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Blue quartz in Brazil: the Rio dos Remédios occurrence – preliminary study

Adrian-Iulian PANTIA[1,2], Danielle Cruz da SILVA[3] & Andra-Elena FILIUȚĂ[1,2]

[1] Geological Institute of Romania, Caransebeș Street 1, 012271, District 1, Bucharest, Romania (padrian.iulian2020@gmail.com, f.e_andra@yahoo.com).

[2] Faculty of Geology and Geophysics Doctoral School, University of Bucharest, Traian Vuia Street 6, 020956, District 1, Bucharest, Romania.

[3] Federal University of Pernambuco, Acadêmico Hélio Ramos, s/n, Cidade Universitária, 5074-530, Recife, Pernambuco, Brazil (dani.cs8@live.com).

Abstract. Rock forming blue quartz occurrences are reported on all continents, but little is known about the specific causes for the coloration, and even less about its geological significance. The present study investigates the occurrence hosted by the metarhyolites of the Rio dos Remédios Group in order to determine the potential causes of the blue coloration and gain insight into the geological significance of the occurrence in the wider context of Brazilian geology. The optical observations have shown that the blue color is likely the result of the Rayleigh scattering of light. The color is stable even at temperatures of 800°C. The FT-IR investigations have not detected the presence of inclusions capable of scattering light. The scanning electron microscopy has shown that there are significant amounts of Fe and Ti in the system, clearly concentrated as Ti/Fe oxides hosted by the matrix, and potentially as light scattering nanometric inclusions. This type of inclusions would likely be syngenetic with the host quartz. The alternation between the blue and colorless areas could be the result of chemical fluctuations in the melt, which may have resulted in disruptions of the crystallization of the light scattering inclusions.
Keywords. Blue quartz, Rayleigh scattering, Rio dos Remédios, syngenetic crystallization.

Resumo. Quartzo azul no brazil: ocorrência Rio dos Remédios – estudo preliminar. Ocorrências de rochas contendo cristais de quartzo azul são relatadas em todos os continentes, mas pouco se sabe sobre as causas específicas para a coloração e menos ainda sobre seu significado geológico. O presente estudo investiga a ocorrência hospedada nos metariolitos do Grupo Rio dos Remédios, a fim de determinar as possíveis causas da coloração azul e obter uma visão mais ampla sobre o significado geológico no contexto da geologia brasileira. As observações ópticas mostraram que a cor azul é provavelmente o resultado da dispersão de luz pelo espelhamento de Rayleigh. A cor é estável mesmo em temperaturas de 800°C. As investigações FT-IR não detectaram a presença de inclusões capazes de espalhar luz. A microscopia eletrônica de varredura mostrou que existem quantidades significativas de Fe e Ti no sistema, claramente concentradas como óxidos de Ti/Fe hospedados pela matriz e, potencialmente, como inclusões nanométricas de espalhamento de luz. As inclusões são provavelmente singenéticas com o quartzo hospedeiro. A alternância entre as áreas azuis e incolores pode ser o resultado de flutuações químicas no magma, o que pode ter resultado em interrupções na cristalização das inclusões de espalhamento da luz.
Palavras-chave. Quartzo azul, espalhamento Rayleigh, Rio dos Remédios, cristalização singenética.

1 Introduction

The scarce literature on rock forming blue quartz attributes the color to the scattering of light by submicron/nanometric inclusions, usually ilmenite or rutile, but also biotite, magnetite, graphite, and others. The consensus is that blue quartz is usually hosted by plutonic and volcanic intermediary to acidic rocks (or their metamorphic equivalent) and that, when hosted by metamorphic rocks, the occurrences are related to amphibolite-granulite facies conditions, as summed up by Pantia & Filiuță (2019). The geological significance of these occurrences is largely undetermined (Zolensky *et al*., 1988; Seifert *et al*., 2011), although potential applications of metamorphic blue quartz in determining the metamorphic history of the area of occurrence have been suggested (Pantia & Filiuță, 2021).

Out of the ca. 290 rock forming blue quartz locations (referred to as locations rather than occurrences, because there are instances where multiple locations intercept the same blue quartz bearing stratigraphic unit) reported worldwide, 19 are reported in Brazil, in close association with regional tectonic features and Au, Sn, and U deposits. When plotted on a geological map, the Brazilian occurrences show a pattern consistent with major geological features such as the Paramirim Aulacogen, Rio Itapicuru and Grão Pará Greenstone Belts, Goiás Tin Province, Brasilia Belt, or Brasiliano/Pan-African orogen (Dom Feliciano Mobile Belt), as shown in figure 1.

The age of the blue quartz bearing rocks ranges from 2760 ± 11 to 2757 ± 7 Ma, to 580 ± 7 Ma, as determined for the felsic (rhyolitic) volcanic rocks of the Grão Pará Group (Trendall *et al*., 1998) and the charnockites of the Porangatu Granulite Complex (Gorayeb *et al*., 2017), respectively. Whether the quartz developed the blue color during the initial crystallization, and therefore the color is the same age as the host rock, or it is the product of subsequent metamorphic events has not yet been determined. All of the Brazilian occurrences are related to major geological events. In some instances, multiple occurrences can be correlated to a single magmatic or tectonic episode, such as the ca. 1700 - 1400 Ma (Marini *et al*., 1986; Moura *et al*., 2014) granitic intrusions of the Goiás Tin Province (Soledade, Serra da Mesa, Sucuri, Mangabeira and Pedra Branca) associated with the evolution of the Arai Rift, or the ca. 1752 - 1746 Ma (Lobato *et al*., 2015; Magalhães *et al*., 2018) intrusions related to the Paramirim Aulacogen (Rio dos Remédios rhyolites and São Timóteo granite). The 1766 ± 4 Ma (Pereira, 2018) blue quartz bearing metavolcanic rocks (dacitic/rhyolitic) in the base of the Central Espinhaço Range are also related to the rifting events which took place ca. 1750 Ma ago.

The rest of the occurrences are related to: _*1 – collisional and metamorphic events*_, such as the 2050 Ma (Rosa *et al*., 2000) Guanambi Batholith related to the Transamazonian Cycle, the 2760 ± 11 - 2757 ± 7 Ma (Trendall *et al*., 1998) felsic volcanic rocks of the Grão Pará Group (Cristalino IOCG deposit) and the 2128 ± 8 Ma (Mello *et al*., 2006) Lagoa do Gato quartz-feldspar porphyry of the Grão Pará and Rio Itapicuru Greenstone Belts respectively, the 580 Ma (Gorayeb *et al*., 2017) Porangatu Granulite Complex charnockites resulted from the Amazon craton collision with the São Francisco - Congo cratons, the charnockites of the Santa Catarina Granulite Complex with a 2720 Ma protolith which underwent granulite facies metamorphic episodes 2680 and 2170 Ma ago (Hartmann *et al*., 2000), the 2552 Ma (Giustina *et al*., 2009) Campinorte Sequence metarhyolite of the Brasilia Belt, the 2157 ± 5 Ma (Cid *et al*., 2000) granite of the Couro de Onça Intrusive Suite related to the Transamazonian Cycle, the Brasiliano/Pan-African 587 ± 8 Ma (Junior *et al*., 2011) Zimbros Intrusive Suite rhyolite, the ca. 2040 - 2060 Ma (Neves *et al*., 2020) Sucuru granite associated with subduction-collisional events linked to the assembly of the Nuna/Columbia supercontinent, or _*2 – within-plate magmatism*_, as is the case with the 1871 ± 5 Ma (Costi *et al*., 2009) Abonari granite of the Mapuera Intrusive Suite, Amazon Craton, or the 2006 ± 24 Ma (Sparrenberger & Tassinari, 1999) Riacho dos Cavalos pegmatite of the Granite-Gneiss Complex (Goiás Tin Province). Similar connections between blue quartz occurrences and regional features have been reported by Pantia & Filiuță (2019).

