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Exceptional metal enrichment caused by deep reaching hydrothermal oxidation

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**FLUID GEOCHEMISTRY OF THE SERRA PELADA Au-Pd-Pt DEPOSIT, CARAJÁS,
BRAZIL: EXCEPTIONAL METAL ENRICHMENT CAUSED BY DEEP REACHING
HYDROTHERMAL OXIDATION**

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ABSTRACT

Serra Pelada is a high-grade hydrothermal Au - Pd - Pt deposit within the Carajás mineral province, eastern Amazon craton, which shows an exceptional metal enrichment at carbon-enriched hosted ores (> 1000 's ppm Au+Pt+Pd). The main orebody is structurally hosted at the hinge of a recumbent syncline, consisting of dolomitic metasandstones at the outer part of the fold, a layer of carbonaceous metasiltstone in which most orebodies are located, and red siltstone in the core of the fold. Distal hydrothermal alteration comprises spatially separated chlorite – carbon (reduced) and hematite (oxidized) alteration zones. The mineralized zones include silicification and argillic (carbon- kaolinite-, and hematite-rich) alteration zones. Fluid inclusions hosted by quartz and dolomite crystals from distal alteration zones and silicified portions within the mineralized zone record a wide range of salinity (5-22 wt. % NaCl_{eqv.}) but relatively constant homogenization temperatures (170 ± 25 °C). Pressure and temperature estimations based in the fluid inclusion microthermometry and selenide thermometry constrain the formation of the deposit at 250-270 °C and 1.2-1.7 kbar. Element concentrations of alkali metals, Pb, Cs and to a lesser extent Ba correlate with salinity, whereas redox-sensitive elements including U and Ce vary widely. Gold concentrations from 50-500 ppb detected in some inclusions are consistent with the highly oxidizing conditions defined by selenide mineralogy. The presence of uranium and gold within fluid inclusions of the distal hematite alteration zone confirms the oxidizing, metal carrier fluids. The occurrence of hydrothermally precipitated carbon implies that a reducing CH₄-bearing fluid was also present, and the structural relationships suggest that mixing between this reducing fluid (and interaction with carbon-upgraded rocks) with an acidic, oxidizing and metal-carrying fluids was the main process driving ore metal precipitation. The genesis of the Serra Pelada Au - Pd – Pt deposit shows similarities with sediment-hosted copper deposits and unconformity-related U ± Au ± PGE deposits, having in common the downflow of highly oxidizing fluids into the hydrothermal domain of the Earth's upper crust. Major differences in metal proportions of these broad classes of ore deposits and other sediment-hosted systems in different ore provinces are primarily caused by differences in source rocks available for oxidative leaching and variations in the geometry of the hydrothermal system.

Keywords: Au-PGE mineralization; Serra Pelada; Carajás; Fluid Inclusion;

1. INTRODUCTION

Serra Pelada is an exceptional world-class Au - Pd - Pt deposit located in the Carajás Mineral Province near the eastern border of the Amazon craton in northern Brazil. The deposit was discovered by artisanal miners in the early 1980s and was the site of the biggest gold rush in South America's late history. Because of the harsh mining conditions in an overcrowded pit, only few exploration drill core data and little descriptive work were published when the mine was active (Jorge João et al., 1982; Meireles et al., 1982; Meireles and Silva, 1988).

An epigenetic mineralization has been suggested by Moroni et al. (1999, 2001), Tallarico et al. (2000), and Grainger et al. (2002). An intrusion-related hydrothermal model has been suggested by Tallarico et al. (2000), and Grainger et al. (2002) proposed a distal association

with the IOCG deposits of the Carajás mineral province. Cabral et al. (2011) suggested that a Late-Cretaceous hydrothermal overprint over the primary Paleoproterozoic mineralization (Grainger et al., 2002) was essential to generate spectacular grades of hundreds of ppm of all three precious metals.

Recently, hydrothermal alteration features and mineralogy of high-grade primary ore were documented by Berni et al. (2014, 2016), and the geology and material of these publications are the basis of the fluid-chemical study presented here. We have selected samples from the distal alteration and ore zones for detailed petrography, microthermometry, and LA-SF-ICPMS analysis of fluid inclusions to constrain the hydrothermal process of ore formation. Based on pressure – temperature conditions indicated by fluid inclusions, selenide mineralogy (~270°C, ~1.2-1.7 kbar) and the unusual trace-element composition (elevated Ni, U, detectable Au, Pt) of fluid inclusions we suggest that highly oxidized, metal carrying fluids reached down to the base of the brittle regime of the crust and mixed there with reduced deeper-crustal fluids of variable salinity to precipitate the high-grade Au-Pd-Pt-ores at Serra Pelada.

2. GEOLOGICAL SETTING

The Serra Pelada Au - Pd - Pt deposit is located in the northern part of the Carajás Mineral Province, a major mining district for world-class iron ore and IOCG deposits. The deposit is hosted by the Águas Claras Formation, formed by metasedimentary rocks which unconformably overlain the meta-volcano-sedimentary sequences from Rio Novo Group (Fig. 1).

The metasedimentary rocks of the Águas Claras formation at Serra Pelada can be divided into three units (Fig. 1). The basal unit 1 is made of metasandstones with layers of mono- and polymictic metaconglomerates. At the western part of this unit, the matrix of metasandstone is dolomitic with local layers of pure marble (Grainger, 2003). These metasedimentary rocks host highly positive C-isotope values (up to +7.6‰ V-PDB), indicating the deposition during the Paleoproterozoic, ~2.2-2.06 Ga Lomagundi carbon isotope excursion (Bekker et al., 2017). These dolomitic metasandstones underlie the red and carbonaceous metasiltstones of Unit 2. The carbonaceous metasiltstone directly overlies the dolomitic sandstone and is the main host rock of the mineralization at Serra Pelada deposit. The unit is 20 m thick layer of a very fine laminated metasiltstone with low carbon contents (< 1 wt. % - Berni et al., 2014). Locally, other layers of carbonaceous metasiltstones outcrop within the sequence but their gold content is unknown. The red siltstone is the predominant lithology within unit 2. It is a red, fine-grained rock, with alternating between silty (redder) and sandy (more yellow) layers. Unit three is made of metasandstones with local layers of metasiltstones and polymictic metaconglomerates with siltstone fragments.

Three intrusive rocks crosscut the metasedimentary sequence at Serra Pelada. Diorite outcrops as a small intrusion at the surface and is intercepted in several exploration drill cores at the Serra Pelada deposit. It is deformed and metamorphosed within the metasedimentary sequence (Grainger, 2003). The age of the diorite intrusion is uncertain. The Cigano Granite intrudes the metasedimentary sequence at its west portion and is of Paleoproterozoic age (1880 ± 2 Ma - U-Pb zircon; Machado et al., 1991). It is a coarse-grained A-type alkaline to sub alkaline granitoid. The latest intrusions within the metasedimentary sequence are gabbro dikes, associated with north-northwest trending fault zones and dated at 198 Ma (Rb/Sr; Meireles et al., 1982). The gabbro dikes are post-mineralization, and the one dike which crosscuts the Serra Pelada Deposit (Fig. 1) does not affect the grade distribution within the deposit.

From stratigraphic base to top, the mine sequence at the Serra Pelada deposit comprises dolomitic metasandstones and metaconglomerates (unit 1), a 20-30 m thick layer of carbonaceous, grey metasiltstone and a red metasiltstone at the fold core (unit 2).

The primary mineralization at the Serra Pelada deposit is hosted by hydrothermally altered metasedimentary rocks of units 1 and 2 (Fig. 2A) and is structurally hosted at the hinge of a recumbent syncline (Moroni et al., 1999, 2001; Tallarico et al., 2000; Grainger et al., 2002).

The hydrothermal alteration at Serra Pelada can be divided into distal and local alteration zones. The distal alteration zones comprise the (1) Hematite and the (2) Chlorite – Carbon alteration zones, while the alterations within the ore zone are divided into different subtypes of (3) Argillic and (4) Siliceous alteration (Berni et al., 2014).

