

EVALUATION OF THE POTENTIAL FOR RARE EARTH ELEMENT (REE) DEPOSITS RELATED TO THE BORBOREMA PEGMATITE PROVINCE IN NORTHEASTERN BRAZIL.

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ABSTRACT

Electron probe microanalyses (EPMA) of rare earth elements (REE) in xenotime, monazite, zircon, thorite and uraninite from different facies of pegmatitic granites supposed to be parental to complex rare element (REL) pegmatites of the Borborema Province (BPP) in Northeastern Brazil indicated monazite and xenotime as most common heavy accessory phases with significant enrichment in these elements. Averages of Nd and Eu in monazite are respectively 13.06 and 0.54 wt. % oxide and mean contents of Dy, Yb, Gd, and Er (respectively 4.77, 3.39, 3.15 and 3.86 wt. % oxide) are found in xenotime. The other common accessory minerals like zircon, thorite, uraninite showed no significant enrichment in REE. Low xenotime and monazite contents in pegmatitic granites and their irregular distribution and reserves in pegmatites, turn the extraction directly from the hard rock impracticable. The REE-enriched minerals could however be enriched in and easily extracted from alluvial placers within the province. Ion adsorption REE enrichment could hypothetically form in parental granites of the pegmatites at the intersection with paleogeomorphologic plateaus culminated by the early Cenozoic siliciclastic Serra dos Martins Formation, because at this unconformity it is possible to observe an intensive kaolinization of the basement and lateritization of the covering sediments.

Keywords: Borborema Province, complex REL pegmatites, parental granites, possibility of related REE-deposits.

RESUMO

Análises de Elementos de Terras Raras (ETR) com microsonda eletrônica em xenotima, monazita, zircão, torita e uraninita de diferentes fácies de granitos pegmatíticos, supostos serem fonte de pegmatitos complexos de elementos raros (REL) na Província Pegmatítica da Borborema (PPB) no Nordeste do Brasil, mostraram que a xenotima e monazita são os acessórios mais frequentes com significativa de (ETR). Conteúdos médios de Nd e Eu em monazita são respectivamente de 13,06 e 0,54 % peso dos óxidos e de Dy, Yb, Gd, e

Er respectivamente de 4,77, 3,39, 3,15 e 3,86 em xenotima, considerando apenas os óxidos de ETR mais caros no mercado. Os outros acessórios analisados, como zircão, torita, uraninita não apresentaram teores significativos de ETR. As reduzidas frequências modais da xenotima e monazita tanto nos granitos pegmatíticos como nos próprios pegmatitos tornam uma exploração das rochas duras impraticáveis. Os minerais com maior enriquecimento de ETR poderiam ser concentrados e explotados em placeres fluviais no âmbito da Província Pegmatítica. Há também a possibilidade de enriquecimento dos ETR a partir dos granitos parentais e pegmatitos na interseção dos plateaus paleo-geomorfológicos culminados pelos sedimentos siliciclasticos e caulinizados da Formação Serra dos Martins. Isto porque, na inconformidade na base da mesma ocorreu lateritização e intensa e profunda caulinização no embasamento cristalino subjacente, em condições similares às das jazidas de “adsorção iônica” (hoje também conhecidas como “hospedadas em regolito caulinado”, como Zudong, maior jazida de terras raras pesadas) na China.

Palavras-chave: Província Borborema, pegmatitos complexos tipo REL, granitos parentais, possibilidade de depósitos de ETR relacionados.

INTRODUCTION

Electron probe microanalyses (EPMA) of rare earth elements (REE) in xenotime, monazite, zircon, thorite and uraninite from different facies of pegmatitic granites supposed to be parental to Rare Element (REL) pegmatites of the Borborema Pegmatite Province (BPP) in Northeastern Brazil were obtained originally for geochronological purposes. Preliminary results were published by Beurlen *et al.* (2009), using the chemical dating method developed by Suzuki & Adachi (1991), Montel *et al.* (1994) and refined by Rhede *et al.* (1996). The results indicated similar ages of these granites and the spatially related gem- and Be-Li-Ta-bearing pegmatites, below 540 Ma (Baumgartner *et al.* 2006, and Beurlen *et al.* 2009).

The exponential increase of the prices of some REE, up to several hundred thousand US dollars per ton of refined rare earth oxide (REO), as consequence of its application in components of a fast growing number of devices of the high-tech industry (e.g. Haxel *et al.* 2002, Campos *et al.* 2015, Statistica 2019, Sreevidya 2017), justifies the use of the available data for

an evaluation if there is a possibility of the existence of economic deposits related to the BPP granites and pegmatites.

