U–Pb geochronology of the Lagoa Real uranium district, Brazil: Implications for the age of the uranium mineralization

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ABSTRACT

The Lagoa Real uranium district in Bahia, northeastern Brazil, is the most important uranium province in the country and presently produces this metal in an open-pit mine operated by Indústrias Nucleares do Brasil. Uranium-rich zones are associated with plagioclase (dominantly albite ± oligoclase) -rich rocks, albites and metasomatized granitic-gneisses, distributed along NNW/SSE striking shear zones. We have used the ID-TIMS U–Pb method to date zircon and titanite grains from the São Timóteo graniteoid, and albite-rich rocks from the Lagoa Real district in order to assess the age of granite emplacement, deformation/metamorphism and uranium mineralization. The isotopic data support the following sequence of events (i) 1746 ± 5 Ma – emplacement of the São Timóteo graniteoid (U–Pb zircon age) in an extensional setting, coeval with the beginning of the sedimentation of the Espinhaço Supergroup; (ii) 956 ± 59 Ma hydrothermal alteration of the São Timóteo graniteoid and emplacement of the uranium mineralization (U –Pb titanite age on an albite-rich sample); (iii) 480 Ma metamorphism, remobilization and Pb loss (U–Pb titanite age for thegneiss sample), during the nucleation of shear zones related to the collision between the São Francisco-Congo and Amazonia paleoplates. The 956 ± 59-Ma mineralization age is apparently associated with the evolution of the Macaúbas-Santo Onofre rift. This age bracket may bear an important exploration implication, and should be included in the diverse age scenario of uranium deposits worldwide.

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1. Introduction

The Lagoa Real granitic-gneiss complex in Bahia State, northeastern Brazil, hosts several uranium-enriched zones and comprises the most important uranium province in Brazil (Figs. 1 and 2). A series of orebodies are known from thirty eight anomalies distributed over an area of 1200 km², roughly along three semi-arched lineaments that cover an approximate extension of 35 km. The orebodies contain a total reserve of ca. 112,000 metric tons of U₃O₈ (Brito et al., 1984; Matos et al., 2003; Matos and Villegas, 2010). Uranium production initiated in 1999 by Indústrias Nucleares do Brasil (INB), and 300 tons per year of uranium concentrate are produced from seven orebodies exploited at the northern Cacheoeira open-pit mine (http://www.inb.gov.br; Matos and Villegas, 2010). Roughly 3.700 tons of U₃O₈ have already been produced by INB.

The uranium deposits are associated with rocks dominated by plagioclase (albite ± oligoclase) and albites, which are the product of extensive, high-temperature metasomatic alteration of granitic-gneissic rocks, mainly in the form of sodium enrichment and silicate depletion, accompanied by oxidation of the original Fe²⁺-dominant phases (e.g., Lobato and Pyke, 1990). Lithotypes dominated by calcium-rich oligoclases host some orebodies along the lineament suggesting Na–Ca metasomatic alteration (Raposo and Matos, 1982; Raposo et al., 1984; Cruz, 2004).
The genesis and age of the uranium mineralization has been the subject of investigation and debate by various authors (e.g., Turpin et al., 1988; Cordani et al., 1992; Pimentel et al., 1994). Previous geochronological studies using Rb–Sr, K–Ar, Sm–Nd and U–Pb methods suggested a polycyclic history for the geological evolution of the Lagoa Real uranium district, with events at ca. 1.7 Ga, 1.3–1.5 Ga and 0.48 Ga (Turpin et al., 1988; Cordani et al., 1992). The formation of the uranium mineralization was assigned to the 1.3–1.5 Ga tectonic event, or Espinhaço collisional event of Turpin et al. (1988) and Cordani et al. (1992), based on U–Pb analyses of heavy mineral concentrates and Rb–Sr whole-rock isochrons.

We carried out a U–Pb geochronological investigation on zircon and titanite grains from granitic rocks and albitites from Lagoa Real, aiming to assess more precisely the crystallization age of the host rocks and subsequent recrystallization episodes, as well as the age of the uranium-enrichment event. Special emphasis is given to the titanite analyses. Due to their relatively low blocking temperature of ca. 600 °C, compared to zircon, titanite can provide good age estimates for metamorphic recrystallization or high-temperature metasomatic alteration processes (Tilton and Grünenfelder, 1968; Mattinson, 1978). The U–Pb data were originally presented by Pimentel et al. (1994) in the form of an extended abstract. However, further comprehensive knowledge of the regional geology of the deposit area was only better unraveled later on by the studies of Arcanjo et al. (2000), Cruz (2004) and Cruz et al. (2007, 2008), allowing a more robust interpretation of the geochronological data, which is put forward in the present contribution.

