

FLUID EVOLUTION AND GOLD DEPOSITION AT THE CUIABÁ MINE, SE BRAZIL: FLUID INCLUSIONS AND STABLE ISOTOPE GEOCHEMISTRY OF CARBONATES

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ABSTRACT The gold-bearing quartz-carbonate vein deposits of the Cuiabá Mine, northern sector of the Quadrilátero Ferrífero, State of Minas Gerais, SE Brazil, are hosted by a sequence of banded carbonaceous metasedimentary rocks within tholeiitic basalt flows of the Archean Rio das Velhas Greenstone Belt. A regional greenschist metamorphic grade characterizes rocks in this section of the Belt. Episodes of hydraulic fracturing enhanced the focussing of fluids percolating along the host primary layering, which promoted pervasive carbonatization and sulfidation associated with crosscutting fractures and quartz-carbonate veins/breccias. The gold mineralization is closely related to the sulfide-rich zones. Fluid inclusion investigations show that the mineralizing fluids were dominantly aqueous, of low salinity (3–6 wt.% NaCl_{eq.}), with variable concentrations of CO₂ (0–11.4 mol%), CH₄ (1.5–10.2 mol%), subordinate N₂ (0–0.7 mol%), and traces of H₂S/HS⁻. Carbonates in the carbonaceous host rock display heavier $\delta^{13}\text{C}$ compositions (–4.3 to –0.2‰) than carbonates in the gold-related, carbonate-sulfide altered zones and associated veins and breccias (–8.1 to –5.5‰). The range of carbonate $\delta^{18}\text{O}$ values in the carbonaceous host rock (12.8 to 16‰) is broadly similar to the carbonates in the mineralized zones (10.6 to 15.2‰), with a slightly larger variation in the latter. Collectively, fluid inclusion and carbon isotope data indicate that: (1) the mineralizing fluid was originally composed of a H₂O–CO₂ solution, with significant concentrations of CH₄ added via hydrolysis of the carbonaceous matter during vein-fluid/wallrock interaction; (2) sedimentary carbonates, probably of marine origin, are readily distinguished from those carbonates formed by reaction between CO₂-bearing vein fluids and wallrocks; (3) gold-related vein-fluid CO₂ was relatively homogeneous in isotopic composition, suggesting equilibration with carbonate and reduced carbon reservoir (e.g. mantle-derived magma); (4) gold deposition may have been triggered by a sharp decrease in $f\text{O}_2$ caused by the CH₄ enrichment of the fluid.

Keywords: hydrothermal alteration, gold mineralization, Cuiabá Mine, fluid inclusions, carbon isotopes.

INTRODUCTION The Cuiabá Mine is one of the most important gold-producing mine in the northern sector of the Quadrilátero Ferrífero, State of Minas Gerais, SE Brazil, with an output of 45000 t of ore monthly, at a mean grade of 8.4 g/t, and reserves estimated at 70t of Au (Vial 1988). The gold mineralization is mainly hosted by a 5 to 15 m-thick sequence of banded carbonaceous metasedimentary rock, metamorphosed in the greenschist facies, within an Archean mafic volcanic sequence forming the base of the Rio das Velhas greenstone belt (Nova Lima Group; Fig 1A). This rock consists of a rhythmic alternation of dark, white, and ocher-colored, millimeter- to centimeter-thick bands, rich in amorphous carbonaceous material, fine-grained quartz, and carbonate, respectively. It has been generally referred to in the literature as a banded iron formation or banded ferruginous metachert (Vial 1980; Ribeiro-Rodrigues 1998; Lobato *et al.* 1998).

The volcano-sedimentary sequence at the mine underwent deformation at different crustal levels, during three successive phases: D₁ and D₂ phases developed in a ductile to ductile-brittle regime, under compressive stress oriented in the SE–NW direction, with tectonic transport from SE to NW; D₃ structures were formed in a brittle-ductile regime and reflect the action of E–W oriented compressive stresses. The structural framework of the deposit is controlled by a large anticline (F₂), with an overturned northern limb outlined by the folding of the primary bedding (Vial 1980). This anticline shows a penetrative S₂ axial plane foliation and presents a tubular geometry with the apex closure pointing to the northwest direction and axis inclined to about 30° to the southeast (Toledo 1997; Fig. 1B).