GEOLOGICAL BACKGROUND

The Borborema Pegmatite Province (BPP) extends along the eastern and southeastern part of the Seridó Belt (SB) in the middle of the Northern Tectonic Sub-Province (NTSP) in the Borborema Tectonic Province (BTP) as defined by Brito Neves & Fuck (2013). The BPP and NTSP are limited in the south by the Patos Lineament, a transcontinental shearzone correlated with the Garoua Lineament in Cameroun-Nigeria. The SB is composed of a Brasiliano (630 Ma, van Schmus *et al.* 2003) supracrustal sequence known as Seridó Group, that overlays a Paleoproterozoic basement dominated by orthogneisses and migmatites (Caicó Group) with some small Archean nuclei.

This basement dominates east- and westward of the SB. The Seridó Group (SG) is composed of basal gneisses, marbles, calc-silicate and amphibolites of the Jucurutu Formation, overlain by quartzites and metaconglomerates of the Equador

Formation and the topmost sillimanite – cordierite – garnet bearing biotite schist of the Seridó Formation. Biotite schists, quartzites and metaconglomerates of the SG are the host rocks of respectively 80% and 10% of the more than 750 known mineralized granitic pegmatites (Da Silva *et al.* 1995) of the BPP(encompassing from simple Be- to complex spodumene and lepidolite subtypes of the REL class-LCT family according to Černý's classification (Černý & Ercit 2005). The remaining

10% of the pegmatites are intruded in Neoproterozoic granites, Jucurutu gneisses or basement rocks (Fig. 1).

The granite types found in the SB were grouped into four main phases, G1 to G4, by Jardim de Sá *et al.* (1981). As summarized in Beurlen *et al.* (2014), only syn- to late- tectonic G3 group granites and late to post-tectonic G4 granites may be considered as possible sources of the mineralized pegmatites.

