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AUTHOR STATEMENT

Cunha, I.RV: Conceptualization, methodology, petrography, and mineral chemistry, writing the manuscript and preparing the illustrations. Dall'Agnol, R: Conceptualization, supervision, reviewing, and editing. Scaillet, B: Supervision, reviewing, and editing. Sousa, L.A.M: Software - Map preparation, collaboration in petrography and iron oxides mineralogy.

MAGMATIC EPIDOTE IN ARCHEAN GRANITOIDS OF THE CARAJÁS PROVINCE, AMAZONIAN CRATON, AND ITS STABILITY DURING MAGMA RISE AND EMPLACEMENT

4

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14 ABSTRACT

15 In the Rio Maria and Sapucaia Domains of the Carajás Province, during the Mesoarchean, tonalite-trondhjemite-granodiorite (TTG) series (2.98-2.92 Ga; Colorado 16 Trondhjemite) and sanukitoid rocks (~2.87 Ga; Rio Maria Suite of Ourilândia do Norte) were 17 formed. They were followed, in the Neoarchean (2.75-2.73 Ga), by numerous stocks of 18 granitoids similar to A-type granites (Vila Jussara Suite). Despite the compositional 19 differences between these granitoids, epidote is a mineral phase common to all of them, with 20 pistacite contents of 26-29 mol% in TTGs, 22-33 mol% in sanukitoids and 25-30 mol% in 21 Neoarchean granitoids. The study of the dissolution kinetics of Archean epidotes of the 22 Carajás Province reveals that the partial dissolution time of their crystals was ~4-10 years, 23 with corresponding magma ascent rates of 4-8 km/year. Magma viscosities at liquidus 24 temperature were estimated at 10^{5.3} Pa s for TTG magma and 10^{2.5} Pa s for sanukitoid magma, 25 whereas monzogranitic magmas of the Vila Jussara Suite exhibited a viscosity of 10^{4.4} Pa s. In 26 contrast, the viscosity of tonalitic magma of the Vila Jussara Suite was 10^{3.5} Pa s. The 27 preservation of magmatic epidote in Archean granitoids requires that the plutons grew in an 28 incremental way, similar to their Phanerozoic counterparts, with the stacking of small sill-29 dykes of about 100 m thickness each intruded in a rather cold crust, allowing fast cooling 30 rates so as to prevent complete dissolution of epidote at the final emplacement level. 31

Keywords: magmatic epidote, Archean, Carajás Province, dissolution, viscosity, incrementalgrowth

34 1. INTRODUCTION

In the Archean, the upper mantle temperature was estimated to be approximately 1600°C, near ~250°C above its current average temperature (Sizova et al., 2015). This was

due to the higher production of radiogenic heat and drastically affected the thickness and 37 composition of the crust (Sizova et al., 2015 and references therein). The onset of the 38 continental crust formation resulted from the cooling and differentiation of the Earth and was 39 combined with continuous magma extraction from the mantle. This caused changes in the 40 crustal growth rates and resulted in an increase of crustal volume. These processes began in 41 the early Archean but were intensified at the end of the Mesoarchean and throughout the 42 43 Neoarchean (Taylor and McLennan, 1985; Kramers and Tolstikhin, 1997; Belousova et al., 2010; Dhuime et al., 2011; Guitreau et al., 2012) when, in addition to tonalite-trondhjemite-44 granodiorite (TTG) associations, larger volumes of sanukitoids, hybrid granitoids and potassic 45 granites were formed, resulting in rocks with more diversified geochemical signatures 46 (Martin, 1987; Moyen et al., 2003; Jayananda et al., 2006; Barros et al., 2009; Feio et al., 47 2012; Moyen and Martin, 2012; Almeida et al., 2013; Laurent et al., 2014; Sizova et al., 2015; 48 Bédard, 2017; Dall'Agnol et al., 2017; Silva et al., 2018). This geochemical variability is 49 evidenced by marked variations in the continental crust. It occurred between 3.0 and 2.5 Ga, 50 when more than 50% of the current continental crust volume was formed (Bédard, 2017). 51

52 TTGs are the oldest and largely dominant felsic plutonic association in Archean times. Accordingly, their origin and relationship with tectonic regimes have been widely discussed 53 54 (Moyen and Martin, 2012; Sizova et al., 2015; Moyen and Laurent, 2018; Smithies et al., 2019). Conversely, high-Mg monzodiorites and granodiorites, known as sanukitoids, are less 55 56 voluminous components of late Archean terranes, formed from interactions between the peridotitic mantle and a component rich in incompatible elements (Martin et al., 2005; 57 Oliveira et al., 2010, 2011; Heilimo et al., 2010; Laurent et al., 2014; Sizova et al., 2015). 58 Considerable volumes of leucogranodioritic and granitic rocks were also formed in the final 59 stages of the Archean (Jayananda et al., 2006; Almeida et al., 2013, 2017; Silva et al., 2018). 60 61 This process of granitoid magma migration enabled the transfer of heat and mass from the lower to upper crust, resulting in the chemical differentiation of the continents and thermal 62 and mechanical interactions during magma rise and emplacement (Brown and Solar, 1988). 63

In this context, it is relevant to determine variations of the viscosity of silicate magmas, because this parameter plays a key role in the transport dynamics, type of eruption and rates of physicochemical processes of natural magmas (Giordano et al., 2008), being crucial for a quantitative understanding of igneous processes (Persikov, 1991). The viscosity strongly depends on the composition of the melt, particularly its SiO₂ content, which determines its

degree of polymerization, but also on temperature, content and nature of volatile species,cation types, among other factors (Cruden and Weinberg, 2018).

71 The Carajás Province (CP) presents a wide diversity of granitoid rocks formed from the Mesoarchean to the Neoarchean (~3.0 to 2.57 Ga). The present study focuses on three 72 73 different types of granitoids that occur in the Sapucaia Domain of this province and at its 74 border with the Rio Maria Domain. Such granitoid rocks include tonalites and trondhjemites 75 of TTG associations (Santos et al., 2013; Teixeira et al., 2013; Silva et al., 2014; Leite-Santos and Oliveira, 2014) and sanukitoids (Gabriel and Oliveira, 2014; Santos and Oliveira, 2016; 76 77 Silva et al., 2018), both of Mesoarchean age (2.93 a 2.85 Ga), as well as ferroan to magnesian Neoarchean granitoids (~2.75 to 2.73 Ga), geochemically akin to A-type granites (Dall'Agnol 78 et al., 2017; Silva et al., 2020). 79

Despite the geochemical and petrological differences among TTGs, sanukitoids and 80 Neoarchean granitoids, magmatic epidote is a common mineral phase in these rocks. It is 81 known that epidote stability strongly depends on the physicochemical parameters prevailing 82 83 during magma evolution (Brandon et al., 1996; Keane and Morrison, 1997; Sial et al., 2008; Brasilino et al., 2011; Nagar and Korakoppa, 2014; Long et al., 2019). Hence, this study aims 84 85 at, on the basis of the textural and chemical-mineralogical characterization of the epidote present in Mesoarchean and Neoarchean granitoids of the CP, estimating epidote partial 86 dissolution time scale, the ascent rates and initial viscosity of each granitoid magma, and the 87 ability of the magma to preserve epidote during pluton emplacement and crystallization in 88 upper crust. Our study allows us to more accurately assess the physicochemical conditions of 89 the formation of these granitoid magmas and to better understand the evolution of magmatic 90 epidote. In the following, we start by a brief overview of factors controlling epidote stability 91 in granitoid magmas, and then present the data obtained in the study area and their 92 implications. 93

94 2.STABILITY OF EPIDOTE IN MAGMAS AND TEXTURAL CRITERIA FOR ITS 95 IDENTIFICATION

96 Epidote is a key indicator of variations in crystallization parameters (Schmidt and 97 Thompson 1996) because it is extremely sensitive to changes in P-T- fO_2 conditions and it 98 may be destabilized and completely dissolved during emplacement (Long et al., 2019). Based 99 on experiments conducted over the past few decades, the role of epidote during magmatic 100 crystallization is relatively well understood, and experimental results have confirmed the 101 paragenesis of natural rocks produced under different P-T- fO_2 conditions (Schmidt and Poli, 102 2004).

103 Experimental studies demonstrate the stability of epidote above 300 MPa; however, depending on the composition and oxygen fugacity, the minimum pressure for epidote 104 105 crystallisation may vary (Schmidt and Poli, 2004). Crawford and Hollister (1982), based on 106 the intersection of the solidus curve with that of epidote (Liou, 1973), proposed that in H_2O saturated granites, magmatic epidote requires a minimum pressure of ~600 MPa. In 107 granodioritic and tonalitic magmas, epidote is stable at pressures between 800 and 1000 MPa 108 at temperatures near the solidus curve (Naney 1983; Liou 1993; Schmidt and Thompson, 109 110 1996). However, epidote may crystallise at pressures as low as ~300 MPa at fO_2 near the Hematite-Magnetite buffer, while at Ni-NiO, it is only stable at pressures \geq 500 MPa (Naney, 111 1983; Schmidt and Thompson, 1996). In general, epidote stability increases under more 112 oxidizing fO₂ conditions (Liou, 1973; Schmidt and Thompson, 1996; Poli and Schmidt, 2004; 113 Schmidt and Poli, 2004; Oliveira et al., 2010), and the pistacite contents characteristic of 114 115 magmatic epidotes observed in experiments are mainly found between hematite-magnetite 116 (HM) and nickel-NNO buffers (Liou, 1973). Oscillatory zones in epidote crystals may reflect changes in composition or in oxygen fugacity (Franz and Liebscher, 2004; Pandit et al., 117 2014). At low-pressures, outside the stability field of epidote, magnetite is usually the main 118 Fe3+-containing phase (Drinkwater et al., 1991) and its modal content increases in rocks 119 without epidote crystallization (Hammarstrom and Zen 1992; Schmidt, 1993). 120

121 The following equilibrium appears to control epidote stability in granitoids (Schmidt 122 and Poli, 2004):

123 hornblende + K-feldspar + plagioclase + magnetite + H_2O = epidote + biotite + quartz

Archean granitoids described below exhibit textural facies consistent with the concomitant formation of epidote and biotite, both of which are magmatic. This relationship can be explained by the peritectic reaction below (e.g., Sial et al. 1999).