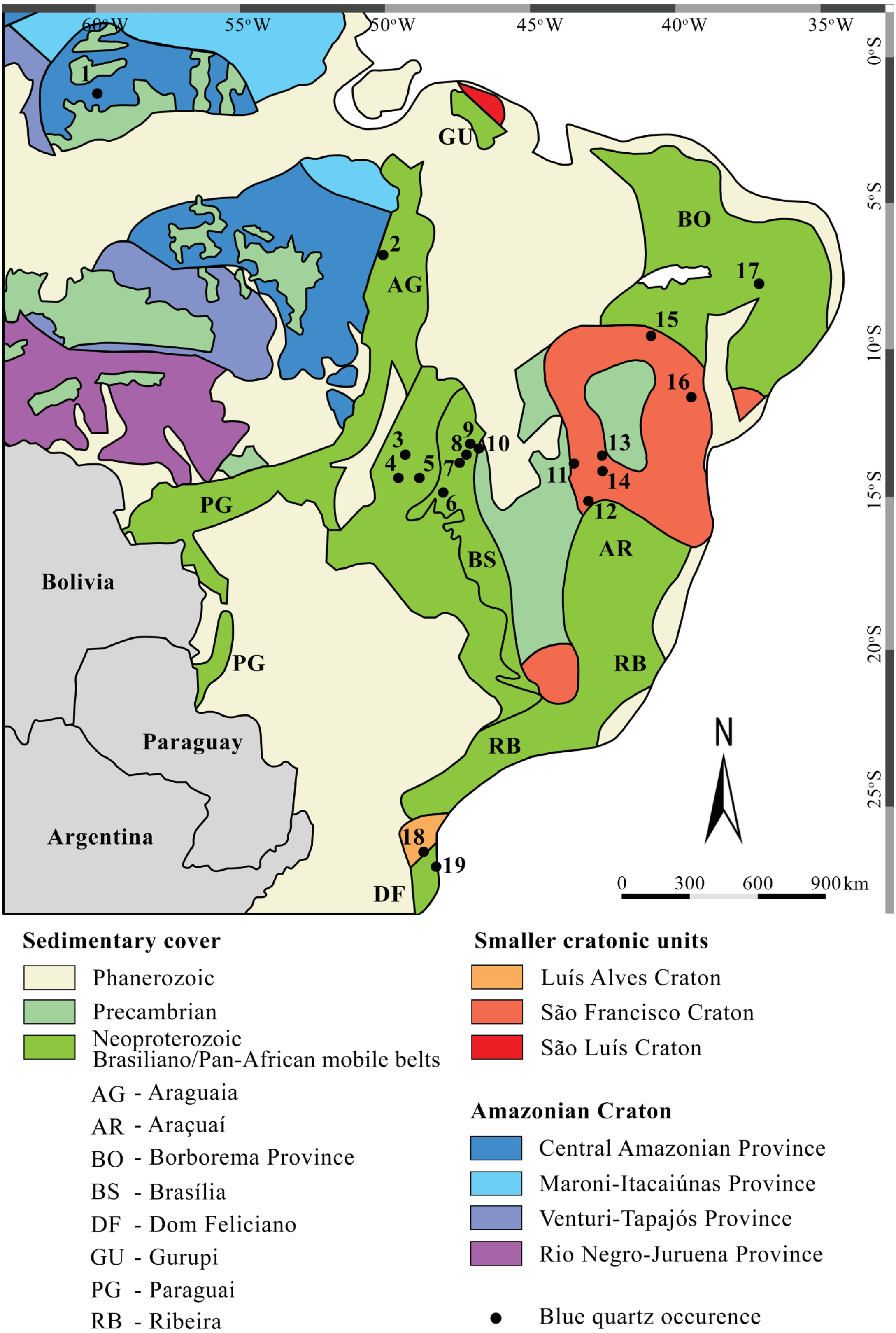

Figure 1. Geological sketch map showing the general location of rock forming blue quartz occurrences in Brazil: 1 – Abonari; 2 – Cristalino; 3 – Porangatu; 4 – Campinorte; 5 – Serra da Mesa; 6 – Sucuri; 7 – Soledade; 8 – Pedra Branca; 9 – Riacho dos Cavalos; 10 – Mangabeira; 11 – Guanambi Batholith; 12 – Central Espinhaço; 13 – Rio dos Remédios; 14 – São Timóteo; 15 – Couro de Onça; 16 – Lagoa do Gato; 17 – Sucuru; 18 – Santa Catarina Granulite Complex; 19 – Zimbros (Modified after Paquette et al., 2015 and Queiroz & Klein, 2018).

Figura 1. Mapa geológico indicando ocorrências de rochas portadoras de quartzo azul no Brasil: 1 – Abonari; 2 – Cristalino; 3 – Porangatu; 4 – Campinorte; 5 – Serra da Mesa; 6 – Sucuri; 7 – Soledade; 8 – Pedra Branca; 9 – Riacho dos Cavalos; 10 – Mangabeira; 11 – Guanambi Batholith; 12 – Espinhaço Central; 13 – Rio dos Remédios; 14 – São Timóteo; 15 – Couro de Onça; 16 – Lagoa do Gato; 17 – Sucuru; 18 – Complexo Granulítico de Santa Catarina; 19 – Zimbros. (Extraído de Paquette et al., 2015 e Queiroz & Klein, 2018).

Out of the 19 Brazilian occurrences, nine show an apparent connection to various mineral deposits as follows: Sn – Soledade, Serra da Mesa, Sucuri, Mangabeira, Pedra Branca (Lenharo *et al*., 2002) and Riacho dos Cavalos (Sparrenberger & Tassinari, 1999); Au – the Cristalino Deposit (Craveiro *et al*., 2019) and Lagoa do Gato (Mello *et al*., 2006); U – São Timóteo (Cordani *et al*., 1992). Similar associations have also been observed in Canada, South Africa and Australia. It is unclear whether the blue quartz is indicative of conditions which enable the formation of such deposits, or the observed association is a mere coincidence.

While limited in scope, the present study aims to lay the groundwork for future research on blue quartz, with special emphasis on the rhyolite hosted occurrence of Rio dos Remédios, and to draw attention to the question of blue quartz in Brazil. The Brazilian occurrence will be compared to the occurrences of Albești (Romania) and Llano (Texas, USA).

2 Geological setting

2.1 The Rio dos Remédios Group

The Rio dos Remédios Group was initially described by Schobbenhaus & Kaul (1971) and is comprised of acidic and intermediate metavolcanic, pyroclastic, and sedimentary rocks, deposited in a continental environment (Schobbenhaus, 1996), which were subsequently metamorphosed. The group is located on the São Francisco-Congo paleoplate (Fig. 2), forming the base of the Espinhaço Supergroup, and represents the bimodal magmatism of the initial phase of a succession of rift/syneclise stages which occurred between 1750 and 670 Ma (Pedrosa-Soares & Alkmim, 2011).

The Group is subdivided into the Novo Horizonte Formation, Lagoa de Dentro Formation, and Ouricuri do Ouro Formation (Guimarães *et al*., 2012). The Novo Horizonte Formation includes volcanic, subvolcanic, pyroclastic, and epiclastic (of volcanic derivation) lithofacies. The Lagoa de Dentro Formation corresponds to the continental sedimentation in the wake of the volcanism/plutonism of the Novo Horizonte Formation (Guimarães *et al*., 2008), and represents the top of the Rio dos Remédios Group. The Ouricuri do Ouro Formation is essentially made up of conglomerates and coarse sandstones which show cross-stratification (Garcia, 1999).

In the study area, near the city of Paramirim in central eastern Bahia, the group is characterized by metarhyolites, alkaline metarhyolites, and metarhyodacites belonging to the Novo Horizonte Formation, characteristically porphyritic with phenocrysts of quartz, potassium feldspar and plagioclase. These rocks were largely affected by metasomatic processes of silicification (Oga, 1997).

2.2 The Albești metagranite

The 481.16 ± 7.3 Ma Albești metagranite is located in the Leaota Metamorphic Suite, part of the Iezer Complex, South Carpathians, Romania. The origin of the granite is still disputed, but crustal anatexis during the middle/late stages of a collision or the emplacement of magmatic material along tectonically enabled pathways are the prevailing models (Negulescu, 2013; Negulescu *et al*., 2018). According to Negulescu & Săbău (2015), the granite underwent an amphibolitic facies metamorphic event, at 600°C and 10 kbar ca. 300.9 ± 11.1 - 318.12 Ma ago (Negulescu, 2013). The metagranite has a peraluminous calc-alkaline character, a predominantly subunitary Na_2O/K_2O ratio, and $Na_2O + K_2O$ ranges from 5 to 8.66 wt%. The CIPW normative composition places the Albești metagranite in the monzogranite field, in close proximity to the granodiorite (Robu & Robu, 1996). It is worth mentioning that not all Albești metagranite outcrops show blue quartz, and it is possible that the outcrops which experienced more intense metamorphic conditions lost the blue coloration in their rock forming quartz as it recrystallized (a metamorphic effect observed in other occurrences as well), as suggested by Pantia & Filiuță (2021).