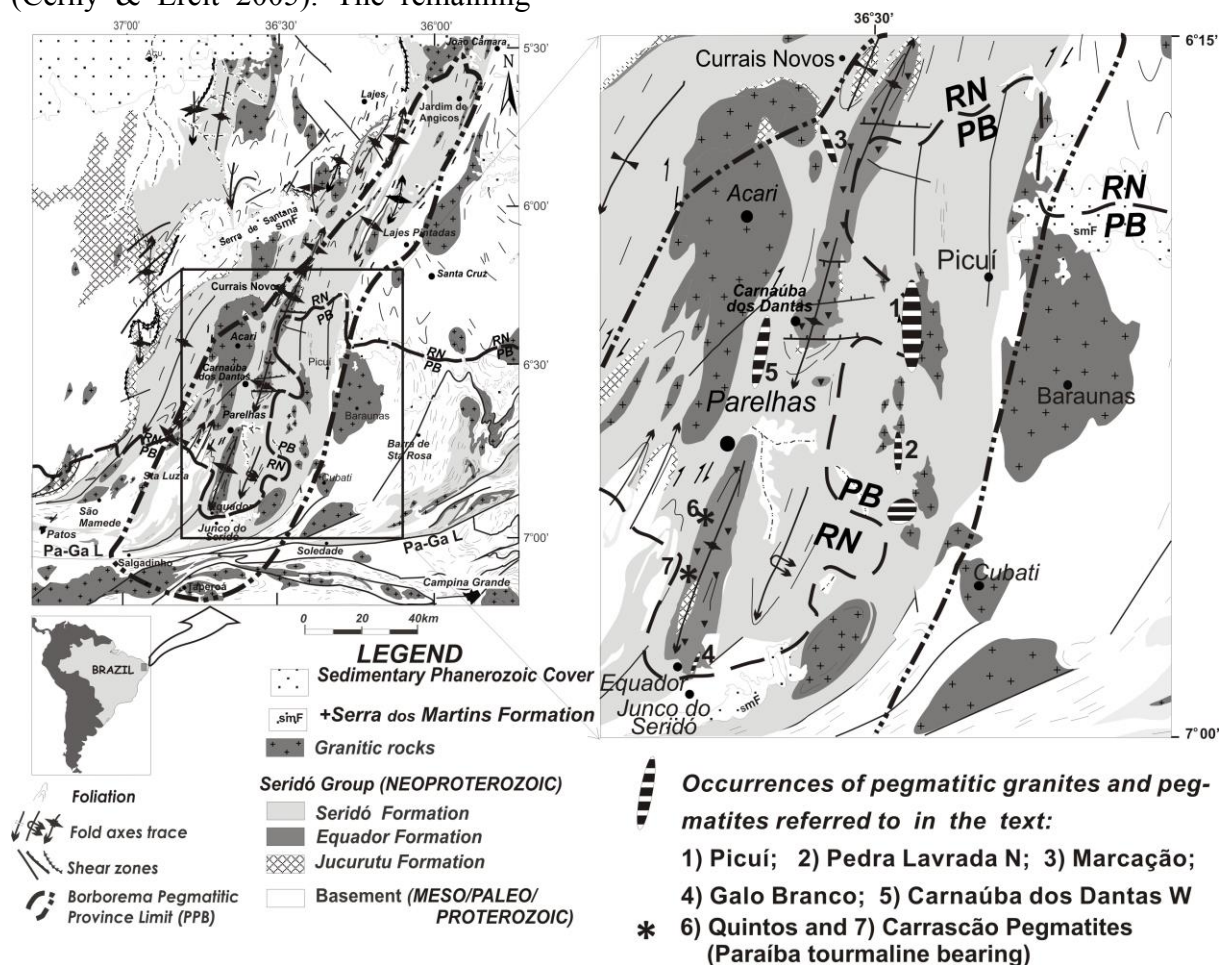


Figure 1 The Borborema Pegmatitic Province on a simplified geological base adapted from Brasil (1998, 2002), and the location of the studied pegmatitic granite intrusions and Quintos e Carrascão, “Paraíba Tourmaline” bearing pegmatites, modified from Beurlen *et al.* (2009).

G3 group granite types can probably also be excluded according to Jardim de Sá (1994) because ages of this group range around 580 ± 30 Ma,

typically related to the main stage of the Neoproterozoic Ediacaran(Brasiliano /Pan-African) Orogeny, much older than the wide range of the first pegmatite age

determinations between 450 and 540 Ma, as discussed by Beurlen *et al.* (2008 and references therein).

The SG and its up to Cambro-Ordovician granites and pegmatites in the BPP, are covered by an unfolded, supposedly early Cenozoic, 5 to 120m thick sequence of sandstones and conglomerates called Serra do Martins Formation (SMF). According to Morais Neto and Alkmin (2001), the SMF occurred along a regional plateau at 500 to 850 m OSL, formed during a Cenozoic period of uplift and, due to subsequent erosion, remains preserved today only as “mesetas”. At the unconformity (at around 750 m OSL in the area of the BPP), it is possible to observe a local lateritization in the covering sandstones and a very intensive kaolinization of the Neoproterozoic basement, including the pegmatites and granites.

PETROGRAPHY OF PEGMATITIC GRANITES

In the Picuí-Pedra Lavrada sub-area of the BPP, da Silva (1993) distinguished several small “pegmatitic-granite” intrusions identified as group GR3 (including a medium grained hololeucocratic facies GR3A and a K-feldspar megacryst bearing facies GR3B), as possible sources of Be-Li-Ta-bearing pegmatites. The occurrence of intrusions with “pegmatitic-granite” facies is also mentioned in the G 4 group granites of Jardim de Sá (1994).

A boom of exploration of these pegmatitic-granites in the early 2000's for ornamental dimension-stones exposed large walls in several quarries (e.g. Figs. 2, 3 and 4) and allowed to distinguish a layered anatomy with four cyclically repeated facies in these granites, namely: 1) fine to medium grained leucogranite; 2) layered usually sodic and garnet bearing aplite from now on shortly “aplite”, identical to the “line rock” as elsewhere described by Webber *et al.* (1997); 3) pegmatitic leucogranite with upward unidirectional growth of graphic K-feldspar/quartz mega-oikocrystals; 4) potassic pegmatite, with internal zoning and centripetal crystal growth, known as inward unidirectional solidification texture (UST). These facies are nearly identical to those described by Černý *et al.* (2005) in the Greer Lake leucogranite, considered to be the source granite of the Cat Lake pegmatite field with tantalum-rich lepidolite subtype pegmatites in Manitoba, Canada. A detailed description of the petrography, geochemistry, geological context and ages of these facies of five pegmatitic granites and related pegmatites of the BPP is available in Beurlen *et al.* (2009 and 2014). The petrographic investigation in these pegmatitic granites allowed verify that the highest modal content and homogeneous distribution of the potential REE bearing accessory minerals was the layered aplite facies.