The gold mineralization occurs closely associated with carbonate-sulfide altered zones that envelop breccias and quartz-carbonate veins. Carbonatization is defined by the precipitation of Ca–Fe–Mg carbonates at the expense of the carbonaceous matter, which is evidenced by the appearance of bleached zones around quartz-sulfide veins, whereas sulfidation is characterized by the formation of banded to massive aggregates of pyrite±pyrrhotite±arsenopyrite that replace carbonates along the sedimentary layering (Toledo 1997).

This paper addresses the question of the fluid-carbon interaction at the Cuiabá Mine, particularly the influence on the chemical and isotopic composition of the fluid involved in the carbonate-sulfide alteration and deposition of the gold, by means of fluid inclusion and carbon stable isotope studies.

SAMPLED SELECTED AND ANALYTICAL PROCEDURES

A fluid inclusion investigation was carried out on 3 doubly polished sections: two of a D₁-related quartz-sulfide breccia and one of a S₂

foliation – parallel quartz-carbonate vein, all representative of the Fonte Grande Sul ore body, level 7 (underground) of the Cuiabá Mine. Microthermometry and Laser Raman microspectroscopy (LRM) measurements of inclusion fluids hosted by quartz were performed on a LINKAM THMSG600 stage and on a Jobin Yvon T64000 Laser Raman Spectrometer, respectively, at the Fluid Inclusion Laboratory, Institute of Geoscience, State University of Campinas (UNICAMP). The stage was calibrated using synthetic fluid inclusions to a precision of $\pm 0.3^\circ\text{C}$ for freezing runs down to -56.6°C and to $\pm 3^\circ\text{C}$ for heating runs up to 500°C .

The stable isotope ratios of $^{13}\text{C}/^{12}\text{C}$ and $^{18}\text{O}/^{16}\text{O}$ were determined on carbonates separated from 3 samples of the banded carbonaceous metasedimentary host, 11 samples of the ore zones (e.g. Fonte Grande Sul, Serrotonho, Galinheiro and Balancão ore bodies), and 2 barren samples, including a quartz-carbonate-sulfide vein and a carbonate veinlet.

X-ray diffraction analyses of the investigated samples identified ankerite, siderite, calcite and dolomite. In the carbonaceous host, calcite and siderite are common, whereas ankerite very subordinate. Within the carbonate-sulfide-altered zones, and associated breccia and veins, ankerite and siderite predominate, accompanied rarely by dolomite and calcite. Calcite seems to be the main carbonate type in the barren veins.

Samples were prepared for isotopic analysis using the standard phosphoric acid technique (at 25°C and 50°C) of McCrea (1950) to release CO₂ for mass spectrometric analysis. As the reaction rates are dependent upon the types of carbonate in the sample, the CO₂ was collected at different times according to the carbonate species present in order to avoid a mixture of CO₂ from different sources, following the procedure in Al-Assam *et al.* (1990). Cryogenically purified CO₂ was analyzed for its isotope composition in a Finnigan MAT 252 mass spectrometer at the Geological Survey of Canada, in Ottawa. The isotope ratios of $^{13}\text{C}/^{12}\text{C}$ and $^{18}\text{O}/^{16}\text{O}$ are reported in the usual d-terminology, expressed in per mil (‰), relative to V-PDB and V-SMOW standards, respectively. The isotopic results are organized in Table 1, with a reproducibility of the d-value in the order of $\pm 0.1\text{‰}$.

FLUID INCLUSIONS Fluid inclusions interpreted of primary nature are abundant in quartz grains of the investigated samples, where they commonly occur as large intragranular groups, independent of any microstructures, and, occasionally, as isolated inclusions. The fluid inclusions generally contain two phases at room temperature, an aqueous liquid and a vapor phase (bubble), which show relatively constant phase-volume ratios in individual groups (5% to 10% variation). A few inclusions also display solid phases, either a