127

plagioclase + amphibole + melt = biotite and epidote

Experimental studies in tonalitic compositions show that epidote + biotite can be formed through peritectic hornblende resorption without completely consuming the hornblende in the system, decreasing only its abundance proportionally to the modal increase in biotite + epidote (Schmidt and Poli, 2004 and references therein).

Textural and compositional criteria used to distinguish between magmatic and secondary epidotes, include (1) euhedral to subhedral crystals with prismatic or hexagonal shapes; (2) mineralogical associations with primary hornblende and biotite; (3) epidote with oscillatory zoning, allanite core and irregular contact with quartz and plagioclase; (4) pistacite content ranging from 25 to 35%; and (5) TiO₂ content $\leq 0.137\%$ (Liou, 1973; Tulloch, 1979; Zen and Hammarstrom, 1984; Zen, 1985; Evans and Vance, 1987; Dawes and Evans, 1991; Schmidt and Poli, 2004; Sial et al., 2008).

In granitic systems, allanite crystallizes at temperatures significantly above solidus, it can be later shielded by epidote rims and finally isolated epidote crystals can also occur (Gieré and Sorensen, 2004). Dawes and Evans (1991) proposed that allanite cores surrounded by epidote form due to magma impoverishment in rare earth elements during crystallization, which inhibits allanite crystallization and enables epidote epitaxial growth. However, the oxidation state of Fe when inserted in the crystalline structure of allanite is controlled by the reaction:

 $\mathrm{Fe}^{2+} + \mathrm{OH}^{-} \leftrightarrow \mathrm{Fe}^{3+} + \mathrm{O}^{2-} + 1/2 \mathrm{H}_2$

Based on this reaction, Gieré and Sorensen (2004) suggested that the stability of allanite in magmas may be more dependent on T and fH_2 than on the content of rare earth elements.

149

3.REGIONAL GEOLOGY

150 Located in the southeast section of the Amazonian craton, the Carajás Province is one of the main Archean terranes of this craton. The CP is inserted in the Archean domain of the 151 152 Central Amazonian Province (Tassinari and Macambira 2004), or alternatively, according to 153 the model proposed by Santos (2003), it represents an independent Archean province (Fig. 154 1a). In recent decades, various tectonic segmentation models have been proposed for the CP 155 (Souza et al., 1996; Althoff et al., 2000; Dall'Agnol et al., 2006, 2013). Vasquez et al. (2008), 156 in line with Santos (2003), proposed naming the southern section of the province as the Rio Maria Domain (RMD) and its central and northern sections as the Carajás Domain (CD). In a 157 more recent model, Dall'Agnol et al. (2013) maintained the RMD but subdivided the CD into 158 the Sapucaia Domain (SD), Canaã dos Carajás Domain (CCD) and Carajás Basin (CB). These 159 160 three domains would have evolved differently from the RMD (Fig. 1b). This interpretation was not followed by Costa et al. (2020), who suggested that dome-and-keel structures 161 162 controlled the evolution of both the RMD and the southern section of the old CD, which 163 would be an extension of that domain.

The Rio Maria Domain is a Mesoarchean terrane (3.0 to 2.86 Ga), predominantly consisting of greenstone belt sequences (3.0 to 2.90 Ga; Macambira and Lancelot, 1996; Souza et al., 2001) and TTG granitoids (2.98 to 2.93 Ga; Almeida et al. 2011, 2013), in addition to sanukitoids, high Ba-Sr granitic-to-leucogranodioritic suites and potassic leucogranites (2.87-2.86 Ga; Althoff et al., 2000; Dall'Agnol et al. 2006; Almeida et al., 2010, 2013; Oliveira et al., 2011; Silva et al. 2018).

The Sapucaia Domain (2.95 to 2.73 Ga) comprises TTG rocks (Santos et al., 2013; Leite-Santos and Oliveira, 2014; Silva et al., 2014), sanukitoids (Gabriel and Oliveira, 2014; Gabriel et al., 2014), high Ba-Sr leucogranodiorites and potassic leucogranites (Teixeira et al., 2013; Leite-Santos and Oliveira, 2016), showing strong lithological similarities to the RMD (Fig. 1c). However, SD rocks were intensely deformed during the Neoarchean (2.75-2.73 Ga) and intruded by granitoids of the Vila Jussara Suite (Dall'Agnol et al., 2017; Silva et al., 2020) and potassic leucogranites (Leite-Santos and Oliveira, 2016), both of Neoarchean age.

The Canaã dos Carajás Domain consists of granites sensu stricto and charnockite 177 178 associations (Fig. 1c). According to Feio et al. (2013), the CCD presents four main magmatic 179 events, three of Mesoarchean age and one of Neoarchean age. The protolith of the Pium 180 Complex (Pidgeon et al., 2000), the Bacaba Tonalite (~3.0 Ga; Moreto et al., 2011) and other rocks with similar ages were formed in 3.05-3.0 Ga. The Canaã dos Carajás Granite and the 181 182 oldest rocks of the Rio Verde Trondhjemite were formed between 2.96 and 2.93 Ga. The 183 Campina Verde Tonalite Complex, the Rio Verde Trondhjemite and the Cruzadão, Bom Jesus, Boa Sorte and Serra Dourada Granites crystallized from 2.87 to 2.83 Ga. The 184 subalkaline granites of the Planalto Suite, the sodic granites of the Pedra Branca Suite and the 185 charnockitic rocks of the Pium Complex were formed in the Neoarchean between ~2.75 and 186 2.73 Ga (Feio et al., 2012; Santos et al., 2013; Rodrigues et al., 2014). 187

188 4. GEOLOGY, PETROGRAPHY AND GEOCHEMISTRY OF THE SELECTED 189 MESOARCHEAN AND NEOARCHEAN GRANITOIDS

190 4.1 TONALITE-TRONDHJEMITE ASSOCIATION OF SAPUCAIA

The Mesoarchean magmatism (2.93 to 2.85 Ga) exposed in the SD is marked by abundant TTG granitoids, predominantly trondhjemitic rocks with subordinate tonalites and granodiorites, grouped stratigraphically as undifferentiated TTGs and Colorado Trondhjemite (2.87 Ga; Silva et al., 2014). These TTG granitoids are distributed from Água Azul do Norte to Sapucaia, at the border between the Amazonian craton and the Araguaia Belt (Fig. 1c; Santos et al., 2013b; Silva et al., 2014; Leite-Santos and Oliveira, 2014). Structurally, these

197 rocks show E-W penetrative foliation and shear zones in the same direction. These rocks also 198 exhibit compositional banding, S-C structures, drag folds and typical features of plastic flow, 199 mostly indicating sinistral kinematics. In general, such rocks show porphyroclastic textures 200 resulting from an intense recrystallization and mineral comminution process, marked by 201 mylonitic-to-ultramylonitic tectonic fabrics (Santos et al., 2013b).

202 In mineralogical terms, the Sapucaia TT essentially consists of plagioclase and quartz, with highly subordinate microcline and with biotite as the main mafic phase, accompanied by 203 204 epidote, opaques, allanite, titanite, apatite and zircon as accessory minerals. Geochemically, the granitoids are calc-alkaline with generally peraluminous compositions, no significant Sr 205 and Eu anomalies, silica content ranging from 59 to 74 wt. %, relatively low ferromagnesian 206 content (Fe₂O₃+MgO+MnO+TiO₂ < 5 wt. %), moderate Mg# (0.27-0.47) and high alumina 207 values (Al₂O₃ > 15 wt. %) (Santos et al., 2013b). The samples selected for analysis derive 208 209 from the eastern section of the SD (Fig. 1c) and belong to the Colorado Trondhjemite. Santos 210 et al. (2013b) reported the presence of magmatic epidote in these rocks and conducted a preliminary mineralogical study based on scanning electron microscopy and EDS analyses. 211

212 4.2 OURILÂNDIA DO NORTE SANUKITOID

The Ourilândia do Norte area is located near the borders of the Rio Maria, Canaã dos Carajás and Sapucaia domains in the CP. This area is marked by a diverse Mesoarchean magmatism, mainly characterized by leucogranites, sanukitoids and associated rocks (Santos and Oliveira, 2016; Silva et al. 2018). The Mesoarchean sanukitoids of the Ourilândia do Norte region are identified as a composite body, predominantly consisting of granodiorites and equigranular tonalites, with subordinate quartz diorite and quartz monzodiorite, which form elongated and deformed lenses along the E-W direction.