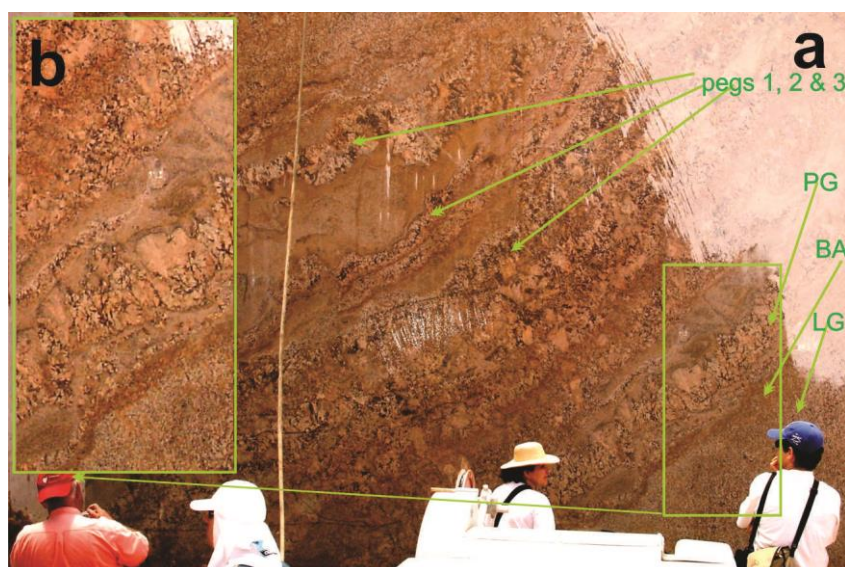


Figure 2 **a**) view of a 6 m high and 8m wide bench of the Marcação pegmatitic granite quarry. It shows decimetric to metric layering formed by the rhythmic alternance of four facies: 1) equigranular fine (to medium) grained leucogranite (LG 1); 2) millimeter sized rhythmic banded sodic aplite or “line rock” with garnet in the pink bands (BA 2); 3) pegmatitic leucogranite (unidirectional upward growth of mega-oicocrysts of graphic feldspar+quartz (PG 3); 4) pegmatite veins with centripetal graphic feldspar crystal growth (“pegs 4”); **b**) detailed insert of **a**; (modified from Beurlen et al. 2014).

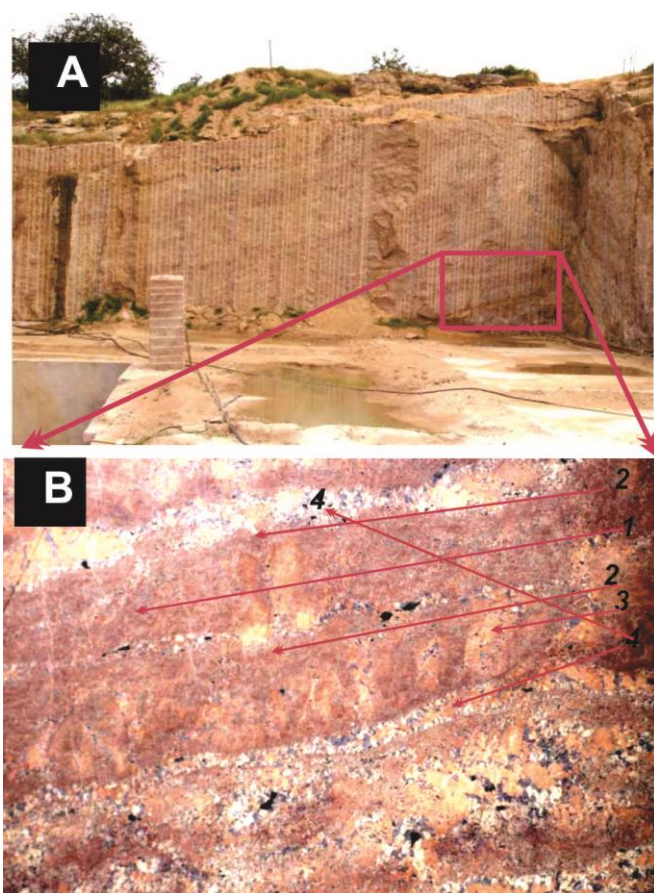


Figure 3**A**) View of a bench of the Picuí pegmatitic granite quarry with similar decimeter to meter sized layering of the four facies as described in Figure 2; **B**) detailed view of the different facies 1 to 4; modified from Beurlen et al. (2009).