According to Cuney (2010), uranium deposits evolved through four major time periods. Of these, the third, from 2.2 to 0.45 Ga, records increasing atmospheric oxygen and encompasses uranium deposits related to sodium metasomatism, which are recorded to have formed mostly between 1.8 and 1.4 Ga, and less importantly during the so-called Pan-African or Brasiliano event (~500 Ma; Cuney, 2010). The age of the Lagoa Real mineralization, as discussed below, departs from these, and may help constrain further the understanding of uranium deposition through time.

2. Geological setting

The Lagoa Real deposit area is located in the Paramirim river valley, in the northern part of the Ediacaran-Cambrian Araçuaí Orogen (Pedrosa Soares and Wiedeman, 2000; Pedrosa Soares et al., 2001, 2007, 2008; Alkmim et al., 2006) (Figs. 1 and 2). The basement of the northern portion of the Araçuaí Orogen is made up of terranes containing granitic-gneiss and granulite of Archean and Paleoproterozoic ages, as well as metavolcano-sedimentary belts (Cordani and Brito Neves, 1982; Bastos Leal et al., 1998; Silva and Cunha, 1999; Barbosa and Sabaté, 2002).

The Paramirim valley separates two morphotectonic domains underlain by Proterozoic sedimentary/metasedimentary units, namely the Espinhaço range (ES, Fig. 1), on the west, and the large plateau of the Chapada Diamantina, on the east (CD, Fig. 1). The Proterozoic covers comprise the Pale-to Mesoproterozoic Espinhaço Supergroup (Figs. 1 and 2) and the Neoproterozoic São
Francisco Supergroup (Schobbenhaus, 1996; Misi and Veizer, 1996; Danderfer Filho and Dardenne, 2002). Both these Supergroups are related to the evolution of the complex, polyphase Espinhaço-Macaúbas basin (Schobbenhaus, 1996; Alkmim, 2012). Periods of extensional tectonics are indicated by Meso- and Neoproterozoic mafic dikes that intrude the Espinhaço Supergroup rocks. The dikes comprise two main age groups: (i) group I, characterized by U-Pb ages of 1492 ± 16 Ma (Loureiro et al., 2008) and 1496 ± 3.2 Ma (Guimarães et al., 2005), which are similar to ages obtained by Babinski et al. (1999) at ca. 1514 Ma; (ii) group II, which has U-Pb ages of 934 ± 14 Ma (Loureiro et al., 2008) and 854 ± 23 Ma (Danderfer Filho et al., 2009). Phanerozoic sedimentary rocks also cover large areas of the Paramirim valley (Fig. 1).

The basement rocks of the northern portion of the Araçuaí Orogen have been affected by tectonometamorphism of various periods: (a) Late Archean age (jequê event), (b) Paleoproterozoic (ca. 2.1 to 1.8 Ga), (c) Espinhaço age (1.3–1.0 Ga), and (d) Neoproterozoic, Brasiliano event (0.6–0.5 Ga) (e.g., Inda and Barbosa, 1978; Brito Neves et al., 1980, 1999; Barbosa and Dominguez, 1996; Cordani et al., 1992; Barbosa and Sabaté, 2002; Corrêa Gomes and Oliveira, 2002; Cruz and Alkmim, 2006; Danderfer Filho et al., 2009). Since the significance of the Espinhaço Event is under debate (e.g., Chemale et al., 2010), it is not discussed in detail in this paper. According to traditional views, the Espinhaço Event was responsible for the deformation and metamorphism of the Espinhaço Supergroup and their sialic basement between ca. 1.4 and 1.1 Ga (Cordani et al., 1992). Other authors, however, demonstrated that the Espinhaço rocks and the Neoproterozoic cover units (São Francisco Supergroup) of the northern São Francisco Craton (Almeida, 1977) underwent the same deformaional history and, consequently, the Espinhaço rocks did not experience deformation and regional metamorphism prior to ca. 600 ± 500 Ma (Caby and Arthaud, 1987; Danderfer Filho, 1990, 2000; Uhlein, 1991; Trompette et al., 1992; Alkmim et al., 2006; Cruz and Alkmim, 2006). Furthermore, Cruz and Alkmim (2006) and Cruz et al. (2007) postulated that tectonic elements affecting both the uranium-bearing Lagoa Real granitic-gneiss complex and the Neoproterozoic São Francisco Supergroup are of the same generation.

The Lagoa Real granitic-gneiss complex is enclosed within the granitic-gneissic basement, east of the Espinhaço Range and west of the Chapada Diamantina (Figs. 1 and 2). The basement gneisses exposed to the north and east of Lagoa Real consist of metagranitoids and migmatites of late Archean and Paleoproterozoic ages (Jardim de Sá, 1978; Bastos Leal et al., 1998).