In petrographic terms, the sanukitoid rocks exhibit well-preserved igneous textures, but 220 221 are also marked by protomylonitic to mylonitic tectonic fabrics. The mineral assemblage 222 essentially consists of quartz, plagioclase, various proportions of K-feldspar and mafic 223 clusters formed by amphibole, biotite and epidote. The main accessory minerals consist of epidote (zoisite and clinozoisite), allanite, titanite, zircon, apatite, magnetite and ilmenite. 224 225 Geochemically, sanukitoids and associated rocks exhibit a highly variable SiO₂ content, moderate K₂O values and high Mg#, Cr and Ni values. They have a predominantly 226 metaluminous character and a calc-alkaline to high-K calc-alkaline signature (Santos and 227 Oliveira, 2016; Silva et al., 2018). Two textural types of magmatic epidote were 228 229 distinguished: crystals with zoned or unzoned allanite cores and euhedral to subhedral crystals

230 associated with biotite and hornblende, partly surrounded by them (Santos and Oliveira, 231 2016).

232 **4.3 VILA JUSSARA SUITE**

233 The Vila Jussara Suite (VJS) granitoids occur in the central and northern sections of the SD, stretching northward to the vicinity of the CCD and eastward to the border between the 234 Amazonian craton and the Araguaia Belt (Figs. 1b, c). The Neoarchean granitic bodies 235 (~2.75-2.73 Ga) of the VJS intrude the Mesoarchean units (Colorado Trondhjemite and 236 Pantanal Leucogranodiorite; Teixeira et al., 2013; Silva et al., 2014; Dall'Agnol et al., 2017). 237 These plutons are associated with shear zones and elongated in the E-W direction. Field, 238 petrographic and microstructural evidence reveal that the VJS rocks were affected by 239 heterogeneous deformational processes of varying intensity, exhibiting structures ranging 240 241 from protomylonitic to mylonitic (Silva et al., 2020).

Four main petrographic varieties were identified in the VJS: biotite-hornblende 242 monzogranite (BHMzG), biotite-hornblende tonalite (BHTnl), biotite monzogranite (BMzG) 243 244 and hornblende-biotite granodiorite (HBGd). The different rock varieties of the VJS are characterized by a similar mineralogy, from essential tectosilicates (quartz, plagioclase and 245 246 potassic feldspar) to mafic minerals (hornblende and/or biotite), and by the main primary 247 accessory minerals, ilmenite, magnetite, apatite and zircon, which can also include titanite, 248 allanite and epidote. Geochemically, the VJS consists of ferroan granitoids (reduced and oxidized A-type granites) and mildly magnesian granites (cf. Dall'Agnol et al., 2017; Sousa, 249 250 2019). The four petrographic varieties of the VJS were formed under different conditions of oxygen fugacity. The reduced to moderately reduced BHMzG was formed under conditions 251 varying from <FMQ to FMQ + 0.5. The oxidized BHMzG was generated under moderately 252 oxidizing conditions (between NNO and NNO-0.5), whereas the magnesian granites were 253 crystallized under oxidizing conditions, with fO2 ranging from NNO to NNO+1 (Sousa, 254 255 2019). Dall'Agnol et al. (2017) described the textural and compositional variations in the 256 magmatic epidote of the specific VJS varieties.

257

5.TEXTURAL ASPECTS OF EPIDOTE IN THE STUDIED GRANITOIDS

Typical magmatic epidote crystals are hexagonal-euhedral and greenish-pink to pinkish-258 259 yellow or colorless, with oblique extinctions (Dawes and Evans, 1991; Schmidt and Poli, 260 2004). In granitoid rocks, magmatic epidote crystals are usually subhedral and generally 261 associated with biotite, hornblende and plagioclase (Pandit et al., 2014).

262 In the Archean granitoids selected for analysis, epidote is a common accessory mineral 263 phase, sometimes present in appreciable modal proportions. In the Sapucaia TT, biotite is the main mafic phase, and the epidote modal content ranges from trace to 4%. The epidote 264 exhibits two textural aspects (Santos et al., 2013b): (a) euhedral to subhedral crystals with 265 well-defined faces, usually with automorphic contacts with biotite and irregular contacts with 266 267 quartz and feldspars (Fig. 2a); and (b) crystals involving allanite cores, either metamictized or preserved (Fig. 2b). The sanukitoids of Ourilândia do Norte (Santos and Oliveira, 2016) have 268 269 a high amphibole content, and epidote is a significant accessory mineral (ranging from trace to 3.9% and reaching 6.2% in the epidote-biotite-amphibole diorite variety). Mafic aggregates 270 271 consisting of hornblende ± actinolite, biotite, epidote, titanite and Fe-Ti oxides are common. Epidote occurs as zoned crystals, with or without allanite cores, or as euhedral to subhedral 272 crystals that are partly or completely surrounded by biotite and/or hornblende (Fig. 2c). 273

In the VJS, oxidized ferroan biotite-hornblende monzogranite is the predominant 274 variety, whose epidote has a modal content $\leq 1.0\%$. In reduced ferroan rocks of this variety, 275 276 epidote occurs in a smaller proportion, is absent from most samples and is predominantly 277 found as anhedral and rarely subhedral crystals with signs of intense corrosion. In the biotite monzogranite variety, epidote is only absent from one sample and exhibits variable modal 278 279 content, albeit generally <1% and peaking at 2.7%. The hornblende-biotite tonalite group has 280 a smaller exposure area than that of other VJS varieties, and when present, the magmatic 281 epidote content ranges from 0.2 to 0.4%.

In general, the epidote present in the study granitoids is commonly found in association 282 with mafic aggregates consisting of biotite \pm hornblende \pm zircon \pm apatite \pm allanite \pm titanite 283 \pm opaque minerals. Based on textural aspects, the following types of epidote can be 284 285 distinguished: (a) euhedral to subhedral crystals that are predominantly associated with biotite that shares straight contacts with epidote (Figs. 2a, 4c, 4d); (b) epidote in rims around allanite 286 287 (Figs. 2b, 4c, 4f); and (c) anhedral to subhedral epidote with irregular contours with 288 amphibole and/or feldspars (Figs. 2c, 2e, 2f). When in contact with biotite, the original shape 289 of the epidote is generally preserved (Figs. 2a, 4c, 4f). In contrast, when in contacts with hornblende and felsic minerals, epidote crystals show signs of marked corrosion (Fig. 2e), 290 291 which sometimes almost completely destroys the original facies. Allanite is also an accessory mineral commonly found in these granitoids, generally exhibiting a subhedral to anhedral 292 293 habit. It is sometimes zoned and generally totally or partly surrounded by epidote (Figs. 2b,

4c). Subordinately, epidote is identified as a post-magmatic phase, forming irregular grainsthat are preferentially associated with altered plagioclase.

The textural aspects observed in the Archean granitoid epidotes of the CP are similar to those interpreted by Zen and Hammarstorm (1984), Bédard (2003) and Schmidt and Poli (2004), as diagnostic of magmatic epidote. However, Keane and Morrison (1997) stressed out that the shape of crystals is insufficient to differentiate the magmatic or subsolidus origin of epidotes because the absence or development of euhedral faces is not exclusive to direct magmatic crystallization. Accordingly, only the combination of textural, compositional and isotopic criteria can reliably indicate the origin of an epidote crystal.

303 6.MINERAL CHEMISTRY

Epidote crystals with textural evidence of magmatic origin were analyzed on a JEOL 304 305 JXA-8230 electron probe microanalyzer (with five spectrometers in wavelength-dispersive spectroscopy (WDS) mode and one in energy-dispersive spectroscopy (EDS) mode) at the 306 307 Microanalysis Laboratory, Institute of Geosciences (Instituto de Geociências - IG), Federal 308 University of Pará (Universidade Federal do Pará – UFPA). The analytical procedure was 309 performed on polished thin carbon coated sections under the following operating conditions: 310 15 kV voltage acceleration; 20 nA current; 5 µm beam diameter; and 20 second analysis times for F, Na, Al, Cr, K, Sr, Ni, Mn, Ba, Ti, Cl, V, Y, Ce, and La, and 10 second analysis times 311 for Mg, Si, Ca, and Fe. The crystals used for the analyses were LIF for Fe, Mn, Ba, and Ti; 312 PETJ for Ca and K; TAP for Na, Mg, Al, and Si; and LIFH for Ce and La. The standards used 313 for calibration were orthoclase (SiO₂ and K_2O), rutile (TiO₂), anorthite (Al₂O₃), magnetite 314 (FeO), diopside (MgO), rhodonite (MnO), wollastonite (CaO), sodalite (Na₂O) and CePO₄ 315 $(Ce_2O_3 \text{ and } La_2O_3).$ 316

In total, 105 analyses were performed (Table 1), with 41 in the Sapucaia TT (PFA-14 and PFR-33A); 42 in the Ourilândia do Norte sanukitoids (NDP-47 and NDP-101); and 22 in the VJS granitoids (ADE-01D, AFD-11A, and AMP-77). The VJS analyses were added to those obtained by Dall'Agnol et al. (2017).