Figure 4 **A)** Outcrop of the Pedra Lavrada N pegmatitic granite outcrop showing the layering of the “line rock” facies (2 – BA2), the equigranular facies (LG 1) and the megacryst facies (3). **B)** detail of A, showing a gradual coarsening of the grain size in the graphic quartz-feldspar megacryst.(PG 3). Modified from Beurlen et al. (2009). Modified from Beurlen et al. (2009).

MINERAL CHEMISTRY

Methodological aspects

Samples were collected of different facies from four pegmatitic-granite intrusions with quarries allowing a good systematic sampling of fresh samples and additionally one from a complex lepidolite-subtype pegmatite. After the petrographic characterization some representative polished thin sections were chosen for electron microprobe analyses of the accessory minerals with potential REE-enrichment. Used standards were of synthetic refined REE phosphates.

Routine chemical analyses and backscattered electron images were performed with the JEOL JXA-8500F (hyperprobe) at the GeoForschungsZentrum Potsdam (GFZ)

- Helmholtz Centre Potsdam, - Potsdam Germany. Operating conditions for quantitative WDS analyses involved an accelerating voltage of 15 kV, a beam current of 40 nA, a beam diameter of 5 μm , acquisition times of 100 to 300 seconds using five spectrometers LDE1 (F), TAB (Na, Mg, Al, Si, Y), LIF (Mn, Fe, REEs), PET (P, Cl, Ti, Ca, Pb, Th, U) and LDEB (Be, B). The counting times on the peaks were 20 s for Ca, Fe, Al, P, and Si, 30 s for Y, 50 s for the REEs, 100 s for Pb, Th, and U. Matrix corrections were performed according to the Armstrong-CITZAF method (Armstrong 1995).

Results

The REE analyses of accessory minerals from the different facies of these supposed parental pegmatitic

granites and pegmatites of the BPP (localization in Fig 1) allowed to recognize monazite and xenotime as main phases (with highest modal content) and with significant enrichments in these elements as summarized in Table 1. The complete dataset is available by request from the first author. Considering the more expensive REE, monazite is enriched in Nd and Eu (respectively with 13.06 and 0.54 wt. % REO) and xenotime is enriched in Dy, Yb, Gd, and Er (respectively 4.77, 3.39, 3.15, 3.86 wt. % REO) with a total of 18 wt. % of HREO, in addition to the other main formula components such as Ce, P, La in monazite and Y, P etc. in xenotime. Similar REE enrichment is known also from pegmatite provinces elsewhere, in their pegmatites and related secondary alluvial deposits. Zircon, thorite, uraninite showed no significant enrichment in REE. Other accessories may occur in still lower modal contents in the studied samples from the BPP.

The distribution of monazite and xenotime in the investigated samples is quite heterogeneous. The highest abundance of xenotime, with estimated modal contents of 0.005 to 0.01 wt. % was observed in the aplite facies ("line rocks") of the Picuí granite (50 to 100 composite zircon+xenotime crystals, 50 to 200 μm in size, were counted in several 22×42 mm large, polished thin sections). This aplite occurs in decimeter- to meter thick sheets repeated

in rhythmical alternation with layers of the other facies of the granite (Fig. 3). The xenotime in this aplite frequently occurs in a very conspicuous coaxial intergrowth with zircon, with compositionally co-variant oscillating crystal-growth zones, attesting the primary simultaneous magmatic crystallization of both (as shown in Fig. 5). Idiomorphic magnetite is another common accessory phase in the aplite and other facies of the Picuí granite.

The same rhythmical facies alternation is observed in the pegmatitic granite intrusions at Marcação (Fig 2), Galo Branco and Nova Palmeira-N (Fig. 4) (for details see Beurlen et al. 2009). In contrast to the Picuí granite, in this pegmatitic granite intrusion, monazite is the dominant REE-bearing accessory phase in the aplite facies instead of xenotime, and garnet (poor in REE) is the most common associated accessory mineral instead of magnetite. As example, in a representative thin section of the aplite facies from the Marcação pegmatitic granite, thirty grains of monazite and only four grains of xenotime, with enough size and polish quality for analyses were identified (sample HB 4201 in Table 1). Many monazite grains were found also in samples from the Nova Palmeira-N and Galo Branco pegmatitic granites and from the contact zones of the Carrascão and Quintos, Paraíba Tourmaline bearing pegmatites.