The blue quartz hosted by the metagranite shows features associated with a color produced by the scattering of light inside the grain. The grains have an opalescent-milky appearance, with no indication of chatoyancy. The color appears to be sensitive to temperatures as low as

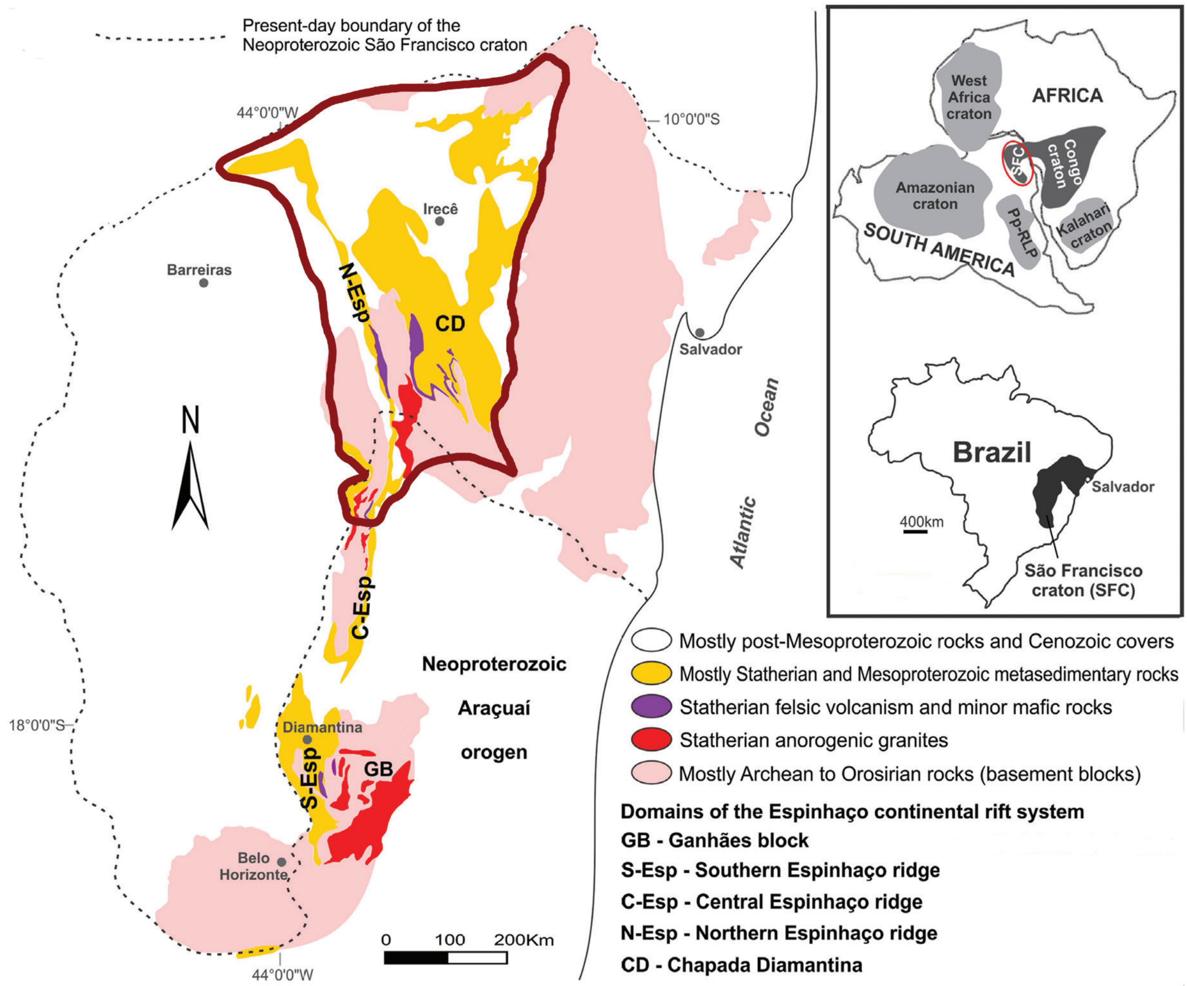

Figure 2. Simplified geological map of the Paramirim Aulacogen and the Rio dos Remédios Group (Modified after Magalhães *et al*., 2018).
Figura 2. Mapa geológico simplificado do Aulacógeno Paramirim e do Grupo Rio dos Remédios (Extraído de Magalhães et al., 2018).

300°C, but new unpublished results suggest that the perceived loss of color only occurs in grains with highly opalescent areas and may actually be an illusion caused by the bleaching and opacization of the aforementioned zones, which overlap with the blue ones. The grains show an apparent color zoning, which is actually the effect of subgrain rotation recrystallization. The marginal recrystallized grains and embayments are colorless, suggesting that recrystallization is incompatible with the blue color in this particular occurrence. The quartz grains which did not recrystallize show strong undulose extinction. Inclusions mostly consist of biotite, but iron oxides, pyrite, and chalcopyrite were also identified under the electron microscope. When present, the biotite/iron oxide inclusions tend to concentrate in the colorless areas of the quartz grains (Pantia & Filiuță, 2021). The identity of the scattering particles responsible for the blue color is still unknown.

2.3 The Llano blue quartz

The Llano blue quartz is hosted by the 1106 ± 6 Ma (DeLong & Long, 1976) Llano rhyolite (also known as llanite), part of the Llano Uplift of central Texas, generated during the collisional Grenville orogeny at the southern edge of Laurentia (Mosher *et al*., 2008) in the larger context of the assembly of the Rodinia Supercontinent.

The matrix makes up ca. 57.2 vol%, quartz ca. 14.2 vol%, and the feldspars ca. 18.6 vol%. As a whole, the llanite is composed of 42 vol% quartz, 27.6 vol% microcline, 27.2 vol% plagioclase, 2 vol% biotite, and 1.2 vol% other minerals (Barker & Burmester, 1970).

According to Zolensky *et al*. (1988), the llanite underwent subgreenschist facies metamorphic conditions which, on account of the low PT conditions (ca. 2 - 3 kbar and 250 - 350°C) would place the blue quartz in the category of pure magmatic/volcanic rock forming blue quartz (i.e. metamorphic conditions had no contribution to the generation of the color).

The quartz grains themselves show an intense blue color, which remains unchanged even at temperatures of 1,000°C (Pantia & Filiuță, 2021), and are strongly chatoyant. The grains show subhedral to euhedral contours, often broken by embayments. The color is produced by light scattering nanometer size (ca. 60 nm) ilmenite inclusions, with a spatial density of up to ca. 125 inclusions/μm^3 which is directly proportional to the intensity of the blue color, and manifests a core-edge zoning caused by an increase in the size (100 nm - 20 μm) of the ilmenite inclusions, which translates into a progressive inability to scatter light (Zolensky *et al*., 1988).

The microscopic observations have not shown strain shadows or recrystallization of the quartz grains. Fluid inclusions are present, as well as fine, needle like, inclusions (possibly ilmenite), which locally form crisscross patterns under a 60° angle and may be responsible for the observed chatoyancy, but with no contribution to the blue color on account of their large dimensions.

Unlike the blue quartz from Albești, which is likely a representative of metamorphic blue quartz, the Llano occurrence may be regarded as the quintessential magmatic/volcanic blue quartz.

3 Materials and methods

3.1 Sampling

The investigated samples from the Rio dos Remédios Group were collected from outcrops located in the southwest of the State of Bahia, in the Canabravinha district, municipality of Paramirim, ca. 670 km west of the city of Salvador, the capital of the State of Bahia (Fig. 3).

The samples collected are composed of three petrographic types, namely alkali rhyolite, metarhyodacite, and metarhyolite porphyry (Guimarães *et al.*, 2008), shown in figure 4A, B and C.

The Llano blue quartz (Fig. 4D) was available courtesy of Professor Mark Helper, PhD, University of Texas at Austin, and was collected at the following coordinates: 30°53'26.3"N, 98°39'30.57"W.

The Albești metagranite (Fig. 4E) was sampled from the Dragoslavele village, upstream the Valea Caselor River, ca. 7 km from its confluence with the Dâmbovița River, Argeș County, Romania (45°20'59.74"N, 25°12'33.48"E).

3.2 Methods

Given the small scale of the light scattering inclusions, generally less than 100 nm, blue quartz is difficult to study by using the commonly available methods, and TEM and EMPA techniques are required. However, it is possible to gather data which, although insufficient for the determination of the inclusions, could provide a starting point for future research.