Tulloch (1979) and Vyhnal et al. (1991) evaluated the origin of epidote based on its chemical composition and concluded that pistacite contents (Ps: molar $[Fe^{3+}/Fe^{3+}+A1]/100$) ranging from 25 to 29% (molecular weight) correspond to magmatic epidotes, differing from those formed by subsolidus alteration of plagioclase (Ps₀₋₂₄) and biotite (Ps₃₆₋₄₈). Experimental studies on synthetic rocks (Liou, 1973) also show that pistacite contents ranging from 25 to
35% correspond to magmatic epidote.

In Sapucaia TT, the pistacite (Ps: molar [Fe³⁺/Fe³⁺+Al]/100) content ranges from 26 to 327 29 mol%, with values primarily clustering between 27 and 28 mol%, thus suggesting a 328 329 magmatic origin of the crystals (Fig. 3; Table 1). In the Ourilândia do Norte sanukitoids, the 330 Ps content of epidote ranges from 22 to 33 mol%, with values concentrating from 29 to 30 331 mol%, that is predominantly in the magmatic origin field as well (Fig. 3; Table 1). The VJS varieties has Ps values ranging from 25 to 30 mol% (Fig. 3; Table 1), corroborating the 332 333 findings of Dall'Agnol et al. (2017) and in line with a magmatic origin. Oliveira et al. (2010) found Ps values ranging from 26 to 33 mol% for the epidotes from the sanukitoid Rio Maria 334 335 Suite, mostly overlapping with those of their analogues from Ourilândia do Norte.

In addition to the molar Ps content, previous empirical and experimental studies (Tulloch, 1979; Vyhnal et al., 1991; Liou, 1993; Sial et al., 1999; Oliveira et al., 2010; Dall'Agnol et al., 2017) suggest that a TiO₂ content in epidote ≤ 0.137 wt. % is in line with its magmatic formation. The Sapucaia TT and VJS granitoid epidotes display TiO₂ values always lower than 0.137 wt. % (Table 1), corroborating the textural and compositional assessments above indicative of magmatic origin. In the Ourilândia do Norte sanukitoids, the epidotes have TiO₂ values predominantly ≤ 0.137 wt. % (Table 1).

Thus, the Sapucaia TT and Vila Jussara granitoid epidotes with textural evidence of magmatic origin exhibit compositional variations compatible with magmatic origin (Fig. 3; Table 1). Comparatively, despite predominantly plotting in the magmatic field, the Ourilândia do Norte sanukitoids epidotes have subordinate compositions compatible with their formation from altered plagioclase (Fig. 3). In this regard, these epidotes diverge from those of the Rio Maria Suite sanukitoids, which only show compositions within the range assigned to magmatic epidotes (Oliveira et al., 2010).

350 Studies conducted so far on Neoarchean granitoid from the Carajás Province (CP) 351 corroborate the results of experimental researches showing that epidote formation depends on fO₂ and pressure, as shown by the Estrela Complex, Serra do Rabo and Planalto Suite 352 granites, which formed under strongly reduced conditions and lack epidote. Conversely, the 353 354 Villa Jussara Suite (VJS) (Dall'Agnol et al., 2017) and Vila União (Marangoanha et al., 2018) granitoids, which are mostly formed under oxidizing conditions, generally present epidote as 355 an important accessory phase. The epidote crystals of the VJS show no considerable 356 variations in Fe³⁺ content between the core and rims of the crystals and rarely display zoning, 357

thus suggesting that in the final stages of crystallization, the residual melt was not impoverished in Fe³⁺ and that fO_2 remained constant, thereby favoring epidote stability.

360 7. EPIDOTE DISSOLUTION AS A MAGMA ASCENT RATE INDICATOR

In general, epidote crystallization occurs near the solidus temperature at relatively high 361 pressure (Schmidt and Poli, 2004). However, epidote phenocrystals in dikes in which the 362 matrix, representative of the residual melt, accounts for approximately two-thirds of the rock 363 volume were described (Dawes and Evans, 1991). The presence of epidote in granitic rocks 364 365 that finished its crystallization at relatively low-pressure conditions indicates that epidote 366 likely started forming at greater depths and then was transported quickly enough to prevent its 367 complete resorption during magma rise (Brandon et al., 1996). The absence of epidote may 368 also result from its complete dissolution by residual magmatic melt. In turn, the presence of allanite cores surrounded by epidote may be due to the magma impoverishment in rare earth 369 370 elements during crystallization, which would inhibit allanite crystallization and enable epidote epitaxial growth (Dawes and Evans, 1991). 371

Experimental evidence shows that the intensity of epidote crystal dissolution is directly related to changes in intensive crystallization parameters during magma rise. Accordingly, magma transport rates have been estimated based on the degree of epidote dissolution (Brandon et al., 1996; Sial et al., 2008; Brasilino et al., 2011; Pandit et al., 2014). This method was used to measure the ascent rate of Archean granitoid magmas from the CP, based on the methodological criteria detailed below:

- 378 (1) Euhedral and subhedral epidote crystals allow us to more accurately infer the original
 379 shape of the crystal and therefore are prioritized when estimating the degree of
 380 dissolution. Generally, these crystals are associated with biotite, amphibole and
 381 residual magmatic melt, which are currently represented by quartz and feldspar
 382 crystals;
- 383 (2) In the absence of isolated epidote crystals, epidote crystals involving allanite cores in
 384 epitaxial growth can be used;
- 385 (3) The pistacite content of the selected epidote crystals should fall within the magmatic
 386 field (Ps: 25-35 mol%; Liou, 1973);
- (4) Calculating the crystal corrosion intensity is crucial, and these calculations must be
 based on maximum extent of dissolution identified in the petrographic microscope and
 always with measurements perpendicular to the crystal faces affected by corrosion;

390 (5) The duration of the corrosion process is calculated using the minimum apparent 391 diffusion coefficient between tonalitic magma and epidote $(5 \times 10^{-17} \text{ m}^2 \text{ s}^{-1} \text{ for Si, Al},$ 392 Ca and F) at a temperature of 750°C, which can also be applied to other granitoid 393 magmas, according to criteria formulated by Brandon et al. (1996), using the equations 394 below:

395
$$d_z = [(D_{app} x t)^{1/2}]$$

396 $t = d_z^{2/}(5x10^{-17})$

397 with

398 $d_z = dissolution zone width (m);$

399
$$D_{app} = apparent diffusion coefficient (5 \times 10^{-17} m^2 s^{-1}); and$$

t = partial epidote dissolution time (s)

400

401 (6) The magma emplacement depth is estimated using the Al-in-hornblende geobarometer
402 and is compared with estimates based on the geological and structural context of the
403 study rocks. The magma generation depth is generally deduced from petrological
404 criteria;

(7) The magma transport rate is calculated using the ratio between the path length
(difference between magma generation depth and emplacement depth) and the average
corrosion time of the epidote exposed to melt. In the original proposal, the path length
corresponds to the difference between 10 kbar, initial epidote crystallization pressure,
and emplacement depth (Brandon et al., 1996):

410 $Tr = L_r/t$

411 Tr = transport rate (m/yr)

412
$$L_r = \text{path length (m), defined by the equation:}$$

413
$$L_r = (10 - P_e) \cdot 10^4 / 3 \text{ (m)}$$

414 $P_e = emplacement pressure (kbar)$

415 t = partial dissolution time do epidote (yr)

The dimensions and length of the maximum dissolution of epidote crystals were measured on thin sections representative of the Archean varieties of interest (Fig. 4). Subsequently, the resulting values were clustered, and the arithmetic mean was assumed to be representative of each sample, enabling calculations of average epidote dissolution and transport rates of each magma (Table 2). 421 The epidote dissolution and magma transport rates were calculated in the PFA-14 422 sample for the Sapucaia TT (Figs. 4a, b; Table 2) and in the NDP-47 and NDP-101 samples for the sanukitoids from the Ourilândia do Norte region (Figs. 4c, d, e; Table 2). Granitoids 423 424 from the VJS contain mineralogical and geochemical evidence of origin of the three study 425 varieties from different magmas. Thus, the epidote crystal dissolution and magma transport 426 rate were estimated separately in each variety. The biotite-hornblende monzogranite variety is represented by the MYF-77 (Fig. 4f) and PFR-16A (Fig. 4g) samples, biotite monzogranite is 427 428 represented by the MDP-02A and LIF-04A samples (Figs. 4h, i, j) and biotite-hornblende 429 tonalite is represented by the MDP-02E sample (Figs. 4k, 1).