Figure 5 Polished thin section HB 460102B, of the banded sodic aplite from the Picuí pegmatitic-granite bench. Black dots are mostly xenotime and zircon and some magnetite. The fine alternance of pink and beige bands simulating the garnet bearing “line rocks” in this case is given by variation of proportions of feldspar and quartz. The pink colored bands are the result of very fine impregnation of limonite at the borders and fractures of well-rounded quartz grains, macroscopically possible to be suspected to be garnet. Beige bands are richer in feldspars and low hydroxide impregnations.

Xenotime and monazite of these pegmatites and of the various coarse-grained facies of the pegmatitic granites show nearly identical REE distributions (and enrichments) than those of the aplite facies in the pegmatitic granites as shown in Fig. 6. Zircon in all facies of the various pegmatitic granite intrusions and in the pegmatites did not show significant REE enrichment. Hafnium values of the zircon grains of both, aplites and border zones of pegmatites, were observed to be nearly the same.

Five whole rock REE analyses of two facies of pegmatitic granite intrusions in the BPP (GR3 A and B) are available in da Silva, (1993) with a range between 50 and 80 ppm total REE. Three, data are from a leucocratic fine to medium grained equigranular facies and the other two from an inequigranular facies with graphic quartz megacrysts. The chondrite normalized data are included in Fig. 7, for comparison with the whole-rock data from the source-rocks in a REL –

pegmatite field in Canada by Černý et al. (2005) and of the source-rock of the regolith-type deposit of Zudong (formerly known as ion-adsorption type) in China by Li et al. (2019). The estimated modal contents of xenotime and monazite and their ETR contents in the sodic aplite of the BPP, agree with whole-rock total REE contents around 100 ppm and confirm the analyses by Da Silva (1993). These values are lower but in the same order of magnitude of the data of the source intrusion of the Zudong deposit, between 300 and 500 ppm (Li et al. 2019). For comparison in Fig. 7 the distribution of the chondrite normalized REE data in xenotime, monazite and zircon in the BPP is also included. The data for xenotime and monazite are two orders of magnitude higher than those of zircon.

REE data obtained by da Silva (1993) of some other granite intrusions (of the G 2 and G 3 groups according to Jardim de Sá, 1999), within the extent of the BPP but without a genetic relation

with the pegmatites, may have even higher total REE contents (ranging between 100 and 300 ppm), but always with high chondrite normalized La/Lu slope and low HREE values. These granite types were not examined in this study for the REE distribution in their accessory minerals, but could also be

candidates for the formation of secondary ion-adsorption deposits if preserved at favorable paleogeographic conditions as already proven for the pegmatitic-granites (G 4, according to Jardim de Sá, 1994, or GR 3A and B, in Da Silva 1993) and the deeply kaolinized pegmatites.

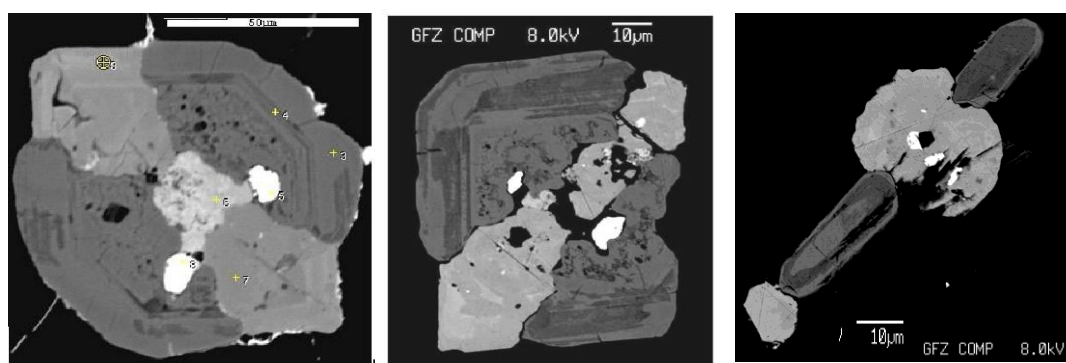


Figure 6. BSEI (Backscattered electron images) of composite zoned grains of zircon (darker gray) and xenotime (lighter gray), with small idiomorphic uraninite inclusions (almost white). Zircon and xenotime crystals in the two images at the left show oscillating clearly covariant compositional variations indicating a co-genetic primary-magmatic growth. In the core of the grain in the left a small grain of metamict thorite can be observed. White scale bar is 50 micrometers long. Ages obtained from both, the uraninite and xenotime are the same. Uranium contents in zircon are too high to allow dating on this mineral with the classical methods.