The most conspicuous geological feature of the Lagoa Real area is the presence of several intrusions of porphyritic rocks known

Fig. 2. Regional geological map of central Bahia state, Brazil, showing the main units discussed in the text and the location of the Lagoa Real district (modified after Cruz et al., 2008). ES – Western Espinhaço range; CD – Chapada Diamantina; RP – Rio Preto.
Fig. 3. Geological sketch map of the Lagoa Real district in the Paramirim river valley (inset of Fig. 2). Towns: C = Caetité; P = Paramirim; IB = Ibiassucê; ST = São Timóteo (after Cruz et al., 2008).
generically as the São Timóteo granitoid (Figs. 3 and 4A). They comprise coarse-grained albitized syenogranite, alkali-feldspar granite and syenite. These are locally converted into protomylonites, augen mylonites and ultramylonites of the same composition, exhibiting gneissic structure, and are here collectively named albitized gneisses (Fig. 4B; Lobato and Fyfe, 1990; Cruz and Alkmim, 2006; Cruz et al., 2007, 2008). The most abundant facies of the São Timóteo granitoid corresponds to a potassic, hastingsite-biotite monzo-to syenogranite of metaluminous and high-K calc-alkaline affinity (Teixeira, 2000). Maruejol et al. (1987) indicate that these are iron-rich, metaluminous, subalkaline plutonic rocks of intracontinental environment.

3. Uranium mineralization

In association with the gneissic rocks of the Lagoa Real granitic-gneiss complex or district, there is a significant number of lenticular albite(±oligoclase)-rich bodies exposed along a ca. 100-km long, arcuate belt roughly in the central portion of the district (Fig. 3). Shear zone-hosted albite(±oligoclase)-rich rocks and albitites may locally contain uranium mineralization (Fig. 4C). They are fine-to medium-grained rocks showing weak to pronounced foliation (Lobato and Fyfe, 1990; Cruz et al., 2007). Where uraninite-bearing, these rocks may contain more than one mineralized zone. Round uraninite crystals are very fine (5–25 µm), chiefly associated with andradite, but also found included and bordering aegerine-augite, albite-oligoclase, hastingsitic hornblende, biotite (Fig. 4D), calcite, and martitized magnetite (Lobato and Fyfe, 1990). Calcium-rich oligoclases may also host uranium ore; these contain epidote, hedenbergite, diopside, grossular and also martitized magnetite (Cruz, 2004). Further detailed mineralogical studies are found in Prates (2008).

Field evidence indicates that the plagioclase(±oligoclase)-rich rocks have been formed by metasomatism and shearing of the São Timóteo granitoid (e.g., Lobato and Fyfe, 1990). These authors point out that all uranium-rich zones in Lagoa Real are associated with metasomatized rocks. Therefore, shearing, metasomatism (sodium enrichment and silica depletion), and mineralization must have been coeval. However, later studies by Cruz et al. (2007) based on microstructural criteria suggested that Na–Ca alteration pre-dates the nucleation of the shear zones that host uranium mineralization.

Other lines of evidence include the presence of flame perthite hosted in K-feldspar of the alkaline São Timóteo granitoid, as well as albite/oligoclase veins and agglomerates, both features found in distal zones to mineralization. Close to shear zones, these plagioclase agglomerates are progressively recrystallized giving place to the albitites (Cruz, 2004; Cruz et al., 2008).

Sodium metasomatism and uranium mineralization, collectively referred to as albitite-type (e.g., Wilde, 2013), metasomatic (Cuney and Kyser, 2008), or albitite-hosted (Polito et al., 2007) uranium deposits are widely distributed in the world, and no single genetic model is consistent with the examples cited in the literature (e.g., Cuney and Kyser, 2008; Wilde, 2013). For the Lagoa Real uranium deposits, no consensus exists regarding their genesis, despite the various studies in the area (e.g., Geisel Sobrinho et al., 1980; Lobato, 1985; Maruejol et al., 1987; Fuzikawa et al., 1988; Maruejol, 1989; Lobato and Fyfe, 1990; Cruz, 2004; Chaves et al., 2007; Souza, 2009; Oliveira, 2010; Oliveira et al., 2012; Chaves, 2013), and a thorough discussion of the subject is beyond the scope of this paper.

According to Lobato and Fyfe (1990), uranium mineralization developed in association with hot (~500–550 °C), oxidizing fluids that had oxygen isotope values somewhat similar for both non-mineralized, from −0.8 to +7.3 per mil, and mineralized plagioclase(±oligoclase)-rich rocks, ranging from −3.7 to +2.6 per mil.