The emplacement pressure of the study granitoids is an important variable for 430 431 calculating the magma transport rate. Dall'Agnol et al. (2017) proposed that the magmas that formed the VJS were generated under pressures between 800 and 1000 MPa, and 432 433 emplacement pressures were estimated to range from 300 to 500 MPa based on Al-inamphibole geobarometer and geological evidence; a value of 400 MPa is assumed here as the 434 435 emplacement pressure of these granitoids. The Ourilândia do Norte sanukitoids exhibit ductile 436 structures and evidence of penetrative deformation associated with mylonitic structures (Santos and Oliveira, 2016; Silva et al., 2018; Nascimento, 2020). Nascimento (2020), based 437 438 on Al-in-hornblende geothermometer measurements, estimated an emplacement pressure of ~200 MPa for those sanukitoids. However, considering the structural features described in the 439 440 present study, a higher emplacement pressure of ~350 MPa corresponding to the mesozone is estimated for the Ourilândia do Norte sanukitoids. The pressures during sanukitoids 441 442 production in the Rio Maria region were estimated at 600 to 900 MPa by Oliveira et al. 443 (2010), a range which we use in the present work. In regard to the Sapucaia TT, Santos et al. 444 (2013b) demonstrated that these rocks show affinity to the TTG with high La/Yb and Sr/Y 445 ratios of the CP, suggesting that they were derived from amphibolite garnet-based sources under high pressures (approximately 1500 MPa). Finally, based on the ductile deformational 446 structures observed in these rocks, we estimate an emplacement pressure of ~500 MPa for the 447 Sapucaia TT magma. 448

Using the equations proposed by Brandon et al. (1996), the average durations of epidote crystal dissolution of the study granitoids are estimated at ~4 years in the Sapucaia TT and ~6 years in the Ourilândia do Norte sanukitoids, whereas these periods are estimated at ~5 years in bt-hb monzogranite, ~10 years in bt monzogranite and ~3 years in bt-hb tonalite for the VJS varieties (Table 2). The results show that the Sapucaia TT magma rose in the crust at an ascent rate of ~4600 m/year, whereas the Ourilândia do Norte sanukitoid magma moved at ~4300 m/year. Finally, in the VJS, the calculations show a comparatively higher magma ascent rate for the three magmas (bt-hb monzogranite at ~8000 m/year; bt monzogranite at ~7900 m/year; and bt-hb tonalite at ~8200 m/year; Table 2).

458 8. DISCUSSION

459 8.1 VISCOSITY AND LIQUIDUS TEMPERATURE IN THE STUDIED GRANITOID460 MAGMAS

The emplacement of a granitoid body in the mid-to-upper crust is the end point of a 461 large-scale mass and energy exchange, which begins with magma generation and segregation 462 463 in the lower crust and is followed by magma ascent and the formation of plutons, batholiths 464 and stocks (Cruden, 1998). During magma ascent, the behavior of the silicate melt is primarily determined by its viscosity, density, composition and volatile content, which 465 466 determine the magma ability to rise when combined with the geometric characteristics of the 467 conduit (see below). Among the physical properties, viscosity is one of the parameters with the strongest effect on the production, transport and eruption of silicate magmas, varying in 468 the range 10^{-1} to 10^{14} Pa s (Giordano et al., 2008). This variation in viscosity by 15 orders of 469 magnitude determines the magmas' resistance to flow, which inversely affects its fluidity. 470

Experimental studies on the viscosity of silicate melts enabled Giordano et al. (2008) to formulate a general model, which includes the effect of H₂O and F, to predict the non-Arrhenius Newtonian viscosity of silicate magmas, with the following equation:

474

VFT $[\log \eta = A + B / (T (K) - C)],$

475 where A is the upper temperature limit for the viscosity of all silica melts, which is -4.55 (+0.2) (for example, log $\eta \sim 10^{-4.6}$ Pa s), and B and C are parameters that describe the 476 compositional control on viscosity (see Giordano et al., 2008). The rationale for the A 477 478 parameter being unique is that at super-liquidus temperatures, all silicate melts become highly 479 dissociated liquids regardless of their lower temperature structure and converge to lower limiting value of viscosity, in accordance with experimental observations and theory. The B 480 481 and C terms embody 17 parameters (oxides content and various combinations of them, including volatiles.) that allow to describe faithfully the viscous behavior of the vast majority 482 of terrestrial magma compositions, over fifteen log units of viscosity (10⁻¹-10¹⁵ Pa s). The 483 model is regressed on more than 1770 experimental measurements, performed either at 1 bar 484 or at high pressure (ie with volatiles dissolved into the melt). Given that the compositions 485 486 dealt with here belong to common magma types, we believe that the application of Giordano et al model retrieves the correct order of magnitude of melt viscosity, provided temperature
and H₂O content are well estimated, as discussed below.

The following representative samples were selected: the MYF-19A sample, containing 69.3 wt. % SiO₂, was chosen for the Sapucaia TT magma, whereas the viscosity of the Ourilândia do Norte sanukitoid magma was calculated based on the composition of the BRM-113B sample, with 65.7 wt. % SiO₂ (Table 3). For the VJS, the PFR-16, MDP-02A and MDP-02E samples with 71.4, 71.2 and 60 wt. % SiO₂ contents, respectively, were chosen as representatives of bt-hb monzogranite, bt monzogranite and bt-hb tonalite magmas, respectively (Table 3).

496 As the magmas of the studied granitoids were derived from different sources, the 497 temperature ranges were estimated from experimental studies conducted on systems with 498 similar compositions than those selected above, so as to constrain initial (liquidus) and final 499 (solidus) crystallization temperatures. For the TTG, the most accepted mechanism for the 500 origin of their parental magmas is the partial melting of hydrated metabasaltic rocks, 501 including greenstones, amphibolites and eclogites, in different tectonic environments (Martin, 1987; Winther, 1996; Condie, 2005; Clemens et al., 2006; Moyen and Martin, 2012). In 502 503 particular, Clemens et al. (2006) proposed that TTG magmas are produced at temperatures 504 between 900°C and 1000°C and that crystallization would start at ~880°C, which was the 505 liquidus temperature estimated for Sapucaia TT magma. In turn, the absence of amphibole in 506 these rocks suggests a low H₂O content (Naney et al., 1983), which was estimated at 2% 507 weight in the present study.

Sanukitoid suites are regarded as key components of late Archean terranes (2.95-2.54 Ga), presenting geochemical characteristics similar to those of mantle- and crust-derived magmatic rocks (Moyen et al., 2003; Condie, 2005; Martin et al., 2005; Oliveira et al., 2010, 2011) with higher temperatures compared to TTG. Oliveira et al. (2010) estimated a liquidus temperature of ~950°C and a H₂O content of ~7% for the Rio Maria sanukitoids, which are used in this study for the Ourilândia do Norte sanukitoids.

The VJS varieties are derived from at least three magmas, two of monzogranitic composition (bt-hb monzogranite and bt monzogranite) and one tonalitic (bt-hb tonalite), but the temperatures estimated by Al-in-hornblende and zircon saturation thermometry (Dall'Agnol et al., 2017) indicate that the granitic and tonalitic magmas were formed at similar temperatures. Thus, a liquidus temperature of ~890°C and a solidus temperature of ~700°C was assumed for the three magmas that formed the VJS. 520 The calculated viscosity of each granitoid magma is listed in Table 4 and its variation as 521 a function of temperature is shown on Fig. 5. The TT magma, with a relatively high content of silica and a low content of ferromagnesian components and water has a viscosity of $\sim 10^{5.3}$ Pa 522 s, which is the highest among all Archean magmas of this study (Fig. 5; Table 4). Conversely, 523 524 the sanukitoid magma, which has a higher liquidus temperature (~950°C), a lower silica 525 content and a higher concentration of ferromagnesian constituents and water, shows a comparatively lower viscosity of $\sim 10^{2.5}$ Pa s (Fig. 5; Table 4). Finally, both monzogranitic 526 magmas of the VJS show a viscosity of $\sim 10^{4.4}$ Pa s based on the liquidus temperature given 527 their geochemical similarities, whereas the tonalitic magma of the same suite has a somewhat 528 lower viscosity of $\sim 10^{3.5}$ Pa s (Fig. 5; Table 4). 529

530 8.2 EFFECT OF PHYSICAL PROPERTIES ON MAGMA ASCENT AND EPIDOTE531 DISSOLUTION

The compositional diversity of igneous rocks reflects the variation in processes and 532 sources involved in magma generation (Pitcher, 1979; Huppert and Sparks, 1988; Whitney, 533 1988; Patiño Douce, 1995; Brown, 2013; Scaillet et al., 2016; Cruden and Weinberg, 2018). 534 Nevertheless, for decades, researchers have assumed that felsic magmas rose from deeper 535 regions of the crust through diapirism essentially because the high viscosities assumed for 536 these liquids seemingly precluded any form of rapid and channeled rise (Cruden and 537 Weinberg, 2018), and also because of the broadly circular shape in map view of most granitic 538 539 intrusions. However, field observations, experimental studies on the viscosity of magmatic 540 liquids (Scaillet et al., 1996; Dingwell, 1999; Caricchi et al., 2007; Giordano et al., 2008) as well as theoretical considerations have demonstrated that felsic magmas can also quickly rise 541 542 to different crustal levels through dikes, faults and shear zones (Clemens and Mawer, 1992; 543 Petford et al., 1994, 2000; Oliveira et al., 2008; Cruden and Weinberg, 2018), in much the same way than more mafic magmas, such as basalts, do. 544

In this context, differences in physical properties and ascent rates between magmas may help to understand the textural differences observed in the epidotes of Archean granitoids of the CP. The millimetric dimension and euhedral character of the epidote grains suggest protracted periods of continuous granitic magma crystallization. Conversely, the textural evidence of corrosion and resorption indicates significant epidote dissolution during magma transport and crystallization in the crust due to changes in the physicochemical conditions (Pandit et al., 2014).