Distal alteration zones of contrasting redox potential occur in distal and spatially separated zones, comprising two main types of regional alteration: (1) Hematite (oxidizing) alteration is the most widespread distal alteration zone within the Serra Pelada deposit, occurring up to a 10 km away from the deposit. It mainly replaces the dolomitic sandstone at the deposit area, forming well-formed hematite, quartz and dolomite crystals within red-dusted dissolution vugs. It can also occur as fine-grained hematite dusting within the dolomitic sandstone matrix or along small cracks. (2) Chlorite – carbon (reduced) alteration zone overprints the red siltstone at the lower limb of the Serra Pelada syncline, and changes the red, hematite-bearing metasiltstone into a green, chlorite-bearing and locally carbon-enriched metasiltstone. Chlorite replaces metamorphic sericite in the red and carbonaceous metasiltstone. The chlorite – carbon alteration is a reduction front where precipitation of quartz locally occurs, and carbon is mainly precipitated along the metamorphic banding, or together with quartz in veins. The hydrothermal alteration related to the ore zone is located at the hinge of the syncline, and minor orebodies occur along the lower and upper limbs (Fig. 2A). The hydrothermal alteration zones comprise: (3) the argillic alteration is characterized by the replacement of muscovite by kaolinite with associated carbon, hematite or amesite. The mineralogy of these alterations determines the carbon-rich argillic, kaolinite-rich argillic and hematite-rich argillic zones, where the first two are the principal Au-Pd-Pt ore. The primary ore mineralogy includes sulfide, selenide, and antimony-arsenide minerals (Berni et al., 2014, 2016). (4) The siliceous alteration replaces both metasandstone and metasiltstones near and within the mineralized zone. The siliceous alteration generally hosts low-grade Au - Pd - Pt mineralization, occurs at the periphery of high-grade ore shoots (Tallarico et al., 2000; Grainger et al., 2002), and is temporally related to the argillic alteration and ore metal introduction in the main orebody. The siliceous alteration occurs as fine- to coarse-grained quartz with hematite and minor pyrite as an envelope around the fold hinge and occurs locally within fine-grained quartz with associated hematite and pyrite replacing the carbonaceous siltstones at the ore zone.

3. SAMPLING MATERIAL

From a much larger number of samples cut for doubly-polished plates, nine samples were studied in detail, providing textural information and quantitative compositional data from 58 fluid inclusion assemblages including 3 to 18 individual inclusions each. Their approximate location with respect to the alteration zones within the deposit is shown in figure 2A. Drill core information, sampling depth, and sample's description are listed in table 1. The sampling was focused on samples containing well-formed crystals and genetically constrained within a specific alteration type.

For characterizing the oxidizing fluid from the distal hematite alteration zones, we have selected quartz and dolomite crystals from vugs within the dolomitic from the periphery of the Serra Pelada deposit (Fig. 2B). The Chlorite – carbon alteration zone rarely contains quartz which can be genetically related to the alteration zone. The selected sample consists of a euhedral quartz crystal with numerous inclusions of amorphous carbon (Fig. 2C). The only sample material suitable for fluid inclusion studies within the ore zone is within the siliceous alteration. The selected samples consist of fine-grained quartz containing vugs within clear and well-formed quartz crystals in vugs which are lined by hematite (Fig. 2D).

4. ANALYTICAL METHODS

Samples with numerous, adequately sized (30 to 80 μm) fluid inclusions within clearly defined fluid inclusion assemblages (FIA) were studied by detailed petrography to assess their relative chronology. Groups of fluid inclusions that are coevally formed (according to the concept of fluid inclusion assemblage; Goldstein and Reynolds, 1994) were analyzed by microthermometry to determine their apparent salinity (from ice melting temperatures) and homogenization temperatures. A Linkam THMSG-600 heating-freezing stage connected to a Leitz optical microscope, calibrated with synthetic fluid inclusions of pure CO_2 , pure water, and $\text{NaCl-H}_2\text{O}$ mixture of eutectic composition and of known homogenization temperature was used for the microthermometry measurements at ETH Zürich.

In-situ analyses of individual fluid inclusion in these FIA were performed by LA-SF-ICPMS with a beam-homogenized 193 nm ArF Excimer laser ablation system (Günther et al., 1997) connected to a high-sensitivity sector field mass spectrometer (ThermoFisher, Element XR; Wälle and Heinrich, 2014). Preliminary runs were analyzed with a quadrupole ICPMS (Perkin Elmer Elan 6100 DRC). After first test-runs, we selected a reduced set of isotopes related to the mineralization at Serra Pelada (^{23}Na , ^{24}Mg , ^{27}Al , ^{32}S , ^{39}K , ^{44}Ca , ^{60}Ni , ^{65}Cu , ^{75}As , ^{121}Sb , ^{133}Cs , ^{137}Ba , ^{140}Ce , ^{197}Au , ^{208}Pb , ^{209}Bi and, ^{238}U) and applied an increased dwell time of 30 ms on ^{197}Au and ^{238}U and 10 ms for all other elements. Individual runs for detecting platinum were performed with a small element menu including ^{29}Si , ^{23}Na and ^{195}Pt with an increased dwell time of 200ms for Pt and 16ms for the other elements. We ablated with a pulse rate of 10 Hz and energy densities per pulse of ca. 30 J/cm^2 for quartz and 20 J/cm^2 for dolomite hosted inclusions. Analytical and standardization procedures were performed according to Heinrich et al. (2003), using NIST SRM 610 as external reference material. Element concentrations of individual fluid inclusion were calculated with the SILLS software (Guillong et al., 2008), using the salt correction equation according to Allan et al. (2005) for K and Mg.

It should be noted that this study pushes the limits of LA-ICPMS analysis of fluid inclusions because all signals were low but complex due to the small inclusion size and the presence of heterogeneously entrapped kaolinite. We exclusively accepted statistically significant and clearly discernable element signals, where several element peaks correlated with that of Na (Schlöglova et al., 2017), and we report estimated overall uncertainties based on several inclusions per assemblage by symbols with error bars in the subsequent diagrams. Data remaining after such rigorous quality selection represent unambiguously detected elements, but quantifications should be considered as order-of-magnitude results only.

5. RESULTS

5.1 Petrography

5.1.1 Hematite alteration (distal)

The hematite alteration occurs as red-dusted recrystallized marble containing vugs with centimeter-scale dolomite (Fig. 3A) and quartz crystals (Fig. 3B). Distinct growth zones within the vuggy crystals are lined with numerous micron-sized inclusions of hematite, chalcopyrite, and Ni-Co sulfides (Fig. 3C) with a small-scale enrichment in Au, Pd and Pt, detected by bulk laser ablation sector field inductively coupled plasma mass spectrometry (LA-SF-ICPMS) analysis of the inclusion-rich zone (Fig. 3D and Fig. 3E). These samples were collected in deep drill holes below the level of supergene leaching of the dolomitic sandstones and are interpreted as feeder veins, located where brittle and steep fracture zones cut the dolomitic metasandstone. The hematite alteration is the most widespread alteration zone of the system, where hematite-altered sandstone with euhedral quartz can be found up to 10 km from the deposit.

Most fluid inclusion from all FIA in this alteration type are aqueous, two-phase fluid inclusion. Well-defined time-relationships in respect to the main growth zones were preserved, allowing the determination of primary, pseudosecondary and secondary fluid inclusion assemblages. Primary fluid inclusion assemblages occur as irregular-shaped, small ($< 40\mu\text{m}$), elongated fluid inclusions parallel to the direction of the growth zones. Pseudosecondary (Fig. 3F) secondary FIA are more common and contain abundant fluid inclusions ($30 - 100\mu\text{m}$) on each assemblage. Quartz-hosted fluid inclusions generally present irregular shape (Fig. 3G), while dolomite hosted samples are often euhedral or with well-defined faces (Fig. 3H). Needle-shaped accidentally trapped crystals are observed in dolomite and quartz hosted fluid inclusions.

5.1.2 Chlorite – carbon alteration (distal)

Fluid inclusion assemblages with measurable size for microthermometry and LA-SF-ICPMS analysis were identified only in one sample from this alteration type. The sample contains a quartz vein with abundant inclusions of amorphous carbon in the silicified wall rock, sub-euhedral fine-grained quartz at the vein walls and euhedral quartz towards the center of the vein (Fig. 4A).

Primary and pseudosecondary fluid inclusions were identified with their location restricted to carbon-rich growth zones at the core of some quartz crystals. The fluid inclusions consist of two-phase aqueous fluid inclusions, (30 to $50\mu\text{m}$ – Fig. 4B), locally with needle-shape and round accidentally trapped crystals. Vapor-only or empty inclusions are found as isolated inclusions, but no consistent vapor assemblages were observed in any sampled material.