Fluid inclusion studies were primarily undertaken by Fuzikawa et al. these authors do not propose (1988) on albite, late-stage quartz and calcite, with indication of aqueous and aquo-carbonic fluids. The authors also imply that a late-stage brine may have existed with oscillating salinities up to 20 percent NaCl equivalent. More recently, Souza (2009) and Oliveira (2010) obtained salinity values that range from 9 to 18 percent NaCl equivalent for the aqueous fluid. The latest fluid inclusion studies coupled with LA-ICMPS analyses (Oliveira et al., 2012) focus on data obtained on
pyroxenes (both metamorphic and metasomatic), garnet and plagioclase of albites, from three of the most important deposits at Lagoa Real. According to these authors, the early-stage aqueous fluids in equilibrium with mafic minerals were saline (12–16% NaCl equivalent), and contained Na, Mg, U, Rb, Ba, Sr, Pb, K, Ca, Fe, Cu, Zn and Li, as well as some Mn, As and Sb. On the other hand, plagioclase precipitated from a more diluted aqueous fluid (0.5–6.4% NaCl equivalent; up to 11% as stated in Fuzikawa et al., 1988), containing Na, Mg, K, Ca, Fe, Cu, Zn, As, Sr, Ba, Pb. Aquo-carbonic fluids are only detected in vein quartz crosscutting mineralization. Despite all these information, these authors have not come to a consensus as to the nature of the fluids involved.

Based primarily on field work, Geisel Sóbrinho et al. (1980) and Stein et al. (1980) anticipated that the Lagoa Real uranium derived from magmatic fluids, without indicating their precise source. Studies by Maruejol et al. (1987), Turpin et al. (1988) and Maruejol (1989) suggest that the mineralizing fluids were derived from the Paleoproterozoic São Timóteo granitoid, which contains uranium-bearing accessory minerals, as the mineralizing agent.

Additionally, on the basis of 87Sr/86Sr isotope ratios in the range of 0.704–0.723 obtained for the albitites, Lobato and Fyfe (1990) discarded the granitic-gneiss (87Sr/86Sr isotope ratios between 0.7038 and 0.814) terrane as the main source of the mineralizing fluids. Alternatively, the authors suggest that the fluids that may have been responsible for the uranium mineralization were derived from the Espinhaço Supergroup rocks, which are adjacent to the Lagoa Real granitic-gneiss complex (Lobato, 1985). Such fluids would be analogous to oxidizing (Hanor, 1979), saline, modernization fluids. Despite all these information, these authors have not come to a consensus as to the nature of the fluids involved.

4. Previous geochronological studies in the Lagoa Real uranium district

The main geological units exposed in the Lagoa Real uranium district have been the subject of geochronological studies using multichronometric approaches (U–Pb, Pb–Pb, Rb–Sr, Sm–Nd and K–Ar methods) in order to constrain the timing of granite emplacement, metasomatism, hydrothermal alteration and uranium mineralization (Turpin et al., 1988; Cordani et al., 1992). These studies produced a wealth of isotopic and geochronological data, which are summarized in Table 1.

4.1. Basement gneiss

Geochronological U–Pb, Rb–Sr and Sm–Nd data for the Para-mirim valley basement rocks suggest crust formation to have been related to at least three Archean plutonic events. The earliest event dates back to the Paleoarchean at around 3.4–3.2 Ga (Martin et al., 1991; Nutman and Cordani, 1993; Bastos Leal et al., 1998; Santos Pinto et al., 1998, 2012), and is represented by tonalite-trondjemitic-granite plutons. A 3.2–3.1 Ga plutonic event, represented by granitic-granodioritic rocks, was probably responsible for the reworking of rocks from the Paleoarchean event (Bastos Leal, 1998; Bastos Leal et al., 1998). The youngest plutonic event was dated at 2.9–2.7 Ga by the Rb–Sr and Pb–Pb systems (Brito Neves et al., 1979; Costa et al., 1985; Cordani et al., 1985, 1992; Santos Pinto, 1996).

Samples that represent neosome of migmatitic gneisses exposed to the northwest and north of Lagoa Real were dated by a Rb–Sr whole-rock. An isotopic Rb–Sr age of 2650 ± 100 Ma was obtained with a high initial 87Sr/86Sr ratio of ca. 0.712 (Cordani et al., 1992). This late Archean date was interpreted as the age of migration, related to the Jequibá tectono-thermal event. Rhyanian (2300–2050 Ma) and Orosirian (2050–1800 Ma) calc-alkaline granitic rocks intrude these gneisses (Bastos Leal et al., 1998; Guimarães et al., 2005; Leal et al., 2005). Two K–Ar analyses in biotite from a gneiss and amphiboles from a migmatite revealed ages of 538 and 491 Ma, respectively, indicating that cooling followed the Neoproterozoic Brasiliano event (Cordani et al., 1992).

4.2. São Timóteo albitized granite

Samples of the São Timóteo albitized granite (Fig. 4A) have been previously investigated by the U–Pb, Pb–Pb, Rb–Sr, Sm–Nd and K–Ar methods (Turpin et al., 1988; Cordani et al., 1992; data in Table 1).

