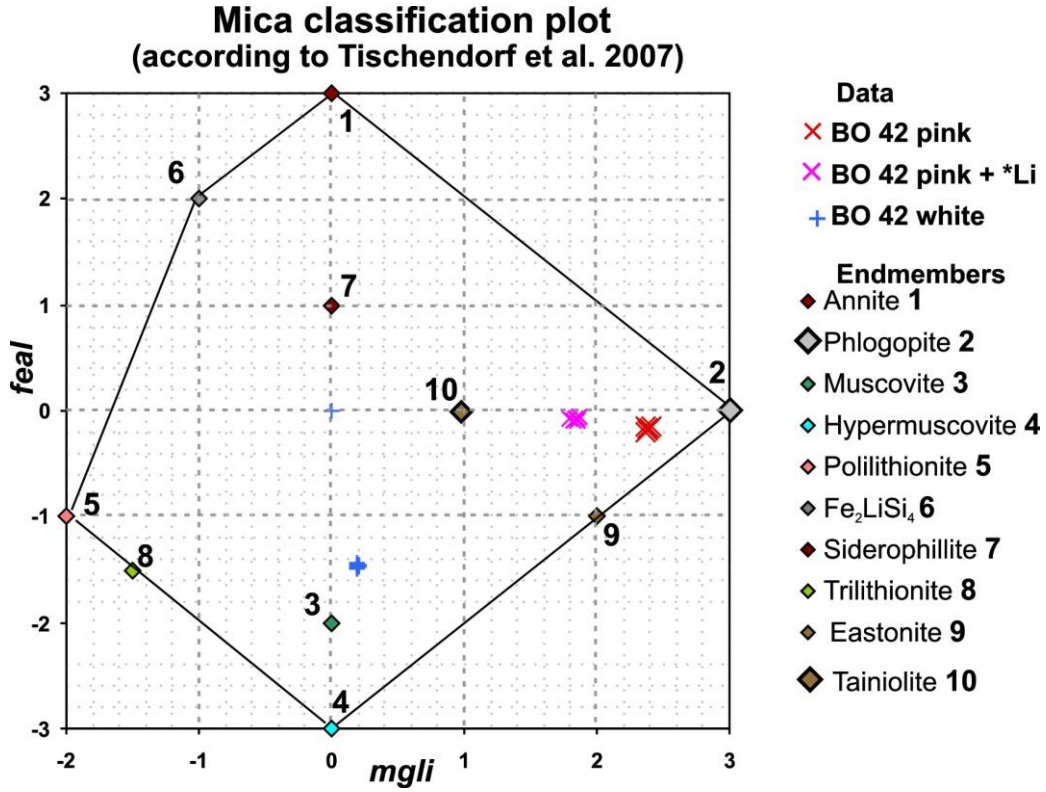

*Figure 5 Plot of the "pink" and "white-mica" data of the pink-colored "PQ" from the BPP in the binary classification diagram of **feal** and **mgli** values (calculated from apfu data) in comparison with most end member compositions of mica species, as proposed by Tischendorf et al. (2007).*

Table 1. Mean formulae in apfu (atoms per formula units) of micas and tourmaline from the "Parelhas Quartzites" (BO42) in comparison with average formulas of mica varieties by (*1) Tischendorf *et al.* (2007) and (*2)Bailey (1984).