5.1.3 Siliceous alteration (ore zone)

The alteration introduces coarse-grained to microcrystalline quartz with inclusions of sericite, kaolinite, and hematite. Fine-grained silicification often contain disseminated hematite and centimeter-sized vugs with well-formed quartz crystals.

Well defined growth zones marked by hematite crystals are preserved (Fig. 4C), and host primary, pseudosecondary and secondary fluid inclusions, made of a two-phase aqueous

fluid inclusion (Fig. 4D), locally with abundant kaolinite as accidentally trapped crystals (Fig 4 E and F).

The needle-shaped accidentally trapped crystals present FIA from all alteration zones are most likely to be kaolinite crystals. During the ablation of a fluid inclusion within the hematite alteration zone, it was possible to identify an aluminum peak which is shifted from elements as the sodium and potassium (Fig. 4E). This shift shows that Al was the main component of the solid, while K and Sodium were in solution. Out of all mineral stable at the Serra Pelada ore mineralogy, kaolinite is the only aluminum-rich phase.

5.2 Microthermometry

Fluid inclusions within one assemblage generally show consistent homogenization temperatures within ± 25 °C, with averages ranging from 140 °C to 185 °C and clustering around 170°C (Fig. 5A). This limited range in homogenization temperatures contrasts with a wide range of salinity, from 2.6 to 23.5 wt. % NaCl_{eqv.} (Table 2). During warming after complete freezing, ice was always the last phase to melt. Cyclic cooling and heating experiments were performed to induce hydrohalite nucleation, which was observable only within few secondary, highly saline FIA (> 20 wt. %), where the calcium content was relatively small, generally between 1-2 wt. % CaCl₂ (estimated according to Schlegel et al., 2012). Fluid inclusions of lower salinities where hydrohalite nucleation was not visible or metastable, were estimated from ice melting temperature and calculated as wt. % NaCl equivalent (NaCl_{eqv.}) using data from Bodnar and Vityk (1994). All inclusions homogenized by bubble disappearance, providing values for minimum temperature.

Primary and pseudosecondary FIA in quartz from the Chlorite – carbon alteration record a salinity variation from 2.6 to 11.9 wt. % NaCl_{eqv.}. Quartz crystals from vugs within the siliceous alteration host fluid inclusion assemblages with salinity variations from 3.3 to 8.8 wt. % NaCl_{eqv.}, although isolated fluid inclusions of higher salinity were observed. Salinities of primary and pseudosecondary inclusions within the siliceous alteration vary from 3.3-6.6 while secondary inclusions record salinities up to 8.8 wt. % NaCl_{eqv.}. Primary FIA hosted by dolomite and quartz in the hematite alteration zone range from 3.2 to 11.0 wt. % NaCl_{eqv.} within different growth zones such as shown in figure 3 A. In samples that showed well time-constrained primary FIA and growth zones, a salinity increase is recorded from earlier to later stages. Pseudosecondary FIA fall in the same range (4.4 to 6.2 wt. % NaCl_{eqv.}), while secondary trails record salinities from 3.9 to 21.6 wt. % NaCl_{eqv.}.

5.3 Major elements

The major components of the analyzed fluid inclusions from all three alteration types contain variable mass ratios but are generally characterized by Na > K > Ca $\geq$ Mg. Compositional data and diagrams are shown in Table 2 and Figure 5B to 5E.

Fluids from the chlorite – carbon alteration zone are sodium dominated (Na >> K > Mg) and the potassium concentration is less than 2000 ppm (Fig. 5B). K to Na ratios range from 0.01 to 0.8, and these fluids record the lower K/Na ratios (Fig 5C) within the deposit. Magnesium concentrations vary widely (100-1000 ppm Mg at Mg/Na mass ratio between 0.1-1 – Figs. 5D and 5E).

The composition of primary and pseudosecondary FIA within the siliceous alteration zone shows a more consistent trend of increasing K concentration (Fig. 5B) and an increase of K/Na

ratios (Fig. 5C) with increasing salinity. Magnesium content (Fig. 5D) is less than 1000 ppm, and it does not show the same correlation with salinity as observed for potassium. Na/Mg ratios (Fig. 5E) are more constant than Na/K ratios at values around 0.02. Calcium was detected only in one fluid inclusion assemblage (~2500 ppm) at a Ca/Na proportion of approximately 0.1, consistent with the more reliable microthermometric observations. Secondary FIA from the siliceous zone display the same salinity range, but higher potassium and magnesium concentrations at similar K/Na or Mg/Na ratios.

The compositional data from the primary and pseudosecondary, hematite alteration zone is subdivided into Au-bearing and barren host rocks (gold not detected) as red and black symbols in Figure 5A to 5E. Within the primary and pseudosecondary from both Au-bearing and barren FIA, the potassium concentration tends to increase with salinity, with K/Na ratios up to 0.8, while high salinity (>11 wt. % NaCl_{eqv.}) secondary fluid inclusion assemblages record overall lower K/Na ratio (Fig. 5C). Magnesium contents are in the order of 1000 to 10,000 ppm from high salinity secondary FIA, at relatively constant ratios with Na (Fig. 5D and 5E). Primary and pseudosecondary FIA have lower magnesium contents (less than 1000 ppm) at the similar Mg/Na ratios.

5.4 Trace elements

Trace levels of As, Cs, Cu, Ba, Pb and U (Fig. 6A-F) were measurable in many individual inclusions of most of the analyzed fluid inclusion assemblages (Fig. 6). Other elements like Ni, Sb, Ce were occasionally measured in some fluid inclusion assemblages hosted within the siliceous and hematite alteration zones. Sulfur was monitored on the first set of analysis and was measurable within two FIA (~4000-1500 ppm) while in most analysis detection limits were on the range of 1000-5000 ppm.

Arsenic and copper (Figs. 6A and 6B) are similarly distributed within the FIA from the different alteration zones. Higher concentrations are observed in FIA from the Chlorite – carbon alteration zone (up to 200 ppm and 300 ppm As and Cu respectively). Within the FIA siliceous and hematite alteration zones, primary and pseudosecondary FIA contain one order of magnitude less copper (~20 ppm Cu) when compared to the fluids hosted by the reduced alteration zone. Secondary fluid inclusion assemblages from both hematite and siliceous alteration zones contain slightly higher Cu but lower As contents when compared to primary and pseudosecondary FIA from the same alteration zone.

Cesium concentration (Fig. 6C) tends to be higher on the FIA from the reduced alteration zone and secondary fluid inclusion assemblages from both siliceous and hematite alteration zones, ranging from 4-10 ppm Cs within FIA hosted by the reduced alteration zone. Secondary FIA from siliceous and hematite alteration zones have Cs concentration values that are more variable but up to 10 ppm Cs. Primary and pseudosecondary FIA from the siliceous and hematite alteration zones contain up to five times less Cs, with about 1-1.8 ppm Cs on average (Fig. 6C).

Barium and Lead display similar concentration with FIA hosted by all alteration zones, ranging from 10's-100's ppm Ba and from 1 to 100 ppm Pb (Figs. 6D and 6E). The Ba concentration on FIA from the reduced alteration zones and secondary inclusion from hematite and siliceous alteration are on average higher than the primary and pseudosecondary FIA's hosted by the same alteration zone.

Uranium was detected, but only in FIA hosted by the hematite and siliceous alteration zones (Fig 6F). Within the hematite (oxidized) alteration zone, uranium was detected in about 50% of the assemblages, with values up to 1 ppm (Fig. 6F). With the siliceous alteration zone, uranium was detected in two FIA, with a concentration of about 0.2 ppm U. By contrast, uranium concentrations in all fluid inclusions of the reduced alteration zone were consistently below the estimated detection limit of 0.1 ppm U.