The Llano (Texas, USA) rhyolite blue quartz is the standard against which Precambrian blue quartz occurrences are compared, since it is the best studied occurrence to date. The blue quartz of the Albești metagranite (Romania) will also be used for comparison.

The Rio dos Remédios blue quartz has been investigated at the Geological Institute of Romania using optical means, heat treatment, infrared spectroscopy, and electron microscopy.

3.2.1 Optical methods

The optical observations were performed, using both binocular and petrographic microscopes, on thin and polished sections, polished hand samples, grains, and thin hand sample slices. When feasible, the investigations were carried out in reflected and transmitted light, in order to assess the chromatic behavior of the samples in connection to the light scattering mechanisms for producing the blue color.

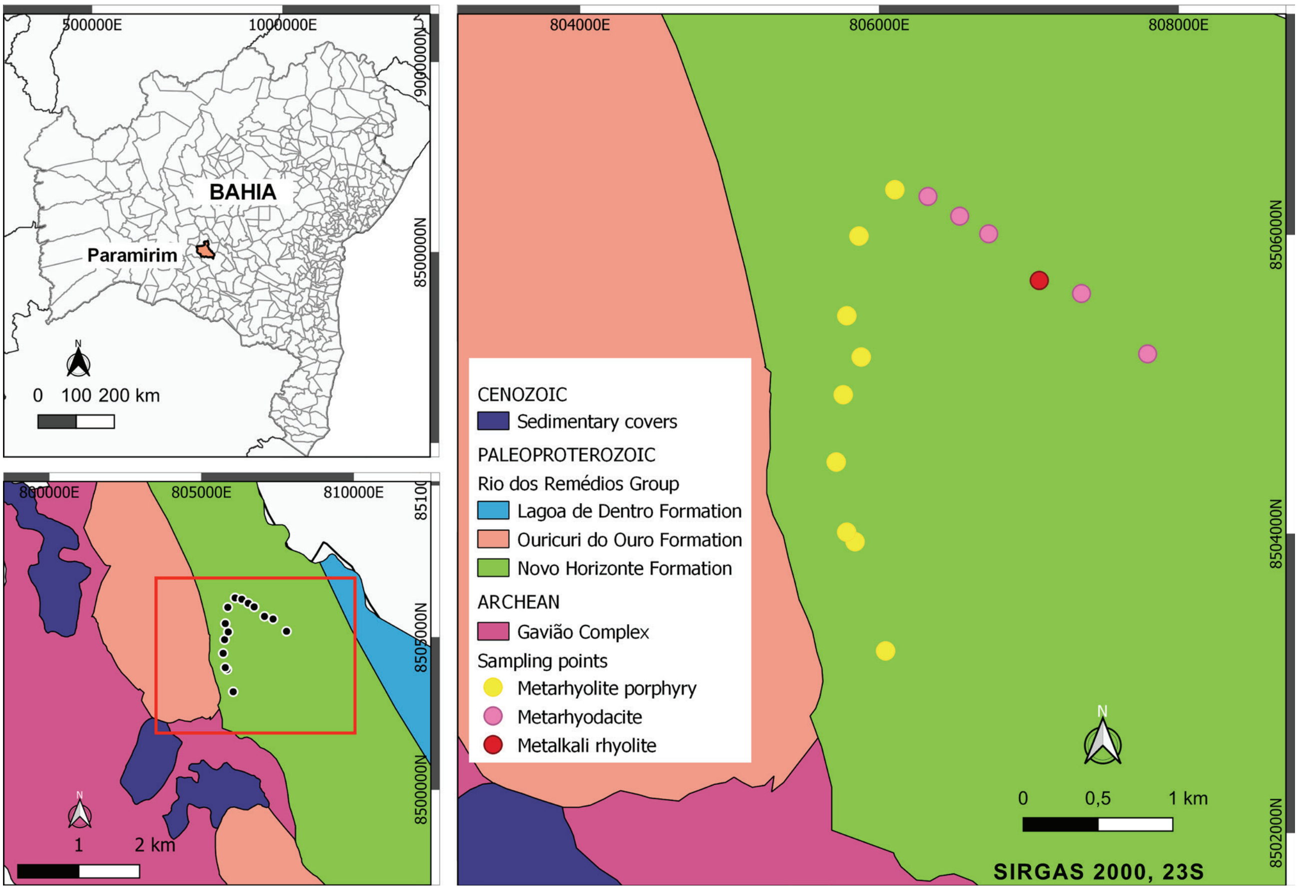

Figure 3. Sampling locations of the Rio dos Remédios rhyolites.
Figura 3. Localização dos pontos de coleta dos riolitos do Grupo Rio dos Remédios.

3.2.2 Heat treatment

The purpose of the heat treatment was to assess the stability of the color with increasing temperature. Both grains and polished samples have been heated to 100°C, 300°C, 500°C, 600°C, and 800°C, with an increment of 1°C/min, and a 24 h isothermal step at the top temperature of each phase, using a Nabertherm Controller P320. The heated grains were observed against both a light and a dark background in order to determine whether any loss of color is owed to heat or changes in the color of the background mineral matrix.

3.2.3 Fourier Transform Infrared Spectroscopy

The Fourier Transform Infrared Spectroscopy (FT-IR) measurements were performed on pellets composed of 2 mg of powdered sample per 148 mg of KBr, using a Bruker Tensor 27 spectrometer. The instrument range is 7500 - 210 cm^{-1}, with a spectral resolution of 1 cm^{-1}. Grains with varying degrees of color intensity have been measured, in order to observe potential correlations between the infrared spectrum and variations in color.

3.2.4 Electron microscopy

Electron microscopy was performed using a Tabletop Hitachi 3030 on Cr covered polished sections. The method was employed for the study of mineral inclusions, and for the preliminary investigation of elemental zoning within the blue quartz grains.

4 Results

4.1 Optical observations

The blue color of the quartz grains from the Rio dos Remédios Group is the most striking feature of the investigated metarhyolite samples. It is not as intense as the color observed in the Llano rhyolite blue quartz, but significantly

Figure 4. Samples of the blue quartz bearing rock referred to in the paper. A) Rio dos Remédios alkali rhyolite; B) Rio dos Remédios metarhyodacite; C) Rio dos Remédios metarhyolite porphyry; D) Llano rhyolite (llanite); E) Albești metagranite.
Figura 4. Amostras da rocha contendo quartzo azul referida no artigo. A) riolito alcalino Rio dos Remédios; B) metariodacit Rio dos Remédios; C) metariolito pórfiro Rio dos Remédios; D) riolito de Llano (llanite); E) metagranito Albești.

stronger than the blue color of the quartz from the Albești metagranite (Fig. 4). When present, chatoyancy is faint, but iridescence (likely caused by closely spaced sets of fractures inside the grains) is often observed.

A color zoning is present, usually manifested as a blue grain core and a progressively fading color towards the edge (Fig. 5A), but circular bands of color (and even two concentric ones) consistent with the overall morphology of the host grain are also observed (Fig. 5B).

The grains are usually fractured and hypidiomorphic, with rare instances of unbroken, hypidiomorphic to idiomorphic quartz. The overwhelming majority of the grains are optically free of inclusions, with rare instances of dark colored flakes, likely Fe/Ti oxides.