552 For the TT magma, with a generation pressure of 1500 MPa, a liquidus temperature 553 estimated at ~880°C, a low H₂O content (~2%) and a relatively high silica content (70%) weight) markedly increase the viscosity, resulting in a magma rise rate of ~4600 m/year 554 (Table 2). However, such transport rates only correspond to the initial stages of magma rise 555 556 because during the process, the magma temperature may decrease, which will be reflected in 557 the increase in the degree of crystallinity and viscosity, thereby decreasing the magmatic flow. 558 In turn, sanukitoid magma, with an estimated generation pressure between 900 and 600 MPa 559 and emplacement in a syntectonic regime linked to transcurrent shear zones (Santos and Oliveira, 2016; Silva et al., 2018; Nascimento, 2020), shows low viscosity (10^{2.5} Pa s), 560 561 reflecting its high temperatures (Fig. 5; Table 4) and relatively high H₂O content (7%, Table 3), with a relatively fast magma rise rate of ~4300 m/year. Moreover, sanukitoids have 562 epidote crystals with a more preserved shapes and a dissolution time estimated at ~6 years 563 (Table 2), whereas TT epidotes exhibit comparatively more intense corrosion and a shorter 564 dissolution time (~4 years; Table 2). In contrast, TT magma transport is fastest compared to 565 sanukitoid magmas analyzed in this study (Table 2). Therefore, for these two magmas, there is 566 no clear correlation between estimated magma ascent rates and mineral dissolution time. 567

Finally, the magma viscosity calculated for the VJS indicates that near the liquidus 568 temperature, the two monzogranitic magmas exhibit a similar viscosity (~10^{4.4} Pa s), which 569 subtly diverges during crystallization (Fig. 5). As the temperature decreases, the viscosity of 570 571 bt monzogranite becomes slightly higher than that of bt-hb monzogranite (Table 4, Fig.5), reflecting the higher magma water content of the latter, which is necessary to stabilize 572 573 hornblende and capable of depolymerizing the magma, thus reducing its viscosity. Accordingly, bt-hb monzogranite exhibits an estimated partial epidote dissolution time of ~5 574 575 years, whereas bt monzogranite has a dissolution time of ~10 years, both with inferred ascent rates of ~7900 m/year (Table 2). In turn, the tonalitic magma has the lowest viscosity among 576 the suite granitoids $(10^{3.5}$ Pa s; Table 3) and therefore the fastest ascent rate (~8200 m/year) 577 and the lowest partial dissolution time of the epidote crystals (~3 years; Table 2). 578

579 It needs to be stressed that the ascent rates calculated above reflect not only the magma 580 viscosity but also the conduit width. This can be appreciated through the following classical 581 relationships which relates both parameters (Furbish, 1996):

$$w = \left(\frac{1}{3\eta}\right) * \left(\rho_c - \rho_l\right)gb^2$$

which gives the average rate of magma flow w (m s⁻¹) in a dyke of width b (m) where ρ_c is the density of host rocks (about 2700 kg m⁻³) and ρ_l the density of melt (about 2350 kg m⁻³), η the magma viscosity (Pa s), and g the gravitational acceleration (9.8 m²s⁻¹). Given that densities of both host rocks and melts are broadly similar, variations in w will mainly arise from those of η and b. A somewhat wider dike width for TT magma could thus explain its faster ascent rates relative to that of Sanukitoid, despite the latter having a lower viscosity than the former.

589

590 8.3 EPIDOTE AS A PETROLOGICAL INDICATOR OF MAGMA EMPLACEMENT IN591 THE ARCHEAN

Recent multidisciplinary studies involving high resolution geochronology and 592 numerical simulations suggest that most plutons are formed through the accumulation of 593 smaller bodies. Thus, plutons result from different pulses of magmatic activity. This factor 594 and the magma composition, emplacement rates of individual pulses, cooling rate and 595 emplacement geometry control the evolution of a pluton and its relationship with the tectonic 596 environment (Annen, 2011; de Saint Blanquat et al., 2011; Zibra et al., 2014). To analyze 597 598 magmatic transport, in addition to the distance travelled by the magma before solidifying, its restite content and the physical properties of the host rocks, the physical and chemical 599 properties of the melt (T, P, fO_2 , XH₂O) and the phases present in the system must also be 600 601 considered (Brown, 1994).

In this context, the epidote kinetics elaborated by Brandon et al. (1996) can be useful in the research of emplacement mechanisms. Those authors have shown that the process of epidote crystal dissolution to be fast: at 700-800 °C, grains with sizes of 0.2-3 mm would only need 2 to 2,000 years to dissolve (Schmidt and Poli, 2004). Another aspect is the size of magmatic intrusions and the construction time of Archean plutons. To a first order, the cooling time (s) of a magma sill of x (m) thickness can be estimated by applying the following standard equation (e.g., Jaupart and Mareschal, 2010):

$$t = x^2 / D_{heat}$$

610 where (D_{heat}) is the heat diffusion $(10^{-7} \text{ m}^2/\text{s})$. A 100 m thick sill would fully crystallize 611 in about 1000 years. This time interval is long enough to dissolve an epidote crystal ~1.2 mm 612 (Brandon et al., 1996). Assuming this calculated value, the formation of an Archean pluton in 613 the CP, with an estimated thickness of ~4 km, would require the juxtaposition of 100-m-thick

1.

-

614 sills fed by successive pulses and ~42,000 years of magmatic injection to form the pluton 615 body. However, geologically, variable times can occur between intrusions, with shorter or longer periods depending fundamentally on the ability of the source to supply magma to the 616 617 surface system. Altogether, the dissolution kinetics of the epidote, which formed grains of 618 varying sizes in the CP, suggests that the mode of emplacement of Archean granitoid magmas 619 was also dominated by a sequential process, i. e. incremental growth, similar to that inferred 620 for their post-Archean counterparts, with crystallization of small individual bodies with an 621 estimated thickness in the order of a hundred meters.

We note that in the case of the Ourilândia do Norte granitoids, Silva et al. (2018) 622 suggested that such granitoids were formed through magma migration and accommodation of 623 several mantle- and crust-derived pulses in a short period, corroborating the above hypothesis 624 of progressive magma amalgamation via small batches. Similarly, the VJS was likely 625 generated by different magmatic pulses from different sources (Dall'Agnol et al., 2017; Silva 626 et al., 2020; Sousa, 2019). In detail, the TT magma appears to have been emplaced at 627 628 somewhat deeper crustal pressures (~500 MPa; Table 5) relative to the sanukitoids (~350 629 MPa; Table 5) and VJS (~400 MPa; Table 5) magmas. This would impart a comparatively slower crystallization rate and therefore a longer epidote reaction with the magmatic melt, 630 631 allowing more intense corrosion of epidote crystals. In contrast, the shallower emplacement of the sanukitoid and Vila Jussara granitoid magmas allowed for a faster crystallization owing to 632 633 slightly cooler host rocks. Accordingly, the process of epidote crystal dissolution was more quickly interrupted because their shielding was favored, resulting in crystals with less 634 635 corrosion than those observed in the TT. We note also that experimental studies have 636 demonstrated that high fO_2 enhances epidote stability (Schmidt and Poli, 2004), which may 637 have been an additional controlling factor of the extent of epidote dissolution (more pronounced at low fO_2 relative to high fO_2 , everything else being equal). Overall, as 638 discussed above, a fast magma rise combined with a more superficial emplacement and a fast 639 crystallization promote epidote preservation. 640

Thus, in addition to providing a more accurate characterization of the nature of magmas, the analysis of physical properties, when combined with experimental studies, epidote dissolution kinetics (Table 2) and viscosity estimates (Table 4), help to understand the magma rise, emplacement and crystallization processes of the Archean plutons in the CP.

645 9. CONCLUSION

Magmatic epidote is commonly found in various Archean granitoids of the CP. By combining a detailed study of the textural aspects with the determination of magma physicochemical properties, epidote dissolution kinetics and viscosity, we reached the following conclusions:

- 650 (1) The epidote of the TT and VJS varieties has pistacite contents indicative of a magmatic 651 origin, whereas that of the Ourilândia do Norte sanukitoid has a Ps content 652 predominantly compatible with primary formation and to a lesser extent with formation 653 through plagioclase and biotite alteration. In addition, the TiO₂ content (≤ 0.137 wt. %) 654 in the epidote of these granitoids also indicates magmatic origin.
- (2) The kinetics showed that the epidote dissolution time of tonalite-trondhjemite was 655 shorter (~4 years) than that of sanukitoid (~6 years). In turn, at the beginning of the 656 657 magma rise, the TT magma transport speed was faster (~4600 m/years) than that of sanukitoid (~4300 m/years). Conversely, in the VJS, monzogranitic magmas exhibited 658 epidote dissolution times of ~5 years for bt-hb monzogranite and ~10 years for bt 659 660 monzogranite, even though the initial magma transport rates were similar (~7900 m/year and ~7906 m/year for bt-hb monzogranite and bt monzogranite, respectively). In 661 662 turn, the VJS tonalitic magma had the shortest epidote dissolution time (~3 years) and the highest magma rise rate (~8210 m/year). 663
- 664 (3) The viscosity of the CP Archean magmas increases from sanukitoid to the tonalitic variety of the VJS, followed by monzogranitic magmas of the same suite and peaks in 665 666 tonalite-trondhjemite. These variations in viscosity, in the case of tonalite-trondhjemite and sanukitoid magmas, show no clear match with magma ascent rates, these magmas 667 having similar ascent rates but different viscosities $(10^{5.3} \text{ and } 10^{2.5} \text{ Pa s, respectively})$. 668 669 This probably reflects variations in conduit width that may have modulated magma 670 ascent rates. In contrast, in the VJS, the correlation between viscosity and magma rise rate seems more coherent. The monzogranitic magmas show similar viscosity values 671 (~10^{4.4} Pa s) and very similar magma rise rates. They contrast with the tonalitic magma, 672 which shows the fastest rise among all studied granitoids and a viscosity $(10^{3.5} \text{ Pa s})$ 673 674 lower than that of the VJS magmas.