5.5 Au and Pt concentrations in fluid inclusions

Unambiguous LA-SF-ICPMS signals for gold were identified in 14 individual inclusions from 6 FIA hosted by the hematite alteration zone. (Fig. 7A and 7B). Gold was not detected in every fluid inclusion within the same FIA, which is more likely to be a consequence of the analytical limitation due to the transient nature of a fluid inclusion LA-SF-ICPMS signal (Rauchenstein-Martinek et al., 2014; Fusswinkel et al., 2018) even though we increased the dwell-time for from the 10 ms of all other elements to 30 ms for gold. Thus, gold was detected in primary and pseudosecondary FIA hosted by the oxidizing, hematite distal alteration zone. These are the same samples where anomalous gold, palladium, and platinum were detected by bulk ablation of dolomite and quartz growth zones containing submicroscopic particles of oxide and sulfide minerals (Fig. 3 3A to 3E). Calculated gold concentration in fluid inclusions varies from 30 to 500 ppb (Table 2, Fig. 7C) whereas all fluid inclusion analyses from FIA in the siliceous and Chlorite – carbon alteration were below their detection limits of 50-100 ppb.

Inclusions from the same two crystals were analyzed with a dedicated LA-SF-ICPMS run for platinum measurement, recording only platinum, sodium, and matrix (Si or Mg). Eight individual fluid inclusions within five different fluid inclusion assemblages yielded notable Pt peaks and calculated concentrations between 150 and 10 ppb (Table 3), unambiguously showing that fluids in these oxidized assemblages transported platinum besides Au and Ni (Table 2, Fig. 7D), even though the precious metal concentrations are order-of-magnitude estimates only.

6. DISCUSSION

6.1 Physicochemical conditions of ore formation

The temperature of formation for the Serra Pelada deposit is around 250-270 °C, using the temperature estimate from selenide mineral stabilities derived by Berni et al. (2016). The fluid pressure during Au - Pd - Pt mineralization at Serra Pelada was estimated by combining microthermometry data and independent mineral geothermometers. Fluid inclusion isochores from microthermometry data measured in this study were calculated using the SOWAT software (Driesner and Heinrich, 2007). The pressure of formation of Serra Pelada deposit was estimated to be about 1.2-1.7 kbar (Fig. 8), by intersecting the isochores of fluid inclusions of 7 and 20 wt. % NaCl homogenizing at 165-185 °C with the mineralization temperature range of 250-270 °C. The stability limit of kaolinite relative to pyrophyllite constrains the temperature to be below 290 °C at 1-2 kbar (Holdaway, 1971), consistent with the ore and gangue mineral stabilities.

The acidic conditions of the fluids have been previously indicated based on the deposit's mineralogy (Tallarico et al., 2000; Grainger et al., 2000). Kaolinite is stable in the primary alteration assemblage of the ore zone and was also identified as accidentally trapped mineral

within some fluid inclusions, as shown in Figure 4E and 4F. This shows that the fluids were in the stability field of kaolinite and therefore their potassium concentration can be used to estimate a minimum pH based on the hydrolysis reaction from muscovite (abundant within the argillic alteration of the ore zone) to kaolinite. The pH of the fluids was calculated using the Geochemist Workbench 9.0 Rxn software and the thermo.dat thermodynamic database therein (Bethke, 2008). For these calculations, the activity of K^+ was constrained by the potassium concentration of fluid inclusions, which range from 1000 to 10'000 ppm within all alteration zones. Equating molal concentration with activity results in K^+ activities between 0.025 and 0.25 provides an estimate of maximum pH between 2.5 and 3.5 for the mineralizing fluid.

Low pH is also indicated by the presence of hydrothermal kaolinite and amesite (Mg-rich chlorite) at the ore zone. While the reaction from sericite to muscovite consumes acidity, the formation of amesite at the expense of muscovite or kaolinite in the wall rock would create acidity according to the following reactions:

The reaction of Mg^{2+} in the fluid with sericite or clay minerals to amesite (Mg-rich chlorite, ubiquitous in the ore zone) would have kept the fluid acidic according to the above buffer reactions.

The estimated fO_2 conditions for some of the primary selenide mineral assemblage of the Serra Pelada deposit is up to 10 log units above the hem/mag buffer (Berni et al., 2016). The redox state of ore fluids needs to be evaluated by compositional data from redox-sensitive elements such as uranium in fluid inclusions. Uranium is a redox-sensitive element and can be effectively transported at geologically realistic concentrations (> 1 ppm) under oxidizing and acidic conditions (Cuney, 2005; Bastrakov et al., 2010) at temperatures around 270 °C. Therefore, the average uranium concentration of the fluid inclusion assemblages from the different alteration zones from the Serra Pelada can be used as a proxy to evaluate the oxidation state of the ore fluids. The uranium concentration of fluid inclusion assemblages of the oxidizing hematite alteration zone is up to 1 ppm U (Fig. 6), two assemblages from the Siliceous alteration within the ore zone had detectable uranium (~ 0.2 ppm) whereas fluid inclusion assemblages from reduced Carbon – chlorite alteration were below detection limits. Detection limits for uranium in fluid inclusion LA-SF-ICPMS analysis was around 0.1 ppm or lower for most of the individual measurements. The distribution of uranium concentration in fluid inclusion assemblages from the contrasting redox alteration zones at Serra Pelada corroborates the mineralogical features observed on the rocks and confirm the high oxygen fugacity conditions estimated from selenide mineralogy (Berni et al., 2016). To solubilize about 1 ppm of uranium at temperatures around 270 °C and acidic conditions, the oxygen fugacity was required to be about ten units above the magnetite/hematite buffer (Bastrakov et al., 2010). Samples from the same oxidizing, hematite alteration zone are also the ones with higher potassium content, with pH estimated at 2.5, and the only fluid inclusions which contained gold (up to 500 ppb). These conditions confirm the occurrence of an acidic, highly oxidizing fluid which transported the precious metals to the ore zone. It is important to highlight that uranium and gold were detected in fluid inclusions from samples which are barren or very low grade. Only traces of gold and PGE were detected on specific growth zones rich in ore minerals (Fig. 3A to 3E). Therefore, the hematite alteration zone within the dolomitic sandstones can be interpreted as feeders of the Serra Pelada deposit.

The reduced fluid (CH₄-bearing) must have been present during metal deposition as there is hydrothermally precipitated carbon associated with a Chlorite – carbon reduced alteration, but the redox state of this fluid cannot be evaluated as there is no available data on the volatile species and their ratios. The fact that there is no detectable uranium at the fluid inclusions assemblages hosted by quartz from the reduced Chlorite – carbon alteration zone is an indicator that these fluids were not at oxidizing conditions. Also, these reduced fluids record lower K/Na ratios and higher Cs, Cu and As, which could be an indicator for different fluid sources or rock-buffer. A reducing rock such as the carbonaceous siltstone would be important for the precipitation mechanism for gold, platinum, and palladium being transported under oxidizing conditions. The reducing fluid which has upgraded the carbon content of the host rock (enhancing their reductant power) and additionally mixing with an oxidizing fluid would be essential to make a very effective precipitation mechanism and explain the extreme high-grade samples from the Serra Pelada deposit.

6.2 Au solubility, metal availability and potential precious metal source rocks

Whenever discussing the formation of hydrothermal ore deposits, a fundamental question is whether metal solubility in the fluid or metal availability in source rocks is the limiting factor controlling the metal concentration transported in an ore fluid. In the case of Serra Pelada, we argue based on thermodynamic estimates that potential solubility was very high due to an unusual transport environment, and that source availability placed the main limit on gold concentration in the fluids forming the Serra Pelada deposit.

The extremely oxidized conditions implied by the hematite and selenide mineralogy in combination with the acidic conditions shown by the low pH estimate from the fluid inclusion data presented here allow very high Au, Pt and U solubilities as metal chloride complexes (Mernagh et al., 1994). Experimental data indicate that at the fO_2 -pH conditions estimated for Serra Pelada, a fluid with a chloride molality of 2-3 and a temperature of 250-300°C, could carry from 1 to 10's ppm of Au dissolved in a hydrothermal fluid (Hayashi & Ohmoto, 1991; Likhoidov et al., 2000). This Au solubility is much higher than the observed Au concentration in fluid inclusions of maximum 0.5 ppm, indicating that the incoming fluid was metal-undersaturated. Extreme gold-undersaturation during hydrothermal transport implies that the Au concentration in the ore-forming fluids may have been limited by metal availability in the source region.

Considering the rock types exposed in the area, the abundant mafic and ultramafic rocks including Mg-rich intrusions like the Luanga complex and Rio Novo group seem to be the most plausible metal source (Moroni et al., 1999, 2001; Tallarico et al., 2000; Grainger et al., 2002). Average precious metals in basalts and ultramafic rocks range between 2-8 ppb of gold (Gottfried et al., 1972; Pitcairn, 2011). The contained metal quantity in the Serra Pelada deposit comprises 110t Au, 35t Pd and 18t Pt (Grainger et al., 2002), which would require 8 km³ of mafic to ultramafic rocks which metal concentrations such as mentioned above. Precious metals in the low-grade metamorphosed ultramafic rocks from the Archean Luanga complex and similar ultramafic intrusions were available as disseminated sulfides and arsenides (braggite and sperrylite), locally enriched to hundreds ppb platinum and palladium in mineralized chromitite and up to 25 ppb in leucogabbro (Diella et al., 1995). The plausible contribution of mafic-ultramafic rocks from the Rio Novo Greenstone and Luanga complex in also indicated by the Ni-sulfides present in the oxidized hematite alteration samples that we interpret as potential fluid feeders to the deposit.

6.3. Physical hydrology of the Serra Pelada ore system

A hydrothermal origin of the Au-Pd-Pt mineralization at Serra Pelada is now accepted by most researchers (Moroni et al., 1999, 2001; Tallarico et al., 2000; Grainger et al., 2002; Cabral et al., 2011), but the mechanism that drives the precipitation of the high-grade Au-Pd-Pt ore and the flow paths of the mineralizing fluids remain debated. The mineralogical and fluid-compositional evidence for highly oxidized fluids suggesting redox levels well above the sulfate - sulfide buffer in the fluid poses serious questions regarding the possible hydrology of this unusual hydrothermal system.

The most challenging question is how highly oxidized fluids can reach depth near the brittle-ductile transition of the crust, without becoming reduced along their infiltration pathway. Free O_2 is poorly soluble in water and would be consumed by reaction with any Fe^{2+} bearing minerals along the flow path. To maintain very high f_{O_2} required for transporting Au, PGE and U fluids must flow rapidly and be channeled within already completely oxidized structures in which the main flow path remains isolated from fresh ferromagnesian minerals. Where such fully oxidized pathways cross mafic and ultramafic rocks, active leaching of precious metals is possible.

Stable hydrostatic conditions consistent with the estimated fluid pressure of >1.2 kbar would require a depth of ore formation of at least 12 km below the surface. However, in an area of active tectonics, as indicated by syn-mineralization reactivation of thrusts and faults (Berni et al., 2014), fluid pressures may fluctuate between times of hydrostatic infiltration and transient return to near-lithostatic conditions. The infiltration of oxidized fluids must have occurred during periods of hydrostatic pressure, possibly driven by enhanced fluid density resulting from high salinity due to evaporation. The entrapment of measurable fluid inclusions in isolated vugs, on the other hand, may have occurred after return to near-lithostatic conditions after events of active fracture opening and brine infiltration from the surface. Such a pressure fluctuation would allow the possibility that ore formation has occurred at a shallower crustal level with only 3-5 km of overburden.

6.4 Fluid mixing at redox boundary drove metal precipitation at Serra Pelada Au-Pd-Pt Deposit.

The Serra Pelada deposit lacks of any mineralogical evidence for a change in temperature at the deposition site, as the absence of massive quartz veining as in orogenic gold deposits. Fluid inclusion homogenization temperatures are relatively constant within all alteration zones despite large variations in salinity.

The most likely driver for ore formation was the chemical contrast between the oxidizing fluids related to the hematite alteration occurring throughout the upper and lower limbs of the Serra Pelada syncline and in brecciated dolomitic sandstones below the deposit, and the reducing chlorite – carbon alteration along the lower-limb of the Serra Pelada syncline. Locally, mutual cross-cutting and overprinting of oxidized and reduced assemblages confirm that they overlapped in time on the larger scale (Berni et al., 2014). Based on fluid inclusion results, general geology and published solubility constraints, we interpret that an acid and oxidized brine transported detectable concentrations of U, Au, and Pt as well as Se. These elements have in common that they are soluble under highly oxidized conditions, even in rather low-temperature hydrothermal solutions (Bastrakov et al., 2010; Cuney, 2005; Mountain and Wood, 1988; Simon et al., 1996 and 1997). Any sulfur present in this fluid would be on the sulfate predominance field (Ohmoto, 1972). This highly oxidized ore fluid mixed at the site of ore deposition with a CH_4 -bearing reduced fluid responsible for the chlorite – carbon alteration

that was buffered by the carbon-bearing metasiltstone. Mixing of the two contrasting fluids will precipitate selenide minerals if a small proportion of reductant is added to the oxidizing fluid, thereby forming the selenide-hematite-kaolinite assemblages at the ore zone. If the same fluids mix with a greater proportion of the CH₄-bearing fluid, precipitation of hydrothermal carbon is expected, together with sulfide minerals including pyrite and native Au-PGE minerals, resulting in the black ore of the main orebody located in the hinge of the Serra Pelada syncline.

6.5 Genetic model and comparison with similar deposits

A schematic interpretation of possible fluid flow paths and processes that generated the metal concentration at the Serra Pelada deposit is presented in Figure 9, which is necessarily speculative due to the lack of detailed structural and stratigraphic information on a regional scale. The reconstruction is based on a mapped, major pre-mineralization (S1) fault (Cutia Fault – Grainger et al., 2002). This regional fault thrust the mafic and ultramafic metavolcanic rocks of the Rio Novo Group over the Águas Claras Formation, host of the Serra Pelada deposit, providing a plausible former source region extending above the deposit, from which the metals were leached. The mineralization is interpreted to be coeval with a regionally mapped crenulation cleavage and steep fractures (S2). The mechanical driving force for fluid flow and fluid focusing is proposed to be a cyclic fracture-scale fluid pumping between two chemically contrasting rock reservoirs.

Oxidizing and acidic fluids containing Au, Pt and Pd (Fig. 9A) would be transferred from upper parts of the crust (overlying mafic, ultramafic rocks) by reactivation of major faults and fracturing (Au-PGE fluid path - Fig 9B). Fluids that would flow down through sedimentary rocks along more dispersed flow paths would not leach any precious metals and reach equilibrium with the reduced siltstones to become the barren reduced fluid at deeper parts of the system (Barren fluid path). The mixing between these two fluids was responsible for the high-grade metal precipitation at the Serra Pelada Deposit.

The ultimate source of the fluids could be a sedimentary basin environment, possibly associated with the Caninana basin (Prado-Pereira, 2009). Basinal brines or evaporitic fluids would have the necessary fluid salinity for metal complexation and may plausibly be enriched in Mg after precipitation of Ca-rich carbonates.

The ore formation model for Serra Pelada derived here has similarities with unconformity-related U ± Au ± PGE deposits (e.g., Coronation Hill – Mernagh et al., 1994; Jabiluka – Hancock et al., 1990) and sediment-hosted copper deposits (e.g., Brown, 2014; Blundell et al., 2003). The essential fluid-chemical requirements of these ore systems are an oxidized source region in a continental basin, with structures of preferred fracture permeability permitting infiltration of surface brines towards a precipitation zone at a sharp redox boundary (reducing rocks in a subjacent basement domain). The physical hydrology of this deposit remains enigmatic, but this is a common challenge to the above ore systems, requiring downflow of highly oxidized fluids to a site of ore deposition by reduction.

While basin and structural architecture vary widely between Serra Pelada and Unconformity-related U and sediment-hosted Cu deposits, common mineralogical features of these deposits include: (1) widespread although usually subeconomic low-T hydrothermal enrichment in Au and PGE and (2) low sulfidation state of the ore mineralogy, and the presence of selenides and oxide ore minerals indicating redox conditions well above the sulphate/sulfide buffer; (3) Mg alteration with the formation of kaolinite and Mg-rich phyllosilicates as main alteration or gangue minerals, indicating highly acid fluids of intermediate salinity according to fluid

inclusion studies; (4) de-silicification and local re-precipitation of quartz consistent with fluids experiencing warming by local down-flow towards the crystalline basement. We interpret the somewhat higher temperature estimated for Serra Pelada (250-270°C) compared to those of unconformity-related U ± Au ± PGE deposits (100-200 °C; Mernagh et al., 1994, 1998) to be a minor difference, compared to their similarity regarding mineralogy and alteration features and more similar to some of the Kupferschiefer deposits (180-290 °C – Blundell et al., 2003; Kopp et al., 2012).

The geometry of the hydrothermal system is considered a secondary difference that seems most evident from entirely different geological architectures but may be genetically less fundamental for the process of ore formation. At Serra Pelada, the Cutia fault as a significant discontinuity probably played the essential role for regional ground preparation, rather than the unconformity between the metavolcanic mafic and ultramafic rocks of the Rio Novo Group and the sedimentary Águas Claras Formation hosting the deposit. This thrust fault is considered critical for establishing the architecture of the Serra Pelada ore system, as it placed the mafic-ultramafic source of Au-PGE to a higher crustal level and allowed the precious metals to be leached by lateral and downward flow of fluids along a major fluid conduit.

The Serra Pelada deposit also shares chemical characteristics with many sediment-hosted ore systems, such as Kupferschiefer Cu deposits and U-PGE unconformity-related deposits. The main differences in these systems, apart from architecture affecting fluid flow patterns, is in their source rocks controlling metal and sulfur availability. Serra Pelada ore fluids contain more or about the same amount of copper that has been estimated for the formation of Kupferschiefer deposits (ca. 60 ppm – Blundell et al., 2003). The lack of sulfur and the extreme redox conditions of this system made it the Serra Pelada high-grade Au-PGE deposit instead of a copper or uranium mineral system.

7. CONCLUSIONS

- Major cation proportion of ore fluids at Serra Pelada is $\text{Na} > \text{K} > \text{Ca} \geq \text{Mg}$.
- The primary Au-Pt-Pd mineralization was formed at 1.2-1.7 kbar and 250-270 °C.
- Gold, Pd, and Pt were transported by oxidizing (uranium-bearing), acidic fluids (with a pH estimated on 2.5) and precipitated upon reduction.
- Gold contents of ore fluids range between 50-500 ppb.
- The combination of (1) oxidizing cover sequence including hematite-rich metasedimentary and thoroughly oxidized mafic and ultramafic along thrust slices; (2) structural focusing for lateral- and downward-penetrating brines along steeply-dipping fractures, and reactivated fault corridors; (3) mixing between reducing CH₄-rich fluids and oxidized metal carrying fluids at the hinge of the Serra Pelada Syncline; were the essential ingredients for generating this exceptionally rich and geologically unusual Au – Pt – Pd deposit.

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763 Figure 1 – Geological map of Serra Pelada region (Modified from Grainger, 2003 and Berni et
764 al., 2014).

765

Figure 3 – Quartz – dolomite veins, vugs textures, and fluid inclusion assemblages (FIA) relationships. (A) Vuggy dolomite with dark growth zones defined by numerous solid inclusions in relation to successive fluid inclusion assemblages. (B) Quartz crystals with growth zones marked by hematite and carbonate precipitation. (C) Backscattered scanning electron microscope image of the hematite-rich growth zone shown in figure 3A, with numerous solid inclusions of sulfide and oxide minerals, and the location of the LA-SF-ICPMS ablation spots shown in figures (D) and (E). (D) Transient LA-SF-ICPMS signal of a 130 μ m ablation pit covering the dark growth zones shown in C, identifying micron-sized mineral inclusions of minerals including elements associated with the Serra Pelada Au - Pd - Pt mineralization such as U, and Bi as well as Au, Pt and Pd. (E) Transient signal of a 130 μ m ablation pit away from the inclusion-rich, showing the normal levels of elements related to the mineralization. (F) Pseudosecondary FIA hosted by dolomite. (G) Detail on pseudosecondary FIA hosted by quartz. (H) Detail on dolomite hosted aqueous fluid inclusions.

Figure 4 – Fluid inclusion assemblages (FIA) from the Chlorite – carbon and Siliceous alteration zones. (A) Fine-grained quartz from the Chlorite – carbon alteration associated with amorphous carbon inclusions and euhedral quartz. (B) Detail of an aqueous fluid inclusion assemblage hosted by quartz within the Chlorite – carbon alteration. (C) Vuggy quartz within siliceous alteration with well-defined growth zones marked by globular hematite inclusions, secondary and pseudosecondary inclusion trails. (D) Detail of an aqueous fluid inclusion assemblage hosted by quartz within the siliceous alteration. (E) Transient LA-ICPMS signal of the ablation of a fluid inclusion with kaolinite crystals. Note the peak offset between soluble elements (Na, K) and Al. (F) Fluid inclusion from siliceous alteration with accidentally trapped kaolinite crystals.

Figure 5 – (A) Average homogenization temperatures (°C) vs. NaCl_{eqv.} (wt. %) from fluid inclusion assemblages (FIA) within the three major alteration types at the Serra Pelada Deposit. (B-E) Summary of average compositions for K, K/Na, Mg and Mg/Na determined by LA-SF-ICPMS microanalysis, sorted by apparent salinity within alteration groups separated by dashed lines corresponding to: (1) distal Chlorite – carbon alteration; (2) siliceous alteration in and around ore zone; (3) Hematite alteration zone. Thin lines separate subgroups between primary/pseudosecondary and secondary FIA, as well gold-bearing (red) and barren FIA (black) from distal hematite alteration. Each symbol represents the average value for a fluid inclusion assemblage (FIA), with the respective 1σ standard deviations shown by the error bars. Within each of the three alteration groups, assemblages are sorted by increasing salinity (NaCl_{eqv.}) indicated by values on the x-axis.

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854

855 Figure 7 –Transient LA-SF-ICPMS signals with detected gold in fluid inclusion hosted by
856 dolomite (A) and quartz (B). (C) Au concentrations (circles) and detection limits (dashes) of
857 individual fluid inclusions microanalysis from FIAs where gold was detected. (D) A transient
858 LA-SF-ICPMS signal with a clear platinum peak in a fluid inclusion hosted by dolomite.

859

Figure 8 – Diagram illustrating pressure and temperature conditions for the formation of the Serra Pelada deposit, based on calculated fluid inclusion isochores (Driesner and Heinrich, 2007), ore mineral geothermometers from Berni et al. (2016) and the stability field of kaolinite/pyrophyllite according to Holdaway (1971).

A - PRÉ-MINERALIZATION

B - MINERALIZATION

892

893 Figure 9 – Schematic model summarizing the proposed mineralization at Serra Pelada Au - Pd
 894 - Pt deposit. (A) Before the deposit was formed, both host and basement sequences were
 895 already deformed and thrust against each other. The Cutia fault thrusts the mafic rocks of the
 896 Rio Novo Group and the Luanga ultramafic complex the over the Águas Claras Formation.
 897 The Águas Claras formation is simplified in (1) upper metasedimentary sequence (mature
 898 sandstones and red beds); (2) red and carbonaceous siltstones, host of the Serra Pelada Au-
 899 PGE deposit; (3) lower metasedimentary sequence (dolomitic sandstones metasiltstones).
 900 Fluid circulation would occur above the hydrostatic/lithostatic boundary, allowing the
 901 oxidizing fluid (red) to leach Au and PGE from oxidized mafic and ultramafic rocks. (B)
 902 During tectonic pulses, fault reactivation and step fracturing would allow oxidizing fluids to
 903 descend below the hydrostatic/lithostatic boundary by fluid focusing structures, either focusing
 904 fluids circulating predominantly through metasedimentary and later by carbonaceous-bearing
 905 rocks (Barren fluid path), or by the metavolcanic rocks of the Rio Novo Greenstone or the
 906 Luanga mafic-ultramafic intrusion (Au-PGE fluid path). At the syncline hinge, extreme
 907 reduction of these fluids would occur by reaction with carbon-enriched lithologies and mixing
 908 with reducing fluids (green), precipitating the Au-PGE ore. After the tectonic pulse, the
 909 pressure would return to the lithostatic regime, pushing the spent ore fluid up through major
 910 thrusts systems.

TABLES CAPTIONS

Sample	Alteration	Drill hole and depth	Description
S.196	Chlorite – carbon	SPD-141 68.2 m	Chlorite altered red siltstone with quartz-carbon vein and euhedral crystals at vein core.
S.064	Siliceous	FD-0126 223.1m	Coarse-grained silicified dolomitic sandstone with abundant hematite inclusions.
S.136	Siliceous	SPD-007 198.6 m	Coarse-grained hematite dusted (red) silicified dolomitic sandstone with vugs filled by well formed quartz and hematite.
S.140	Siliceous	SPD-051A 214.5 m	Coarse-grained silicified dolomitic sandstone with vugs filled by well formed quartz and hematite.
S.166	Siliceous	SPD-007 195.3 m	Brown, clay-rich breccia with free standing quartz crystals within the outer silicification zone.
S.145	Hematite	SPC-022 320.0 m	Recrystallized dolomitic sandstone with vugs containing centimeter scale quartz crystals.
S.147	Hematite	SPC-022 351.6 m	Recrystallized dolomitic sandstone with vugs filled by centimeter scale coexisting quartz and dolomite crystals and abundant hematite inclusions.
S.141	Hematite	SPC-022 351.7 m	Recrystallized dolomitic sandstone with vugs containing centimeter scale dolomite and quartz crystals.
S.142	Hematite	SPC-022 268.3 m	Recrystallized dolomitic sandstone with vugs containing centimeter scale dolomite crystals.

Table 1 – Drill hole, depth, and alteration zone of the samples used for fluid inclusion analysis.

916 Table 2 – Average elemental composition of fluid inclusion assemblages (FIAs) from LA-SF-
917 ICPMS microanalysis.

918 Footnote table 2: Values shown as <X correspond to the lower detection limit within the FIA.
919 Data labeled as the matrix are related to elements which could not be quantified due to host
920 contamination. Abbreviations: Pr: primary, Ps: pseudosecondary; Sec: secondary; Un:
921 unknown/unidentified FIA. n.a.; not analyzed.

922

N°	Sample / Fia Type	NaCleqv. wt %	Th °C	Na ppm	Mg ppm	S ppm	K ppm	Ca ppm	Ni ppm	Cu ppm	As ppm	Sb ppm	Cs ppm	Ba ppm	Ce ppm	Au ppm	Pb ppm	U ppm
Reduced	196/PS	2.57	164	10445	<310	n.a.	560	n.a.	<0.3	<10	152	<0.085	<0.1	5.2	<0.01	<0.1	<0.05	<0.068
	1σ std	0.26	14	1365			555				12			2.6				
	196/PS	8.66	160	35025	125	n.a.	355	n.a.	<60	277	98	<0.1	4.1	59	<0.1	<0.01	39	<0.1
	1σ std	0.41	17	805	35		15			181	59		0.6	47			17	
	196/PS	8.81	170	35455	565	n.a.	1505	n.a.	<2	72	43	<0.4	5.9	27	<1	<1	7.5	<0.05
	1σ std	0.59	19	680	350		1690			19	18		2.7	3.5			6.7	
	196/PS	9.84	152	380570	<170	n.a.	950	n.a.	<0.5	133	19	<0.1	6.3	15	<0.1	<0.1	9.8	<0.1
	1σ std	0.28	1	1215			150			120	20		3.9	5.3			6.8	
	196/PS	11.81	133	46935	2730	n.a.	940	n.a.	<55	157	215	<10	9.5	294	<0.1	<0.1	165	<0.1
	1σ std	0.28	26	1215	1685		205			54	225		10.1	87			8	
	136-2/Pr	3.36	159	12695	180	n.a.	1430	n.a.	<5	4.4	7.0	0.3	0.35	14	<0.1	<0.1	5.2	<0.1
	1σ std	0.55	7	2135	120		1350			3.4	3.9	0	0.31	7.6			4	
	136-2/PS	3.79	161	14240	395	n.a.	470	n.a.	<2.5	6.1	31	<0.5	0.62	19	<0.1	<0.1	18	<0.1
	1σ std	0.49	7	1555	510		520			2.6	43		0.21	21			21	
	136-2/Pr	3.99	158	15570	460	n.a.	835	n.a.	<10	<5	13	<1.5	0.86	21	<0.1	<0.2	8.5	0.3
	1σ std	0.27	8	955	120		670			7.2	7.2		0.77	12			8	0.3
	136/PS	4.40	n.a.	16290	<160	n.a.	1435	n.a.	<20	<5	24	2.1	1.31	22.0	<0.5	<0.2	<0.5	<0.1
Slicaceous	1σ std	0.32		1290			325			2.4	2.4	1	0.29	2.6				
	140/PS	5.32	n.a.	17550	289	n.a.	4975	n.a.	<70	85	38	<2	1.9	49	<0.2	<0.3	56	<0.1
	1σ std	0.20		1695	55		2125			59	3.5		0.1	22			58	
	140/PS	6.59	n.a.	23735	365	n.a.	2920	n.a.	<2	<1	6	<1	0.0	9	<2	<0.1	114	<0.1
	1σ std	0.55		3635	215		2610			1.3	1.3	0.0	0.0	6			14	
	136-2/Un	3.30	n.a.	12500	195	n.a.	975	n.a.	6.5	<5	9.4	<1	<0.5	12	<0.2	<0.3	7.9	<0.1
	1σ std	0.20		885	155		1235		3.3		1.9			13			6	
	64/Un	3.54	120	10435	n.a.	<360	295	<630	n.a.	<2.5	<3.0	<1	0.15	6.46	<0.3	<0.3	<0.3	<0.2
	1σ std	1.01	17	1380			40						0.08	0.36				
	64/2/Un	3.94	166	15500	n.a.	<800	2690	<5030	n.a.	188	<10	<5	<1	77	<1	<1	<2	<3
	1σ std	0.72	12	2955			2335			46				29				
	0.64-2/Sc	4.02	145	150060	120	n.a.	1150	n.a.	<20	3.26	12	<1	0.95	56	<0.5	<0.5	<1	<0.1
	1σ std	0.31	11	1670	80		1215			0.83	0.1		0.46	22				
	136-2/Un	5.00	n.a.	18870	665	n.a.	1405	n.a.	<5	<5	<5	<1	<0.2	11.3	<0.1	<0.2	19.0	0.2
	1σ std	0.08		490	75		445							8.5			2.5	0.1
	166/Un	5.71	n.a.	19265	280	<2200	4580	<2600	<85	<20	<10	<5	4.7	321	<1	<1	<8	<1
	1σ std	0.36		1585	50		1190						1.2	198				
	140/Un	5.73	n.a.	21095	<1620	n.a.	4920	n.a.	<190	<100	<100	<20	<10	85	<5	<5	<5	<0.5
	1σ std	0.21		315			350							20				
	166/Un	5.92	n.a.	21140	360	<1600	4380	2595	<35	39	<5	<4	5.8	704	<1	<1	<5	<1
	1σ std	0.27		1350	20		2000	335		7.5			0.6	55				
	64/Un	9.02	151	32245	n.a.	<1200	6950	<11250	n.a.	<11	<17	<5	<2	34	<2	<2	<3	<1
	1σ std	1.60	19	8200			4940							19				

