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## Classification of rare earths mineral resources using the Jequié/BA complex rock weathering chemical index

### Classificação de recursos minerais de terras raras utilizando o índice químico de intemperismo de rochas do complexo Jequié/BA

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### ABSTRACT

The most common economic mineral deposits of Rare Earths are associated to magmatic calc-alkaline to alkaline rocks and their products. This work presents data from drilling carried out in rocks of the Jequié/BA Complex, mineralized in rare earths, which were subjected to an intense weathering process, similar to what occurred in the Ion Adsorption Clays deposits in southern China. The interpretation of the survey data allowed the classification of rare earths mineral resources with similar, sometimes higher, levels than those of Chinese deposits, contained in the lateritic regolithic profile studied. The chemical index of rock alteration (CIA) was adopted as a classification tool, horizontally separating the mineralized weather profile into six zones. The data indicate that there was supergenic enrichment REO in the lower portions of the profile and sometimes in the upper zones. The average levels ranged between 0.09% and 0.22% REO, with maximum values of 1.6%. A model section representing the deposit of rare earths minerals of supergenic origin in rocks of the Jequié/BA Complex was developed.

**Keywords:** Rare earths oxides; Classification of mineral resources; Jequié/BA Complex; Ion adsorption clays.

## RESUMO

Os depósitos minerais econômicos de Terras Raras mais comuns estão associados a rochas magmáticas calcálicas a alcalinas e seus produtos. Neste trabalho são apresentados dados de sondagem realizada em rochas do Complexo Jequié/BA, mineralizadas em terras raras, que foram submetidas a intenso processo de intemperismo, semelhante ao ocorrido nos depósitos do tipo argilas de adsorção atômica (ou ion adsorption clays) no sul da China. A interpretação dos dados de sondagem permitiu a classificação os recursos minerais de terras raras com teores semelhantes, por vezes superiores, aos dos depósitos chineses, contidos no perfil regolítico laterítico estudado. O índice químico de alteração de rocha (CIA) foi adotado como ferramenta de classificação, separando horizontalmente o perfil intempérico mineralizado em seis zonas. Os dados indicam que houve enriquecimento supergênico dos OTR nas porções mais inferiores do perfil e por vezes nas zonas superiores. Os teores médios variaram entre 0,09% e 0,22% de OTR, com valores máximos de 1,6%. Uma seção modelo representando o depósito de mineral de terras raras de origem supergênica em rochas do Complexo Jequié/BA foi desenvolvida.

**Palavras-chave:** Óxidos de terras raras; Classificação de recursos minerais; Complexo Jequié/BA; Depósitos do tipo argilas de adsorção atômica.

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## INTRODUCTION

Rare Earth Elements, or REE, can be found in several types of crustal rocks and are contained mainly in minerals most commonly found in the form of carbonates, fluorocarbons, phosphates, silicates, fluorides and oxides or even adsorbed on neoformed clays, in this case sometimes forming the so-called Ion Adsorption Clays (IAC) deposits (MARIANO & MARIANO Jr., 2012). The main geological processes associated with REE are carbonate magmas, CO<sub>2</sub>-rich alkaline magmas, and carbonate complexes. Carbonatite magmas are commonly associated with plutonic complexes, which form plugs, cones, sheets, dikes and more rarely sills (WARNER & BARKER, 1989). They are generated in the lithospheric mantle by ascension and positioning giving rise to primary carbonatites (volcanic or plutonic) (PERHAC, 1970). Alkaline magmas rich in CO<sub>2</sub> are generated in the lithospheric mantle by ascension, emplacement and differentiation in a magmatic chamber, generating alkaline-carbonatite complexes (dunites, pyroxenites, bebedourites, ijolites, syenites, foscrites, carbonatites), or by extrusion, generating alkaline volcanism (nephelinites, kamafugites, phonolites, lamprophyres, melilitites) (WOOLEY & KJARSGAARD, 2008). Carbonatite complexes are generated from the complex evolution of magmas, through recurrent, sometimes simultaneous processes, such as fractional crystallization, immiscibility of liquids, degassing and/or metasomatism and crustal assimilation (BARKER, 1989).

The main processes that form REE deposits are igneous (magmatic + hydrothermal), associated with syenites and granites, sedimentary, secondary and/or supergenic and hydrothermal, associated or not with alkaline and calcialcaline rocks.