(4) The sanukitoid and Vila Jussara granitoid magmas are emplaced closer to the surface
and therefore tend to crystallize more rapidly than the TT magma, which displays
epidote crystals more intensely corroded than those of the first two magmas.

(5) The short epidote crystal dissolution time in the CP suggests that these Archean
magmas were emplaced through a mechanism involving incremental stacking of silllike bodies with a thickness of about a hundred meters, each cooling in approximately
1000 years. In such a view, approximately 42,000 years of magmatic feeding would
have been required through several pulses to form ~ 4-km-thick plutonic bodies,
assuming no time interval between each injection.

(6) Magmatic epidote is generally considered to be a phase absent from granitoids of
different Archean cratons worldwide (Schmidt and Poli, 2004), despite being relatively
common in the CP. This study shows that magmatic epidote crystals can be preserved,
albeit rarely, in Archean granitoids under fast magma rise and crystallization conditions.

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1029 FIGURE CAPTIONS

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Figure 1. (a) Tectonic provinces of the Amazonian Craton with the location of the Carajás Province shown (based on Santos et al., 2003); (b) Simplified geological map of the Carajás Province highlighting the division into the Rio Maria, Sapucaia, Canaã dos Carajás domains and Carajás Basin (Source: Silva et al., 2018, modified); (c) Geological map of the Sapucaia Domain and its transition into the Canaã dos Carajás and Rio Maria Domains, with the location of the studied samples from the Vila Jussara Suite granitoids, the tonalite-Colorado Trondhjemite association of Sapucaia and the Ourilândia do Norte Suite sanukitoids.

Figure 2. Textural relationship of the magmatic epidotes in the CP Archean granitoids. 1038 1039 Trondhjemite-tonalites of Sapucaia: (a) subhedral epidote partly surrounded by biotite and hornblende, exhibiting an irregular shape in contact with alkali-feldspar; (b) allanite crystal 1040 with an epidote rim, showing generally regular contacts with hornblende and irregular 1041 contacts with quartz: Ourilândia sanukitoids: (c) epidote with an irregular shape and signs of 1042 resorption through residual magmatic fluid in contact with hornblende, quartz and 1043 plagioclase; dissolution is apparently more intense when in contact with felsic minerals; (d) 1044 mafic aggregate of epidote, hornblende, biotite, titanite opaque minerals and apatite; (e) 1045 epidote crystal with irregular contacts with hornblende, showing more intense dissolution in 1046 contacts with plagioclase and alkali-feldspar; and (f) anhedral epidote surrounded by 1047 1048 hornblende, with irregular contacts resulting from partial resorption. Abbreviations according 1049 to Kretz (1983).

Figure 3. Histogram of compositional variation in the pistacite component (Ps: molar [$Fe^{3+}/Fe^{3+}+Al$]/100), in molecular percentage (mol.%), in Sapucaia TT epidotes, Ourilândia do Norte sanukitoids and Vila Jussara granitoids. Ranges of compositional variation in Ps in magmatic and secondary epidotes formed by plagioclase and biotite alteration are also indicated.

1055 Figure 4. Textural aspects of thin sections used in the dissolution kinetic study of epidote in the Archean granitoids from the CP. The original shapes of the corroded epidote crystals are 1056 indicated by black lines; dotted lines indicate the largest dimension of the crystal, and red 1057 lines delimit the current contour of the corroded crystals. Line segments delimited by arrows 1058 show where were defined the maximum dissolution values for each crystal. The PFA-14 1059 sample represents the Sapucaia tonalite-trondhjemite; the NDP-47 and NDP-101 samples 1060 represent the Ourilândia do Norte sanukitoid; and the MYF-77 sample represents the VJS 1061 biotite-hornblende monzogranite variety. (a) and (b) epidote crystals are best preserved when 1062 in contact with biotite and partly reabsorbed by the residual magmatic fluid currently 1063 represented by feldspars; (c) epidote involving an allanite euhedral core partly surrounded by 1064 biotite and exhibiting maximum dissolution in contact with felsic minerals; (d) subhedral 1065 epidote crystal heavily corroded when in contact with feldspars; (e) epidote with marked 1066 resorption at the edges, which is less marked in contacts with biotite; and (f) allanite with an 1067 epidote rim, preserved when in contact with biotite and corroded along those with felsic 1068 1069 minerals that formed from residual melt.

1070 Figure 4 continuation. Textural aspects of epidote in the VJS granitoids. Sample PFR-16A is representative of the biotite-hornblende monzogranite variety; samples MDP-02A and LIF-1071 04A are representative of the biotite monzogranite variety and sample MDP-02E is 1072 1073 representative of biotite-hornblende tonalite. (g) anhedral epidote in irregular contact with 1074 hornblende showing a more pronounced resorption in contacts with feldspars and quartz; (h) subhedral epidote crystal with intensely corroded portions; (i) epidote with rectilinear contacts 1075 1076 with biotite and irregular contacts with opaque minerals and feldspars; (j) allanite surrounded 1077 by epidote and in association with opaque minerals, corroded in contacts with feldspars; and 1078 (k) and (l) heavily corroded epidote crystals in contact with hornblende and felsic minerals. Abbreviations according to Kretz (1983). 1079

Figure 5. Diagram showing the variation in the calculated magma viscosity of TT, sanukitoid and the three varieties from the VJS from the CP as a function of temperature. Viscosity was calculated based on the model by Giordano et al. (2008). The red circle at the beginning of the curve represents the estimated liquidus temperature of each magma.

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	Sapucaia tonalite-trondhjemite						Ourilândia do Norte Sanukitoid					Vila Jussara Suite											
Sample		PFA-14			PFR-33A					NDP-47	7				NDP-10	1	ADE	E-01D	AFD	-11A		AMP-77	1
Analyses	C1.1-4	C1.1-5	C1.1-6	C1.1-1	C2.1-3	C2.1-4	C2.1-2	C2.1-3	C3.1-1	C3.1-3	C3.1-4	C3.1-5	C3.1-6	C3.1-4	C3.1-6	C2.1-8	C3.1-1	C3.1-2	C2.1-1	C2.1-3	C2.1-6	C2.1-7	C2.1-8
SiO_2	37.74	38.23	37.82	37.62	38.2,503	38.05	38.21	38.16	37.93	37.53	37.61	37.93	37.51	37.60	37.86	37.92	37.59	37.76	37.55	37.82	37.96	37.82	37.59
TiO_2	0.10	0.09	0.07	0.10	0.04	0.05	0.10	0.11	0.09	0.05	0.04	0.09	0.13	0.09	0.04	0.08	0.01	0.10	0.09	0.09	0.06	0.00	0.07
Al_2O_3	23.10	23.74	23.01	22.63	23.18	23.22	24.90	24.48	23.84	21.37	21.28	24.31	20.64	22.04	22.24	23.28	23.04	22.68	23.45	23.28	24.08	23.96	22.34
Fe_2O_3	13.49	12.98	13.92	12.83	13.50	13.49	9.73	10.19	12.72	15.39	15.61	11.78	15.71	13.60	14.84	13.41	14.04	14.64	12.37	13.83	12.76	13.13	15.12
MnO	0.19	0.19	0.25	0.38	0.36	0.37	0.18	0.16	0.10	0.13	0.16	0.16	0.14	0.10	0.16	0.21	0.18	0.20	0.19	0.23	0.21	0.22	0.18
CaO	23.70	23.53	23.45	23.23	23.13	23.16	23.48	23.52	23.42	23.28	22.45	23.35	22.69	23.25	23.23	23.67	23.36	23.04	23.27	23.31	23.48	23.29	23.08
Na ₂ O	0.00	0.00	0.00	0.00	0.00	0.02	0.01	0.01	0.01	0.02	0.00	0.00	0.01	0.00	0.00	0.00	0.02	0.00	0.02	0.00	0.01	0.01	0.01
K_2O	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.02	0.00	0.00	0.00	0.00	0.01	0.01	0.00	0.02	0.00	0.01
Ce_2O_3	0.05	0.02	0.01	0.00	0.00	0.08	0.03	0.03	0.01	0.00	0.00	0.06	0.00	0.01	0.04	0.04	0.00	0.01	0.03	0.00	0.00	0.05	0.01
La_2O_3	0.00	0.04	0.04	0.00	0.00	0.02	0.03	0.00	0.01	0.02	0.00	0.03	0.02	0.00	0.01	0.00	0.05	0.07	0.04	0.00	0.00	0.00	0.00
Total	98.37	98.83	98.59	96.80	98.23	98.47	96.67	96.66	98.14	97.79	97.15	97.72	96.86	96.70	98.42	98.61	98.30	98.51	97.01	98.54	98.58	98.47	98.43
Number of	cations	on the ba	usis of 13 o	oxygens																			
Si	3.120	3.134	3.122	3.121	3.140	3.138	3.141	3.142	3.127	3.140	3.160	3.132	3.169	3.126	3.137	3.124	3.113	3.124	3.103	3.118	3.117	3.112	3.118
Ti	0.006	0.006	0.005	0.006	0.002	0.003	0.006	0.007	0.006	0.003	0.002	0.005	0.008	0.006	0.003	0.005	0.001	0.006	0.006	0.005	0.004	0.000	0.004
Al	2.251	2.294	2.239	2.213	2.256	2.258	2.413	2.376	2.317	2.107	2.108	2.366	2.055	2.159	2.172	2.260	2.249	2.211	2.283	2.262	2.330	2.324	2.183
Fe ⁺³	0.839	0.800	0.865	0.890	0.839	0.837	0.669	0.702	0.789	0.969	0.987	0.732	0.999	0.946	0.926	0.832	0.875	0.911	0.855	0.858	0.788	0.813	0.944
Mn	0.013	0.013	0.018	0.026	0.025	0.026	0.013	0.011	0.007	0.009	0.011	0.011	0.010	0.007	0.011	0.014	0.013	0.014	0.014	0.016	0.015	0.015	0.013
Ca	2.099	2.067	2.073	2.064	2.047	2.047	2.068	2.075	2.068	2.087	2.021	2.066	2.054	2.071	2.063	2.089	2.073	2.042	2.060	2.059	2.066	2.053	2.051
Na	0.000	0.000	0.000	0.000	0.000	0.004	0.002	0.002	0.001	0.003	0.000	0.000	0.001	0.000	0.000	0.000	0.003	0.000	0.003	0.000	0.001	0.002	0.002
Κ	0.000	0.001	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.002	0.000	0.000	0.000	0.000	0.001	0.001	0.000	0.002	0.000	0.001
Pistacite	27	26	28	29	27	27	22	23	25	32	32	23	33	30	30	27	28	29	27	28	25	26	30