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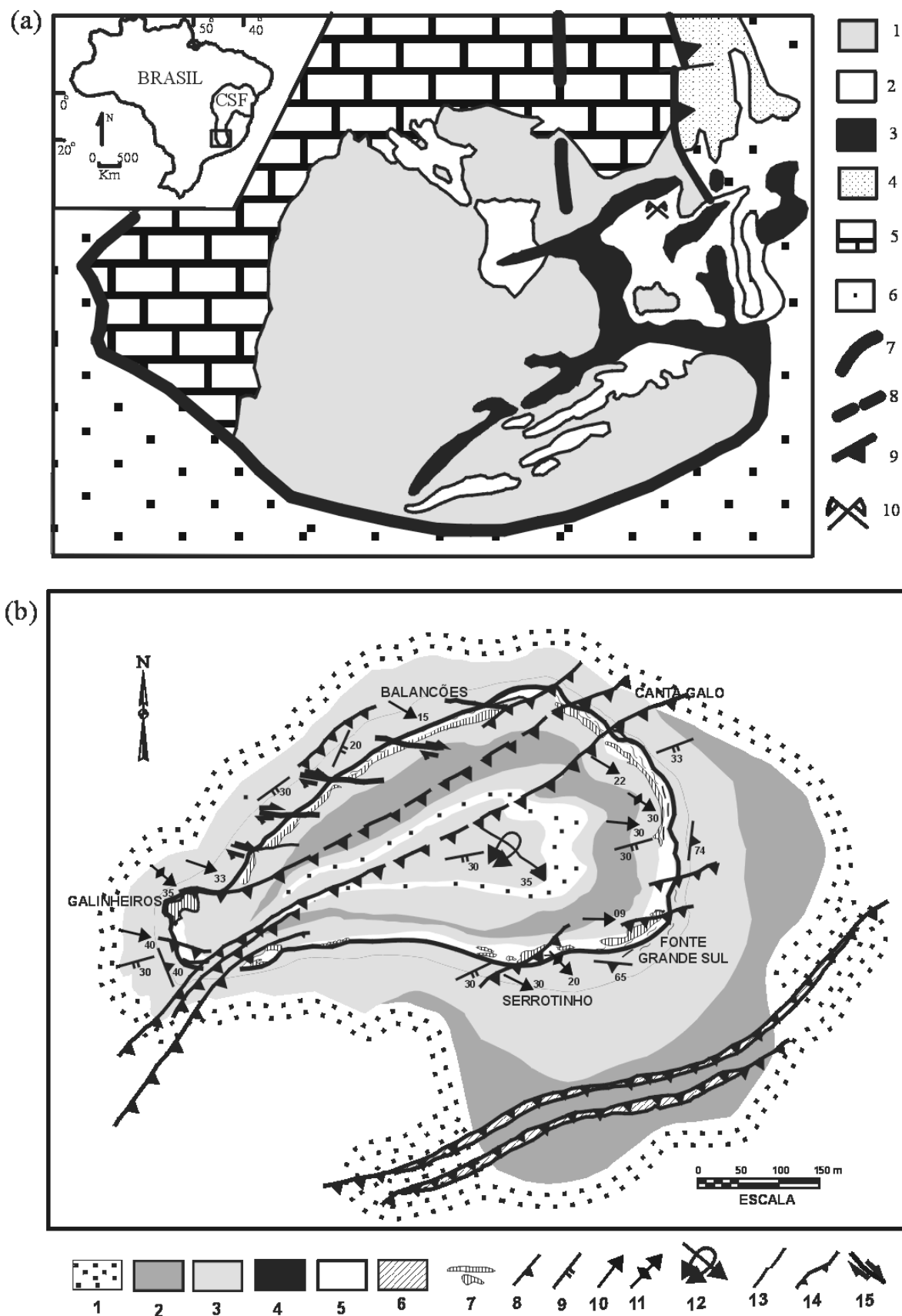


Figure 1-(a) Simplified geologic map of the Quadrilátero Ferrífero. Legend: 1. Granite-gneiss terrains; 2. Archean greenstone belts; 3. Minas Supergroup; 4. Espinhaço Supergroup; 5. São Francisco Group; 6. Terrains strongly affected by the Brasiliano (Pan-African) Event; 7. Limit of the Brasiliano Event influence zone; 8. Normal faults; 9. Thrust faults; 10. Cuiabá Mine; CSF – São Francisco Craton. Modified from Marshak and Alkmin (1992). (b) Simplified geologic map of the Cuiabá Mine, level 3.1. Turbidite sequence; 2. Metabasalt; 3. Altered zones in metabasalts; 4. Carbonaceous phyllites; 5. Banded carbonaceous metasedimentary rock; 6. S₂ foliation-parallel quartz veins; 7. Orebodies; 8. S₁ foliation parallel to primary bedding; 9. S₂ foliation; 10. Stretching and mineral lineation Lm₂; 11. F₂ fold axis; 12. Axis of tubular fold; 13. Shear zones parallel to primary bedding; 14. Thrust faults; 15. Strike-slip faults. Modified from Vial (1980) and Vieira (1992).

carbonate mineral or solid carbonaceous matter (?), probably due to accidental capture during the fluid trapping.