In Brazil, REE, lithium and silicon make up a group of substances considered strategic for the country by the federal government, dubbed "substances that carry the future". These chemical elements are the basis for the technological development of the high-tech industry and, together with so-called agromineral, integrate a group of priority substances in the Science, Technology and Innovation Plan for Strategic Minerals 2018-2022.

Chemical rock weathering indexes are widely used for characterization of weathering rock profiles. They are based on the combination of percentage ratios of the largest chemical elements contained in the rocks and that are considered more mobile in a natural leaching system to which the rocks (protoliths) were subjected over geological time during the pedogenetic process (PRICE & VELBEL, 2000). Considering the main chemical weathering indexes evaluated by Price & Velbel (2000) regarding their applicability in the evaluation of weathering profiles of rocks, we chose to use the Chemical Index of Alteration (CIA), developed by Nesbitt & Young (1982). This index has been widely used for geochemical evaluations of paleosols (SUTTON, RITGER & MAYNARD, 1989, SUTTON & MAYNARD, 1992, SUTTON & MAYNARD, 1993). It is the most important chemical index currently used to aid interpretations related to the weathering profiles of mineralized rocks in REE (WU, HUANG & GUO, 1990), especially in

REE deposits of IAC type, as is the case in the Muzishan, Keshutang e Getengzui deposits, in southern Jiangxi, China (ZHAO et al., 2017), the Jiangxi, Guangdong, Guangxi and Yunnan deposits in southern China (WANG et al., 2018), the Minaçú/GO deposits in central Brazil (ROCHA et al., 2013) and the southeast São Paulo state deposits in southeastern Brazil (FARIA, 2018). According to Zhao et al. (2017), the study of weathered layers and/or horizons mineralized in REE with the aid of the CIA has been shown to be an efficient tool in the characterization of potential targets to contain IAC-type mineralizations. However, the CIA should be used integrated with the geological knowledge of the area under assessment and should be treated in conjunction with other also important physicochemical parameters to improve the use of this geochemical resource as a prospective tool. The author also points out that studies carried out on granitic rocks from southern China from weathered and mineralized rock samples, when they present CIA values lower than 85%, the degree of weathering is positive for correlation with the REE content of the IAC type, and in the case of  $CIA > 85\% < 100\%$ , the degree of weathering has a negative correlation with the REE content of the IAC type.

Fernandes et al. (2019) proposed a new geological and stratigraphic classification for the Jequié complex rocks that occur in the municipalities of Jequié, Jitaúna and Itagi in southeastern Bahia. However, data on the mineralization of Rare Earths in the Jequié Complex, located in the southeastern region of Bahia, Brazil, are still scarce in the literature. This work presents a vertical classification of the mineralized weathered profile in REE from rocks in said complex through the analysis of the distribution and vertical geochemical behavior of CIA and its relations with the observed mineralogy and with the REE contents in the ore.

## METHODOLOGY

A mineral exploration campaign was carried out in the region between Jequié and Jitaúna, in southeastern Bahia. The work was conducted with the purpose of seeking a mineral deposit with economic potential of REE for subsequent evaluation of resources and reserves. Geological mapping, surface geochemistry (soil and rock), exploratory drilling, terrestrial gamma spectrometric survey, mineralogical and technological characterization tests, among others, were carried out. The holes drilled had an average depth of 9.12 meters, with a maximum depth of 26 meters. A total of 450 boreholes were drilled by mechanical auger in an area of approximately 2,800 hectares. All holes were vertical and performed on an exploratory basis on the weathering profile of the mineralized rocks in REE, with sample collection at each meter drilled. The holes chosen for analysis were those that crossed at least three horizons of the weather profile, namely: horizon A (residual and/or colluvial soil), horizon B (mottled zone) and horizon C