Turpin et al. (1988) reported a zircon U–Pb age of 1724 ± 5 Ma, and interpreted it as the best estimate for the age of emplacement of the São Timóteo granitoid. Zircon grains from deformed granite and gneissic samples lie on the same discordia, and these rocks were considered to be the recrystallized facies of the São Timóteo granitoid. An additional discordia line was traced based on analyses of morphologically different zircon grains indicating upper and lower intercept ages of ca. 1726 Ma and 524 Ma, respectively. The latter age suggests a period of Pb loss during the Cambrian that may be related to shear zones truncating the São

Table 1

<table>
<thead>
<tr>
<th>Rock unit</th>
<th>Material</th>
<th>Method</th>
<th>Age (Ma) and isotopic Parameters</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Basement gneiss and migmatite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>2650 ± 100 (i. r. = 0.712)</td>
<td>2</td>
</tr>
<tr>
<td>Basement gneiss</td>
<td>Biotite</td>
<td>K–Ar</td>
<td>538</td>
<td>2</td>
</tr>
<tr>
<td>Migmatite</td>
<td>Amphibole</td>
<td>K–Ar</td>
<td>491</td>
<td>2</td>
</tr>
<tr>
<td>Granite</td>
<td>Zircon</td>
<td>U–Pb</td>
<td>1724 ± 5</td>
<td>1</td>
</tr>
<tr>
<td>Granite</td>
<td>Whole rock</td>
<td>Pb–Pb</td>
<td>1706 ± 107</td>
<td>2</td>
</tr>
<tr>
<td>Granite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1710 ± 45 (i. r. = 0.7135)</td>
<td>2</td>
</tr>
<tr>
<td>Granite</td>
<td>Biotite</td>
<td>K–Ar</td>
<td>573</td>
<td>2</td>
</tr>
<tr>
<td>Lineated granite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1280 ± 20 (i. r. = 0.775)</td>
<td>2</td>
</tr>
<tr>
<td>Gneiss</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1629 ± 30 (i. r. = 0.7104)</td>
<td>1</td>
</tr>
<tr>
<td>Orthogranite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1000 ± 60 (i. r. = 0.722)</td>
<td>2</td>
</tr>
<tr>
<td>Orthogranite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1220 ± 130 (i. r. = 0.723)</td>
<td>2</td>
</tr>
<tr>
<td>Gneiss</td>
<td>Amphibole</td>
<td>K–Ar</td>
<td>503</td>
<td>2</td>
</tr>
<tr>
<td>Albitite</td>
<td>Acid-soluble fractions of whole-rock, heavy mineral, and uraninite concentrates</td>
<td>U–Pb</td>
<td>1397 ± 9</td>
<td>1</td>
</tr>
<tr>
<td>Albitized orthogranite</td>
<td>Whole rock</td>
<td>Rb–Sr</td>
<td>1520 ± 20 (i. r. = 0.7221)</td>
<td>2</td>
</tr>
</tbody>
</table>
Timóteo granitoid; these shear zones are associated with the deformational history of the northern portion of the Araçáui Orogen, and result from the collision between the São Francisco-Congo and Amazonia paleotplates during the Ediacaran (Cruz and Alkmim (2006; Alkmim et al., 2006). According to Turpin et al. (1988), the gneiss formation took place during a late stage of the Brasiliano orogenic event, at ca. 480 Ma.

Cordani et al. (1992) also analyzed samples of the São Timóteo granitoid. They report the historical whole-rock Rb–Sr and Pb–Pb isochron ages of 1710 ± 45 Ma and 1706 ± 107 Ma, respectively, in agreement, within errors, with the U–Pb magmatic age of Turpin et al. (1988). However, Rb–Sr analyses on samples collected from the gneissic facies of the São Timóteo granitoid yield isochron ages at ca. 1.6, 1.2 and 1.0 Ga (Turpin et al., 1988; Cordani et al., 1992), probably reflecting the disturbance of the isotopic system since these ages have no geological significance. The high initial 87Sr/86Sr ratio (0.7135) and the model 1 value of 8.4 are compatible with an important crustal component in the parental magma. This is also supported by the Sm–Nd isotopic data of Turpin et al. (1988), which give TDM model ages between ca. 2.2 and 3.0 Ga, and by the high 87Sr/86Sr values reported by Lobato et al. (1983) in quartz samples from the Lagoa Real granite. A considerably younger Rb–Sr age (1629 ± 30 Ma) was interpreted as a disturbance of the Rb–Sr isotopic system during the Brasiliano tectonothermal event (Turpin et al., 1988).

Magmatic titanite grains of the São Timóteo granitoid yielded an Pb–Pb age of 1743 ± 28 Ma (Cruz et al., 2007). Amphibole and biotite K–Ar cooling ages for samples of the São Timóteo granitoid are 503 Ma and 573 Ma, respectively, which indicate a strong Brasiliano event overprint in the area (Cordani et al., 1992).

4.3. Albitized gneisses and albitites

The U–Pb data for uraninite grains extracted from the Lagoa Real albitized gneisses yielded a concordant age of ca. 820 Ma (Stein et al., 1980), interpreted to represent the time of the metasomatic alteration caused by alkaline fluids generated during a basin-forming event.