	"Pink mica" BO42 *4				***Average apfu mica varieties*** [1*] ***Tischendorf***					[2*] ***Bailey***	**"white mica"BO42**		**Tourmaline BO42 *3**		
	no *Li (n=6)		*with *LI (n=6)*		Magnesian ="pink", n>20			***"white" (n>>20)***			no *Li, n = 6 *4		formula for 31 anions		
	Average apfu *4	1 Sdev	Avera-ge	1 Sdev	Tainio-lite	Phlogo-pite ss	Al-rich phlogo	Mg-Fe-muscovit	musco-vite ss	*phengi-te*	Avera-ge apfu	1 S d		Avera-ge,n=13	1 S d
Si	5.637	0.017	5.512	0.017	*7.726*	*5.736*	*5.520*	*6.882*	*6.148*	7.000	6.299	0.022	T: Si	5.937	0.030
Al iv	2.363	0.017	2.488	0.017	*0.274*	*2.242*	*2.480*	*1.118*	*1.852*	1.000	1.701	0.022	Al	0.063	0.030
													B	3.000	0.000
Al vi	0.679	0.027	0.485	0.027	*0.028*	*0.000*	*0.738*	*2.690*	*3.600*	3.000	3.347	0.022	Z: Al	5.998	0.005
Ti	0.086	0.006	0.084	0.006	*0.020*	*0.130*	*0.048*	*0.020*	*0.036*		0.066	0.008	Mg	0.002	0.005
Fe	0.205	0.016	0.201	0.016	*0.138*	*0.494*	*0.020*	*0.688*	*0.164*	0.500	0.336	0.012	Y: Al	0.149	0.126
Mn	0.050	0.003	0.049	0.003	*0.034*	*0.018*	*0.054*	*0.016*	*0.006*		0.010	0.004	Ti	0.035	0.007
Mg	4.769	0.032	4.662	0.031	*4.014*	*5.180*	*4.640*	*0.690*	*0.240*	0.500	0.385	0.013	Fe^{2+}	0.422	0.084
Zn	0.008	0.005	0.008	0.005					*0.001*		0.003	0.002	Mn	0.020	0.006
Cu	0.005	0.004	0.005	0.004							0.005	0.005	Mg	2.211	0.068
Li*	0.000	0.000	0.980	0.044	*1.576*	*0.006*	*0.006*	*0.022*	*0.096*		0.000	0.000	Zn	0.004	0.004
Ca	0.000	0.000	0.000	0.000	*0.038*	*0.014*	*0.016*	*0.022*	*0.008*		0.000	0.000	Cu	0.002	0.003
Na	0.039	0.011	0.038	0.010	*0.11*	*0.124*	*0.044*	*0.048*	*0.192*		0.051	0.007	Li*	0.157	0.022
K	1.868	0.022	1.827	0.022	*1.858*	*1.798*	*1.612*	*1.808*	*1.702*	2.000	1.869	0.016	Σ [VI]R	3.000	0.000
													X: Ca	0.322	0.053
OH*	3.997	0.002	3.997	0.002	*0.648*	*3.288*	*3.844*	*3.856*	*3.798*		3.997	0.003	Na	0.343	0.039
F	0.000	0.000	0.000	0.000	*3.348*	*0.702*	*0.152*	*0.134*	*0.190*		0.000	0.000	K	0.018	0.006
Cl	0.003	0.002	0.003	0.002	*0.004*	*0.010*	*0.004*	*0.010*	*0.012*		0.003	0.003	r	0.318	0.065
Σ cat	15.71		16.34		15.82	14.12	15.18	14.00	14.05		14.071		OH	3.999	0.001
TOTAL	19.71	0.020	20.340	0.030	*19.816*	*18.124*	*19.178*	*18.004*	*18.045*		18.071	0.014	F	0.000	0.000
Σ [IV]R	8.000		8.000		8.000	8.000	8.000	8.000	8.000		8.000		Cl	0.001	0.001
Σ [VI]R	5.800	0.009	6.475	0.038	5.810	5.828	5.506	4.126	4.143	4.000	4.151	0.007	Bi_2O_3	0.009	0.010
Σ [XII]R	1.910	0.023	1.865	0.024	2.006	1.991	1.672	1.878	1.902	2.000	1.920	0.017			
Best fit	Al-rich *4	phlogopite	Al-rich phlogopite								Mg-Fe-muscovite			*3 Mg-foitite to dravite & uvite	

Observations :*3 In the case of tourmaline BO42 the average (dravite) of thirteen data is not representative. A significant variation is observed from the core (Mg-foitite) to the border (uvite) of three zoned crystals. *4 Mica compositions are homogeneous.