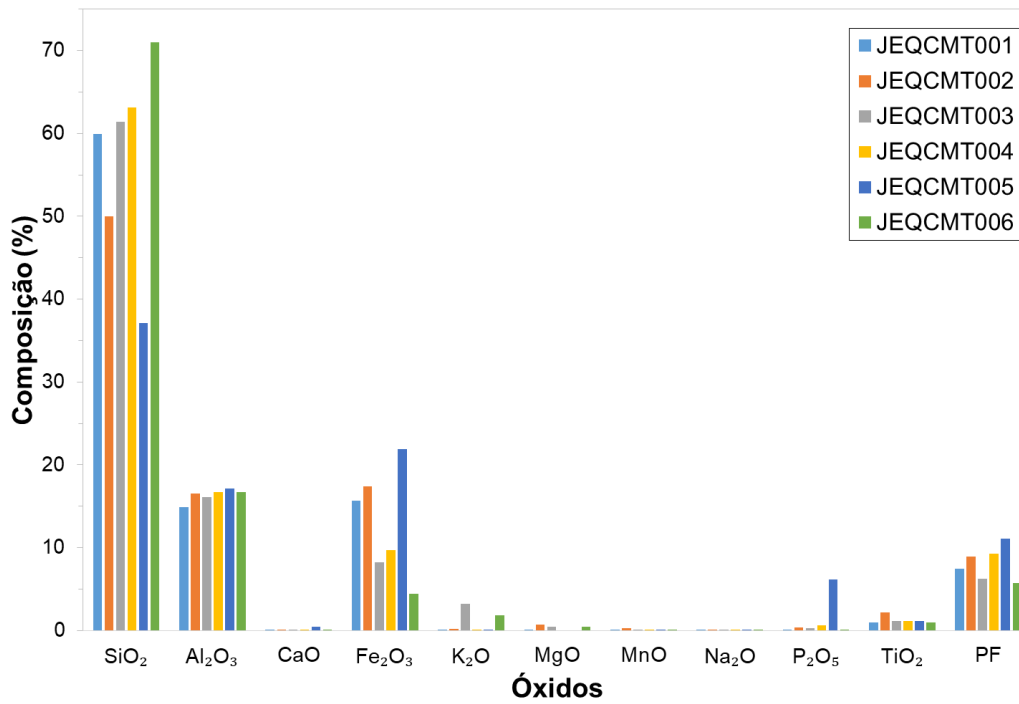
(saprolite + saprock), so that in each hole it was possible to obtain information from all important weather horizons for data analysis and also so that the results obtained with the analysis of these horizons had the best possible horizontal representativeness.

Of the 450 holes drilled, 103 were chosen for this work, corresponding to 953 samples. Six samples belonging to the mottled and saprolytic horizons were subjected to mineralogical characterization studies by scanning electron microscopy with coupled imaging system (MLA) and by X-ray diffraction in the Technological Characterization Laboratory (LCT). Samples were identified with codes JEQCMT001 to JEQCMT006. The chemical composition of the samples was determined by plasma emission spectroscopy (ICP-OES and MS) at SGS Geosol Laboratórios Ltda, considering the main oxides of interest (major elements, rare earths,  $\text{ThO}_2$  and  $\text{U}_3\text{O}_8$ ).