Heavy minerals separated from albitite samples were dated by the U–Pb TIMS method (Turpin et al., 1988). The zircon fractions studied show extremely positive as well as negative discordances (sample 95 of their work is ~20% discordant). These authors interpreted the unusual behavior of the albitite zircon grains as the result of relative mobility of U, Pb and 238U series products. Nevertheless, the albitite zircon grains align along a discordia line with an upper intercept age of 1504 ± 12 Ma, which is in agreement with an 1520 ± 20 Ma Rb–Sr whole-rock isochron age reported by Cordani et al. (1992) for variably albitized augen orthogneisses from localities near the uranium mineralization.

A discordia with seven analytical points for acid-soluble fractions of whole-rock albitite samples, heavy mineral concentrates and uraninite concentrates is also reported by Turpin et al. (1988). The upper and lower intercept ages of 1397 ± 9 Ma and 480 Ma were interpreted as the ages of uranium mineralization and reworking of the uranium deposit, respectively. However, these ages are not robust considering that distinct U-bearing mineral phases, with different closure temperatures, were analyzed together.

5. Analytical procedures

For the present work, heavy mineral fractions were obtained from ca. 20 kg samples by panning at the laboratories of Vale (previously Rio Doce Geologia e Mineração S.A.), Belo Horizonte, Brazil. These fractions were further processed by sieving, magnetic separation using a Frantz isodynamic separator, and conventional heavy liquid separation at the GEOTOP, Université du Québec a Montréal. A combination of the following techniques was applied to reduce discordance: (i) hand-picking of the most transparent, least-altered and crack-free zircon grains from the least-magnetic fractions, and (ii) air abrasion following the procedure of Krogh (1982).

Despite severe abrasion, many zircon fractions in this study are still discordant, reflecting the difficulty in obtaining closed U–Pb system behavior. Titanite fractions were not abraded. The selected zircon and titanite grains were homogeneous in size, color and shape.

Zircon dissolution and chemical extraction of U and Pb followed the general procedures described by Krogh (1973). A 206Pb/236U(238U–235U) mixed spike was used. For titanite, the chemical procedures outlined by Curf and Stott (1986) were followed. Analyses were performed on a VG Sector 54 mass spectrometer at the Centre de Recherche en Géochimie et en Géochronologie (GEOTOP) of the Université du Québec a Montréal, Canada. Errors at the 1-sigma level are generally around 0.5% and 0.1%, respectively for the U/Pb and 207Pb/206Pb ratios. In samples with higher amounts of initial common lead, errors in U–Pb and 206Pb/208Pb ratios are ca. 1.0% and 0.3%, respectively. Regression lines were calculated using the method described by Ludwig (2008) and quoted errors represent the 95% confidence level.

6. Results

6.1. São Timóteo granitoid (sample HBU-872)

Sample HBU-872 is a coarse-grained, isotropic granite, exhibiting an incipient and poorly defined foliation. Other than perthitic K-feldspar, plagioclase and weakly recrystallized quartz, this granite also contains hastingsite, iron biotite, and traces of almandine, zircon and opaque minerals.

Three zircon fractions from this intrusion were analyzed, and the results are shown in Table 2. Although core-overgrowth relationships were not observed in the fractions investigated, we analyzed the tips of large (200–300 μm), colorless, good quality prismatic crystals (fractions HBU-872.1 and HBU-872.2; Fig. 5) to ensure that no inherited Pb was included. Fraction HBU-872.3 comprised only whole crystals. Analyses of fractions HBU-872.1 and HBU-872.2 produced nearly identical results. They are both ca. 1.0% discordant and yielded 206Pb/207Pb ages of 1747 and 1745 Ma, respectively (Fig. 5). Fraction HBU-872.3 has a much larger common lead content and required a larger correction, which resulted in a larger error ellipse. Nevertheless, this fraction indicated a similar 206Pb/207Pb age (1746 Ma), being ca. 0.7% discordant. The three fraction analyses are collinear, defining a discordia line with an upper intercept age of 1746 ± 5 Ma (Fig. 5) considered as the best estimate for the crystallization age of the São Timóteo granitoid.

6.2. Mineralized albitite (sample HBU-871)

Sample HBU-871 is a mineralized andradite albitite. It is fine-to-medium-grained, granoblastic to polygonal granoblastic, exhibiting albitite, andradite, diopside (aegirine-augite), uraninite, plus traces of titanite, zircon, martitized magnetite and apatite. Relicts of hastingsitic amphibole (after pyroxene), biotite (after garnet), carbonate and epidote are rare. Uraninite forms inclusions in garnet and pyroxene.