Dark monophasic inclusions, at room temperature, frequently coexist with the above two-phase aqueous inclusions, but in a much lower frequency than the latter.

Both types of inclusions show polyhedral outlines or irregular shapes, with their longest dimension ranging from 10 μm up to 40 μm .

Three main types of microthermometric behavior are distinguished in the bubble of the two-phase aqueous inclusions during the low temperature runs: (1) nucleation of a CH_4 gas phase within the bubble at temperatures $< -100^\circ\text{C}$ and its subsequent homogenization, to the liquid state, at temperatures ranging from -134.2 to -82.6°C , accompanied occasionally by the sublimation of a solid CO_2 phase at temperatures between -90 and -77.8°C ; (2) the same previous behavior at temperatures $< -100^\circ\text{C}$, but with melting of solid CO_2 preceding the CH_4 homogenization (-97.6 to -84.7°C) and varying from -108.3 to -85.3°C ; (3) melting of solid CO_2 varying from -74 to -61.2°C , followed by homogenization of CO_2 in the liquid state, between -67.8 and -18.2°C , in the presence of clathrates, or between -19.9 and 5°C , in the absence of clathrates. Very rarely, individual inclusions showed a sequence of CH_4 homogenization, melting of solid CO_2 , and CO_2 homogenization. Based on the above microthermometric behaviors, a mixture of variable proportions CH_4 and CO_2 may be expected for the bubble of the two-phase aqueous inclusions (van den Kerkhof 1990). This was confirmed by LRM analyses in the bubble of individual inclusions with different microthermometric behaviors, which additionally yielded subordinate amounts of N_2 and traces of $\text{H}_2\text{S}/\text{HS}^-$ (Fig. 2A).

Salinity values for the aqueous phase of these inclusions were roughly estimated between 3 and 6 wt.% NaCl eq., based on the combination of the melting temperatures of clathrates, which took place within the interval 12.4 to 22.3°C , and representative CH_4 isochores for these inclusions, as explained in Thomas and Spooner 1988. A few measurements of the total homogenization of the two-phase aqueous inclusions yielded values ranging from 160 to 340°C , in the liquid state.

The only phase change shown by the monophasic inclusions was that of homogenization, into the vapor state, at temperatures between -81.2°C and -82.7°C . This behavior, in conjunction with LRM data, constrains the composition of this inclusion fluid to $\text{CH}_4 \pm \text{CO}_2$ (H_1 inclusions; van den Kerkhof 1990).

CARBON AND OXYGEN ISOTOPE RESULTS Carbon Isotopes

Carbonates in the banded carbonaceous metasedimentary host typically show markedly heavier carbon isotope compositions, compared to carbonates in the gold-related carbonate-sulfide altered zones and gold-sulfide breccia and veins. The $\delta^{13}\text{C}$ values of siderite and calcite in the carbonaceous host range from -4.3 to -1.7‰ , with one value at -7.1‰ for ankerite, whereas ankerite from carbonate-sulfide altered zones has values of -8.0 and -7‰ and siderite -7.6 and -6‰ (Table 1; Fig. 2B). Rare calcite and dolomite in the carbonate-sulfide zones showed similar values of $\delta^{13}\text{C}$ of ankerite and siderite: -7.1‰ (calcite) and -8.0‰ (dolomite).

Carbonates in the gold-sulfide breccia and veins were isotopically similar to those in adjacent carbonate-sulfide altered zones: -8.1 to -6.9‰ (ankerite), -6.3 to -6.2‰ (siderite), -5.5‰ (calcite), and -7.1‰ (dolomite). Additionally, a $\delta^{13}\text{C}$ value of -7.8‰ was obtained for calcite from a barren quartz vein.

The heaviest value of $\delta^{13}\text{C}$ (-0.2‰) was obtained for calcite from veinlets that crosscut the carbonaceous host, away from the mineralized carbonate-sulfide zones (Table 1; Fig. 2B).

Oxygen Isotopes The range of $\delta^{18}\text{O}$ values obtained for carbonates from the banded carbonaceous metasedimentary rock and from an enclosed barren carbonate veinlet, between 12.8‰ and 16‰ , overlaps the interval of 10.6 to 15.2‰ for carbonates from mineralized zones, although there is a slightly larger variation in the latter (Fig. 2B). A $\delta^{18}\text{O}$ value of 13.2‰ for calcite from a barren quartz vein (sample CURPX3; Table 1) falls within the above range of $\delta^{18}\text{O}$ values for carbonates from the mineralized zones and host rock.

DISCUSSION The primary nature of the two-phase $\text{H}_2\text{O}-\text{CH}_4-\text{CO}_2-\text{N}_2-(\text{H}_2\text{S}/\text{HS}^-)$ inclusions imply that they may be regarded as the closest representative of the fluid which interacted with the

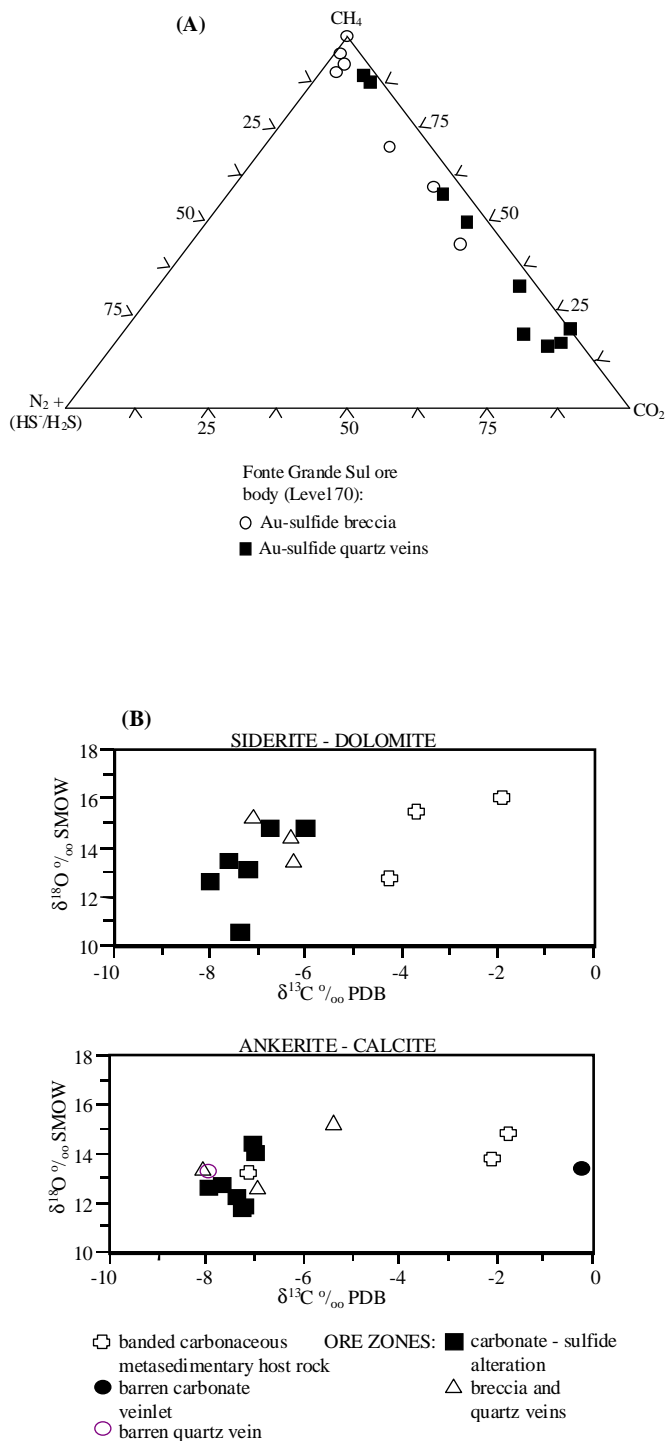


Figure 2—(A) Composition of the bubble obtained by Laser Raman micro-spectroscopy in the two-phase aqueous inclusions investigated in gold-bearing quartz-carbonate veins and breccias within the carbonate-sulfide altered zones. (B) $\delta^{18}\text{O}$ versus $\delta^{13}\text{C}$ values for siderite, ankerite, calcite and dolomite in the carbonaceous host rock, ore zones and barren vein samples of the Cuiabá Mine.

carbonaceous wallrocks and probably precipitated gold at the Cuiabá Mine. Bulk composition estimates for these inclusion fluids defined this fluid as dominantly aqueous, of low salinity (< 1.5 mol%), with variable concentrations of CO_2 (0 – 11.4 mol%), CH_4 (1.5 – 10.2 mol%), subordinate N_2 (0 – 0.7 mol%), and traces of $\text{H}_2\text{S}/\text{HS}^-$.

Table 1- $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values measured in ankerite (ank), siderite (sid), calcite (cc), and dolomite (dol) in the carbonaceous metasedimentary host, orebodies and barren samples of the Cuiabá Mine. Cb= carbonates; sulf= sulfides; qtz= quartz.

Samples	Rok type/Location	$\delta^{13}\text{C}_{\text{Ccc-ank}}$ ‰ V-PDB	$\delta^{18}\text{O}_{\text{Ccc-ank}}$ ‰ V-SMOW	$\delta^{13}\text{C}_{\text{Csid-dol}}$ ‰ V-PDB	$\delta^{18}\text{O}_{\text{Csid-dol}}$ ‰ V-SMOW
CB247	Host/Galinheiro ore body-N7	-2.11 (cc)	13.72 (cc)	-4.27 (sid)	12.76 (sid)
CB28C	Host/Serrotinho ore body-N5	-7.14 (ank)	13.24 (ank)	-3.70 (sid)	15.44 (sid)
CB95	Host/Cantagalo ore body-N11	-1.74 (cc)	14.80 (cc)	-1.85 (sid)	15.99 (sid)
CB67A	Cb-sulf zones/Fonte Grande Sul ore body-N7	-7.22 (ank)	11.87(ank)		
CB67B	Cb-sulf zones/Fonte Grande Sul ore body-N7	-6.99 (ank)	14.05 (ank)	-7.40 (sid)	10.60 (sid)
CB77C	Cb-sulf zones/Fonte Grande Sul ore body-N7	-7.98 (ank)	12.60 (ank)	-8.00 (dol)	12.60 (dol)
CB49C	Cb-sulf zone/Fonte Grande Sul ore body-N7	-7.68 (ank)	12.73 (ank)	-6.00 (sid)	14.80 (sid)
CB-BAL/N11	Cb-sulf zones/Balanção ore body-N11	-7.31 (ank)	11.76 (ank)	-7.20 (sid)	13.14 (dol)
CB-SERR/N5	Cb-sulf zones/Serrotinho ore body-N7	-7.07 (cc)	14.44 (cc)	-6.74 (sid)	14.84 (sid)
CB245	Cb-sulf zone/Galinheiro ore body-N11	-7.42 (ank)	12.26 (ank)	-7.61 (sid)	13.45 (sid)
CURPX1A	Breccia/Fonte Grande Sul ore body-N7	-6.94 (ank)	12.62 (ank)	-7.08 (dol)	15.24 (dol)
CURPX 1B	Breccia/Fonte Grande Sul ore body-N7	-8.07 (ank)	13.41 (ank)		
CB202	Qtz-cb-Au vein/Fonte Grande Sul ore body-N7			-6.24 (sid)	13.44 (sid)
CB202	Qtz-cb-Au vein/Fonte Grande Sul ore body-N7	-5.49 (cc)	15.26 (cc)	-6.33 (sid)	14.47 (sid)
CURPX3	Barren qtz-cb vein/Fonte Grande Sul ore body-N7	-7.95 (cc)	13.25 (cc)		
CB247	Barren cb veinlet/Fonte Grande Sul ore body-N7	-0.22 (cc)	13.37 (cc)		

Considering that field and petrographic evidence suggests that the carbonaceous matter was generally consumed during carbonate alteration, and that CH_4 and CO_2 are ubiquitously present in significant concentrations in the inclusion fluids studied, the possibility exists that hydrolysis/oxidation of the carbonaceous matter in the wallrocks, upon interaction with the hydrothermal fluid, may have influenced the composition of the fluid, subsequently trapped during alteration and veining. CO_2 and CH_4 may have formed, for example, via hydrolysis reactions of the carbonaceous matter (1) $2\text{C} + 2\text{H}_2\text{O} = \text{CO}_2 + \text{CH}_4$ or, at a lower oxidation state, by the reaction (2) $\text{C} + 2\text{H}_2\text{O} = \text{CH}_4 + \text{O}_2$, which could have provided the requisite CO_2 for carbonatization (reaction 1) and/or enriched the ore fluid in $\text{CH}_4 \pm \text{N}_2$ (reaction 2).

The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ data obtained on carbonates from the host rock, mineralized and barren zones of the Cuiabá Mine, allow a further assessment of this question. If the hydrothermal fluid contained no original CO_2 , and were of sufficiently high oxidation state and quantity (i.e., water/rock ratio) to oxidize reduced wallrock carbon, then the $\delta^{13}\text{C}_{\text{CO}_2}$ should approximate the $\delta^{13}\text{C}_{\text{WALLROCK CARBON}}$. The $\delta^{13}\text{C}$ values of carbonate precipitated from the resulting fluid, and detected in the wallrocks, would then reflect temperature-dependent fractionation factors. Alternatively, if the oxidation state of either the original hydrothermal fluid, or the evolved hydrothermal fluid, was sufficiently low to stabilize CH_4 , then the $\delta^{13}\text{C}$ value of carbonate precipitated from the fluid would depend also on the CH_4/CO_2 ratio of the fluid; values of $\delta^{13}\text{C}$ would have been higher, even positive, with increasing CH_4/CO_2 . If the oxidation state of the fluid happened to be buffered by graphite-bearing wallrocks according to reaction (1) above, then the $\delta^{13}\text{C}$ of isotopically-equilibrated CO_2 would most likely have been < -12 ‰ (e.g., Bottinga 1969). In all cases, the $\delta^{18}\text{O}$ values of the carbonate in veins and hydrothermally altered wallrocks and breccias depended on the temperature and isotopic composition of the fluids, as determined by previous water/rock interaction and isotope exchange.

As Figure 2B demonstrates, the $\delta^{13}\text{C}$ compositions of carbonate in sedimentary wallrocks and in altered and mineralized veins and wall rocks are markedly different, suggesting an external source(s) for the fluid-derived CO_2 , different modes of carbonate formation, and/or carbon isotope variation of CO_2 due to equilibration with CH_4 in the fluid. If equilibration with CH_4 in the reacted fluid were a factor, then the carbon isotope composition of precipitated carbonate must reflect a maximum value for $\delta^{13}\text{C}$ for the carbon source, inasmuch as CH_4 is

depleted in ^{13}C relative to CO_2 at equilibrium (e.g., Richet *et al.* 1977).

The most straightforward interpretation of the marked differences in $\delta^{13}\text{C}$ between carbonate in the metasediments and that in the veins, breccias, and altered zones is that the formers are essentially marine carbonates. The near-zero permil $\delta^{13}\text{C}$ values are certainly in agreement with this suggestion, whereas the lower values (-7.1 and -4.3 ‰) could represent some isotopic exchange with hydrothermal fluids. The rather narrow variation in $\delta^{13}\text{C}$ of gold-related alteration and breccia/vein-associated carbonate, with significantly lower values of $\delta^{13}\text{C}$, including that of calcite in a barren vein, than that of the metasedimentary carbonate, suggests a relatively homogeneous isotopic composition for the hydrothermal fluid CO_2 and either equilibration with carbonate and reduced carbon reservoir (e.g. mantle-derived magma; Taylor 1987).

CONCLUSIONS Fluid inclusion and carbon isotope data indicate that the original fluid responsible for carbonate-sulfide alteration and gold deposition at the Cuiabá Mine was probably of a low salinity $\text{H}_2\text{O} - \text{CO}_2$ solution, typical of fluids related to the formation of greenstone-hosted mesothermal lode-gold deposits, which incorporated significant concentrations of CH_4 related to hydrolysis of the carbonaceous matter during vein-fluid/wallrock interaction. On the basis of the carbon isotope data, sedimentary carbonates, probably of marine origin, are readily distinguished from those carbonates formed by reaction between CO_2 -bearing vein fluids and wall rocks. Vein fluid CO_2 was relatively homogeneous in isotopic composition, suggesting either equilibration with carbonate or reduced carbon reservoir (e.g., mantle-derived magma).

The addition of CH_4 during fluid-rock carbon interaction may have caused a sharp decrease in $f\text{O}_2$ of the fluid which, as a consequence, destabilized gold-bearing sulfur complexes, liberating S^{2-} for the formation of Fe sulfides, and triggered gold deposition.

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