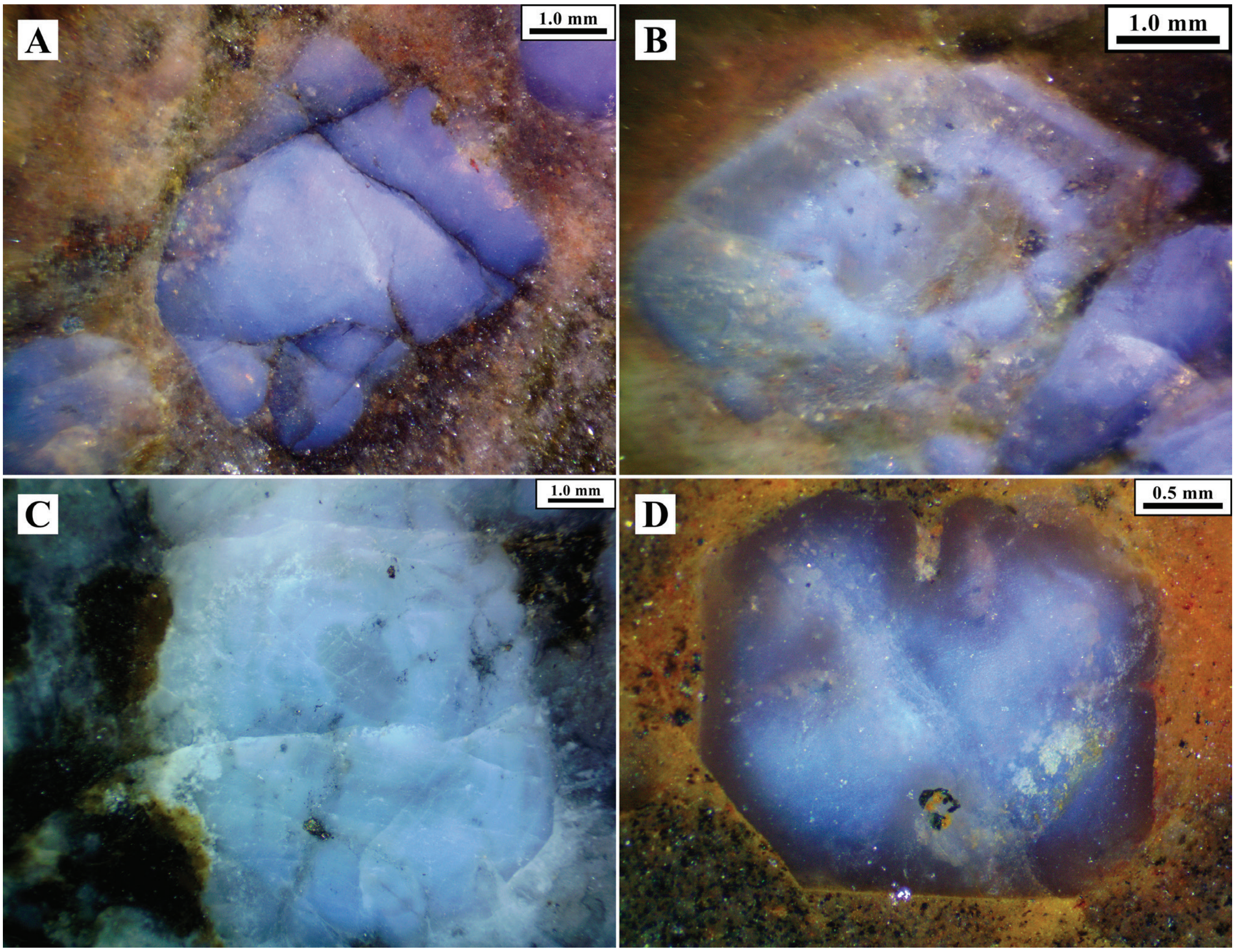

Figure 5. Color zoning observed in reflected light in the Rio dos Remédios blue quartz (A and B), Albești (C), and Llano (D), using a binocular microscope.
Figura 5. Zoneamento observado em luz refletida por meio de microscópio binocular, no quartzo azul Rio dos Remédios (A e B), Albești (C) e Llano (D).

The observations performed on thin metarhyolite slices in reflected and transmitted light indicate that the blue color of the quartz grains is produced by light being scattered inside the grains. The quartz zones which are blue in reflected light turn brownish-yellow when light passes through them; this is a feature generally associated with the scattering of light. It has been observed that the variations in color mentioned above clearly apply to the colored areas of the grains (Fig. 6), suggesting that the brownish-yellow color is indeed complementary to the blue color and not the exclusive effect of light absorption inside the quartz grains (although some absorption in the colorless areas does occur, as seen in Fig. 6B). The Albești (Fig. 6C and D) and Llano (Fig. 6E and F) blue quartz grains observed in the same circumstances as the Rio dos Remédios occurrence yielded the same results.

Under the petrographic microscope, the quartz grains occur as single crystals, with recrystallization along the edges and internal fractures (Fig. 7B). Undulose extinction is present in the original quartz grains, and absent in the recrystallized areas. Fluid inclusions are preferentially located along healed fractures and the smallest ones measure 3 µm on average. The solid inclusions are mainly present as opaque phases along fractures (Fig. 7A), but local concentrations of rutile needles are also present (Fig. 7C). The needles are ca. 10 - 20 µm in length and ca. 0.5 µm thick, and therefore the observed rutile exceeds the requirements for Rayleigh scattering (< 0.1 µm), but its presence explains the slight chatoyancy mentioned in section 4.1.

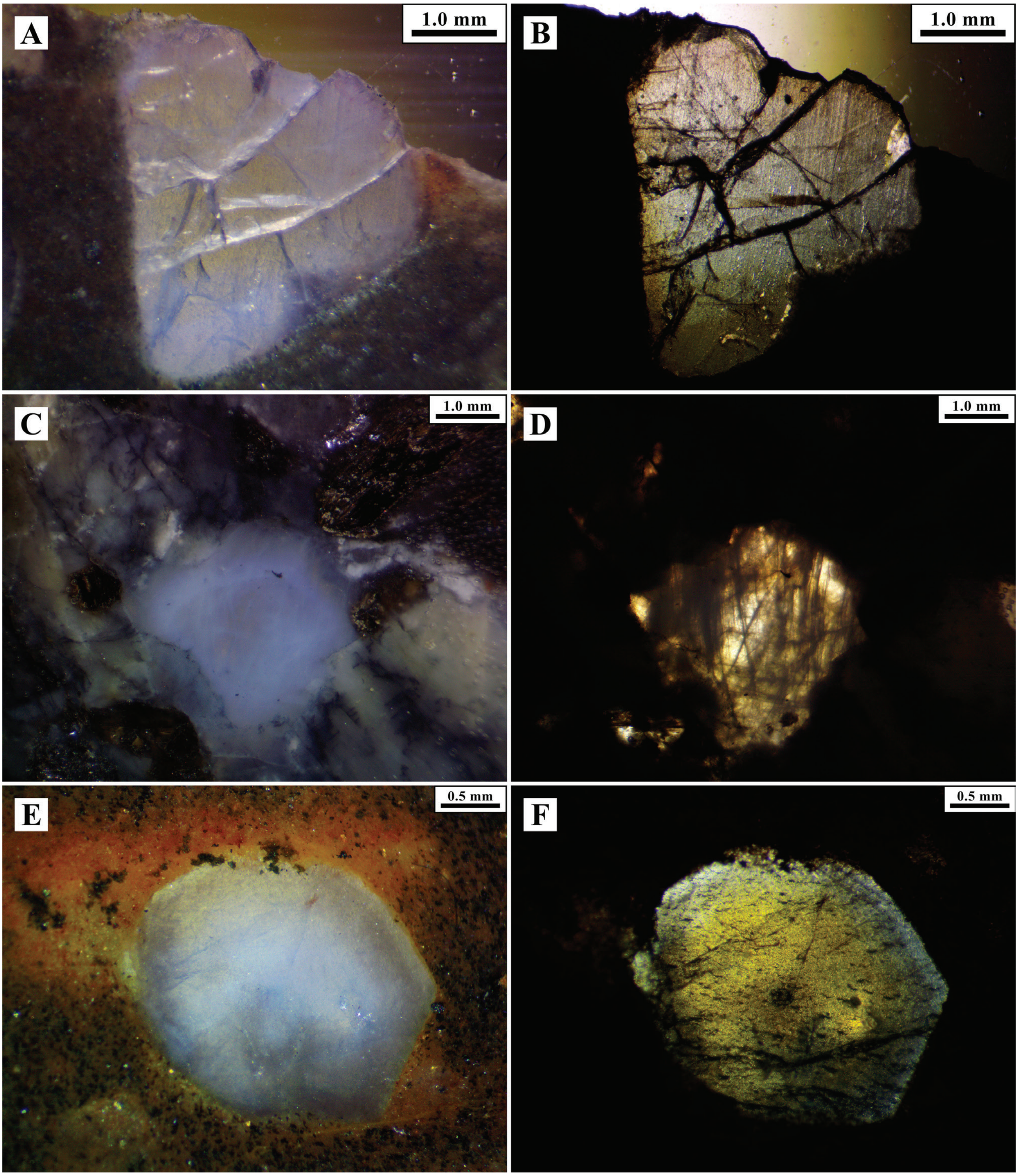

Figure 6. Color zoned quartz grains from the Rio dos Remédios metarhyodacite, Albeşti metagranite, and Llano rhyolite porphyry in reflected (A, C, E, respectively), and transmitted light (B, D, F, respectively). Notice the spatial overlap between the blue and brownish-yellow areas.
Figura 6. Grãos de quartzo azul zonados do metariodacito Rio dos Remédios, metagranito Albeşti e riolito pórfiro de Llano em luz refletida (A, C, E, respectivamente) e luz transmitida (B, D, F, respectivamente). Observe a sobreposição espacial entre as áreas azul e amarelo acastanhado.

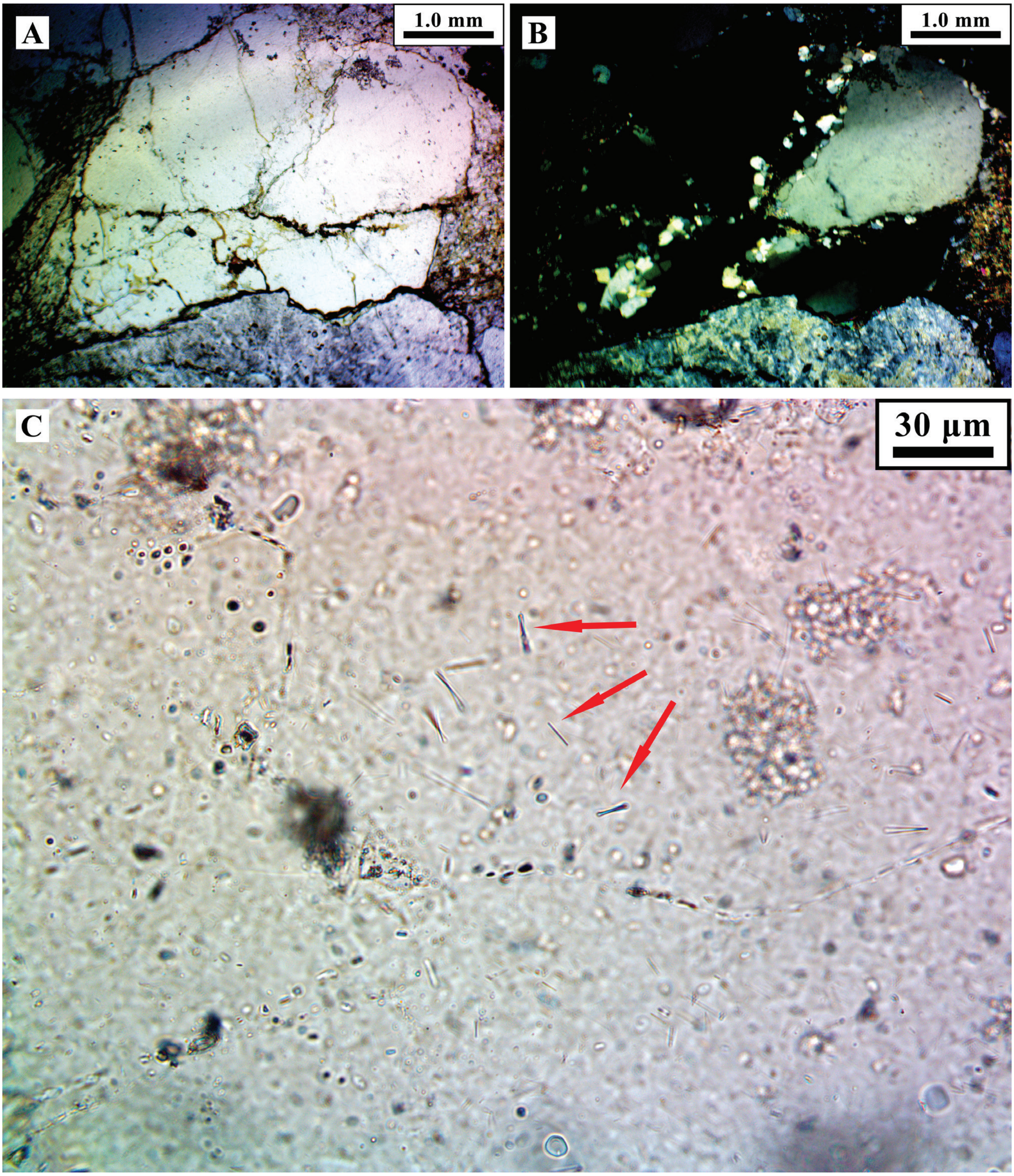

Figure 7. Petrographic microscope images of a quartz crystal from an alkali rhyolite from Rio dos Remédios. A) parallel nicols; B) crossed nicols; C) Rutile needles in a quartz crystal from an alkali rhyolite from Rio dos Remédios observed with parallel nicols. The red arrows indicate some of the needles.

Figura 7. Fotomicrografia de um cristal de quartzo de um riolito alcalino do Rio dos Remédios. A) nicóis paralelos; B) nicóis cruzados; C) Agulhas de rutilo em um cristal de quartzo de um riolito alcalino do Rio dos Remédios observadas com nicóis paralelos. As setas vermelhas indicam algumas das agulhas.

4.2 Heat treatment

The polished samples showed little loss of color when exposed to temperatures of up to 500°C, but after 600°C the discoloration becomes more obvious in all the heated samples (Fig. 8). However, the mineral matrix itself also bleaches significantly, potentially contributing to the observed loss of color. Grain fracturing was expected, but no new significant fissures were observed in the wake of the heat treatment.

In order to determine whether the loss of color is real or it is an effect of the light colored background, blue quartz grains from all three varieties of metarhyolite were observed against a white and black background, then heated to 800°C at an increment of 1°C/min, maintained at the top temperature for 24 h, and then observed against a black background for a second time. The unheated grains are virtually colorless on a white background, and blue on a dark one. Observing the heated grains against a black background shows that the blue color is essentially preserved. Some bleaching is noticeable, likely resulted from the scattering of light by microscopic stress features which can appear during heating (Hajpál & Török, 2004; Chernov & Khadzhi, 1968). However, the aforementioned features were not observed over the course of the present study, and it is uncertain at this point whether the observed bleaching is caused by the thermal stress features or actual changes in the color producing mechanism (Fig. 9). These results are in stark contrast to the thermal behavior of the blue quartz grains from Albești, which shows a definite loss of color for most grains (probably caused by the thermal transformation of the opalescent areas which in turn conceals the blue color), but similar to those gathered for the quartz from Llano.

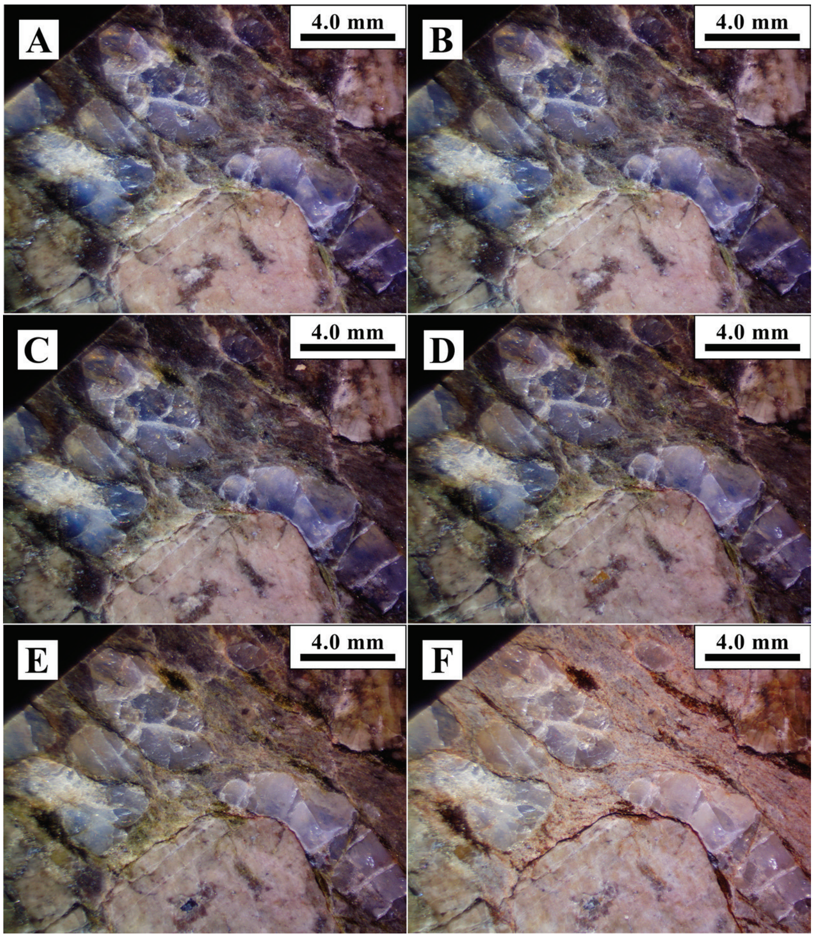

Figure 8. Representative incremental color loss in the Rio dos Remédios rhyolite varieties. The figure illustrates blue quartz grains from an alkali rhyolite sample at room temperature (A), 100°C (B), 300°C (C), 500°C (D), 600°C (E), and 800°C (F).

Figura 8. Perda de cor incremental representativa nas variedades de riolito do Rio dos Remédios. A figura ilustra grãos de quartzo azuis de uma amostra de riolito alcalino à temperatura ambiente (A), 100°C (B), 300°C (C), 500°C (D), 600°C (E), e 800°C (F).

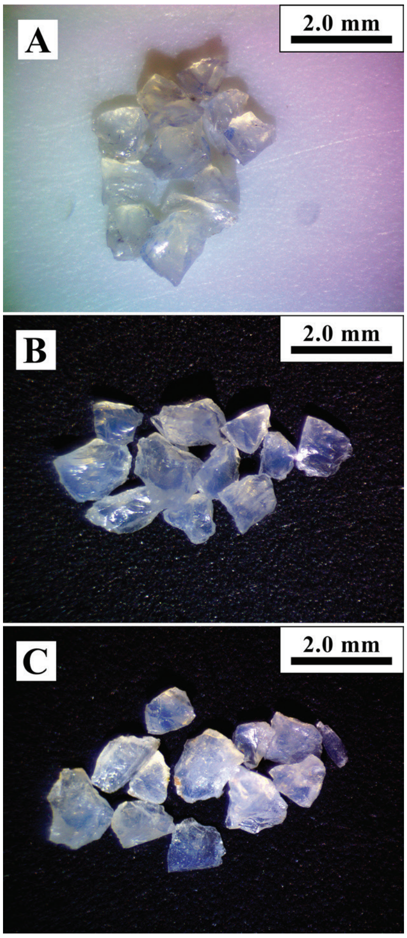

Figure 9. Unheated blue quartz grains from a metarhyodacite sample observed against a white background (A) and black background (B); the same grains observed against a black background after being heated to 800°C. Notice how the color of the background can influence the perceived color (A and B).
Figura 9. Grãos de quartzo azul não aquecidos de uma amostra de metariodacit observada contra um fundo branco (A) e fundo preto (B); os mesmos grãos observados contra um fundo preto após serem aquecidos a 800°C. Observe como a cor do plano de fundo pode influenciar a cor percebida (A e B).

4.3 FT-IR

The FT-IR spectra obtained do not show any differences between the intensely blue grain fragments and the colorless ones, regardless of the sample investigated. The peaks present in the infrared absorption spectra are centered on 1081 cm^{-1}, 797 cm^{-1}, 694 cm^{-1}, and 459 cm^{-1}, corresponding to the Si-O $\upsilon 3$ asymmetrical stretching, Si-O $\upsilon 1$ symmetrical stretching, Si-O $\upsilon 2$ symmetrical bending, and Si-O $\upsilon 4$ asymmetrical bending vibration modes, respectively. The peak centered on 1879 cm^{-1} is also attributed to quartz (Sivakumar *et al*., 2012).

The quartz samples from the Rio dos Remédios metarhyolite, both colored and colorless/less colored, do not differ significantly from those of the Albeşti metagranite or Llano rhyolite in terms of FT-IR signal (Fig. 10). The frequency of the diagnostic peaks is mostly the same, and the difference in the relative intensity of the highest peak and its shoulder (Shoval *et al*., 1991), and the ca. 778 and 695 cm^{-1} peaks (Razva *et al*., 2014) is indicative of various degrees of crystallinity between the measured quartz samples.

4.4 Electron microscopy

The most striking feature observed from the electron microscope imaging is the sharp contrast between the quartz grains and the feldspathic matrix in terms of inclusions (Fig. 11A). The inclusions in question are the white patches observed in figure 11A, G and H, which correspond to phases composed of atoms with high atomic numbers (conversely, the lower the atomic number, the darker the color). When present within the boundaries of the quartz grains, the inclusions are usually associated with fractures filled with feldspar like material (Fig. 11). The inclusions identified in the matrix are mostly iron oxide with up to 10 wt% Ti, zircon, and rarely rutile. Rutile and iron oxide, ca. 1 - 3 μm across, have also been observed in quartz grains, independent of features indicative of material transportation subsequent to crystallization.

The embayments observed in the blue quartz grains, particularly visible in the grains of the metarhyodacite (Fig. 11G and H) indicate quartz resorption caused by either the early crystallization of the quartz grains at disequilibrium with the melt, rapid changes in the pressure and temperature of the magma, or magma mixing.

5 Discussion of results

The results point to a blue color produced by Rayleigh scattering, a feature shared by all investigated blue quartz samples. The grains are blue only in reflected light against a dark background, much like the blue color of the sky

Figure 10. Stacked FT-IR absorption spectra of blue quartz grains the Rio dos Remédios alkali rhyolite, Albeşti metagranite, and Llano rhyolite.
Figura 10. Espectros de absorção FT-IR de grãos de quartzo azul do álcali riolito Rio dos Remédios, metagranito Albeşti, e riolito Llano.

against the dark background of outer space. In transmitted light, the blue areas of the grains turn brownish-yellow. This indicates that certain volumes of the quartz grains contain light scattering nanometric solid particles or, as suggested by Zolensky *et al*. (1988) and Wise (1981), light scattering fluid inclusions or sets of subparallel microfractures, respectively, which cause the color difference in reflected and transmitted light associated with Rayleigh scattering. The different intensities of blue observed in the three studied samples can be caused by variations in the size and spatial density of the light scattering inclusions/features from each occurrence, based on the data presented by Zolensky *et al*. (1988).

All of the investigated blue quartz occurrences show color zoning. The color zoning observed in the Llano blue quartz rigorously follows the contour of the grains, regardless of embayments, and only one blue color zone was observed for each grain. The Rio dos Remédios blue quartz presents a color zoning similar to Llano, but locally it also shows multiple color bands within the same grain, which have a circular shape most of the times, and loosely follow the contour of the grain. This feature is reminiscent of the type of color zoning observed in the Albeşti blue quartz. In the case of Llano, the observed morphology of the colored areas is attributed by Zolensky *et al*. (1988) to light scattering ilmenite which was added to the growth surfaces of the quartz grains during crystallization. The ordered pattern of inclusion resulted in the correlation observed between the morphology of the color zones and the contour of the quartz grains.

The color zoning observed in the Rio dos Remédios blue quartz provides insight into the origin of the scattering inclusions. Seifert *et al*. (2011) have suggested exsolution, syngenetic

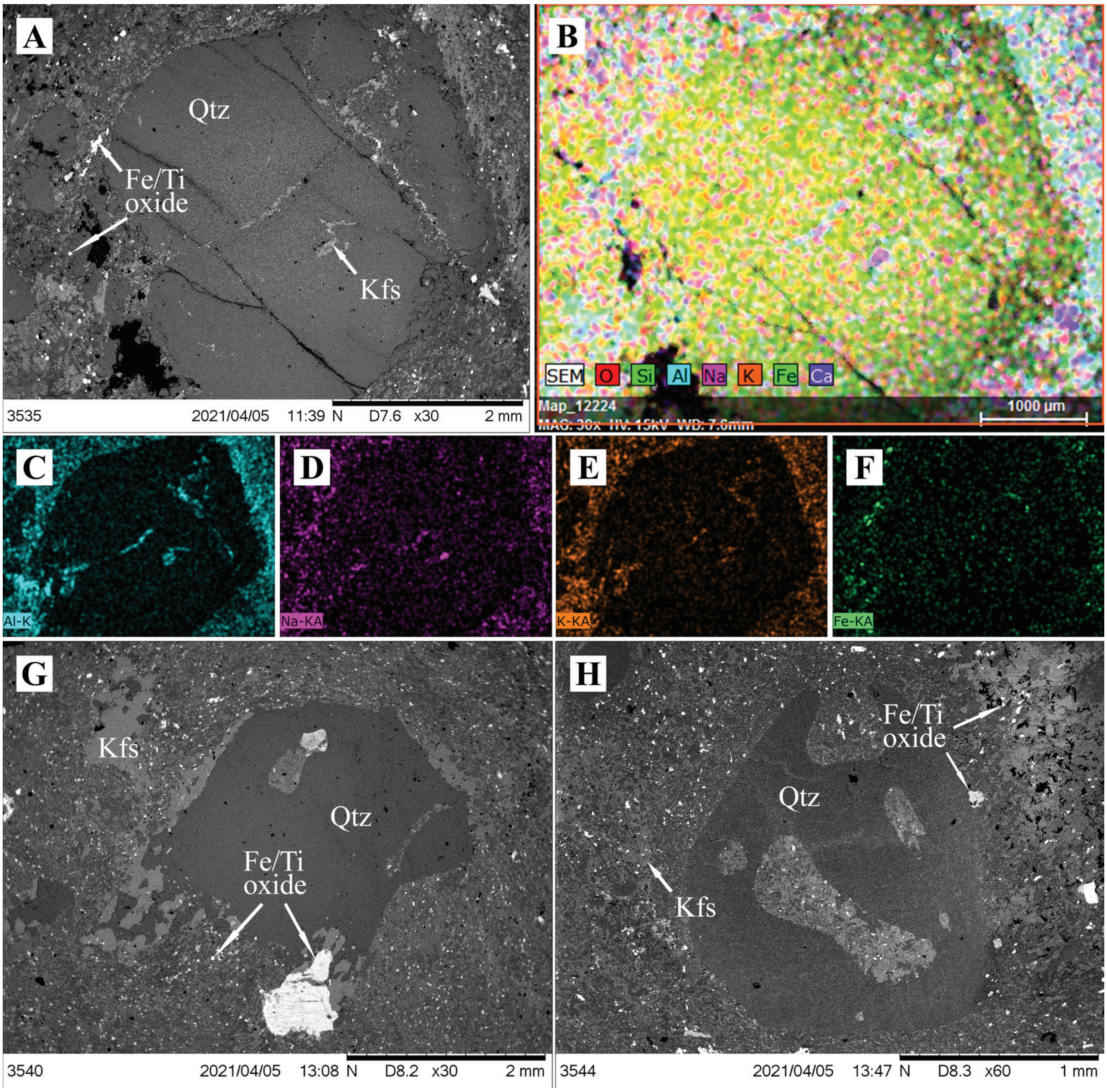

Figure 11. Electron microscope image of a blue quartz grain hosted by an alkali rhyolite sample of Rio dos Remédios. A) image showing the lack of inclusions in the quartz grains by comparison to the matrix; B) EDS mapping of the quartz grain and matrix from figure A; C), D), E), F) the spatial distribution of Al, Na, K, and Fe, respectively, in the quartz grain from figure A; G), H) matrix embayments in the blue quartz grains of the Rio dos Remédios metarhyodacite.
Figura 11. Imagem do microscópio eletrônico de um grão de quartzo azul em uma amostra de álcali riolito do Rio dos Remédios. A) imagem mostrando a falta de inclusões nos grãos de quartzo em comparação com a matriz; B) mapeamento EDS do grão e matriz de quartzo; C), D), E), F) a distribuição espacial de Al, Na, K, e Fe, respectivamente; G), H) embainamentos de matriz nos grãos de quartzo azul do metariodacito Rio dos Remédios.

formation, and inclusion as the mechanisms by which the presence of light scattering inclusions inside the quartz can be explained. Considering the complex and, at times, geometrically rigorous color zoning (Fig. 5B), the syngenetic origin of the quartz and scattering inclusions is the most plausible. The crystallization of the inclusions at the boundary layer between the growing quartz crystal and generating melt explains why the colored zones tend to follow the contour of the host grain. Furthermore, fluctuations in the chemistry of the melt as it crystallizes can explain the alternating blue and colorless zones, as it is possible that the crystallization of the inclusions was not continuous throughout the volume of the melt.

Blue quartz in general does not differ from regular quartz under the petrographic microscope. The thickness of the thin section is too small to allow the manifestation of the blue color or the complementary brownish-yellow tones, and the size of the scattering particles themselves is prohibitive to observations performed under the petrographic microscope. The color zoning observed in the grains is locally disrupted by recrystallization (Fig. 5A). The recrystallized domains are usually related to fractures (and to a lesser extent to grain boundaries), and are randomly distributed. The randomness of the recrystallized domains is in stark contrast with the geometry of the color zones (Figs. 5B, 6A and 6B). These observations point to the fact that the blue color zones formed during the crystallization of the quartz grains and while they can be interrupted by recrystallization, they are not the result of regularly alternating zones of primary and recrystallized quartz, as such instances have not been observed.

FT-IR has not shown the presence of lattice bound or fluid inclusion OH. Fluid inclusions have been observed under the petrographic microscope, but in the process of preparing the sample they were destroyed, as the average size of the smallest fluid inclusions is 3 μm and the grains in the powders used for analysis average 1.5 - 4 μm at their widest. The fact that the measurements have not shown the presence of OH in quartz does not necessarily imply that the quartz grains are free of fluid inclusions. If these inclusions are present, but their size is smaller than the wavelength of the analyzing infrared radiation (as required by Rayleigh scattering), they will be undetectable.

The heat treatment of the quartz grains has shown that the color is stable at temperatures of up to 800°C. If the scattering of light is indeed the color producing mechanism, then the light scattering inclusions themselves are stable at the same temperature. On the other hand, since the scattering of light is mainly dependent on the size of the scattering particles, it will still be preserved even if phase transitions do occur, provided that there are no significant subsequent changes in the dimensions of the inclusions.

Electron microscopy has shown an abundance of iron oxides in the matrix and feldspars, in stark contrast with the quartz grains. To a much smaller extent, Fe and Ti oxides are also present in quartz, but their size is too large (>1 μm) to scatter light. These observations point to high Fe and Ti contents which could potentially concentrate as nanometric (>0.1 μm) inclusions capable of Rayleigh scattering.

EDS chemical mapping was unable to detect any type of zoning which could be correlated to the color zoning clearly observable in the polished samples. This is also true for the Albești and Llano blue quartz (Pantia & Filiuţă, 2021). The underlying causes for the coloration are likely beyond the detection capacity of the instrument. On the other hand, this could be a result in itself, suggesting that the color is not caused by large concentrations of chemical impurities or micrometric inclusions, but rather by impurities which are small enough to elude detection using this method, in agreement with the results of previous studies. In the case of Zolensky *et al*. (1988), only TEM methods were able to actually detect and provide an image of the light scattering ilmenite inclusions.

6 Conclusions

The present study has shown strong similarities between the Rio dos Remédios and Llano blue quartz. Both occurrences are hosted by Proterozoic rhyolites, manifest various degrees of chatoyancy, have largely similar color zoning, and their color is stable at elevated temperatures.

Considering the data available to date (the presence of significant Fe and Ti in the system, the color stability at elevated temperatures, and the chromatic behavior in reflected and transmitted light), it is likely that the blue color of the Rio dos Remédios rhyolite hosted quartz is caused by light scattering inclusions with iron oxide, ilmenite, or rutile like chemistry.

Similarly to Llano, the Rio dos Remédios blue quartz occurrence represents a form of magmatic blue quartz. The low metamorphic grade (greenschist facies) experienced by the studied sample was insufficient to cause the exsolution of light scattering phases although theoretically, recrystallization could cause the remobilization of impurities in structural defects, thus giving rise to molecular colloids and nanometric inclusions capable of scattering light. The conclusion of the present study is that the blue coloration of the investigated quartz grains from Rio dos Remédios is caused by solid inclusions trapped by the quartz grains during their initial crystallization from melt. The quartz was thus capable of scattering light once it crystallized. The fact that the color is magmatic in origin and not the result of metamorphism is supported by the fact that similar rhyolites in the area (near Rio de Contas), which have experienced intense deformation and a low to medium grade metamorphism (as shown by the quartz subgrain rotation recrystallization and the kyanite-garnet equilibrium temperatures) during the Brasiliano orogeny (Santos *et al*., 2019), now host regular quartz, and that recrystallization disrupts the color (as shown by the optical observations) rather than creating it. The effect of metamorphism on the blue color in quartz has been reported before, as summed up by Pantia & Filiuţă (2021). If the Rio dos Remédios blue quartz is indeed an expression of the same mechanisms which produced the blue color in the Llano blue quartz, then according to Zolensky *et al*. (1988) it may reflect particular chemical characteristics of the Proterozoic magmas from which it crystallized.

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