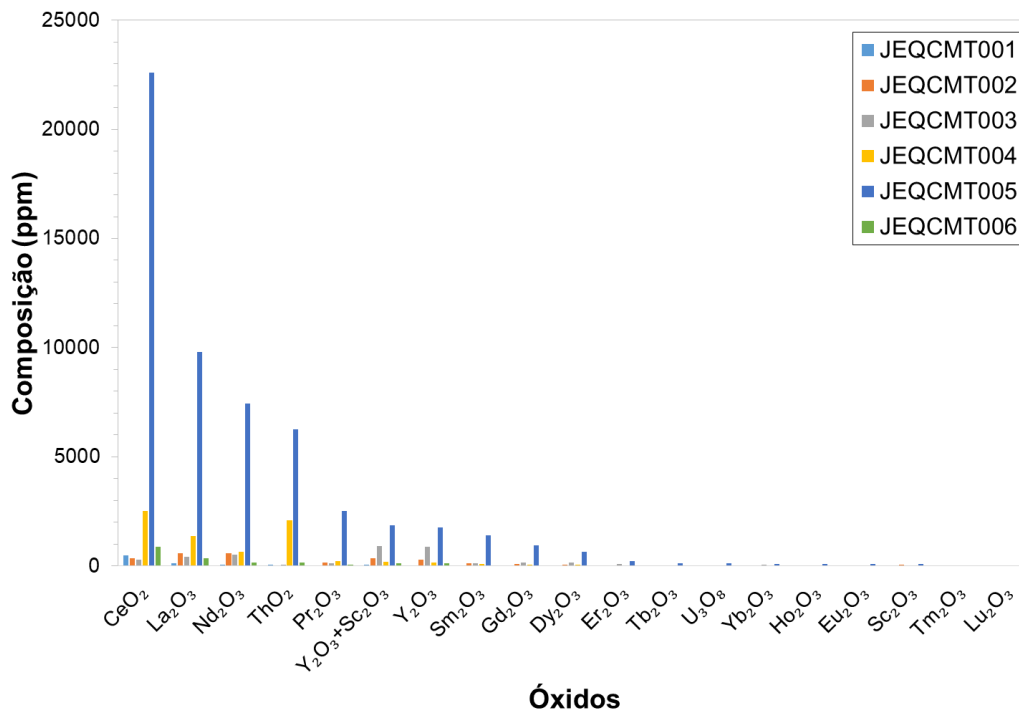
## RESULTS AND DISCUSSION

Figure 1 shows the results of the chemical composition by ICP of the six samples considered. Among the larger constituents, the samples present significant contents of  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ , in varying proportions. The  $\text{SiO}_2$  content ranges from 37.1% (sample 5) to 71.0% (sample 6), that of  $\text{Al}_2\text{O}_3$  is between 14.9 and 17.1%, while that of  $\text{Fe}_2\text{O}_3$  ranges from 4.40 (sample 6) to 21.9% (sample 5). Sample 3 also has a high  $\text{K}_2\text{O}$  content of 3.24% (compared to 0.04 to 1.86% in the other samples), and sample 5 has a 6.15%  $\text{P}_2\text{O}_5$  content (between 0.09 and 0.62% in the other samples). The REO content (sum of rare earths, from  $\text{CeO}_2$  to  $\text{Lu}_2\text{O}_3$ ) varied significantly in the samples studied, from 699 ppm in sample 1, increasing to values between 1,567 and 5,009 ppm in samples 2, 3, 4 and 6 and reaching 46,012 ppm in sample 5. Similar behavior was observed for  $\text{Y}_2\text{O}_3$ , higher in sample 5 (1,766 ppm).  $\text{Sc}_2\text{O}_3$ ,  $\text{ThO}_2$  and  $\text{U}_3\text{O}_8$  contents were generally less than 100 ppm, with the exception of  $\text{ThO}_2$  in sample 4 (2,086 ppm) and  $\text{ThO}_2$  and  $\text{U}_3\text{O}_8$  in sample 5 (6,246 and 128 ppm, respectively).

**Figure 1** – Chemical composition of the samples.



(a)



(b)

Figure 2 shows the results obtained by MLA. In mineralogical terms, the samples consisted basically of the same minerals, varying only in relative proportion between them. The rare earth-bearing mineral in all samples was monazite, corresponding to 0.1% by mass in samples 2 and 3, 0.2% in samples 4 and 6, and a maximum of 1.2% in sample 5, and less than 0.1% in sample 1. Cerianite and xenotime were also identified, but do not exceed 0.1% (few grains identified).

**Figure 2** – Mineral phases present in the samples determined by MLA.

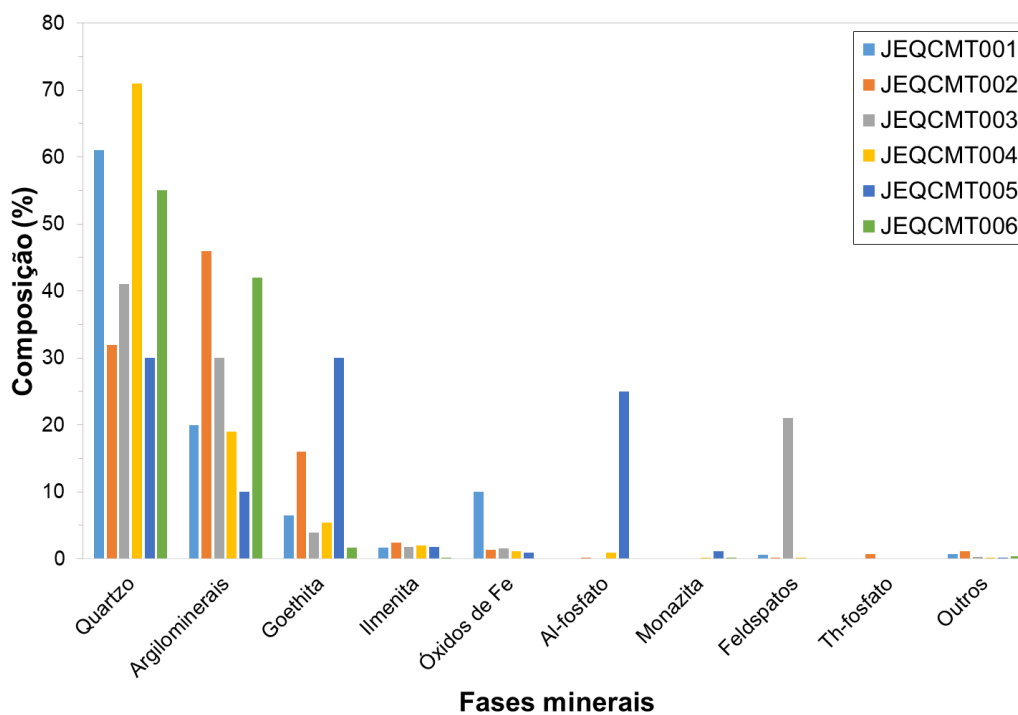
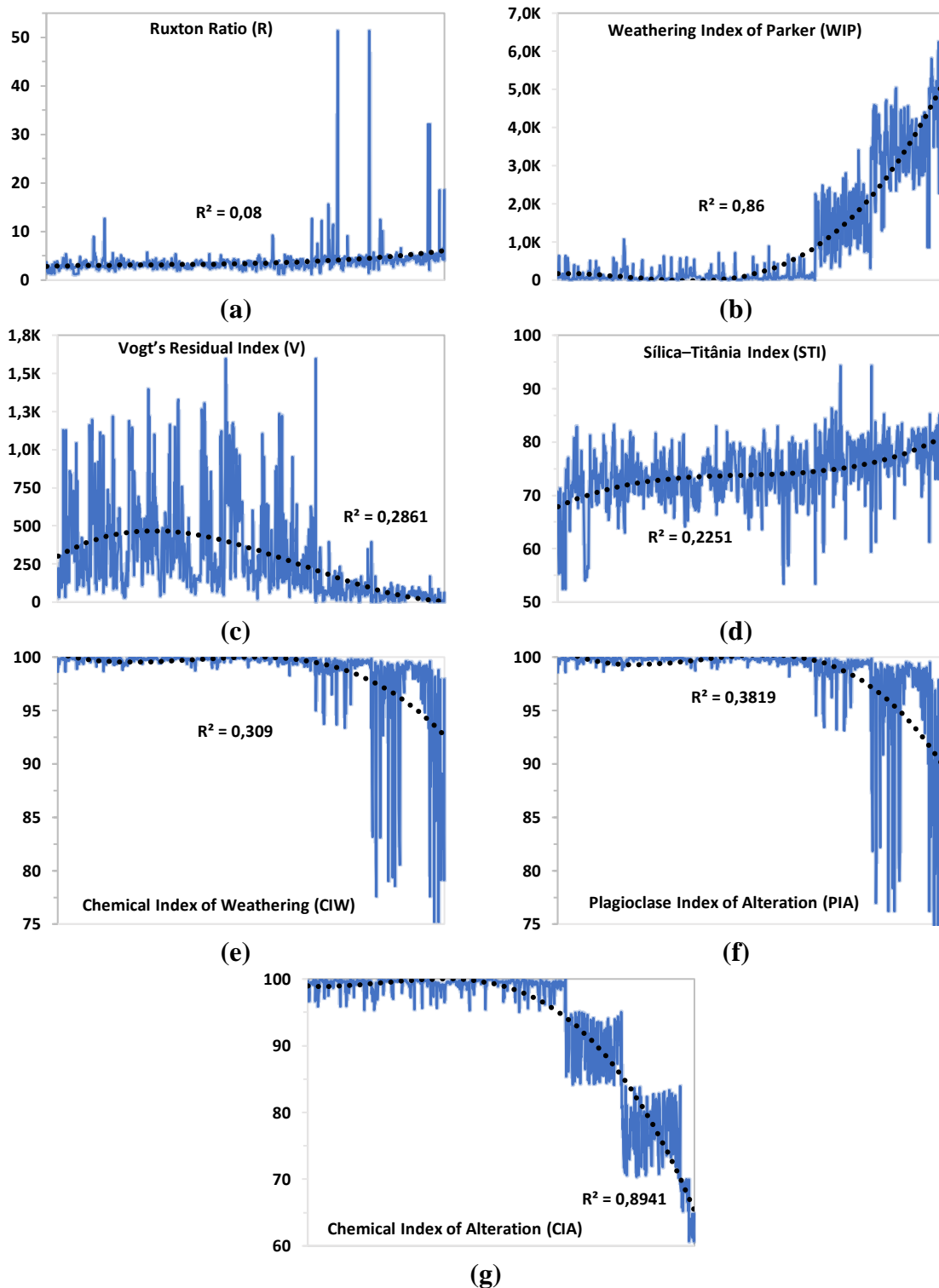


Figure 3 shows the distribution of the rock chemical weathering indexes as well as the geochemical behavior of the 103 rig holes chosen, and the data of the 953 samples generated from these holes. The indexes were plotted according to each composition of the main chemical weathering indexes defined by Price & Velbel (2000). In addition to the CIA, the R, WIP, V, STI, CIW, and PIA indexes were calculated, since they could be useful in the classification of REE resources from the type of environment studied.

**Figure 3** – Calculated rock weathering chemical indexes for each of the 953 samples: (a) R, (b) WIP, (c) V, (d) STI, (e) CIW, (f) PIA, and (g) CIA. Polynomial trend curves of 3rd order.



One of the main assumptions for the correct choice of a rock chemical weathering index is the (positive or negative) uniform or gradual tendency towards increasing the depth of the profile, consequently towards greater proximity to the fresh rock (SUTTON & MAYNARD, 1992). Thus, the curves were adjusted in search for the one with the best



$R^2$  correlation, opting for a third-degree interpolator polynomial. Apart from the CIA and WIP indices, which presented  $R^2$  values above 0.85, all other index obtained  $R^2$  correlation below 0.4, that is, a weak correlation.

Comparing the composition of the chemical formulas of the WIP and CIA weathering indexes, the absence of  $Al_2O_3$  sesquioxide in the WIP was observed. This compound is fundamental for the analysis of weathering rock profiles in situations of intense natural leaching, when bauxitization and/or laterization tend to occur, as is the case of the weathering profile analyzed in this study. Therefore, the applicability of WIP in this study is not recommended (ESWARAN, STOOPS & DE PAEPE, 1973). After performing a comparative analysis between chemical indexes based on treatment of data obtained by the surveys, and after alignment with current concepts on the use of the CIA as an auxiliary tool for understanding REE deposits of the IAC type as described by Sanematsu & Watanabe (2016), Zhao et al. (2015), Zhao et al. (2017a), and Zhao et al. (2017b). It was found that this is the most appropriate chemical index to classify potential REE resources contained in the weathering profile of rocks belonging to the Jequié Complex, southeast of Bahia, Brazil.

The geochemical classification used to represent the different compartments and/or lithochemical horizons was established from the analysis of the vertical distribution of data obtained with the boreholes carried out in the mineralized weathered profile in REE, as can be seen in figure 4. Notably, it is observed that there is a grouping of data in populations which demonstrate geochemical similarity to the CIA measurement, maintaining the uniform trend of decreasing CIA values with increasing depth and/or toward greater proximity to fresh rock (lower limit of the weathering profile). From this interpretation, the CIA intervals were defined to represent the horizontal compartmentalization of the mineralized weathered profile in REE.

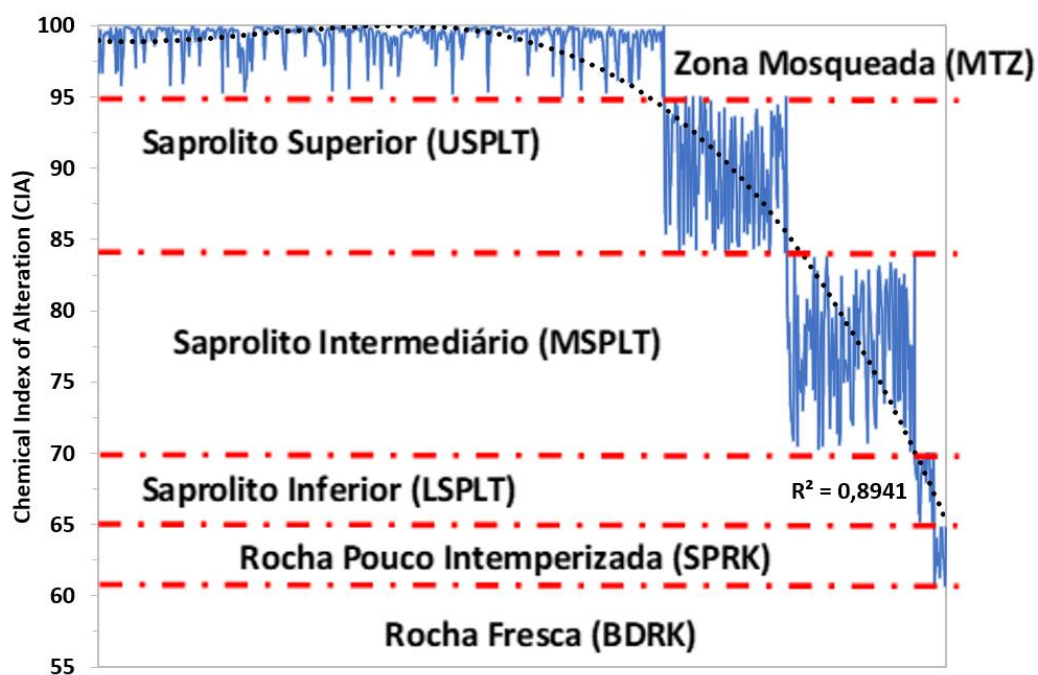
The mineralized weathered profile can be compartmentalized horizontally by CIA intervals called lithochemical horizons into 3 large groups:

- i. Mottled Zone (MTZ): most leached zone of the alteration profile with CIA values  $\geq 95$ ;
- ii. Saprolitic zone: zone of lower weathering in relation to MTZ, with values of  $61 \leq CIA < 95$ ;
- iii. Fresh rock: with CIA values less than 61%.

The saprolite zone can be further subdivided into upper saprolite (USPLT) with  $84 \leq \text{CIA} < 95$ , intermediate saprolite (MSPLT) with  $70 \leq \text{CIA} < 84$ , lower saprolite (LSPLT) with  $65 \leq \text{CIA} < 70$  and the saprock ( $61 \leq \text{CIA} < 65$ ), consisting of a transition zone between the saprolite and the fresh rock, where minerals are only partially weathered.

In addition to the tendency of the CIA to decrease towards the base of the profile (fresh rock), the data are grouped into distinct populations in different horizons of the weathering profile. Thus, these populations deserve to be individualized for a better understanding of this tendency of geochemical grouping and how relevant this is in the study of vertical variability of REE, Th and U elements.

**Figure 4** – Grouping of distinct geochemical populations according to a trend defined by the composition of the CIA in each meter of drilling carried out for the 103 holes drilled (equivalent to 953 meters drilled).



Although Vogel (1975) and Harnois & Moore (1988) mentioned that weathering rates that use iron in their composition are not suitable for analysis of regolithic profiles, as is the case here, the weathering horizons which presented  $\text{CIA} > 95$  and  $\text{Fe}_2\text{O}_3 \geq 15\%$  were called Mottled Zone Laterized (MTZL). These horizons had a higher  $\text{P}_2\text{O}_5$  average than the other litho-geochemical horizons, which may characterize a greater presence of phosphate rare earth-bearing minerals in these horizons. However, this relationship with the laterization process may be masked by the metamorphic character of the context

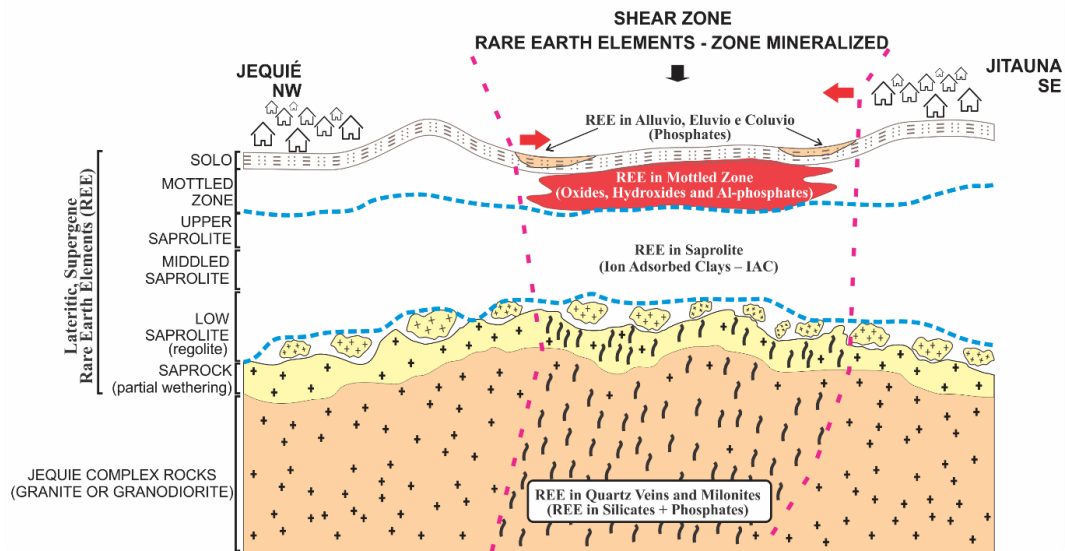
surrounding the Jequié Complex rocks (FERNANDES et al., ) and the enrichment of  $\text{Fe}_2\text{O}_3$ ,  $\text{P}_2\text{O}_5$  and REE, in this case, may be associated with enrichment processes of primary origin and would not necessarily be attributed in their entirety to supergenic processes (exogenous processes).

In general, when observing the mean REO values in the different lithogeochemical compartments presented, it is observed that there is an abrupt enrichment of REE in the MTZL lithogeochemical horizon, reaching average contents of 0.13%, followed by depletion in the MTZ where  $\text{Fe}_2\text{O}_3$  is not greater than 15%. When the average concentrations of all lithogeochemical horizons are observed, the enrichment of REO of the weathering profile toward lower layers becomes evident, ranging from 0.09% to 0.22% of REO in the SPRK, lithogeochemical horizon where the rock is poorly weathered.

Notably, the lithogeochemical horizon that presented the sample with the highest REE content was the MTZ horizon, which reached 1.6% REE. Observing the maximum REO values found between lithogeochemical horizons, there was variation from 0.3% to 1.6%. Another important point was the concentration of  $\text{P}_2\text{O}_5$  in the samples represented by the MTZL horizon, highly leached and with  $\text{Fe}_2\text{O}_3 > 15\%$ .  $\text{P}_2\text{O}_5$  contents were significantly higher in relation to the other weathering chemical horizons. This phenomenon may be associated with laterization processes and/or hydrothermal processes that deserve to be further investigated in the future, as this may be an important characteristic for evaluating mineral processing routes related to the greater or lesser amount of phosphate minerals in the ore matrix (minerals in grains and not in the form of REE ions adsorbed on clays).

Based on the analysis carried out in the different compartments of the mineralized weathered profile in REE defined in this study, the lithogeochemical horizons established through CIA ranges integrated with the geological information presented, a typical section of the supergenic REE deposit in the rocks of the Jequié Complex, southeastern Bahia, was elaborated. Figure 6 shows the proposed section.

**Figure 5** – Typical section of the supergenic REE deposit in rocks of the Jequié/BA Complex according to the lithogeochemical classification defined in this work, integrated with field geological data.



## CONCLUSIONS

The found results demonstrate that there is a group of rocks not yet well defined in literature inserted in the geological context of the rocks of the Jequié Complex, southeastern of Bahia, Brazil, which presents REE concentrations compatible with those of other deposits known worldwide as potentially economic in REE, revealing an economic potential in exploring the region, still unknown for this type of deposit model. Although the studies carried out so far do not allow us to clearly establish the genesis of the primary origin of REE mineralization present in this region, it was possible to verify at least four different types of important REE metallotects. They are:

- i. REE in quartz and mylonite veins (REE silicates and phosphates);
- ii. REE in alluvium, eluvium and colluvium (REE phosphates);
- iii. REE in mottled zones (REE oxides, hydroxides and Al-phosphates);
- iv. REE in saprolite (REE Ion Adsorbed Clays – IAC).

The work and field suggest that these rocks underwent an extensive metamorphism process predominantly in granulite facies. This event may have caused remobilization of REE contained in these rocks and their reconcentration through recrystallization of some rare earth-bearing minerals, giving rise to ore shoots (high REE content zones). It is also a conceptual model that lacks works of greater detail linked to a deeper study of the complex geochronology of the rocks of this region, in order to identify

and more assertively associate this succession of geological events with these occurrences. Therefore, the model suggested here can be considered in concept still.

The data from the survey allowed the establishment of important geochemical relationships regarding the use of the rock alteration index (CIA) to characterize litho-geochemical horizons, making it possible to establish relationships of vertical variability of REE concentration and other elements.

Through the use of this classification tool integrated with particular geological knowledge of each mineral deposit with these characteristics, it is expected that this study contributes to the development of future prospective works in the region, in other REE mineral deposits of supergenic origin, and also to research in general, in view of the complexity of scientific challenges surrounding the subject.

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