Table 1. Representative electron microprobe analyses of Archean granitoid epidotes from the Carajás Province.

		Sapucaia tonalite-trond	hjemite						
Sample	Grain width	Maximum width of	Partial dissolution	Transport rate					
Sample	(µm)	dissolution (µm)	time (years)	(m/year)					
PFA-14		22							
Spot 1	519	80	4.1	4109					
Spot 2		73	3.4	4935					
Spot 3	259	82	4.3	4693					
Average			4	4579					
	Carrier and dela	Ourilandia do Norte Sai	Dertial discolution	Turner and under					
Sample	Grain width	Maximum width of	time (veers)	(m/yoor)					
NDP 47	(μπ)		time (years)	(III/year)					
Spot 1		85	16	1732					
Spot 2	440	79	4.0	5478					
Spot 3	0	99	62	3488					
Spot 4		68	2.9	7393					
Spot 5	498	122	9.4	2297					
Spot 6		88	4.9	4415					
NDP-101			\mathbf{N}						
Spot 1		121	9.3	2335					
Spot 2	609	81	4.2	5211					
Spot 3		98	6.1	3560					
Average			6	4323					
	Granitoids of the Vila Jussara Suite								
Sample	Grain width	Maximum width of	Partial dissolution	Transport rate					
Bumple	(µm)	dissolution (µm)	time (years)	(m/year)					
MYF-77			_						
Spot 1	415	65	3	7469					
Spot 2		63	3	7951					
PFR-16A		47	1	14296					
Spot 1 Spot 2	252	4/	l 15	14280					
Spot 2 Spot 3	235	133	15	1314					
Average		00	2 5	7957					
Average	Grain width	Maximum width of	Partial dissolution	Transport rate					
Sample	(um)	dissolution (um)	time (years)	(m/vear)					
MDP-02A	()			(111) (111)					
Spot 1	010	110	8	2608					
Spot 2	812	279	49	405					
LIF-04A									
Spot 1		132	11	1811					
Spot 2	464	107	7	2756					
Spot 3		65	3	7469					
Spot 4		60	2	8766					
Spot 5	600	34	1	27299					
Spot 6		51	2	12133					
Average	<u>a</u>		10	7906					
Sample	Grain width (µm)	Maximum width of dissolution (µm)	Partial dissolution time (years)	(m/year)					
MDP-02E									
Spot 1	5/11	74	3	5763					
Spot 2	571	96	6	3424					
Spot 3	337	74	3	5763					
Spot 4	551	42	1	17890					
Average			3	8210					

Table 2. Estimation of the transport rate of magmas based on the kinetic dissolution study of the Archean granitoid epidotes from the Carajás Province.

Lithology	TT	Sanukitoid		Vila Jussara Suit	e
Sample	MYF-19A	BRM-113B	PFR-16A	MDP-02A	MDP-02E
SiO ₂	69.30	65.68	71.44	71.20	60.00
TiO ₂	0.30	0.41	0.36	0.49	1.06
Al_2O_3	16.20	15.52	12.89	13.90	13.90
FeOt	2.79	3.49	4.28	2.97	8.18
MnO	0.04	0.04	0.06	0.03	0.12
MgO	0.77	2.07	0.30	0.80	2.78
CaO	2.85	3.43	1.91	2.23	5.56
Na ₂ O	5.20	4.90	3.46	3.42	3.38
K ₂ O	1.50	2.30	3.93	3.91	1.75
P_2O_5	0.09	0.16	0.07	0.13	0.31
H_2O^*	2.0	7.0	4.0	4.0	4.0

Table 3. Whole rock geochemical composition used to calculate the viscosity of the Sapucaia tonalite-trondhjemite⁽¹⁾, Ourilândia do Norte sanukitoid⁽²⁾ and Vila Jussara Suite⁽³⁾ magmas.

* H₂O wt. % estimate based on rock mineralogy and experimental data. Data sources: ⁽¹⁾Santos et al.(2013b); ⁽²⁾Santos and Oliveira (2016); ⁽³⁾Dall'Agnol et al. (2017) and Silva (2019).

Table 4. Viscosity⁽¹⁾ of Archean granitoid magmas from the Carajás Province at different liquidus⁽²⁾ to solidus temperatures

Melt	TT	Sanukitoid	I	iite	
Sample	MYF-19A	BRM-113B	PFR-16A	MDP-02A	MDP-02E
Temperature (°C)		Viso	cosity (10 ^x Pa	s)	
950		2.5			
940		2.6			
930		2.7			
920		2.7			
910		2.8			
900		2.9			
890		2.9	4.4	4.4	3.5
880	5.3	3.0	4.5	4.5	3.6
870	5.4	3.1	4.6	4.6	3.7
860	5.6	3.2	4.7	4.7	3.8
850	5.7	3.2	4.8	4.8	3.9
840	5.8	3.3	4.8	4.9	4.0
830	5.9	3.4	4.9	5.0	4.1
820	6.0	3.5	5.0	5.1	4.2
810	6.1	3.6	5.1	5.2	4.3
800	6.2	3.6	5.2	5.3	4.4
790	6.4	3.7	5.3	5.4	4.6
780	6.5	3.8	5.4	5.5	4.7
770	6.6	3.9	5.5	5.6	4.8
760	6.8	4.0	5.7	5.7	4.9
750	6.9	4.1	5.8	5.8	5.0
740	7.0	4.2	5.9	5.9	5.2
730	7.2	4.3	6.0	6.0	5.3
720	7.3	4.4	6.1	6.2	5.4
710	7.5	4.5	6.2	6.3	5.6
700	7.6	4.6	6.4	6.4	5.7

(1) Viscosities calculated on the basis of the model proposed by Giordano et al. (2008); (2) The liquidus temperatures of the various magmas are given on Table 5.

Table 5. Estimates* of intensive crystallization parameters of representative samples of Sapucaia tonalite-trondhjemite, Ourilândia do Norte sanukitoid and Vila Jussara Suite magmas

Lithology	Sapucaia TT ⁽¹⁾	Ourilândia do Norte Sanukitoid ⁽²⁾	Vila Jussara Suite ⁽³⁾			
Sample	MYF-19A	BRM-113B		MDP- 02A	MDP- 02E	
Generation Pressure (P						
MPa)	1500	600-900	800-1000	800-1000	800-1000	
Emplacement P (MPa)	500	350	400	400	400	
Liquidus Temperature (T°C)	880	950	890	890	890	
Solidus T (°C)	700	700	700	700	700	
Saturation in Zr T (°C)	794	750	856	888	778	
XH ₂ O (wt. %)	2	7	4	4	4	
fO ₂	NNO±0.5	NNO±0.5 and NNO±2.5	FMQ±0.5	NNO±0.5	NNO±1	

*Estimates based on (1) Clemens et al. (2006) and Santos et al. (2013b); (2) Oliveira et al. (2010) and Santos and Oliveira (2016); (3) Dall'Agnol et al. (2017); cf. text.

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HIGHLIGHTS

- Epidote is relatively common in the Archean granitoids of the Carajás Province.

- The variations in the viscosity of the Archean magmas do not show a clear match only with magma ascent rates.

- The epidote crystal dissolution time in the Carajás Province suggests that Archean magmas were emplaced through a mechanism involving incremental growth.

- A fast magma rise combined with a more superficial emplacement and a fast crystallization promote epidote preservation.

- Epidote dissolution kinetics and viscosity estimates, help to understand the magma rise, emplacement, and crystallization processes of the Archean plutons.

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Declaration of interests

 \boxtimes The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

□ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: