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ORE DEPOSITS

Titaniferous magnetite and barite from the San Gregorio de Polanco dike swarm, Paraná Magmatic Province, Uruguay

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ABSTRACT

The San Gregorio de Polanco Dike Swarm (Tacuarembó Department, Uruguay) is the southernmost set of dikes in the Paraná Magmatic Province of Uruguay. Five major dikes have been identified with two main structural trends: N140º–N170º and N50º–N80º. The dikes have tholeiitic affinities and are composed of plagioclase (An55), augite and augite-pigeonite, relicts of olivine and opaque minerals. These rocks have high contents of Fe–Ti oxides (titanomagnetites), the mineralogical and textural characteristics of which have been studied using scanning electron microscopy and energy dispersive spectrometry techniques (SEM – EDS). These features, along with other mineralogical and textural relationships, have been used to propose the following crystallization sequence for the dikes: (i) crystallization of olivine, plagioclase and Ca-rich pyroxene phenocrysts; (ii) precipitation of the first population of Ti-magnetite; (iii) crystallization of plagioclase and pyroxene in the groundmass; (iv) partial dissolution of Ti-magnetite by reaction with magmatic fluids; (v) crystallization of the second population of Ti-magnetite and finally, (vi) crystallization of interstitial barite.

Key words: titanomagnetite, barite, mafic dikes, Mesozoic, Uruguay

RESUMEN

El Haz de Diques de San Gregorio de Polanco (Departamento de Tacuarembó, Uruguay) es la ocurrencia más meridional de diques pertenecientes a la Provincia Magmática Paraná en Uruguay. Fueron identificados cinco 5 diques principales con dos direcciones estructurales principales: N140º - N170º y N50º - N80º, respectivamente. Son diques de afinidad tholeítica compuestos por plagioclasa (An55), augita y augita-pigeonita, relictos de olivina y minerales opacos. Estos diques se caracterizan por el alto contenido de óxidos de Fe y Ti (titanomagnetitas), cuyas características mineralógicas y texturales fueron estudiadas con microscopio electrónico de barrido y espectrometría de energía dispersiva (SEM-EDS), incluyendo mapeos composicionales. Estas características junto con otras relaciones mineralógico-texturales presentes en estas rocas permitieron proponer la siguiente secuencia de cristalización: (i) cristalización de fenocristales de olivina, plagioclasa y piroxenos cálcicos; (ii) precipitación de una primera población de titanomagnetita, (iii) cristalización de plagioclasa y piroxenos conformando la matriz; (iv) disolución parcial de la primera población de titanomagnetitas por reacción con fluidos magmáticos; (v) cristalización de la segunda población de titanomagnetitas y finalmente, (vi) cristalización de barita intersticial.

Palabras clave: titanomagnetita, barita, diques máficos, Mesozoico, Uruguay

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Introduction

The Paraná Magmatic Province (PMP) is located in central South America, and it is one of the largest igneous provinces in the world (Figure 1a). It is mainly composed of flood basalts, dike swarms and sills, and other intrusive bodies related to the breakup of Gondwana and the opening of the South Atlantic Ocean (Peate 1997).

The age of this magmatism has been constrained to 138–120 Ma, with most ages between 132–129 Ma (Renne et al., 1992; 1996; Turner et al., 1994; Stewart et al., 1996; Deckart et al., 1998). Concomitant dike swarms (132–131 Ma; Féraud et al., 1999) distributed across the province are interpreted to be potential feeders of the sills and lava flows (Bellieni et al., 1984; Peate et al., 1992; Turner et al., 1994), providing a view of the upper levels of the plumbing system of the PMP. In Uruguay, the PMP covers an area of approximately 40,000 km^2 and consists of lava flows of basaltic composition and related intrusions (Figure 1b). The intrusive rocks resulting from mafic dike swarms and sills are known as the Cuaró Formation (Preciozzi et al., 1985), and they can be grouped according to their three main areas of occurrence: Melo, Tacuarembó and San Gregorio de Polanco. The first studies of this sub-volcanic unit only involved the post-Permian doleritic dikes that outcrop near the city of Melo (Walther, 1927; 1938). Féraud et al. (1999) were the first to relate these sills and dikes to the PMP, providing them with ^{40}Ar/^{39}Ar ages of approximately 132 Ma. Additional background information on the petrographic aspects and regional distribution of the Cuaró Formation can be found in Bossi and Shipilov (1997; 2007), Bossi (2006), Masquelin et al. (2009) and Muzio et al. (2012). The main petrographic features of these intrusions are the presence of glomeroporphyritic textures, plagioclase phenocrysts (An$_{55}$), a groundmass with plagioclase and clinopyroxene (augite – augite pigeonite) and partially iddingsitized relicts of olivine and titaniferous magnetite (Muzio et al., 2012).

Among all of the dike swarms in the Cuaró Formation, the San Gregorio de Polanco dike swarm (SGPDS) is the least-known, and it contains the southernmost mafic dikes related to the PMP in Uruguay (Figure 1c). Detailed petrologic studies concerning the SGPDS have been published recently by Masquelin et al. (2009) and Scaglia (2010). A remarkable mineralogical feature observed in these studies is the abundant presence (close to 10% of the modal volume) of two generations of iron-titanium oxides (Scaglia, 2010), as well as evidence of hydrothermal activity (Masquelin et al., 2009; Scaglia, 2010). The available lithochemical data outlined by Muzio et al. (2010) allow these dikes to be classified as basalts and andesitic basalts that correspond to the low-TiO$_2$ suite within the sub-alkaline tholeiitic series. Based on trace-element analyses, the parental composition of the magma indicates that these dolerites are related, with at least some contribution of a subcontinental lithospheric mantle (Bossi, 2006; Muzio et al., 2010; 2012). This is consistent with other Gondwana-related flood basalts and dikes in the province (Marques and Ernesto, 2004).

This work is focused mainly on characterizing the mineral content in the SGPDS by means of scanning electron microscopy (SEM) and presenting the main mineralogical and textural features of the iron-titanium oxides, as well as their relationship with other mineral phases.

Geological setting and petrography of the investigated dikes

The northwestern region of Uruguay is geologically underlain by the southern extreme of the Paraná basin, with ca 100,000 km^2 of exposed surface (Figure 1a) and sedimentary infill since the Devonian (Fúlfaro et al., 1997). The sedimentary pile, which has been identified in both outcrops and boreholes, is represented by 1) marine Devonian sediments (Durazno Group; Bossi, 1966); 2) a late Carboniferous to late Permian sequence composed, from base to top, of glacial sediments, deltaic to intra-cratonic marine sandstones, organic-rich claystones and aeolian sandstones (de Santa Ana et al., 2006); and 3) a late Jurassic intra-continental sedimentary sequence composed of siltstones and aeolian sandstones (Bossi et al., 1998).

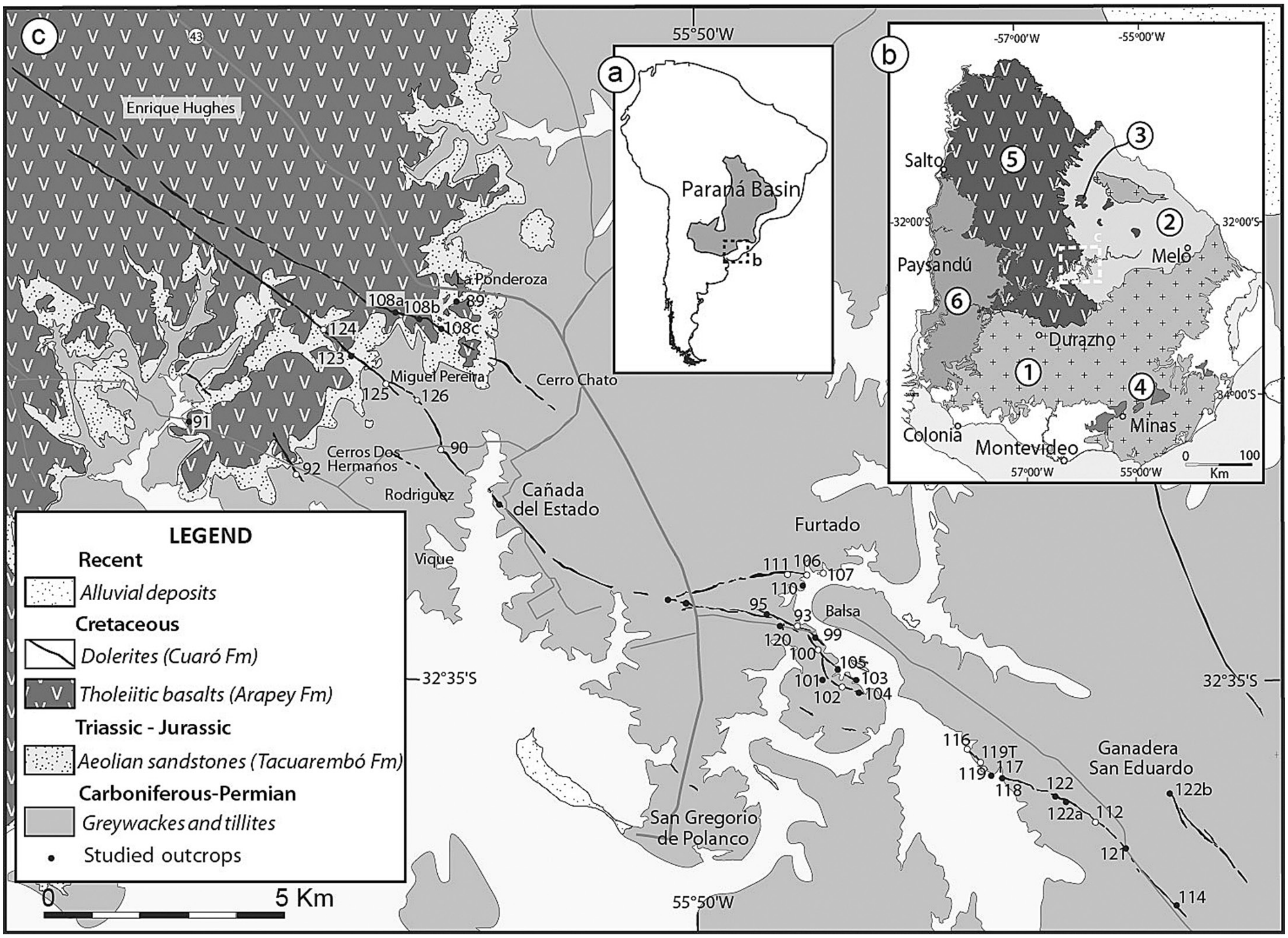

Figure 1 - A) Regional distribution of the Paraná basin; **B)** Location of the study area in Uruguay. References: 1 – Crystalline basement; 2 – Sedimentary infill of the Uruguayan extension of the Paraná Basin; 3 – Intrusions (sills) related to the Paraná Magmatic Province (Cuaró Fm); 4 – Early Cretaceous rhyolitic magmatism; 5 – Tholeiitic basalt flows of the Paraná Magmatic Province (Arapey Fm); 6 - Late Cretaceous sediments (Cuenca del Litoral del Rio Uruguay); c – study area, detailed in Figure 1c; **C)** Simplified geological framework of the San Gregorio de Polanco area.

These units are covered by Jurassic – Cretaceous Paraná basalt flows of the PMP, which are grouped under the name of the Arapey Formation (Bossi, 1966) and are over 1000 m thick (de Santa Ana et al., 2006). Later, sedimentation continued until the late Cretaceous, forming alluvial and fluvial siltstones and sandstones that are locally referred to as the Cuenca Litoral del Río Uruguay (Goso and Perea, 2003).

Based on geophysical and structural geological surveys, the regional structural framework of the Paraná basin is dominated by NW and EW alignments that controlled the depositional history of the basin and the associated magmatism in the early stages of Gondwana break up (Almeida, 1972; Fúlfaro et al., 1982; Hasui et al., 1993).

The target area is located near the geographic center of Uruguay (Figure 1b and c) in the southern portion of Tacuarembó Department and the northern portion of Durazno Department (between latitude 32°24’02”–32°51’07”S and longitude 56°05’52”–55°28’55”W), bordering the southern extreme of the basin. The SGPDS consists of doleritic dikes trending N140°–N160° that cross-cut Devonian, lower Permian and lower Jurassic sediments, as well as coeval basalts of the PMP. It follows the same general direction (N100°–N140°) as the eastern Paraguay and Ponta Grossa dike swarms, which are also part of the PMP (Deckart et al., 1998; Raposo and Ernesto, 1995).

Good exposures are infrequent and occur mainly near the village of San Gregorio de Polanco (Figure 1c). This paper focuses on 17 dike segments identified by Masquelin et al. (2009) and Scaglia (2010). They correspond to five major dikes classified as macro-dikes (after Rickwood, 1990). Their thicknesses vary from 5 to 20 m, and they are 15 to 60 km in

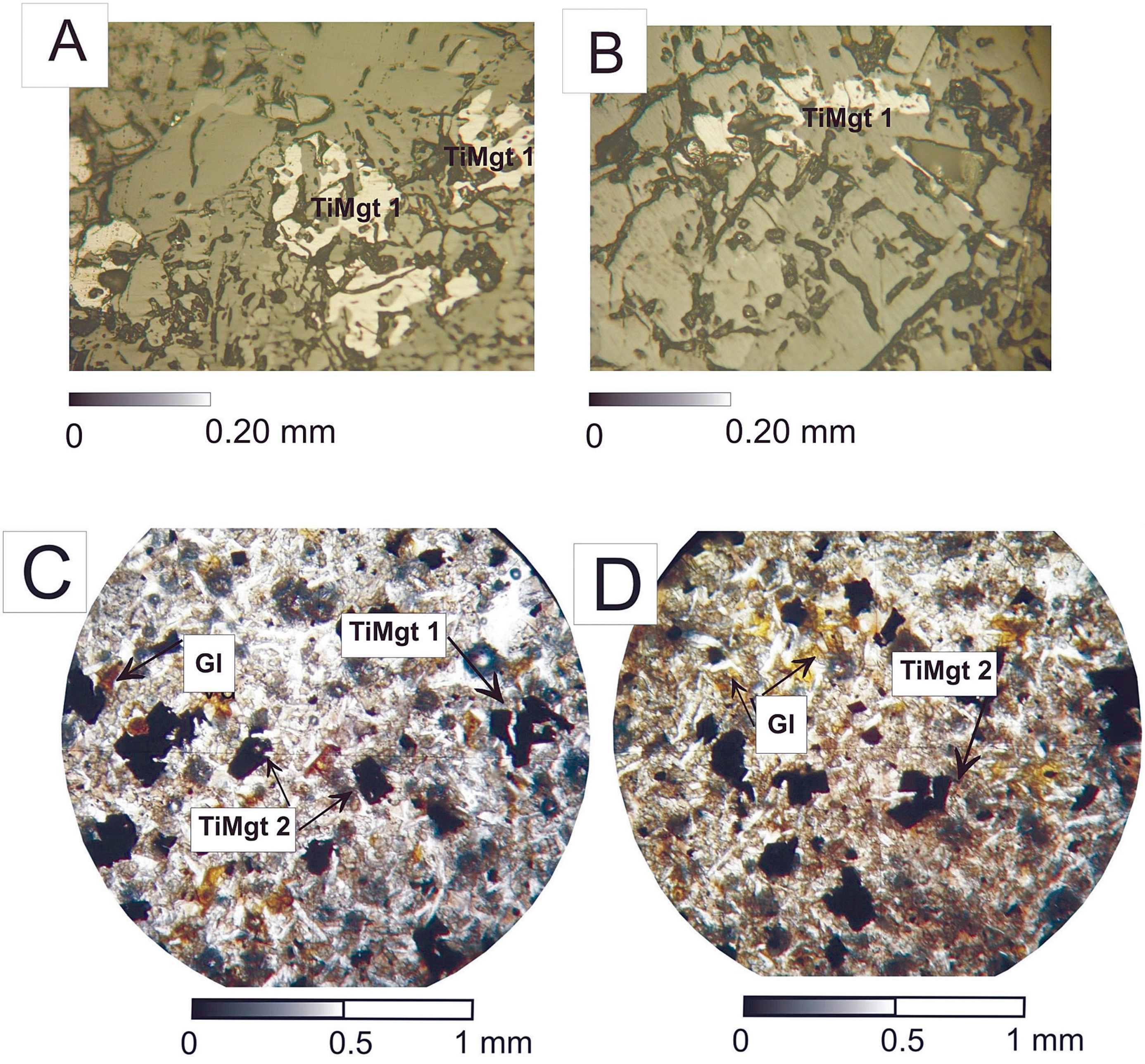

Figure 2 – Photomicrographs showing the textural arrangement and the mineral content of the dikes. **A** and **B**) Skeletal crystals of the first population of titanomagnetite (reflected light microscopy); **C)** Distribution of titanomagnetite with mainly skeletal shapes (first population). **D)** Titaniferous magnetite (small euhedral crystals of the second population) distributed in intersertal groundmass. Abbreviations: TiMgt 1 – titanomagnetite (first population); TiMgt 2 – titanomagnetite (second population); Gl - glass.

length. Field observations and thin-section descriptions of specimens from the SGPDS reveal a broad petrographic similarity. They crop out as typically grey, moderately altered dolerites characterized by glomeroporphyritic textures, with clusters of plagioclase up to 5 mm long and occasional clinopyroxene (augite), set in a fine-grained groundmass. The matrix frequently grades from a medium-fine grain in the center of the dikes to aphyric near the wall contacts, depending on the dike's thickness. No evidence of multiple intrusions has been observed in the field or in the thin sections (such as internal chilled margins), despite evidence of this process in some similar Mesozoic dike swarms to the northeast near the city of Melo (Masquelin et al., 2009). A zigzag emplacement pattern has been observed for most of the dikes, with minor dike segments trending N110°–120° to E-W and NNE-SSW. Regionally, the basement fractures and major lineaments comprise two main structural trends: N140°–N170° and N50°–N80°. Both are vertical to sub-vertical and affect some of the sedimentary rocks intruded by the dikes. The low dispersion of the dike segments relative to the surrounding material adjacent to the dikes suggests that the main orientation (N140°–N170°) is coincident with previous fracturing that was reactivated during the late Jurassic - early Cretaceous extensional events (Almeida, 1983; Hoeck, 1994 *in* Castaño and Druguet, 2008). Most of the structures found both in the host rocks and dike walls and the general orientation of the dike arrays suggest a NS to NNE–SSW finite extension direction, synchronous with diking, as postulated by Masquelin et al. (2009) for similar structural patterns in the northeastern part of the basin. Silicified sandstones near the dike walls that were produced by contact metamorphism, as well as chilled margins, have been identified in many dike segments. Hydrothermal veining is also observed, based on the presence of hematite, calcite or silica veinlets within the dikes and their host rocks.

Methodology

The objective of this paper is to deepen petrographic studies, and the results obtained from the various analytical techniques are discussed in separate sections.

Analytical techniques

The samples for petrographic descriptions and subsequent studies using Scanning Electron Microscopy – Energy Dispersive Spectrometry (SEM – EDS) techniques were selected from 37 outcrops (Figure 1c). Whenever possible, samples were collected at the center and edge of the dikes, depending on the state of weathering and fracturing.

Petrographic descriptions of 60 thin sections (textural arrangement and mineralogical content) were studied using a NIKON Eclipse – 50i polarizing microscope at the Faculty of Sciences, Uruguay. To avoid textural or mineral relationships that could be susceptible to misinterpretation, each sample was cut in different directions in order to locate a three-dimensional view of the texture in a single thin section. The textural interpretation criteria follow those of Hibbard (1995).

Later, 12 representative samples were analyzed and described using SEM – EDS (Jeol 5900 - Low vacuum equipment at the Faculty of Sciences, Uruguay), with particular emphasis on the opaque minerals. Analytical techniques that are routine for this type of equipment (Reed, 2005) were used. Due to the ubiquitous presence of similar mineralogical arrangements of interest, two representative samples were selected for compositional mapping either in the middle or close to the walls of the dikes.

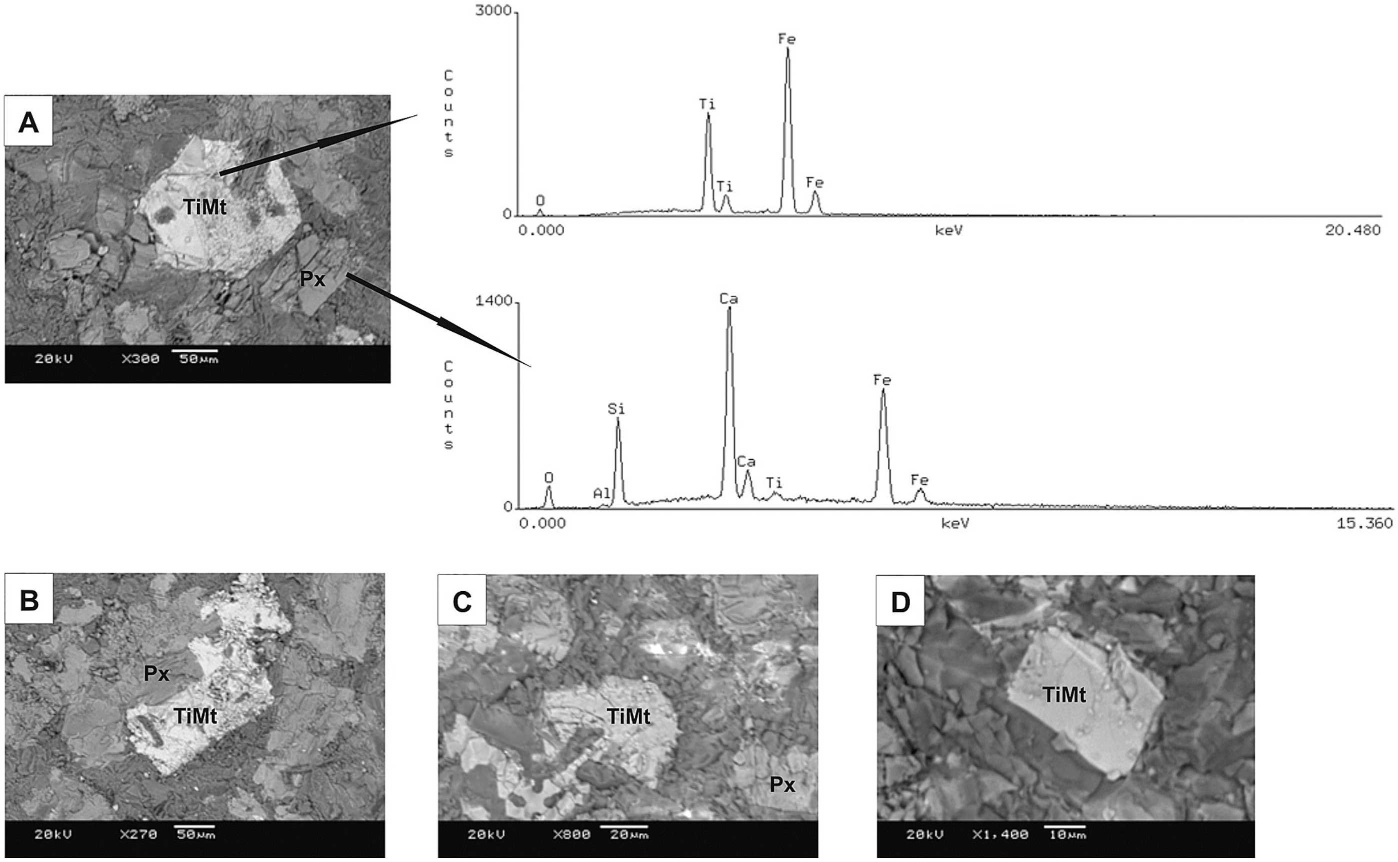

Figure 3 – (A) Well-shaped titanomagnetite (first population), partially surrounding an augite phenocryst; BSE image with EDS spectra. **(B)** Euhedral titanomagnetite with corroded borders, partially including an augite phenocryst; BSE image. **(C)** Skeletal titanomagnetite; BSE image. **(D)** Second population of titaniferous magnetite. Abbreviations: Px - pyroxene; TiMgt - titanomagnetite.

Petrography and SEM – EDS results

The mineral assemblage of the groundmass consists of plagioclase (An_{55}), clinopyroxene (augite), and relicts of olivine (iddingsitized, with celadonite also present) and opaque minerals. The presence of some interstitial glass has been observed, which causes the matrix mineral array to vary from sub-ophitic to intersertal textures (Figure 2). Comparison with previous petrographic descriptions in Peate (1997), Deckart et al. (1998), Bossi and Schipilov (2007) and Masquelin et al. (2009) indicates that similar dolerites are common throughout the PMP.

Two populations of opaque minerals have been noted by Scaglia (2010). SEM-EDS analysis shows that the opaque minerals are Fe–Ti oxides, with a homogeneous distribution in the matrix that corresponds to titaniferous magnetite (also known as ulvospinel) and a chemical composition of Fe_2TiO_4 (Figure 3; Sial and McReath, 1984). These iron-titanium oxides represent approximately 10% of the modal mineralogical composition, and they are characterized by different petrographic features. One population occurs as euhedral crystals, with a mean size of 0.5 mm and frequently embayed borders leading to skeletal shapes. The second population consists of euhedral grains ranging in size from 0.1 to 0.3 mm. The Ti – magnetite crystals from the centers of the dikes mainly have skeletal shapes, most likely as a result of dissolution processes. They have rhombic, rectangular, square and skeletal shapes, and they sometimes include pyroxene crystals. On one hand, the presence of two populations of titaniferous magnetite indicates that precipitation occurred at different stages during magmatic crystallization. On the other hand, the presence of Ti-magnetite crystals along the cleavage of plagioclase and clinopyroxene phenocrysts is indicative of relatively early crystallization.

The geometry of the opaque minerals described here is found both in the center of the dikes and on the borders, with no significant differences. However, the titanomagnetites in the centers of the dikes (mainly from the earlier population), are skeletal but partially preserve the original shape (rhombic, square or prismatic; Figure 3a – c). This may be due to interaction and reabsorption processes between early Ti-magnetite crystals and the residual magmatic fluid (Hibbard, 1995).

Interstitial light-colored (bright white) xenomorphic crystals were also identified. Further EDS-BSE (backscattered electrons) analyses indicate that they are barite crystals ($BaSO_4$), Figure 4a. The barite crystals in the centers of the dikes are up to 50 µm in diameter, and they have not been identified near the dike walls. Samples from the contact between the dolerite and the host rock contain sandstones of the Tacuarembó Formation (Bossi, 1966), and they have also been analyzed to observe possible metamorphic assemblages. The petrographic studies revealed the presence of fine grains of illite between quartz and feldspar, implying a very low degree of contact metamorphism. The analyses and interpretations of SEM images and X-ray spectra also indicate the penetration of magma through the host rock due to the porosity of the sandstone, which is verified by the crystallization of pyroxenes and titanomagnetite crystals in the sandstone.

Compositional mappings

The petrographic study using SEM analysis was complemented with compositional mappings (SEM – BSE images). These images show the chemical distribution based on the main element abundances in the analyzed section, in this case, Si, Ca, Fe and Ti. These results were used to complement the interpretation of the petrogenetic processes. Two re-

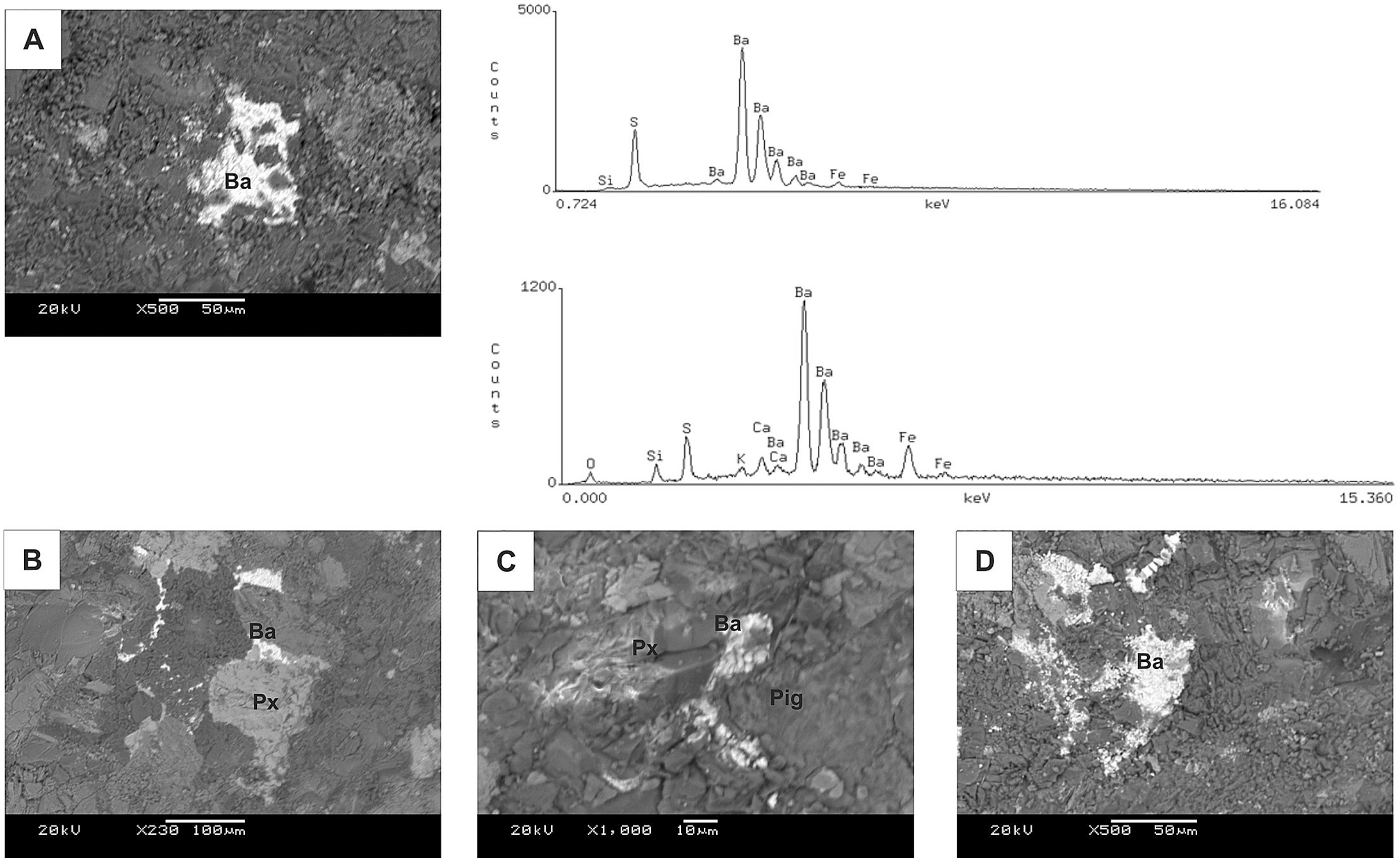

Figure 4 – (A) Pyroxene and titanomagnetite crystals (second population), including barite crystals; BSE image and EDS spectrum. **(B, C** and **D)** Interstitial barite between plagioclase and pyroxene crystals; BSE images. Abbreviations: Ba - barite; Plg - plagioclase; Px - pyroxene.

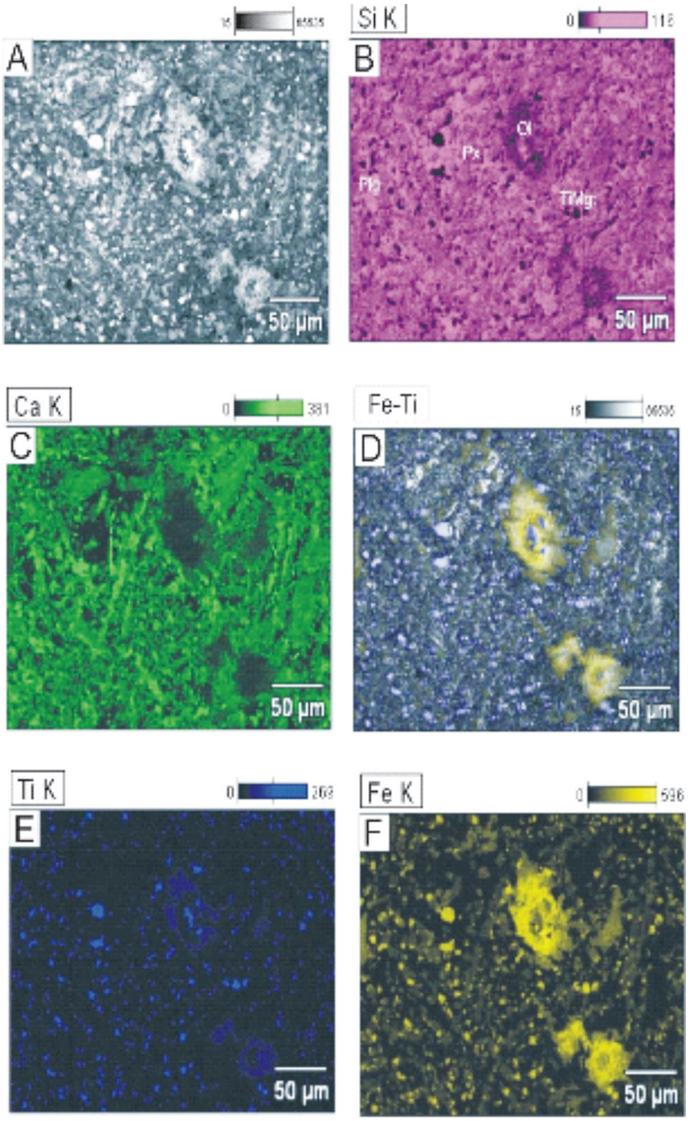

Figure 5 - (A) Compositional mapping of a dike; BSE images. (**B, C, E** and **F**) Distribution of the main elements, Si, Ca, Ti and Fe, in different mineral phases in the studied section. (**D**) Distribution of Fe and Ti using superimposed BSE images.

presentative samples were selected based on conventional petrography and SEM studies: one corresponding to the center of the dike and one near the contact with the doleritic host rock. The compositional maps are shown in Figures 5 and 6. The main mineral phases were identified in the compositional images, and the main concentration of Si is observed to be distributed in plagioclase crystals, with minor amounts in olivine and pyroxene. Areas with no Si correspond to the Ti-magnetite crystals (Figure 5b).

Figure 5c shows a similar distribution for Ca, with the highest concentrations in plagioclase and pyroxene crystals. Figure 5e and f shows that the highest concentrations of Ti are limited to some portions of the sample, coincident with the sites of titanomagnetites. The distribution of Ti is also concordant with the distribution of Fe (Figure 5d). Although most of the Fe and Ti corresponds to the Fe-Ti oxides, some is also present in olivine and pyroxene. The distributions of Ti, Fe and Si observed in Figure 6 (c, d and e, respectively) are similar to those in Figure 6a. However, this sample corresponds to the center of the dike and has some areas that are not well defined by BSE images, indicated by bright white (Figure 6a). These areas correspond to high Ba concentrations, described previously as barite.

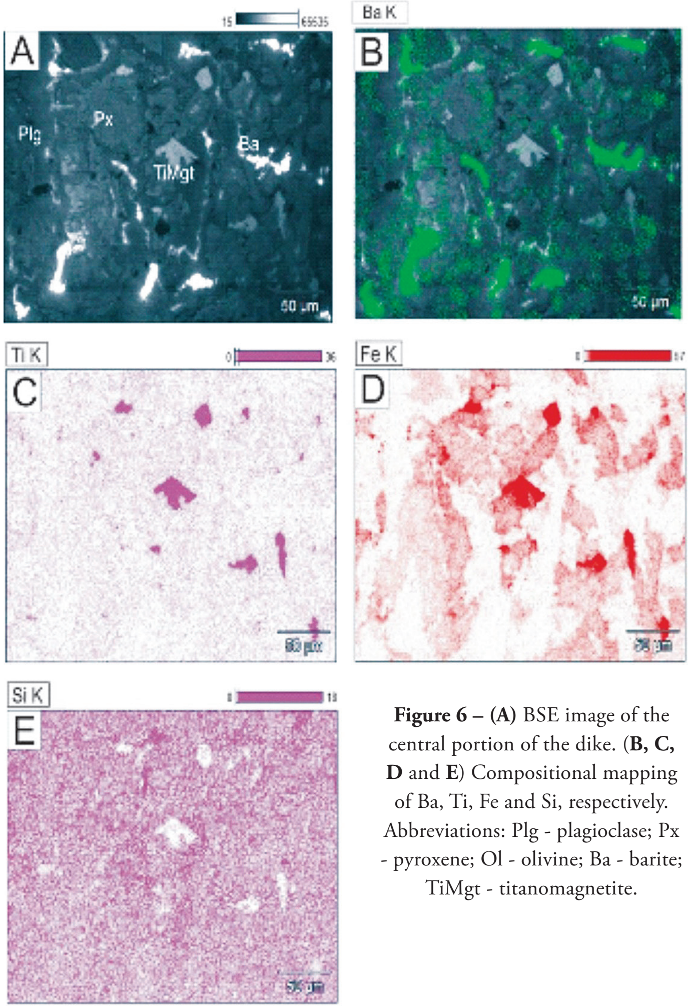

Figure 6 – (A) BSE image of the central portion of the dike. (**B, C, D** and **E**) Compositional mapping of Ba, Ti, Fe and Si, respectively. Abbreviations: Plg - plagioclase; Px - pyroxene; Ol - olivine; Ba - barite; TiMgt - titanomagnetite.

Discussion

The presence of barite, the high percentage of titanomagnetite crystals with skeletal shapes, the existence of reabsorption borders in plagioclases and olivines and the occurrence of oscillatory zoning in plagioclase phenocrysts, partially altered to sericite, are clear evidence of the concentration of fluids in the centers of the dikes, which is most likely driven by differentiated flow processes (Hibbard, 1995; Rickwood, 1990). This would create a "wall effect" with low permeability that causes the migration of residual fluids to the center of the dike. Consequently, the high concentration of fluids in the centers of the dikes over a long time period would have allowed the reaction with early formed crystals. After a gradual decrease in temperature, the second population of titaniferous magnetite would have crystallized in the late stages of magmatic crystallization.

Textural variations of the dikes can also be attributed to the disparity in the rate of flow, as proposed by Komar (1976). As the viscosity increased, phenocrysts would not have been displaced, but would rather have clumped together in a hypocrystalline matrix. These variations, as well as differences in phenocryst abundances, can be observed in several samples.

Additionally, unmixing processes affecting augite-pigeonite are common in plutonic and hypabyssal rocks (Sial and McReath, 1984). Their presence as exsolution lamellae, both in the groundmass and phenocrysts, suggest the presence of metastable solid solutions under well-constrained temperature and pressure conditions. Therefore, the presence of augite-pigeonite can be used as a geothermometer, indicating that unmixing processes occurred at 950°–1100°C (Brown, 1967). Finally, the genesis of Fe-Ti oxides may have occurred as magmatic segregation, partially crystallizing with the other silicate phases (Guilbert and Park, 1986). Crystallization temperatures of 590°C and 450°C are estimated for the first and for the second population, respectively.

As noted previously, SEM-EDS analyses also highlight the presence of barite ($BaSO_4$) as interstitial xenomorphic crystals, ranging in size from 10 to 50 µm. As crystallization continued, the residual liquid enriched in sulfate and Ba, either as a product of crustal assimilation or from the contribution of hydrothermal fluids crystallizing as barite at low P – T conditions. The source of these hydrothermal fluids can either be magmatic (juvenile waters) or related to connate waters and juvenile waters (Guilbert and Park, 1986). According to our field data and as noted by Montaño et al. (2006), these barite-bearing dikes are mainly hosted in the San Gregorio Formation, which is characterized by SO_4^{-2}, Cl^-, F^- and Ba^{+2} anomalies. Hence, the S content could be magmatic, in association with the groundwater contribution and/or assimilation processes. However, hydrothermal fluids from mafic magmas usually have low pH values, but they progressively increase while the magma rises and assimilation processes occur (Hibbard, 1995). These pH variations in the SGDS could have been controlled by the oxidation of the organic matter available in Devonian sediments (3 to 3.6% organic matter; Bossi, 1966). According to Pons et al. (2009), the organic content may change the pH in fluids, favoring the precipitation of dissolved metals (barite, in this case). Furthermore, this pH variation in the residual fluids may have also favored the crystallization of the second population of Ti-magnetite. For the samples in this study, there would have been reducing conditions with temperatures of approximately 450ºC, while the temperature would have varied between 350ºC–100ºC for barite precipitation (Guilbert and Park, 1986).

Conclusions

The information obtained by SEM–EDS/BSE analysis indicates that both populations of opaque minerals correspond to titaniferous magnetite.

The mineralogical and textural features observed suggest that fluids were concentrated in the centers of the dikes.

SEM-EDS analyses indicate the presence of interstitial xenomorphic barite crystals, which were produced by the crystallization of hydrothermal fluids. The source of these hydrothermal fluids is postulated to be either magmatic or related to the mixing of connate waters with juvenile waters.

A crystallization sequence for the SGDS is proposed, inferred from textural observations and mineralogical relationships: a) crystallization of olivine, plagioclase and calcium-rich pyroxene phenocrysts; b) precipitation of the first population of titaniferous magnetite at approximately 600ºC; c) crystallization of plagioclase and pyroxene from residual magmatic fluids, developing the groundmass; d) partial dissolution of the first population of titaniferous magnetite crystals (resulting in skeletal shapes) through the reaction with magmatic residual fluid; e) crystallization of the second population of titaniferous magnetite from Ti present in the residual magmatic fluid, whose source was the previous dissolution of the first population of Ti-magnetite; the low fugacity of oxygen and a temperature close to 450°C would have been necessary for the second generation of titanomagnetite; and f) crystallization of barite from hydrothermal fluids rich in S and Ba. The precipitation of barite indicates stable reducing conditions and temperatures between 100°C and 350°C.

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