Non-magnetic zircon grains in the albitite sample are morphologically identical to those of the São Timóteo granitoid. This observation agrees with the interpretation of Turpin et al. (1988)
that the albitite zircon grains are relict crystals from the original granite. Two zircon fractions were analyzed and the results are included in Table 2. Fractions HBU-871.6 and HBU-871.7 are slightly more discordant (1.2% and 1.8%, respectively; Fig. 6) than those of the São Timóteo granitoid zircon grains (Fig. 5), but show $^{207}\text{Pb}/^{206}\text{Pb}$ ages of 1741 and 1744 Ma, very similar to the crystallization age of the São Timóteo granitoid. When zircon fractions for the granite and the albitite are plotted together on a concordia diagram, they define a discordia line with an upper intercept of 1744 ± 4 Ma (Fig. 7), in agreement with the crystallization age obtained from the granite zircon grains alone. This corroborates field evidence that the albitite-oligoclase-rich rocks and albitites represent deformed and metamorphosed portions of the São Timóteo granitoid.

Two distinct titanite populations with contrasting color characteristics (colorless and dark brown) are found in the albitite sample. Fractions HBU-871.1 and HBU-871.3 are made of colorless to very pale brown, good quality titanites, whereas fractions HBU-871.2 and HBU-871.5 represent the dark brown population. The latter revealed higher U contents (114 and 421 ppm, respectively). The average U content of titanite grains in fraction HBU-871.5 is high, and somewhat similar to that of zircon grains (134–185 ppm) from the same rock sample.

The four titanite fractions were variably discordant (from 17% to 150%; Fig. 8), and there is no relationship between the degree of discordance and the color or U contents of the different mineral fractions. Reverse discordance in U-bearing minerals is explained by U-loss or more likely by an excess $^{206}\text{Pb}$. It is suggested that the titanites went through a complex history of Pb and U loss after their crystallization. Alternatively, one could imagine that this was caused by minute U-poor, Pb-rich mineral inclusions in titanite. Nevertheless, the four analytical points form a discordia line with a 4% probability of fit, which is here understood to be a good fit, if one considers the very large spread of the analytical points. The discordia defines an upper intercept age of 956 ± 59 Ma and a lower intercept age of 384 ± 100 Ma (Fig. 8).

The high U content of some of the titanite grains is unusual in this kind of mineral and suggests that they are coeval with the uranium mineralization. The upper intercept age of 956 ± 59 Ma (Fig. 8) is, therefore, the best estimate for the age of the hydrothermal activity that gave origin to the uranium mineralization.

### 7. Discussion and conclusions

The age of 1746 ± 5 Ma for the São Timóteo granitoid is similar to the $\text{U-Pb}$ zircon crystallization ages, in the range of 1711 Ma and 1770 Ma of acid volcanic rocks from the basal portion of the Espinhaço Supergroup dated in several areas of Bahia and Minas Gerais states (Brito-Neves et al., 1979; Machado et al., 1989; Dossin et al., 1993; Schobbenhaus et al., 1994; Babinski et al., 1999).

The 1746 ± 5 Ma São Timóteo magmatic event is roughly coeval with this rift-type acid magmatism (1711 Ma and 1770 Ma) that occurs both in Minas Gerais state and further to the west in the state of Goiás, represented by rhyolites at the base of the Espinhaço (Minas Gerais) and Arari (Goiás) Groups (e.g., Fuck et al., 1988; Trompette et al., 1992). It is associated with A-type plutonic rocks in Minas Gerais, known as the Borrachudos suite (Dorr and Barbosa, 1963), comprising a series of sub-alkaline granitic bodies that cut the basement rocks.

Chemale et al. (1998) and Fernandes et al. (1997) presented $\text{U-Pb}$ zircon ages for two the Borrachudos plutons at 1670 ± 32 Ma and 1777 ± 30 Ma, respectively, whilst a much more precise $\text{U-Pb}$ SHRIMP age of 1740 ± 8 Ma is reported by Silva et al. (2002). In Goiás, these granites have been dated between ca. 1769 and 1770 Ma (Pimentel et al., 1991). The emplacement of the São
Timóteo granitoid is, therefore, part of a regional extensional event during which the deposition of the Espinhaço Supergroup took place in Minas Gerais and Bahia states, as well as the Araí Group in Goiás state.

Our study failed to recognize a Mesoproterozoic event of hydrothermal alteration and uranium mineralization, as suggested by Turpin et al. (1988) and Cordani et al. (1992). Our data indicate that the processes above occurred at the beginning of the Neoproterozoic. The discordia line with the upper intercept age of ca. 1397 Ma presented by Turpin et al. (1988) was defined by analyses of acid-soluble fractions derived from heavy mineral concentrates and albite whole-rock samples. As observed in our study, these heavy mineral concentrates may contain minerals which are ca. 1742 Ma old (e.g. relict zircon grains from the original granitic rock), as well as minerals crystallized or reset at 956 ± 59 Ma (e.g. titanite grains). The age of 1397 Ma is intermediate between ca. 1742 Ma and 956 ± 59 Ma, and it is here suggested to represent a mixed age, obtained by mixing U and Pb extracted from minerals with different ages. This age range of Turpin et al. (1988) has been inferred as the Lagoa Real mineralizing event in a contribution by Cuney et al. (2012).

