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GEOCHEMICAL ASPECT OF CAAPUCU HEIGHT AT SOUTHEASTER PARAGUAY BY X-RAY FLUORESCENCE AND NEUTRON ACTIVATION ANALYSIS

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Abstract. *The Precambrian in eastern Paraguay is present mainly in two areas in the north and in the south; at the Apa High in the former and at the Caapucu Height in the latter which is constituted by Rio Tebicuary complex; to the north of this river is exposed the Caapucu Hight (Jaguarete Kua plug). Recently, geochemical studies were performed in the Precambrian plug of Fuerte San Carlos, near the Apa River providing analytical data in this regard. In this work hand specimen of granitoid rocks from the southern Jaguarete kua outcrops are studied in some of their major, minor and trace elements aiming to look for relationships with the northern outcrops and their provenance as well as the granitoid type. The analyzed elements were Na, Al, Si, K, Ca, Ti, Mn, Fe, Cu, Zn, Ga, Pb, Rb, Sr, Th, Y, Zr, Nb, Ba, La, Ce, Pr, Nd. Analyses were carried out by EDXRF except for sodium that was analyzed by Neutron Activation. The XRF experiments were carried out using the facility of Josef Stefan Institute in Ljubljana, whereas NAA was done with an Am- Be neutron source with a flux of 5 x10*^{7} *n s*$^{-1}$ *at the Facultad de Química (UNA), Paraguay. The results of analysis allow to establish inter alia indexes, ratios which are related with crystallization, granitoid type etc. The spidergrams standardized to primordial mantle of refractory elements content, show an enrichment of incompatible elements in the samples. Besides, they resemble to those found in Precambrian (Neoproterozoic) outcrops from the northern area (eastern and the Paraguayan Chaco), as well as from Brazil.*

Keywords: *granitoids, spidergrams, Precambrian Neoproterozoic, Continental shelf, Eastern Paraguay, outcrops, plug*

1. Introduction

1.1. Granites and granitoids

Granites and granitoids constitute the most relevant and extended rocks on the continental shelf of the upper crust; they form the crystalline basement of the Precambrian.

These rocks originate by fusion at temperature equal to and greater than 1000ºC, at a given pressure (depth depending) from hydrous sediments from the erosion of said crust. Like any magmatic rock, their chemical/geochemical constitution depends on the source rocks. They therefore show a great variety of chemical elements; however, the variability can be narrow due to the interaction liquid/melt/mineral especially at eutectic points (irreversible processes, precipitation, dissolution solid state reactions, etc.), concurrent into metasomatism phenomena [1-6].

In Eastern Paraguay, the Precambrian is present mainly in two structural highs: the Apa High in the north and Caapucu Hight in the south [7-10]. The outcrops in the former, distributed along the Apa River, belong to the Brazilian Shield. The latter is constituted by Rio Tebicuary complex in which is distinguished the north and the south developments [7,8]; to the north of that river is exposed the Caapucu Group (See Maps). It should also be mentioned that there are also small glimpses in San Bernardino and between Cerro León and Escobar [8].

The North zone includes the area that extends west to east from Puerto Fonciere to Caracol; south forward direction it continues until about 2 km from Villa Sana. The first to study this area was Prof. R. Boettner [11] who identifies a Formation of metamorphic rocks partially covered by Quaternary sediments and constituted of gneisses, micaceous schists, bended phyllites and quartzites, outcropping from San Luis de la Sierra to Ferreira Cué. This metamormic formation borders another one of granite, which develops toward west. Near Caracol and Puentecinho the metamorphic rocks are frequently crossed by veins of quartz and pegmatite bodies, sometimes with tourmaline and beryl. Towards the east of the metamorphic formation, the granitic rocks appear again, a good one of them can be seen on the banks of the Quién Sabe stream [8,11].

In regard to the South of Easter Paraguay, details will be given in the next section.

The present work studies selected major, minor and trace elements of a massif that crops out near Caapucu in the southern region as follows: Si, Al, Na, K, Ca, Ti, Mn, Fe, Cu, Zn, Ga, Rb, Sr, Y, Zr, Nb, Cs, Ba, La, Ce, Pr, Nd, Th, Pb, all of them were analyzed by XRF techniques except Na which was studied by neutron activation.

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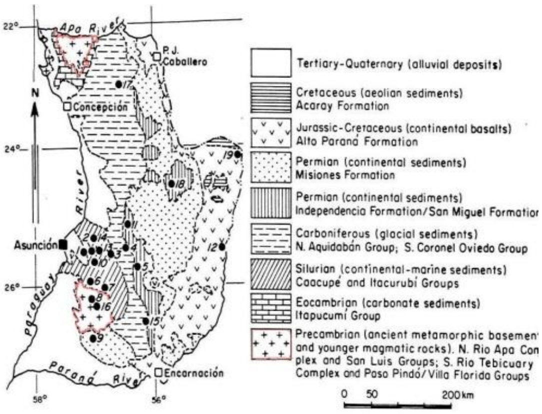

Figure 1. Eastern Paraguay geological map from V.J. Fulfaro, 1996

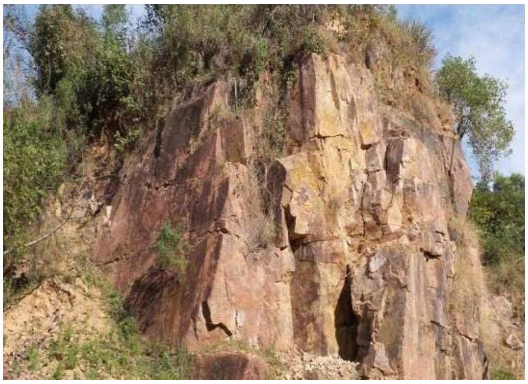

Figure 2. Jaguarete kua outcrops
courtesy of Prof. J.C. Velazquez (Paraguay)

1.2. Geological settings

The Precambrian in the south [7,8] outcrops along the Route Py1 from Quiindy City to just north of the city of San Juan Bautista. Outcrops extend west to about 15 km from Caapucú and ca 30 km from Itayuru. At East, extend to approximately 36 km from Caapucú. Westward, the Precambrian is covered by recent meta-sediments. From Quiindy to a few kilometers south of Villa Florida, there are large outcrops of granitic rocks. These begin with normal biotitic granite strips immediately south of Quiindy and proceeds to porphyritic granite and porphyry granitoids to reach a better development in the area of Caapucú. In general, these rocks are just little altered and exempt of metamorphism. They are crossed frequently by aplite streaks as well as by pegmatites; from southern Villa Florida to the Itayuru mountain range, just north of San Juan Bautista outcrop, a series of regional metamorphic rocks formed by gneisses, migmatites, chloritic quartzites, micaseos schists; like as in the north, this metamorphic series also is crossed by quartz veins and aplite dykes. To them, it was attributed Upper Proterozoic/Neoproterozoic age [8] which also were found in central Brazilian granitoids [12-15].

They are well correlated [8] from one side, to upper Proterozoic/Neoproterozoic Brazilian granitic rock (Central, SW Matto Grosso, etc) [12-15]. From the other side, to the well studies granites granitoids from San Carlos Apa River [9]. In both of that granites types indicator/indexes, as well as their multi diagrams are very close to that here mentioned for Jaguarete kua. These was confirmed more recently by isotopic dating method [16].

2. Methodology

2.1. Samples preparation

In this work hand specimens granitoids rock from the Jaguarete Kua plug were studies. The samples were prepared for neutron irradiation as well as for XRF analysis by quartering of finely pulverized materials [17].

2.2. Neutron bombardment

a) Irradiations

Sodium elements contents were analyzed by neutron bombardment. The used nuclear reaction is ^{23}Na (n, γ) ^{24}Na ($T_{1/2}$ = 15.06 h) [18]. The amount of irradiated samples material oscillates between 5 to 8 g. They were irradiated with a 25 Ci ^{241}Am-Be neutron source from Amersham. According to the maker, the total flux is 5×10^7n.s^{-1} ± 20% [19], in good agreement with the known yield of 80 n/10^6 Bk. The flux per area unit was estimated ~ $3.4{\times}10^5$ n.s^{-1} cm^{-2}; the thermal flux is about 7×10^4 n.s^{-1} cm^{-2}. The neutrons energy spectra are complex averaging 4-5 MeV with net spikes at 4.7, 6.5 MeV. [20].

b) Measurements

Irradiated samples were measured in a 3”. 3” Na I (Tl) crystal. The 1.37 MeV γ ray of ^{24}Na was used for the analysis. The analysis of the samples was performed in two steps: in the first, an aliquot of sample was irradiated and afterwards followed the half-life of appropriate photo peaks, in order to check the absence of any significant tail; in the second step, after irradiation, a certain number of counts was collected, on a fixed time basis and extrapolate to t = 0. For calculations, calibration curves were employed.

2.3. X-ray Irradiation and Fluorescence Analysis

From the pulverized material, pellets pressed were prepared; the weight of them was between 0.1 and 0.3 g cm^{-2}.

For the excitation of fluorescence radiation, the X-ray tube (at 40 kV and 20 mA) with the Mo anode and Mo secondary target was used. The energy dispersive X-ray spectrometer was based on a Si (Li) semiconductor detector (FWHM ~140 eV at 5.9 keV) coupled to a spectroscopic amplifier and a multi-channel analyzer.

The analysis of complex spectra was performed by the AXIL [21] software which is based on iterative nonlinear least square fit of the spectra by the gaussian shaped spectral lines. The resulting intensities of pure K_α and L_α lines of measured elements were then utilized in quantitative analysis, employing the quantification software of QAES (quantitative analysis of environmental samples) designed by Kump [22]. Details have been given elsewhere [23].

3. Result and discussion

Metasomatism

The melting of the source rock, according temperature and depth (pressure), occurs in the sediments/meta sediments of the continental Crust.

And together with magma, liquid granite is originated, particularly in the presence of H2O.CO2 [1-3], [24,25], It should be taken in account that granites often are mixture of melts from mafic and crustal rock [25,26]. The interaction of liquid granite, residual minerals and magma can give rise to various types of granite [1-3]. It is customary to designate them with alphabetic letters; one of them is S-type granite which is formed primarily when sediments and equivalent metamorphism, work at depth in the continental Crust [1-5],[27].

The results are broken down in Table 1. It shows the values obtained for major, minor and trace elements and consequently some aspect of the geochemistry of the analyzed materials. They allow to explore aspects that suggest the type of granite present, according indexes that are used for such effects.

Table 1. Results from analyzed major and trace elements

%	1a	1b	2a	2b	3a	3b
Al_2O_3	12.5	13.1	12.9	13.1	13.8	13.0
SiO_2	72.0	71.6	73.10	77.3	71.0	73.4
K_2O	4.50	4.13	4.0	4.26	4.1	4.30
Na_2O	3.1	3.2	3.30	3.45	3.30	3.20
CaO	0.38	0.61	0.58	0.57	0.52	0.43
µg/g						
Ti	6.01	6.38	5.23	5.52	9.17	8.42
Mn	139	142	146	148	114	217
Fe	6550	5630	6530	5770	7580	7550
Cu	61.5	25.6	27	29.7	31.6	34.1
Zn	46.5	32.6	74	39.6	24.7	28.4
Ga	32.4	6.7	19.1	16.1	18.5	17.8
Rb	236	170	134	184	232	247
Sr	30.8	51	57.9	50.7	33.9	26.6
Y	28.9	26.3	22.1	28.3	41.8	39.9
Zr	187	106	110	102	184	179
Nb	92.3	39.4	48.7	13.3	49.6	29.9
Cs	1.64	3.36	2.0	1.5	1.6	1.5
Ba	315	321	551	265	343	265
La	44.8	47.4	30.3	42.3	46.7	42.3
Ce	97.8	106	71.4	93.6	101	93.6
Pr	7.5	7.27	3.9	6.1	7.0	6.1
Nd	34.9	39.4	29.2	34.1	36.3	34.1
Th	14.1	7.7	8	11.4	16.4	17.0
Pb	30.1	16.0	24.2	16.7	18.3	27.0

*SD: from 10 to 25%

They show relatively high values for major elements concentration remarkably at SiO_2, Al_2O_3, K_2O, Na_2O and low CaO usually generally used to distinguish the type of granites [6,27].

The various indexes/indicators, given in ratio used here, appears in Table 2; [1,3, 5,6,25,27].

i) Si_2/Al_2O_3: ratio >1; values from 3.39 to 3.47: This index compliments granite S, I and A- types.

ii)$Al_2O_3/(Na_2O+K_2O+CaO)$ ratio >1; in this work the range found of values are 2.07 to 2.26: compliments only S type granite.

iii)Na_2O/K_2O ratio <1; values here found= 0.43 to 0.55; compliments S-type; something A -type granite.

iiii) $Al_2O_3/(Na_2O+K_2O+0.5CaO)$ > 1; values range from 2.07 to 2.26, S- type granites.

v)$K_2O/(Na_2O+CaO)$ ratio > 1. Values found 2.05 to 2.26 suggesting pelitic rock sources. S- types granites.

vi) K/Rb ratio >75; values found, from 172.4 to 299. Compliments by S- type granite.

Table 2. Ratio index calculated for major and trace elements

Major elements	1a	1b	2a	2b	3a	3b
Si2/Al2O3	3.39	3.22	3.33	3.47	3.16	3.32
Al2O3/ (Na2O+K2O+CaO)	2.05	2.15	2.26	2.06	2.0	2.15
Al2O3/ (Na2O+K2O+0.5CaO)	2.07	2.15	2.26	2.12	2.26	2.19
Na2O/K2O	0.43	0.51	0.54	0.54	0.55	0.46
K2O/(Na2O +CaO)	2.06	1.67	1.82	1.62	1.6	1.92
K/Rb	179.23	243	299	231.5	172.4	182.9
Trace elements						
Rb/Sr	7.61	3.33	2.31	3.70	6.84	9.15
Nb/Y	3.18	1.44	2.20	0.46	1.18	0.79
Rb/(Y+Nb)	1.94	2.58	1.89	1.4	2.9	3.53

The above index, very important, it is a "*marker*" of the fractionation of geochemical types of rocks: K/Rb "fractionated" granites and are defined by K/Rb >75. In this effort, ratios values found range from 172.4 to 299.

The ratio K/Rb is considered by several writers, as one of the most useful indexes in analyses of the discrimination of the various granotoid types [6,16].

It must be mentioned that mineralogically samples containing inter alia red biotite, muscovite Al-silicates, ilmenite, apatite fulfil S-type [5].

Petrologically: aplite very frequently, pegmatite, rhyolites, enclave of metasediments. Complimented by S-type granite, as well as gneisses, migmatites, chloritic quartzites, micaseos schists, etc.

Ga content has been found an important ratio in the S-type granite geochemistry. Ga follows Al group 13 of the periodic system. Al is relatively reach peraluminous in S-type granite. This ratio is therefore important in the relationship of magma fractionation. In grosso mode, it ranges from 1.2 to 5.9 in granite/rhiolites in Bohemian S-type granites [6,28].

The Fe^{+3}/Fe^{+2} ratio is another important index to discriminate the S-type granite, since for this type of rock the Fe_3/Fe_2 ratio is usually very low in this type of granite, indicative of low oxygen concentration in the meta sediments/clays as found here [1-6,25,27].

The values of these calculated indexes are close to those characteristics of S- type granite.

4. Multielement diagrams

In regard to the refractory elements, their spidergrams normalized to PM (Primitive Mantle) [29] (see Figures 1,2,3) of Jaguarete Kua samples show strong negative anomaly for K, Sr, Ti; positive spikes for Rb, Nb, Ce, Nd, Zr. Of high importance, they display strong resemblances to the granitoids from the Apa Hight (San Carlos) as well as Fuerte Olimpo (in Western Paraguay) [9,23-25].

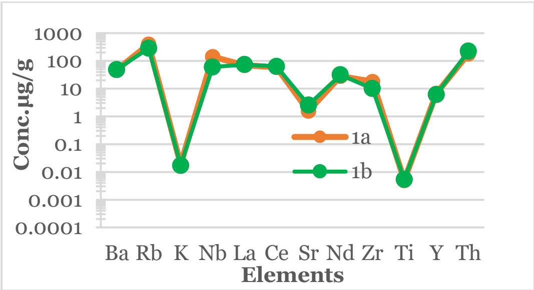

Figure 1. Aragnograms PM normalized 1a & b

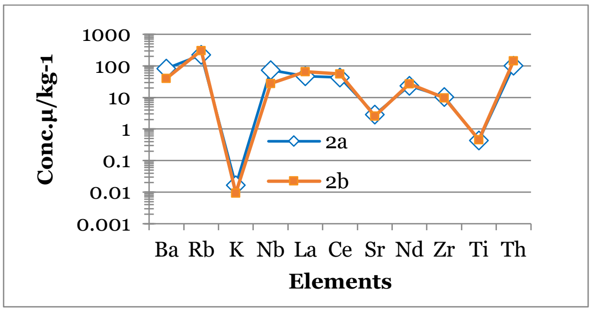

Figure 2. Aragnograms PM normalized 2a & b

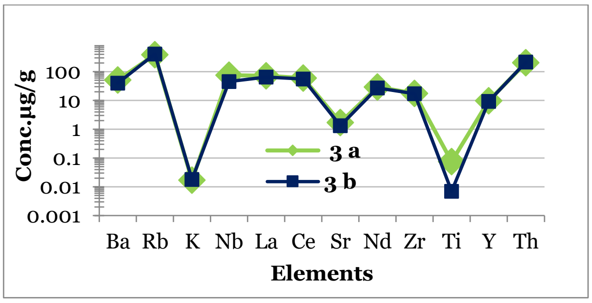

Figure 3. Aragnograms PM normalized 3a & b

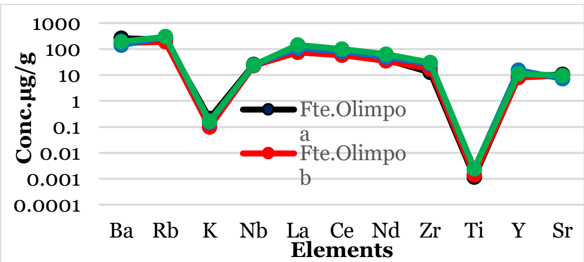

Figure 4. Aragnograms PM normalized Fuerte Olimpo and San Carlos

The spidergrams of the samples are very similar among them.

Incompatible elements enrichment may be associated with metasomatic processes involving fluids and/or small volume melt [2]. It seems that the source was in upper asthenosphere and that such fluids are likely, from volatile melt.

For comparison, it was included in Figure 4 from data taken at random from the spidergrams of San Carlos and Fuerte Olimpo at the North Precambrian show strong resemblances with those of from samples of Jaguarete Kua –Caapucu Hight [9,30].

The variation in Nb relative to Zr in the source could be due to a particular $CO_2/H_2O + CO_2$ ratio. Different $H_2O + CO_2$ proportions in different depth might explain the concentration differences [24]. Figure 5 neoproterozoic from Rio Branco [30], Brazil also taken for comparison, presents a strong resemblance to samples from the north of eastern Paraguay as well as those from Jaguarete Kua.

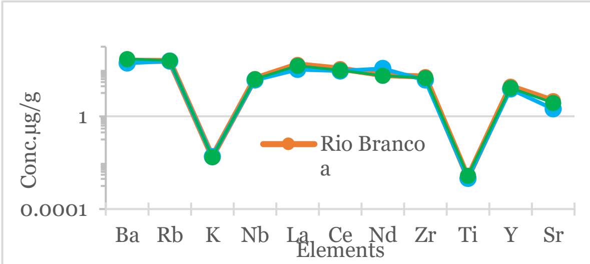

Figure 5. Aragnograms PM normalized Rio Branco

5. Conclusion

Characteristic of S-type granite is coincidence with the result obtained in this work. In addition, PM normalized incompatible elements diagrams here obtained, are very similar to those of Fuerte Olimpo, Fuerte San Carlos of Neoproterozoic in the north, coincident with similar diagrams of Rio Branco suite in Amazon Craton also from the Neoproterozoic Precambrian. Both in son way confirm Neoproterozoic for granites of Caapucu.

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