TOURMALINES

The average formula for all 13 tourmaline analyses, if the presence of Li is supposed to complete the occupancy of the Y site (Selway & Xiong 2002), is:

$(Na_{0.34},Ca_{0.32},X_{0.32})(Li^*_{0.16}, Al_{0.15}, Mg_{2.28}, Fe_{0.43}, Mn_{0.02}, Ti_{0.03})\ Al_{6.00}\ (Si_{5.94}, Al_{0.06})\ (B^*_3O_2)\ (OH)_4$

or,

$(Na_{0.34}, Ca_{0.32}, x_{0.32}, K_{0.02})(Al_{0.15}, Mg_{2.28}, Fe_{0.43}, Mn_{0.02}, Ti_{0.03})\ Al_{6.00}\ (Si_{5.94},Al_{0.06})\ (B^*_3O_2)\ (OH)_4$

if an absence of Li and an incomplete occupancy of the Y site is supposed (Beurlen *et al.* 2011).

A systematic variation for the X and Y sites occupancy is observed for data of the analized crystal cores: $^X[vac_{0.46}, Na_{0.31}, Ca_{0.22}]\ ^Y[Mg_{2.1}, Al_{0.4}, Fe_{0.3}, Li^*_{0.2}]$, (magnesiofoitite) grading to
$^X[vac_{0.29,}\ Na_{0.44}, Ca_{0.34}]\ ^Y[Mg_{2.2}, Al_{0.3}, Fe_{0.4}, Li^*_{0.1}]$ (dravite) and to
$^X[vac_{0.30}, Na_{0.30}, Ca_{0.40}]\ ^Y[Mg_{2.3}, Al_{0.1}, Fe_{0.4}, Li^*_{0.2}]$ (uvite) for the crystal rims.

The obtained data for tourmaline analyses from the PQ are plotted in the first step classification diagram of the tourmaline-supergroup species into three primary groups according to the X-site occupancy according to Henry *et al.* (2011), which are respectively X-vacant, Alkali and Calcic-groups (Fig. 6). In the figure only a few (most common and of interest for the present case) of the more than 20 IMA validated species (found in nature) of the supergroup are referred. Other well known species are fluor-uvite, fluor-liddicoatite, darrelhenryite, of the calcic group, elbaite, olenite, oxischorl, oxidravite, povondraite, buergerite and many other, less common species in the alkali group.

For comparison with the PQ hosted tourmaline, data from other magnesio-foitite (and associated foitite and alkali group) occurences in the literature are also shown in Fig. 6. Also included are data of tourmaline hosted in schists of the Seridó Formation in the BPP. It can be seen easily that the pink-colored, PQ hosted tourmaline compositions are quite different from all other occurrences.

The presence of calculated Li* up to 0.2 apfu in the Y-site of the tourmaline formulas above may easily be explained by an erroneous supposition that the Y-site must be completely filled (with a total of 3.00 cations, formula calculated for 31 anions), as presumed by the used formula calculation work sheet.

The possibility of Y-site vacancies up to 0.2 apfu (instead of Li*) was demonstrated in praxis by several authors (eg. Ertl *et al.* 2003, 2006, Beurlen *et al.* 2011), and by theoretical approaches (eg. Bosi 2010). In the present case the inexistence of a correlation between the ^{Y}Al and ^{Y}Li* obtained by EMPA, clearly demonstrates that the usual substitution mechanism for elbaite formation [^{Y}(Mg,Fe = Li+Al)] does not operate in the present case, e.g. there is probably no significant Li present in this tourmaline. Instead, the charge compensation for the entry of ^{Y}Al,Fe^{3+} may be explained in part by the olenite $[(Al^{3+}O)(R^{2+}(OH)^{-1})_{-1}]$ or oxi-schorl $(Fe^{3+}O)(R^{2+}\ OH)_{-1}$ substitution mechanisms, or also by small Y site vacancy.

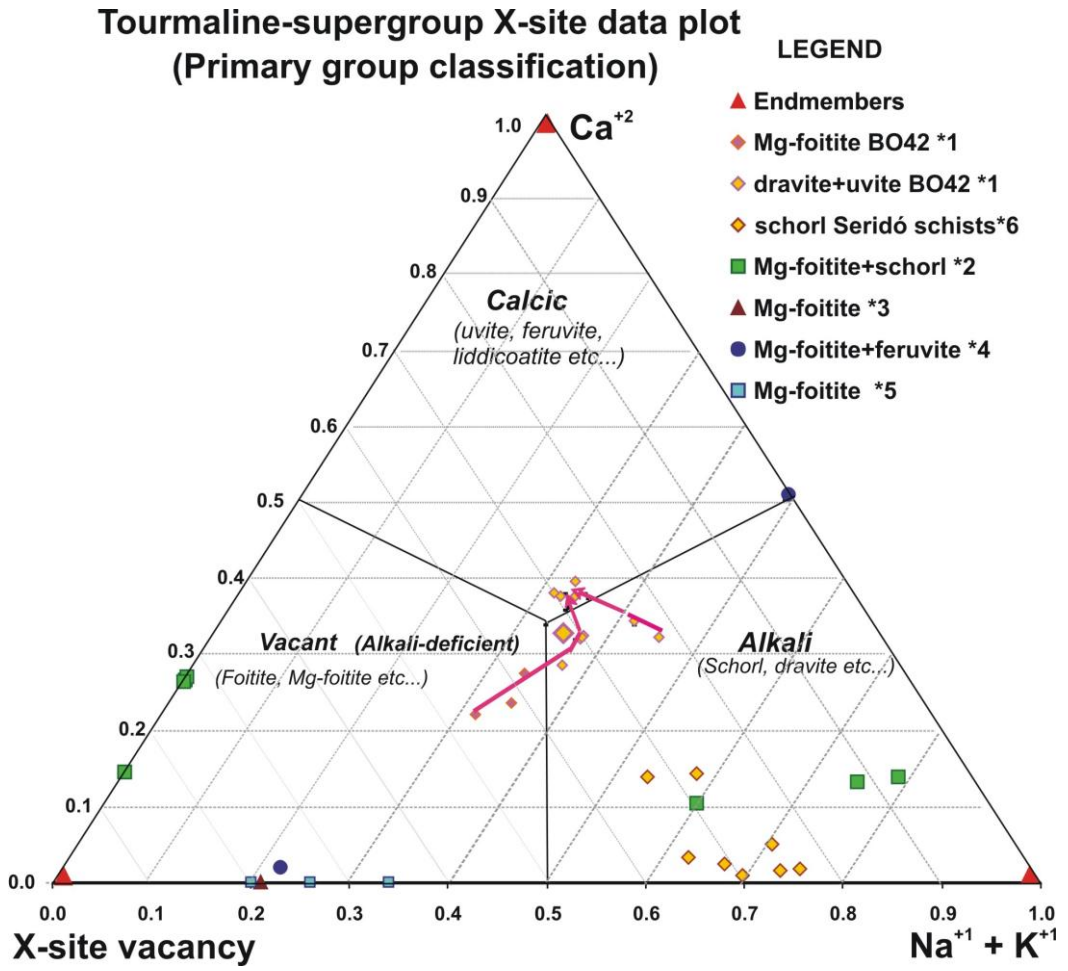

*Figure 6 Compositional data of "magnesiofoitite" crystals from the pink-colored "PQ" (number*1 in the legend) in the data-plot for tourmaline-supergroup classification into three primary groups according to the X-site occupancy. For comparison data of other magnesiofoitite occurrences as described by, Hawthorne et al. (1999), Garda et al. (2003), Roseberg et al. (2003) and Medaris et al. (2006), respectively numbered with *3, *2, *4 and *5 in the legend are shown. Note the very uncommon position of the data of the zoned crystals of the "PQ" in the BPP (*1), varying from magnesiofoitite cores through dravite to uvite rims, all of them very close to the limit between the fields of calcic, alkali- and vacant primary groups of tourmaline species. Note also the difference of the"PQ" hosted crystals with those of the dravites hosted in the Seridó Schists (*6).*

DISCUSSION

The mineral chemistry data of the examined micas and tourmaline indicate that they do not have significant Li-contents. Their macroscopic determination as "lepidolite" "rubellite", respectively for the pink mica and tourmaline is actually incorrect. Instead, the identification as Al-rich phlogopite) for "pink-colored mica" and Mg-rich alkali-deficient tourmaline (magnesiofoitite and dravite-uvite with high X-site vacancy) is suggested. In addition, the associated "white mica" in the same pink PQ can be identified as Mg-Fe-rich muscovite variety (or formerly named *phengite).*

These data are in disagreement with the statements in the literature, where the quartzites of the Equador Formation are referred as common muscovite quartzites (Ebert 1970, Roy *et al.* 1964, Da Silva *et al.*

1995, Da Silva 1993,) and sometimes as "rubellite and lepidolite" bearing quartzites (Da Silva 1991, 1993).

Phlogopite-bearing quartzite and magnesian quartzites are uncommon rocks and are found either as (1) intercalations in metamorphosed volcano-exhalative sequences or (2) as remnants of evaporite facies incursions in dominantly mature continental or coastal sandstone sequences. Type 1 quartzites should occur intimately interlayered with calcic-silicate gneisses, skarns and marbles (sometimes associated with sulfide ore occurrences, e.g., Höy 1985, Mruma & Basu 1986). This type of sequences is found in the Jucurutu Formation underlying the Equador Formation, and also at the transition between the Equador Formation and the overlying Seridó Formation. However, in the immediate neighborhood of the PQ such kind of interlayering with marbles, skarns or amphibolites is not observed. In the studied case, the PQ occurs completely (stratigraphically and laterally) enclosed in massive quartzites, meta-arkoses and meta-conglomerates. The enhanced B-, Mg- and Fe-contents of the PQ indicated by the presence of Mg-rich tourmalines and micas would therefore be better in agreement with their formation by metamorphism of mature (no feldspar) coastal sandstones with sporadic evaporitic incursions. Similar magnesian evaporitic sandstones were documented by Fenton & Fenton (1937), with their metamorphic equivalents (Hietanen 1984, Mora & Valley 1989) in the Belt Series in NW-USA. Such an origin was also admitted by Da Silva (1991, 1993) as a possible alternative to the proposed origin by "exomorphic alteration" related to pegmatites.

The pre- to syn-deformational formation of the micas and tourmaline in the PQ, based on the field and petrographic evidences, is also in disagreement with the supposition of their genesis by pegmatite or granite related exomorphic-metassomatic or hydrothermal activity. The outcrop relations between quartzites and pegmatites indicate that the pegmatites were emplaced after the main peak metamorphic-deformational ductile stage, along fractures formed during a late extensional brittle stage, precluding a direct origin of pegmatites as products of partial melting or static recrystallization of the quartzites as advocated by Ebert (1970). However, boron assimilation from the quartzitic wall-rocks of the Equador Formation by pegmatite melts during ascent or emplacement can not be discarded.

CONCLUSIONS

The pink-colored mica and tourmaline flattened within the mica rich strongly foliated laminae in the "Parelhas Quartzites" are not *"lepidolite"* and *"rubellite"* as frequently supposed, but respectively phlogopite and magnesiofoitite, dravite and uvite, with insignificant Li-contents.

Considering that the pink-colored mica and tourmaline were the only minerals supposedly indicative of enhanced Li-contents, it could be concluded that the PQ in particular and, by extension, the quartzites of the Equador Formation are not candidates, neither as potential lithium-ore, nor as lithium enriched source-rocks for (i) indirect (with intermediate) formation of anatectic granite plutons (e.g. Ebert 1970), either (ii) "in situ" anatectic pegmatite forming melts (as proposed by Müller *et al.* 2015, 2016 for Scandinavian pegmatites), or (iii) Li-enrichment in pegmatites by metasomatic assimilation by ascending melts.

The pink-colored bands of the quartzites are enriched in magnesium and boron, probably due to evaporitic, or less probably, to syn-sedimentary hydrothermal exhalative activity during the formation of mature coastal clastic sediments on continental margins.

The conclusions, so far valid only for the pink intercalations of the PQ, need to be confirmed with similar studies in other quartzites of the Equador Formation. The results are in at least partial agreement

with experimental approaches of anatexis of the Equador Formation (and other possible source rocks) by Sallet *et al.* (2015), indicating that only high temperature infracrustal anatexis, would be able (with additional contamination by other sources) to produce source magmas similar to those of the pegmatitic granites. Furthermore, use of additional techniques such as $^{11}B/^{10}B$ and other isotopic studies may be helpful to constrain petrogenetic issues.

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