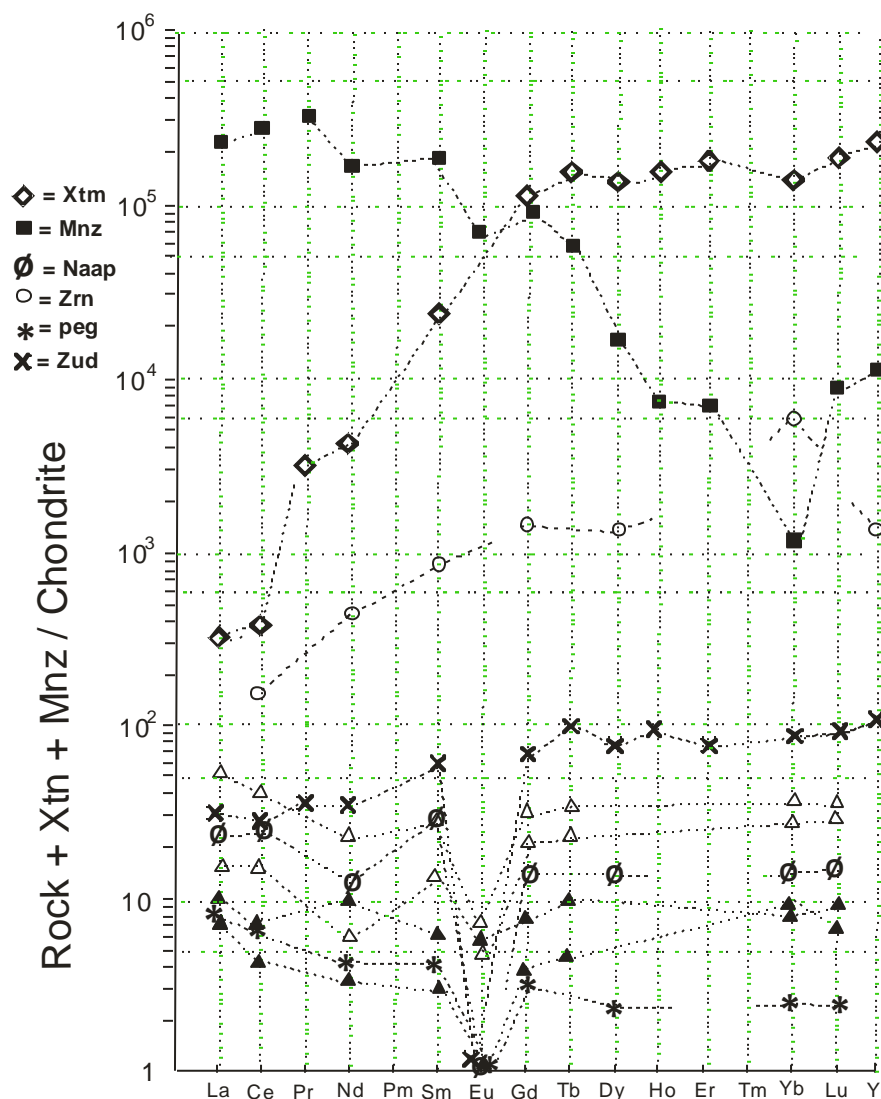


Figure.7 Chondrite-normalized REE distribution in xenotime (diamonds, Xtm) monazite (full squares, Mzt) and zircon (circles, Zrn) in comparison with whole rock G3a (medium grained leucogranite facies-empty triangles) and b (pegmatitic facies full triangles) by da Silva (1993) and sodic banded aplite facies (dashed circle, Naap) and pegmatitic facies (stars, peg) by Cerny et al. (2005b), and (x-crosses, zun) Zundong granite, Jiangxi-China (by Li et al 2019).

Table 1: Summary of representative REE analyses from the BPP, NE-Brazil.

	Mindata Type Xtm	HB460102 Xtm Mean (n=38)	HB460101 Xtm Mean (n=52)	HB460101 Xtm Std. Dev.	HB4201 Xtr HB4201 Mnz Mean (n=4)	HB4201 Mnz Mean (n=30)	HB460102 Zrn Mean (n=16)	HB460102 Zrn Std. Dev.
La₂O₃	–	0.013	0.014	0.021	0.004	9.184	–	–
Ce₂O₃	–	0.040	0.052	0.029	0.062	25.758	0.016	0.019
Pr₂O₃	–	0.037	0.028	0.032	0.049	3.323	–	–
Nd₂O₃	–	0.327	0.382	0.047	0.305	13.056	0.031	0.038
Sm₂O₃	0.49	0.629	0.644	0.054	0.596	4.284	0.022	0.036
Eu₂O₃	0.07	bdl	bdl	bdl	bdl	0.537	–	–
Gd₂O₃	2.69	3.252	3.169	0.189	3.182	2.797	0.045	0.038
Tb₂O₃	0.58	0.677	0.647	0.069	0.815	0.243	–	–
Dy₂O₃	4.93	4.935	4.714	0.366	5.783	0.658	0.053	0.073
Ho₂O₃	1.27	1.005	0.963	0.083	0.863	0.044	–	–
Er₂O₃	4.05	3.978	3.908	0.218	2.920	0.144	–	–
Yb₂O₃	4.36	3.473	4.058	0.236	3.190	0.027	0.125	0.091
Lu₂O₃	0.87	0.790	0.904	0.057	0.764	0.037	–	–
Y₂O₃	46.49	44.204	44.282	1.156	45.498	2.137	0.227	0.320
P₂O₅	34.37	31.120	31.858	0.907	32.635	27.592	0.230	0.186
SiO₂	0.31	1.297	1.314	0.520	1.468	1.809	32.523	0.809
ThO₂	–	0.857	1.049	0.706	1.057	6.540	0.036	0.040
UO₂	–	0.954	1.277	0.508	1.473	0.342	0.308	0.187
PbO	–	0.090	0.113	0.051	0.137	0.186	0.009	0.013
CaO	–	0.050	0.040	0.047	0.040	0.816	0.084	0.140
Al₂O₃	–	0.000	0.000	0.000	0.000	0.190	0.138	0.127
FeO	–	0.202	0.140	0.271	0.401	0.089	0.274	0.214
ZrO₂	–	–	–	–	–	–	63.716	1.156
HfO₂	–	–	–	–	–	–	2.481	0.251
TiO₂	–	–	–	–	–	–	0.007	0.009
Sc₂O₃	–	–	–	–	–	–	0.022	0.012
Tm₂O₃	0.70	–	–	–	–	–	–	–
Total	101.18	97.932	99.556	0.974	101.240	99.795	100.347	1.302

. Abbreviations: Xtm = xenotime, Mnz=monazite, Zrn=zircon. Columns: **Mindata** refers to a single analysis of a typical pegmatite Xnt from Norway; **HB460102** refers to analyses of a sample of the aplite facies in the Picuí pegmatitic granite; **HB460101** another sample of the same aplite facies. **HB4201** refers respectively to Xtm and Mnz results of a polished thin section of the aplite facies from the Marcação granitic-pegmatite; last column HB460102 are Zrn data of the same section in column 2. Hyphens = not analysed.

DISCUSSION AND CONCLUSIONS

The analytical results confirmed that xenotime and monazite in aplites and pegmatites of the BPP are the only mineral groups with significant enrichment in highly prized REE (Fig. 7), in proportions similar to that observed in other pegmatite provinces or alkaline granites. Both minerals however occur in low mean modal concentrations in these rocks (around 0.01 % or less) even in both, the enriched aplitic facies

of the parental granites and the border zones of the complex pegmatites. The REE concentrations are far below the mining grade for hard-rock exploitation. There is however a good chance of secondary REE deposits as: **a)** alluvial placer deposits rich in xenotime as those reported by Sengupta & van Gosen (2015) from some Southeast-Asian countries; and **b)** ion-adsorption (or regolith) type deposits as reported by Voncken (2016) and Li et al. (2019) from China. Both deposit types could be

expected within or close to the extent of the BPP. Type **b**) occurrences could be related to clay- and iron-hydroxide-rich alteration along the unconformity between the Proterozoic BPP granites and the plateau formed by an early Cenozoic siliciclastic couverture (Serra dos Martins Formation), later uplifted and eroded during the younger Cenozoic, and today preserved only as “mesetas” (Moraes Neto & Alkmin 2001). Coastal heavy mineral Ti-deposits exploited 250 km eastward of the BPP, occur close to the mouth of rivers draining the BPP and older basement. Probably due to the large distance from the BPP, they contain only insignificant contents of monazite and no xenotime.

Acknowledgements

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