Cordani et al. (1992) based their conclusions on Rb–Sr whole-rock isochrons on albitized gneisses and albites. They report several isochrons displaying ages of ca. 1520 Ma, 1280 Ma, 1220 Ma and 1000 Ma. We suggest that these isochron ages could alternatively be interpreted as the result of partial re-homogenization of older rocks, due to the younger (Neoproterozoic) tectonothermal event.

Age determinations performed by Babinski et al. (1999) and Danderfer Filho et al. (2009) on basic rocks exposed in the Chapada Diamantina and northern Espinhaço range point toward a 1.51 ± 1.57 Ga magmatic event, probably associated with a renewed rifting episode in the Espinhaço basin.

The significance of the 956 ± 59 Ma hydrothermal event is still not completely understood. Results obtained by Loureiro et al. (2009) and Danderfer Filho et al. (2009) in the regions of the Chapada Diamantina and northern Espinhaço, respectively, indicate the existence of a Tonian (1.0–850 Ma) mafic magmatism. A single outcrop belonging to the upper part of the Chapada Diamantina metasedimentary pile yielded an age of about 960 Ma, while the lower part furnished an age of 920 Ma (Cordani et al., 1992); these latter rocks discordantly overlie the São Francisco Supergroup. In other areas of the São Francisco Craton, an extensional event marked by the emplacement of mafic dikes and sills has been dated at ca. 1.0 Ga (Machado et al., 1989; Renne et al., 1990), and is interpreted as the initial stages of extension associated with the formation of the Neoproterozoic Macaúbas basin (e.g., Schobbenhaus, 1996), which covers large areas of the interior and of the eastern part of the São Francisco Craton. The Salto da Divisa granitic rocks from the northern Craton region were dated by Silva et al. (2008), and their ages fall in the interval between 875 and 850 Ma. These rocks are interpreted as a manifestation of the Macaúbas rifting event (Pedrosa Soares et al., 2008; Danderfer Filho et al., 2009).

The titanite age at 956 ± 59 Ma falls, therefore, into the time interval related to the rifting stage of the precursor basin of the Araçuaí-West Congo orogen. This stage started at the onset of the Tonian and lasted until ca. 875 Ma, with the intrusion of the Salto da Divisa anorogenic plutonic suite in northeast Minas Gerais and
southwest Bahia states (Silva et al., 2008). However, thick rift-related volcano-sedimentary units older than 912 Ma are only found in the African side, suggesting an asymmetrical rift with the thermal–magmatic axis slowly migrating westward (Pedrosa Soares et al., 2008).

As mentioned earlier, Lobato (1985) and Lobato and Fyfe (1990) pointed out that their data suggest the Lagoa Real uranium mineralization derived from the interaction with basinal brines (see also Cuney, 2009), a model that is yet to be fully established. The 956 ± 59 Ma mineralization age presented in this paper helps corroborate this assumption, since this time period represents a major rifting stage. This age bracket may stand as an important exploration implication, and should be included in the diverse age scenarios proposed by Cuney (2010).

The importance of the Brasiliano tectono-thermal event in the region has been demonstrated by K–Ar data obtained by various authors. For instance, Jardim de Sá et al. (1976) indicate an age of 492 ± 25 Ma for a saussuritized gabbro intrusion in the Chapada Diamantina sequence. Bastos Leal (1998) dated biotites of the Orosirian Caculó, Espirito Santo and Iguatemi granites, some 35–50 km east of the Lagoa Real complex, at 551 ± 6 Ma, 490 ± 12 Ma and 483 ± 5 Ma, respectively. The K–Ar ages by Cordani et al. (1992) on amphibole (503 Ma) and biotite (573 Ma) of the Lagoa Real Granite are further indicative of the Neoproterozoic overprint.

The Ar–Ar age of 497 Ma on white mica of sheared gneisses in the Paramirim valley region (Guimarães et al., 2005) also point to a Neoproterozoic reworking event. Moreover, Cruz (2004), Cruz and Alkmim (2006) and Cruz et al. (2007, 2008) have demonstrated that the deformation indicators in the Lagoa Real granitic-gneiss district are related to shear zones that also truncate the Proterozoic Espinhaço and São Francisco Supergroups, constraining the age of these structural features to the Neoproterozoic overprint.

In summary, the data presented in this study support the following sequence of events for the geological evolution of the Lagoa Real uranium district:

(i) 1746 ± 5 Ma — emplacement of the São Timóteo granitoid (U–Pb zircon age);
(ii) 956 ± 59 Ma — hydrothermal alteration, albite formation and age of the uranium mineralization (U–Pb titanite age for the albite sample);
(iii) Cambrian age of 480 Ma — final recrystallization, remobilization and Pb loss at the final stages of the Brasiliano tectono-thermal event.

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