## GEOLOGY AND PETROGRAPHY OF NEPHELINE SYENITE FOR ORNAMENTAL USE AT MARAPICU PEAK, MENDANHA INTRUSIVE COMPLEX, STATE OF RIO DE JANEIRO, BRAZIL

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> Introduction Natural stones and the deposits Regional geology Marapicu nepheline syenite quarries Erosive resistance of the nepheline syenite body Petrographic characteristics Comparative visual aspects of the three types Quantitative colour specification Conclusion Acknowledgement References

**RESUMO** - *Motoki, A., Vargas, T., Motoki, K.F., Sichel, S.E. – Geologia e petrografia das rochas ornamentais de nefelina sienito no Pico do Marapicu, complexo intrusivo de Mendanha, Estado do Rio de Janeiro, Brasil.* Este trabalho apresenta geologia e petrografia das rochas ornamentais de nefelina sienito no Pico do Marapicu, complexo intrusivo de Mendanha, RJ, Brasil, conhecido comercialmente Granito Cinza Ás de Paus. Existem três tipos de produtos de nefelina sienito: Tipo Clássico, Tipo Moderno e Tipo Anfibólio Prismático. O Tipo Clássico é caracterizado pelo aspecto visuais suaves de cor cinza de tonalidade ondulante. A textura é equigranular e homogênea sem orientação dos minerais. O Tipo Moderno tem aspectos similares, porém sem tonalidade ondulante da cor cinza. O Tipo Anfibólio Prismático é caracterizado por cristais prismáticos de anfibólio com notável orientação. Os minerais constituintes principais são feldspato alcalino de anortoclásio, nefelina, anfibólio e biotita. Apesar da vulnerabilidade ao intemperismo químico, as rochas são muito frescas devido aos efeitos da passividade intempérica. O nefelina sienito é mecanicamente resistente, com a compressão uniaxial para a ruptura superior àquela de granitos ornamentais comuns. O corpo rochoso é maciço sem notáveis sistemas de fratura e falhas de origem tectônica, o que facilita a extração de grandes blocos. A especificação quantitativa de cores revela que a tonalidade suave da cor cinza é devida principalmente à abundância de anfibólio, com o parâmetro B em torno de 15, relativa a biotita, com o B próximo ao zero. A ondulação na tonalidade de cinza do Tipo Clássico é originada do metassomatismo.

Palavras-chave: nefelina sienito, Granito Cinza Ás de Paus, Marapicu, jazida de rocha ornamental, passividade intempérica, especificação de cor.

**ABSTRACT** - This paper presents geology and petrography of the nepheline syenite for ornamental use at Marapicu Peak, Mendanha intrusive complex, State of Rio de Janeiro, Brazil, known commercially Granito Cinza Ás de Paus. There are three types of nepheline syenite products: Classic Type, Modern Type, and Prismatic Amphibole Type. The Classic Type is characterised by soft visual aspect of undulated grey tone. The texture is equigranular and homogenous without mineral orientation. The Modern Type has similar aspects, but has no undulant the grey tone. The Prismatic Amphibole Type is featured by acicular amphibole crystals with notable orientation. The main constituent minerals of these rocks are anorthoclase alkaline feldspar, nepheline, amphibole, and biotite. In spite of the vulnerability to chemical weathering, the rocks are quite fresh because of the effects of weathering passivity. The nepheline syenite has high mechanical strength, with uniaxial compression for rupture superior to common granites for ornamental use. The rock body is massive without notable systems of fractures and faults of tectonic origin, which helps large block extraction. The quantitative colour specification reveals that the soft grey tone is due mainly to the abundance of amphibole, with brightness ca. 15, relative to biotite, ca. 0. The undulated grey tone of Classic Type is originated form metasomatism.

Keywords: nepheline syenite, Granito Cinza Ás de Paus, Marapicu, natural stone deposit, weathering passivity, colour specification.

#### **INTRODUCTION**

Nepheline syenite and alkaline syenite are attractive products of Brazilian natural stones for ornamental uses (Vargas et al., 2002). Such rocks are rare in world occurrence and found generally in extensional tectonic fields. The most common case is the nepheline syenite and alkaline syenite in the continental break-up region. Another case is the alkaline syenite and quartz-bearing alkaline syenite in back-arc regions of continental collision zones.

In the south-eastern Brazilian coast region, there were syenitic magmatisms of the Cretaceous to early Cenozoic related to the Pangea break-up (Woolley, 1987). They are constituted mainly by grey nepheline syenite and alkaline syenite, such as of Itatiaia (Brotzu et al., 1997), Mendanha (Motoki, A. et al., 2007a), Morro de São João (Brotzu et. al., 2007), and Cabo Frio Island (Motoki, A. et al., 2013). The nepheline syenite of the Marapicu Peak, State of Rio de Janeiro, Brazil, is an example of active extraction in rock blocks shipped to dimension stone factories, with commercial name of *Granito Cinza Ás de Paus*, meaning Spade Ace Grey Granite.

In addition to the grey syenitic rocks, there are green alkaline syenitic rocks with few modal percents of quartz, such as of Tunas (Fuck, 1973), State of Paraná, Vitória Island (Motoki, A., 1986), Búzios Island (Gomes & Alves, 2001), and São Sebastião Island (Bellieni et al., 1990), State of São Paulo. The green rock of Tunas is in exploitation, named commercially *Granito Verde Tunas*.

At Caldas, in State of Minas Gerais, quartz-bearing alkaline syenites occur in the back-arc region of the Pan-African continental collision zone. The *Granito Marrom Caldas* is characterised by amphibole and 2 to 3% of quartz, and *Granito Café Royal*, by biotite and aegirine-augite without quartz (Meyer et al., 2003; Artur et al., 2005). A similar rock is in exploitation at Riacho de Santana, central region of State of Bahia, with commercial name of *Granito Café Bahia*.

There is a general relationship between macroscopic colours and geotectonic background of alkaline syenite and quartzbearing alkaline syenite. Deep green and dark blue rocks are found in continental break-up region and brown ones in back-arc regions of continental collision zones. The macroscopic colours are originated from slight tone of alkaline feldspar. The genetic relation between colour and tectonic field is still in discussion.

In addition to the igneous nepheline syenite, there also is metamorphic nepheline svenite, so-called litchfieldite. This rock is very scarce and an example occurs at Canaã Village, Duque de Caxias Municipal District, State of Rio de Janeiro. The rock is characterised by banded texture and high modal amount of of muscovite nepheline. presence and lepidomelane, perthitic texture of substitution, and absence of clinopyroxene and amphibole. They have very low Ti, Mg, Mn, and Ca and high Na, K, and Al. It is considered that the nepheline syenite gneiss is originated from igneous intrusive nepheline syenite and later Pan-African high-grade submitted to metamorphism with strong influence of fluids.

The nepheline syenite which occurs at Marapicu Peak is widely known in the natural stone market with commercial names of *Granito Cinza Ás de Paus*, *Granito Cinza Campo Grande*, and *Granito Cinza Carijó*. This rock has peculiar visual aspects of undulating grey tone with soft visual aspects (Figure 1). It is an important non-metallic mineral deposits of Nova Iguaçu Municipal District, together the special quality gravel of alkaline syenite at the eastern limit the Mendanha massif (Petrakis et al., 2010).



**Figure 1**. The *Granito Cinza Ás de Paus*, a natural stone of nepheline syenite for ornamental uses extracted from Marapicu Peak, western limit of Mendanha alkaline massif, State of Rio de Janeiro: A) Outcrop at the quarry; B) Boulder extraction; C) Block to be transported; D) Polished rock slab at local dimension stone factory.

This article presents geologic observations, petrographic descriptions, and quantitative surface colour specification for the nepheline syenite of Marapicu Peak. Based on the data, the authors consider the genesis of the peculiar visual aspects of this rock and the geology of this nepheline syenite natural stone deposit.

#### NATURAL STONES AND THE DEPOSITS

The exploitation target material of natural stone quarries is entire rock, and not, a specific mineral. Different from metallic ore deposits, no physical selection and chemical concentration are available. Natural stone deposits with commercial value grades into the adjacent valueless rocks, without clear boundary.

There are three main types of rocks for ornamental use, called commercially *granite*, *marble*, and *slate*. The commercial name *granite* indicates holocrystalline cause-grained silicate rocks of igneous and metamorphic origin, such as alkaline granite, granite, granodiorite, diorite, gabbro, nepheline syenite, augen gneiss, and charnockite. The *marble* is carbonaceous metamorphic and sedimentary rocks, as marble and limestone. These rocks are submitted generally to surface polish in dimension stone factories.

The slate is silicate rocks with welldeveloped cleavages, such as slate, schist, and quartzite. The *porphyry* is a special type natural stone originated from pyroclastic flow deposits of ultra-high grade welding, such as Roman Stone of northern Italy and Patagonian stone of southern Argentina. Basalto da Serra Gaúcha is a Brazilian product of this rock. Basalto is the commercial name used in dimension stone market. In fact, these rocks are ignimbrite of ultra-high grade welding with rhyolitic to dacitic composition (Petrakis et al., 2010) and have very high mechanical strength, with uniaxial compression rupture stress of 350 MPa. Most of them are used without surface polish, but some of them are transformed into polished dimension stone slabs.

The deposits of natural stone for ornamental use with economic feasibility occur in limited cases. The important social factors are: 1) There is cheep and easy transport means from the quarry to the consumption areas; 2) The extraction locality is situated out of urban zones and environmental protection zones; 3) Mining permission cab be obtained without difficulty. The natural factors are: 1) The rock has enough mechanical strength; 2) Free from fractures; 3) Large and homogeneous rock body; 4) High resistance to urban weathering; 5) Aesthetic beauty.

Most of the granits of the Brazilian dimension stone market have uniaxial rupture stress, of about 120 MPa, which is higher than the standard of orthogneiss road gravel, of about 90 MPa. The rocks in fracture zones and hydrothermal alteration areas are not available. The natural stone for ornamental uses are extracted in blocks of metric sizes and sent to the factories to be submitted to slice and polish processes. If total loss of the extracted material is larger than 70%, the commercial operation becomes unfeasible. Faccio Quarry of Basalto da Serra Gaúcha (commercial name) at State of Rio Grande do Sul has exceptionally high yield rate with less than 10% of loss. As an industrial product, dimension stone have must homogeneous quality and stable supply.

Many dimension stone slabs are used in urban zones for building wall coating and floor paving. The urban atmosphere has high SOx and NOx, which causes chemical corrosion of certain rock-forming minerals, such as pyrite, calcite, nepheline, and mafic minerals. Pyrite decomposes easily into iron hydroxides and black spots appear on the rock surface within years. Marbles exposed in outdoor few environment lose the lustre of polished surface within 10 years. In the same way, gabbro and diorite lose lustre. On the other hand, the rocks with quartz, such as granite, granodiorite, augen gneiss, and charnockite, maintain the lustre in a long time. However, surface oxidation can change the macroscopic colour of charnockite.

Visual aesthetic beauty is a most important factor for the commercial value of natural stones for ornamental use. This factor is highly subjective and difficult to be discussed by scientific means. For this purpose, the authors have been developed the method for quantitative specification of colour for while rock and each minerals using scanners, establishing an objective and quantitative colour parameters, with the help of the original software Wilbur 1.0 (Motoki, A. et al., 2006).

#### **REGIONAL GEOLOGY**

The Marapicu Peak is situated at the western limit of Mendanha svenitic intrusive complex, which is located at about 40 km to the west-northwest of the city of Rio de Janeiro. Three laser-spot Ar-Ar datings for the biotite for the alkaline syenite at the eastern limit of Mendanha intrusive body showing ages of the Early Cenozoic, ca. 60 Ma (Motoki, A. et al., 2007a). On the other hand, the laser-spot Ar-Ar datings for the nepheline syenite of Marapicu Peak, at the western limit of the intrusive complex has Late Cretaceous age, ca. 80 Ma (unpublished data of the authors). The intrusive complex forms a massif of 15 x 3 km of extension with a relative height of 900 m, called Mendanha felsic alkaline complex.

The basement of this area is constituted by the Pan-African metamorphic rocks of Oriental Terrane (Heilbron & Machado, 2003) with metamorphic age of ca. 550 Ma (Valladares et al., 2008). The metamorphic basement is cut by the silicified tectonic breccia of the last phase of Pan-African Orogeny (Motoki, A. et al., 2011; 2012a) and early Cretaceous mafic dykes (Guedes et al., 2005; Motoki, A. et al., 2009a).

In State of Rio de Janeiro, there is a dozen of the large felsic alkaline rock bodies with distribution area larger than 1 km<sup>2</sup>: Itatiaia (Brotzu et al., 1997), Morro Redondo (Brotzu et al., 1989), Tinguá, Mendanha (Motoki, A. et al., 2007b), Canaã, Itaúna (Motoki, A. et al., 2008a), Tanguá, Soarinho, Rio Bonito (Motoki, A. et al., 2010), Cabo Frio Island (Sichel et al., 2008), and Morro de São João (Brotzu et al., 2007). They constitute a WEW-ESE linear sequence, called Pocos de Caldas-Cabo Frio alkaline magmatic alignment (Figure 2: Thomáz Filho et al., 2005; Riccomini et al., 2004). They are made up mainly of nepheline syenite and alkaline syenite (Sichel et al., 2012), with local occurrences of phonolite (Motoki, A. & Sichel, 2008) and trachyte (Motoki, A. et al., 2008b). The syenite and monzonite of the Morro dos Gatos (Motoki, A. et al., 2012b; Geraldes et al., 2013) is a rare example. Seven of them have strongly welded vent-filling subvolcanic breccia (Motoki, A. & Sichel, 2006; Motoki, A. et al., 2007c; 2008c).



Figure 2. Felsic alkaline rock bodies of State of Rio de Janeiro, modified from Sichel et al. (2012). The Canaã body (CAN) is exceptionally made up of nepheline syenite gneiss.

With the exception of Canaã body, all of the above-mentioned alkaline rocks form shallow intrusive rock bodies. According to the K-Ar and Ar-Ar datings (*e.g.* Sonoki & Garda, 1988), the intrusive ages range from the Cretaceous to the Early Cenozoic. The K-Ar age of the Itatiaia body is 72.6 Ma. The Rb-Sr and Ar-Ar datings show similar ages, that is, 66.8 Ma for the Tanguá complex (Motoki, A. et al., 2010) and 54.8 Ma for the Cabo Frio Island (Motoki, A. et al., 2013). These intrusions could be related to the late state of the Pangea break-up and the Atlantic Ocean opening (Woolley, 1987).

The present-day exposure of the Mendanha intrusive body corresponds to root of

an elongated funnel-shaped pluton with the intrusive depth of ca. 3 km (Figure 3; Motoki, A. et al., 2007a; 2008d). The contact plane with the host gneiss is subhorizontal (Figure 3: 4A; 22°50.99'S, 43°35.84'W). Loc. 1. The difference of the constituting rocks, alkaline syenite and nepheline syenite, and the Ar-Ar ages, ca. 60 Ma and ca. 80 Ma, indicate that the eastern and western parts of the Mendanha massif are originated from two different alkaline intrusive bodies. The syenitic bodies are intruded by trachytic dykes (Figure 3B) and pyroclastic conduits filled by welded tuff breccia.



Figure 3. Geologic map and the cross sections of Mendanha felsic alkaline intrusive rock body, State of Rio de Janeiro, indicating the locations of the Marapicu Peak and the nepheline syenite quarries, modified from Motoki, A. et al. (2007a).



**Figure 4**. Adjacent bodies of Marapicu nepheline syenite: A) Basement gneiss at the foot of the Marapicu Peak (Loc. 1, 22°50.99'S, 43°35.84'W); B) A rolling stone showing phonolite dyke intrusion into the nepheline syenite.

#### MARAPICU NEPHELINE SYENITE QUARRIES

The nepheline syenite quarries occur on the western slope of the Marapicu Peak at the altitude ranging from 200 to 400 m (Figure 5). The nepheline syenite with undulating greyscale tone patches is a representative commercial product. It is called Classic Type (Figure 6A) and occurs at a limited area of Marapicu Peak. The extraction front is situated at the sites of about 350 m of altitude (Figure 7A; Loc. 2, 22°50.61'S, 43°35.75'W), so-called Upper Front. The quarry outcrops correspond geologically to bottom zone of the flattened coffee filter-shaped nepheline syenite pluton. Close to the extraction front, an orthogneiss megaxenolith of about 300 m in size is found (Figure 7).



**Figure 5**. Extraction fronts of the *Granito Cinza Ás de Paus*, Marapicu Peak: A) Upper Front, Loc. 2 (22°50.61'S, 43°35.75'W); B) Classic Type rock at the upper front; C) Lower Front, Loc. 3 (22°50.61'S, 43°35.75'W); D) Modern Type rock at the lower front with intercalation of Prismatic Amphibole Type. The optical contrast of the photo B is exaggerated.



**Figure 6.** Three types of polished dimension stone of *Granito Cinza Ás de Paus*: A) Classic Type applied to wall coating; B) Modern Type constituting a table counter; C) Prismatic Amphibole Type used for an office tool.



Figure 7. Schematic N-S geologic cross section of Marapicu Peak (A) and the contact outcrop with the orthogneiss megaxenolith at the Loc. 4, 22°50.74'S, 43°35.86'W (B).

During the 20th Century, Classic Type nepheline syenite was intensely extracted (Figure 1D) and widely used (Figure 6A), becoming one of the representative Brazilian natural stones for ornamental use (Azambuja & Silva, 1997; Erthal et al., 2003). However, its extraction is no more active. Today, an alternative type of nepheline syenite is exploited from the fronts of altitude ranging from 150 to 200 m (Figure 3, 4; Loc. 3, 22°50.61'S, 43°35.75'W), so-called Lower Front, extracting Modern Type nepheline syenite. The rock has homogeneous grey colour without undulated patches, (Figure 6B).

The rock body of these quarries is massive without notable fractures and faults of tectonic origin. Therefore, large rock blocks can be easily extracted. On the other hand, along subsurface of the massif slope, there are fractures and faults (fractures with small displacement) parallel to the surface plane, which are originated from overload release and gravitational accommodation, such as rockslides and mountain creeps.

There is one more type of nepheline syenite product characterised by oriented prismatic hornblende crystals, called Prismatic Amphibole Type (Figure 6C). This type occurs in flow bands of 50 cm to 1 m of width scattered in Lower Front (Figure 5D). In spite of the attractive visual aspects, Prismatic Amphibole Type is unfavourable for dimension stone industries because of the discontinuous and limited outcrop occurrences, so it is used for small office tools (Figure 6C).

### EROSIVE RESISTANCE OF THE NEPHELINE SYENITE BODY

For the exploitation of high-quality natural stone blocks, weathering and erosive resistance is an important factor. *Granites* (commercial name) contain generally more than 20 modal percent of quartz. This mineral has high hardness and cannot be altered by weathering. The dimension stone slabs of rocks with quartz are resistant to urban weathering and can maintain the brightness of surface polish during decades.

On the other hand, nepheline syenite is composed mainly of alkaline feldspar and nepheline. These minerals are vulnerable to chemical weathering under humid tropical climate transforming into clay minerals and amorphous materials. Especially, nepheline is easily leached out by HCl and the cavities originated from the nepheline leaching are found frequently on the rock surface (Figure 8A).

Although nepheline syenite and alkaline syenite are rare in world (Sørensen, 1974), they are commonly found in the dimension stone quarries, market. At the these rocks demonstrate unexpected high erosive resistance (Motoki, A. et al., 2008d; Petrakis et al., 2010). The surface of nepheline syenite boulders is covered by weathered crust of dark-colour with 1 to 2 cm of thickness (Figure 8B), which is much thinner than granitic, orthogneissic, and paragneissic rocks.



**Figure 8**. Weathered surface of nepheline syenite: A) Cavities originated from the nepheline leaching (brown spots pointed by arrows) by means of the reaction with rain water observed on weathered surface of the Cambrian nepheline syenite gneiss of Canaã, Duque de Caxias, State of Rio de Janeiro; B) Thin weathered crust on the surface of the nepheline syenite boulder at Upper Front of the Marapicu Peak.

The felsic alkaline intrusive bodies of this region form morphologic elevations with relative height of 300 m to 900 m, so-called alkaline massifs. In spite of the weathering vulnerability of the constituent minerals, the alkaline rock bodies are resistant to erosion. In fact, rock surface is strongly weathered, but its subsurface is free from the weathering. This apparently controversial weathering behaviour is attributed to the phenomenon called weathering passivity (Motoki, A. et al., 2008d). That is, the impermeable clay-rich regolith originated from chemical weathering of nepheline syenite covers the surface of the rock, preventing surface water infiltration and percolation into the rock body. Therefore, the syenite nepheline body is resistant to

weathering and erosion under the humid tropical climate of Rio de Janeiro (Motoki, A. et al., 2014a; b; c).

The weathering passivity is observed notably in morphologic features of the Mendanha massif. The Figure 9 shows the summit level map (*seppômen*) of the Mendanha massif based on the grid interval of 960 m, based on the satellite-derived topographic data of ASTER GDEM. The summit level map is the geomorphologic analyses technique that reproduces the palaeogeomorphology before the erosion filling virtually the valleys and drainages narrower than the grid interval, (Motoki, A. et al., 2008d; 2009b; Couto et al., 2012; Aires et al., 2012).



**Figure 9**. Summit level map and its geomorphologic principle (Motoki, A. et al., 2008d; 2014d): A) Summit for Mendanha felsic intrusive massif of the grid interval of 960 m; B) Geomorphologic principle of summit level surface.

This map shows the marginal scarp (MS) with declivity of  $30^{\circ}$  to  $35^{\circ}$ , virtual plateau of 700 m of altitude (VP), and the top

swell (TS) of 250 m of relative height. The massif is characterised by general morphology of convex form, which is a geomorphologic

feature of a massif highly resistant to erosion. This general form is highly contrasted with the general concave form of young volcanic edifices and sedimentary massifs.

The Figure 10 shows the diagram of the MCI (massif concavity index; Aires et al., 2012; Motoki, A. et al., 2014a; b; c; Motoki K. et al., 2014), which are elaborated from the data of the summit level surface and base level

surface (*sekkokumen*; Figure 9B). A deeply eroded massif of concave general form has a positive MCI, and a slightly eroded massif of convex general form has a negative MCI. The alkaline intrusive massifs, including the Mendanha Massif, have strongly negative MSI, showing clear contrast with the young volcanic edifices, which are constituted by porous lava flows and unconsolidated pyroclastic deposits.



Figure 10. Comparison between alkaline intrusive bodies and young stratovolcanoes on the MCI (massif concavity index) diagram. Relief amount (*kifukuryo*) corresponds to the difference between summit level (*seppômen*) and base level (*sekkokumen*). The data are originated from Motoki, A. et al. (2012c). MDN - Mendanha felsic alkaline intrusion; MSG - Morro de São João alkaline intrusion; CBG - Cabugi subvolcanic neck; DMN - Volcán Cerro de Diamante; NVD - Volcán Cerro Nevado; PYN - Volcán Payún Liso; OSR - Volcán Osorno.

Mechanical strength of these rocks also is an important factor. The uniaxial compression failure stress for granite, granodiorite, granulite, and charnockite is about 120 MPa, but that of the alkaline syenite of the Mendanha Massif is about 170 MPa (Petrakis et al., 2010). Because of the fresh rock exposure by the weathering passivity, rare fractures, and peculiar visual aspects, the nepheline syenite of Marapicu Peak is favourable for natural stone block extraction and industrial production of dimension stone slabs.

#### **PETROGRAPHIC CHARACTERISTICS**

The Figure 11A shows thin section image of Classic Type obtained by high resolution scanning of 2400 DPI, showing equigranular and homogenous texture without mineral orientation. The mafic minerals are brown and green hornblende and biotite with inclusions of alkaline feldspar and apatite (Figure 11B). Mafic and opaque minerals form aggregations of 2 to 3 mm. Individual crystals of the aggregations are of 1 to 2 mm (Figure 11C). Amphibole is idiomorphic and of short prismatic habit. The aspect ratio is low, ranging from 1.5 to 3.0, suggesting that the amphibole

crystals could be pseudomorph after clinopyroxene.

The mineral inclusions of apatite have diameter of ca. 0.05 mm and length of 0.2 to 0.3 mm. The alkaline feldspar inclusions are 0.1 mm thick and 0.3 mm wide, and biotite inclusions are 002 x 0.05 mm (Figure 11B). Along the border and fracture of the crystals, the amphibole crystals are partially substituted by biotite (Figure 11C). Idiomorphic primary biotite also is commonly found (Figure 11D). Some grains of the opaque minerals have skeletal or dendritic shape.



Figure 11. Photomicrography of the *Granito Cinza Ás de Paus*, the nepheline syenite of Marapicu Peak: A) Scanner image of thin section; B) Amphibole with mineral inclusions of biotite, alkaline feldspar, and apatite; C) Amphibole and biotite; D) Idiomorphic biotite and skeletal opaque mineral; E) Interstitial nepheline and auto-metasomatic alkaline feldspar; F) Small interstitial crystal of plagioclase. The photos A to D are under parallel nicol and the E and F are under crossed nicol. Af - alkaline feldspar; Afs - alkaline feldspar altered into muscovite micro-crystals; Amp - amphibole; Bi - biotite; Ne - nepheline; Opq - opaque minerals; Pl - plagioclase.

The felsic minerals are constituted mainly by alkaline feldspar with interlocking perthite, indicating that, it is not potassic feldspar but intermediate anorthoclase (Figure 11E). The crystals of the alkaline feldspar have thick tabular form, which are 3 mm wide 1 mm thick, and constitute interstitial frameworks. More than 40% of the alkaline feldspar show partial transformation into micro-crystals of muscovite (Figure 11E). The size of the

muscovite is relatively large, from 0.05 to 0.2 mm. This size suggests that the alteration of could alkaline feldspar be caused by metasomatism, being different from the sericite hydrothermal originated from alteration (Motoki, A. et al., 2007d; Novais et al., 2008). Nepheline occurs in the interstitial triangular spaces of 0.5 x 1.2 mm in size. Plagioclase is rare and smaller than 0.3 mm, filling interstitial spaces of alkaline feldspar (Figure 11F).

#### **COMPARATIVE VISUAL ASPECTS OF THE THREE TYPES**

The Classic Type is characterised by: 1) Grey colour with soft visual aspect; 2) Equigranular and homogenous texture without phenocrysts and mineral orientation; 3) Mafic minerals constituted mainly by short and wide prismatic crystals of amphibole; 4) Undulation of the grey tone (Figure 1D, 12A). The Modern Type is featured by homogenous grey colour without tone undulation (Figure 12B). The Prismatic Amphibole Type has acicular hornblende with orientation by magma flowage (Figure 12C). According to the nomenclature of Streckeisen (1973), all of them are classified as nepheline syenite of the field 11.

Most of the Brazilian grey granites, such as Granito Cinza Andorinha e Granito Cinza Co rumba, are characterised by porphyritic texture with oriented alkaline feldspar phenocrysts of 1 x 3 cm to 2 x 8 cm. They fall on the field 3b and classified as monzogranite. In comparison with the other grey granites, the Granito Cinza Ás de Paus is slightly darker in whole rock macroscopic colour (Motoki, A. et al., 2006).



**Figure 12**. Polished surface of the *Granito Cinza Ás de Paus*: A) Classic Type featured by grey nepheline, amphibole, and biotite; B) Modern Type characterized by homogeneous grey colour; C) Prismatic Amphibole Type with acicular hornblende.

The amphibole crystals of Classic Type are 1.5 x 3 mm to 2 x 5 mm in size. The nepheline is generally grey, but sometimes pinkish. The Modern Type has mafic minerals of 2 x 4 mm to 3 x 6 mm, constituted by biotite and amphibole (Figure 12B). Amphibole grains of Prismatic Amphibole Type are smaller, 1 x 4 mm in size (Figure 12C). The alkaline feldspar is the main constituent mineral generating grey macroscopic colour.

The most highlighted macroscopic feature is grey tone undulation of Classic Type with an interval of 10 cm (Figure 1D). As far as the authors concern, no other natural stone for ornamental use with similar aspect are present in the dimension stone market.

### QUANTITATIVE COLOUR SPECIFICATION

The authors have performed macroscopic colour specification for the dimension stone slabs of Marapicu nepheline syenite based on the method of Motoki, A. et al. (2006). The quantitative determination of the rock colour is important for high-precision quality control and genetic consideration of the natural stone for ornamental use ore deposits (*e.g.* Motoki, A. et al., 2006; 2007e; Campello, 2006; Urano et al., 2006; Lópes et al., 2010).

The scanner adopted for the colour analyses is HP Scanjet G4050<sup>™</sup> with optical

resolution of 4800 DPI. The obtained images are compacted into 300 DPI for the colour analyses in order to neutralise the *Feldspar Problems* of hardware origin (Motoki, A. et al., 2006). All of the automatic adjusting functions of the scanner software are put inactive, such as: highlight=0, shadow=0, and meantone=0. For colour balancing, white colour standard plate for photo-optical calibration of the Nihon Denshoku<sup>™</sup> K.K. has been adopted.

The Table 1 presents the whole rock colour in the HSB chromatographic parameters.

The average values of S parameter (saturation) of Classic Type, Modern Type, and Prismatic Amphibole Type are, respectively, 4.84, 5.33, and 5.53. They are classified to be of slight colour (SC) category (Figure 13). The average H parameters (hue) are, respectively, 115, 100, and 117, which correspond to green tone. Because of the low saturation, general

macroscopic colour is almost grey. The B parameters (brightness) are, in average, 58, 60, and 50. According to the brightness, Classic Type and Modern Type are classified to be of the light grey category (LG), and Prismatic Amphibole Type is classified to be of the dark grey (DG).

**Table 1.** Whole rock physical colours in HSB chromatographic system for the three types of the Granito Cinza Ás dePaus, that is, nepheline syenite of the Marapicu Peak. CL - Classic Type; MD - Modern Type; PA - Prismatic

Amphibole Type. The other greyscale *granites* are: Corumbá - *Granito Cinza Corumbá*, Brazil; Ceará - *Granito Branco Ceará*, Brazil; Bandai - *Bandai Granite*, Japan; Inada - *Inada Granite*, Japan;

Sample	Н	S	В
CL1	107	5.0	61
CL2	109	4.9	60
CL3	125	4.7	55
CL4	120	4.8	56
MD1	101	6.1	68
MD2	104	5.1	57
MD3	98	5.2	59
MD4	98	5.2	59
MD5	100	5.1	58
PA1	112	5.5	55
PA2	119	5.6	49
PA3	120	5.6	49
PA4	117	5.5	46
Corumbá	153	3.8	69
Ceará	76	3.3	88
Bandai	117	5.5	47
Inada	121	2.7	78



Figure 13. Colour classification diagram of saturation vs. brightness diagram for the samples of the Granito Cinza Ás de Paus, modified from Motoki, A. et al. (2006). The categories are: W - white; LG - light grey; DG - dark grey; B - black; GS - greyscale; SC - slight colour; LC - light colour; MC - medium colour; HC - high colour.

The whole-rock greyscale tone histograms (Figure 14) show two peaks for the nepheline syenite of Marapicu Peak. The higher peak has B parameter (brightness) ranging from 60 to 65, which corresponds to alkaline feldspar and nepheline. The lower peak has the B parameter from 10 to 20, being attributed to amphibole. The diagram patterns characterise the differences of greyscale tone of white, grey, and black *granites*. This well-distinguished bimodal peak pattern is widely different from the other grey *granites*, working as greyscale fingerprint of this nepheline syenite (Figure 14D).



**Figure 14**. Whole rock greyscale tonality histogram for the *Granito Cinza Ás de Paus*, the nepheline syenite of the Marapicu Peak: A) Classic Type; B) Modern Type; C) Prismatic Amphibole Type; D) Comparison with other grey and white ornamental *granites*.

The Granito Cinza Corumbá (Espírito Santo, Brazil) and the Bandai Granite (Fukushima, Japan) have an acute peak at B=0, which corresponds to biotite (Figure 14D). However, the Granito Cinza Ás de Paus has no such a peak and amphibole shows a gentle rise on the diagram at B~15. In addition, this rock contains no quartz grains, which have strong surface lustre. Presence of amphibole and absence of quartz generate "soft" grey tone of the nepheline syenite. On the other hand, granite and granodiorite contain quartz and biotite. These minerals have strong lustre, which makes "hard" grey tone. This difference is well represented by the standard deviation of the B parameter (Motoki, A. et al., 2006).

The B parameter of the mafic minerals of Prismatic Amphibole Type (B~7) is lower than those of Classic Type and Modern Type (B~15). That is, the hornblende colour of these rocks is not homogenous. A possible genesis is the oxidation degree of Classic Type and Modern Type is higher than Prismatic

Amphibole Type. For the determination of whole rock  $Fe^{3+}/Fe^{2+}$ , the chemical analyses by atomic absorption is required, which is now rarely practiced. If clinopyroxene were present in this rock, the  $Fe^{3+}/Fe^{2+}$  could be calculated from EMPA data based on the ion charge balance (e.g. Motoki, A., 1986). However, the rock has no clinopyroxene and for hornblende no such a method is available. The highly alkaline amphibole, as riebeckite, have high  $Fe^{3+}/Fe^{2+}$ , but the optical observations show that the amphibole is hornblende to barkevikite and  $Fe^{3+}/Fe^{2+}$  is not so high. Whole-rock  $Fe^{3+}/Fe^{2+}$  has a correlation to (Na+K)/Al(Motoki, A. et al., 2010). However, the rocks are not peralkaline but meta-alkaline with low estimated  $Fe^{3+}/Fe^{2+}$ . Due to the effects of weathering passivity, the rocks are quite fresh without notable oxidation originated from weathering. In this moment, there is no direct evidence that supports the idea that  $Fe^{3+}/Fe^{2+}$  of the amphibole of Classic Type and Modern Type is higher than Prismatic Amphibole Type.

The sample CL1 and CL2 of Classic Type have the peak of felsic minerals (B~60) little higher than that of the CL3 and CL4 (B~56). This difference represents the undulating greyscale tone patches (Figure 14A). Under thin section, the light grey patches exhibit strong alkaline feldspar alteration into muscovite (Figure 11E). The alkaline feldspar of the dark grey areas is unaltered. Therefore, the unique visual aspect of Classic Type is attributed possibly to the heterogeneous metasomatism originated from deuteric alteration. This phenomenon is observed only along the border of the megaxenolith (Figure 5A) and the metasomatism could be attributed to the  $H_2O$  extracted from the megaxenolith.

#### CONCLUSION

The above-mentioned geologic, lithologic, and petrographic observation, and surface colour specification of the nepheline syenite of Marapicu Peak, Mendanha Massif, State of Rio de Janeiro, Brazil, called commercially *Granito Cinza Ás de Paus*, lead the authors to the following conclusions.

1. The nepheline syenite rock body has three types of natural stone for ornamental use products: 1) Classic Type, which is characterised by equigranular texture with grey tone undulation; 2) Modern Type, with homogenous grey tone; 3) Prismatic Amphibole Type, featured by acicular crystals of amphibole with orientation;

2. The Classic Type was extracted from Upper Front. It was the main product during the 20<sup>th</sup> Century. The Modern Type and Prismatic Amphibole Type are in active extraction from Lower Front. The latter is an accessory product and occurs in small and discontinuous flowbands;

3. In spite of the chemical weathering vulnerability of the constituent minerals, such as alkaline feldspar and nepheline, this rock is

very fresh. The apparently controversial phenomenon is attributed to the effects of weathering passivity;

4. The rock has high mechanical strength with high uniaxial compression stress for the rupture. The tectonic fractures and faults are rare. Therefore, large blocks can be extracted easily from the massive outcrops;

5. The main constituent minerals are alkaline feldspar, nepheline, amphibole, and biotite. Alkaline feldspar is not potassic, but intermediate anorthoclase. The amphibole of the Classic Type and Modern Type has short prismatic habit. The accessory minerals are magnetite, ilmenite, apatite, titanite, and plagioclase;

6. The soft tone of the grey colour is an important visual feature of this rock. It is originated from the abundance of amphibole relative to biotite. The peculiar aspect of undulated greyscale tone of Classic Type would be caused by the heterogeneous deuteric metasomatism. The  $H_2O$  for the metasomatism is originated from the megaxenolith of the country orthogneiss of 300 m of size.

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