N°	Sample / Fia Type	NaClevq.	Th	Na	Mg	S	K	Ca	Ni	Cu	As	Sb	Cs	Ba	Ce	Au	Pb	U
		wt %	°C	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
Oxidized - Gold bearing	147-2/Ps	4.26	n.a.	14605	755	n.a.	2454	n.a.	254	34	24	1	1.2	18	<0.2	0.325	5.5	<2
	1σ std	0.39		2090	330		870		150	15	13	0	0.6	7.1		0.090	3.7	
	147-2/13/Ps	4.98	171	16195	1020	n.a.	2315	n.a.	10.9	10	72	5	2.8	4.4	<2	0.255	22	<1
	1σ std	0.18	9	1550	585		220		1.0	2	32	3	1.3	1.4		0.145	28	
	147-2/15/Ps	5.12	163	17000	1830	n.a.	665	n.a.	11.2	42	8	3	1.4	19	0.8	0.210	9	1
	1σ std	0.21	12	2045	1290		465		12.4	36	4	2	1.1	5.6	0.4	0.165	6	1.11
	145/3.1/Ps	6.07	178	19595	Matrix	n.a.	5770	Matrix	<10	3	38	0	2.1	11	<0.5	0.080	Matrix	0
	1σ std	0.22	5	1085			1795		2	24	0	0.8	7.3		0.050			0.01
	145/0.1-2B/Ps	6.15	n.a.	16310	Matrix	n.a.	10480	Matrix	<5	7	46	<0.2	1.7	47	2.3	0.345	Matrix	<0.1
	1σ std	0.05		1370			1825		3	21		0.6	74	4.0	0.030			
	145-2/Ps	3.25	n.a.	13080	Matrix	n.a.	1650	Matrix	<10	<35	<15	<1	1.5	31	<0.2	<0.3	Matrix	<0.1
	1σ std	0.62		2745			1570					0.4	36					
	145-2/Ps	3.83	n.a.	12790	Matrix	n.a.	2110	Matrix	<15	<5	<5	<2	1.1	<1	<0.2	<0.2	Matrix	<0.1
	1σ std	0.36		1005			1140					0.2						
	145-2/Ps	3.83	n.a.	12835	Matrix	n.a.	2935	Matrix	<5	<3	36	<1	1.5	32	1.3	<0.2	Matrix	0.80
Oxidized - Barren	1σ std	0.08	n.a.	1105			1120			30		0.6	5.6	0.8				0.1
	147-2/Un	4.20	185	14755	Matrix	n.a.	2590	Matrix	<40	17	43	<1	2.3	52	<0.2	<0.2	Matrix	0.72
	1σ std	0.25	5	810			1635		9	39		1.1	8					0.6
	145-2/Pr	4.31	n.a.	14920	Matrix	n.a.	2560	Matrix	<15	<15	<15	<3	1.0	19	<2	<0.2	Matrix	<0.1
	1σ std	0.13		1925			2740					0.0	5.6					
	145-2/Un	4.49	n.a.	13305	Matrix	n.a.	5660	Matrix	<5	3	7	<0.2	1.8	13	<0.05	<0.05	Matrix	0.26
	1σ std	0.13		415			315		1	6		0.2	11					0.22
	147/2/Ps	4.62	166	16455	n.a.		3855	1470	2465	28.6	71	12	<0.5	1.3	4.1	<0.1	<0.1	16
	1σ std	0.28	12	1075			95	1245	1355	9.4	26	6		0.6	0.9			17
	147/1/Ps	4.92	168	16225	n.a.	<2000	1230	2655	<2	10	2	<0.2	1.5	17	<0.2	<0.1	4	<0.1
	1σ std	0.35	4	1830			375	570		5	1		1.0	14				3
	147-2/18/Ps	5.04	n.a.	17950	410	n.a.	475	n.a.	23.3	4	40	1	2.3	11	<0.2	<0.1	8	1
	1σ std	0.08		1115	2250		480		7.3	2	12	0	0.3	4.5				7
	145/4/Ps	5.10	169	15280	Matrix	n.a.	6966	Matrix	30.2	10	24	<0.1	2.1	141	<0.5	<0.1	Matrix	0
	1σ std	0.14	10	1485			1325		8.4	10	8		0.6	81				0.05
	147/3/Ps	5.39	154	19340	n.a.	<3500	1795	980	25.7	73	20	<0.5	1.2	7.5	<0.2	<0.3	5	<0.1
	1σ std	0.40	12	1655			300	190	26.1	68	11		0.4	6.7				2
	145-2/01B/Ps	5.18	n.a.	16320	Matrix	n.a.	8710	Matrix	<2	38	42	<0.3	2.4	25	<1	<0.2	Matrix	<0.1
	1σ std	0.15		1190			1595			42	14		1.0	37				
	145/1/Ps	5.94	177	16650	Matrix	<1050	8970	Matrix	<1.5	5	36	n.a.	2.2	9.3	<0.1	<0.1	Matrix	<0.1
	1σ std	0.13	7	670			780			5	13		0.4	3.6				
	145-2/0.1D/Ps	5.98	n.a.	18315	Matrix	n.a.	6980	Matrix	<1.02	<2.5003	35	<0.165	1.8	10.1	<0.1	<0.2	Matrix	0
	1σ std	0.21		1510			2445			21		0.4	7.8					0.04
	145/3/Ps	6.26	185	17590	Matrix	n.a.	9635	Matrix	<2	6	46	<0.2	2.5	11.2	<0.1	<0.05	Matrix	0
	1σ std	0.25	2	615			355			4	22		0.4	10.9				0.04
	145/4.1/Ps	6.51	n.a.	17585	Matrix	<1200	10725	Matrix	<4	4	48	n.a.	2.5	10.0	<0.2	<0.1	Matrix	0
	1σ std	0.07		663			865			3	12		0.4	6.8				0.42
	145-2/0.1-2D/Pr	7.30	n.a.	20345	Matrix	n.a.	11200	Matrix	<15	<10	44	<2.16	2.6	51.7	<0.2	<0.4	Matrix	<0.031
	1σ std	0.20		1635			1100			9		0.3	70.0					
	145-2/2/Pr	11.01	n.a.	42755	Matrix	n.a.	1030	Matrix	<30	<20	<25	<5	<2	13.9	<0.3	<0.4	Matrix	<1
	1σ std	0.22		775			1190						4.6					

Oxidized - Barren

N°	Sample / Fia Type	NaCleqv. wt %	Th °C	Na ppm	Mg ppm	S ppm	K ppm	Ca ppm	Ni ppm	Cu ppm	As ppm	Sb ppm	Cs ppm	Ba ppm	Ce ppm	Au ppm	Pb ppm	U ppm
	141/Un	3.95	174	12870	740	<4460	4555	1936	<30	21	31	<5	7.1	26	<2	<1	9.0	1.3
	1σ std	0.42	8	3650	1730		3595	693		5	19		6.9	16			2.14	0.1
	147-2/Un	4.06	169	15930	830	n.a.	625	n.a.	<10	11	18	<0.3	0.8	13	0.4	<0.1	8.5	<0.5
	1σ std	0.53	5	1315	630		575			7	6		0.3	14.0			4	
	145-2/Un	4.31	n.a.	14920	Matrix	n.a.	2560	Matrix	<15	<15	<15	<3	1.0	19	<2	<0.2	Matrix	<0.1
	1σ std	0.13		1925			2740						0.0	5.6				
	141/Un	4.32	162	14365	n.a.	<320	4460	<6000	<5	142	<40	<2	9.3	8.9	<8	<2	1	<1
	1σ std	0.23	11	4245			4890			57			2.9	1.7			0.4	
	142/Un	4.43	177	12570	Matrix	<1300	6490	Matrix	<15	<10	26	<1	2.9	90	<2	<0.5	22	<0.1
	1σ std	0.18	6	1115			1340			14	14		2.3	54			23	
	142/Un	4.49	174	13480	Matrix	<1200	5890	Matrix	<35	48	46	<4	2.4	90	<2	<2	33	<2
	1σ std	0.09	10	2315			2670			21	13		0.5	79			17	
	141/2/Sc	4.55	165	16475	n.a.	<1400	2035	<3130	<15	<25	18	<6	5.4	<15	<3	<5	<6	<3
	1σ std	0.42	9	1030			675				3		4.4					
	147-2/2/Un	14.29	175	50535	2500	n.a.	2430	n.a.	<40	93	<10	<2	0.5	28	<0.2	<0.2	7	<0.1
	1σ std	0.13	6	3000	1750		750			108			0.1	5.9			5	
	147-2/7/Un	15.72	166	50730	4244	n.a.	6170	n.a.	<5	43	18	<1	6.7	156	<1	<0.2	57	0
	1σ std	0.74	16	2450	1535		1035			19	13		2.4	55			17	0.14
	147-2/16/Sc	16.67	141	54150	3915	n.a.	8350	n.a.	<5	38	10	<45	9.3	252	<4	<0.1	126	<0.1
	1σ std	0.19	3	3260	1410		1455			4	1		0.3	248			14	
	147-2/3/Un	17.80	n.a.	61300	4895	n.a.	2700	n.a.	<30	49	7	<2	0.3	25	<0.2	<0.3	34	<0.1
	1σ std	0.29		5935	2940		435			9	4		0.2	10.7			24	
	147-2/8/Un	18.39	131	52285	9115	n.a.	7530	n.a.	<20	32	<20	<3	5.0	136	<0.5	<1	38	<1
	1σ std	0.21	11	4210	2315		1020			14			0.7	8.4			15	
	141/6/Sc	19.96	160	71745	n.a.	<3500	15015	<6000	<50	183	<80	<18	<5	200	<6	<15	23	<7
	1σ std	2.32	10	6115			4295			92				208			13	
	145-2/3E/Sc	20.70	n.a.	79820	Matrix	n.a.	2455	Matrix	<4	4	4	<0.2	2.6	8.5	<0.5	<0.032	Matrix	0
	1σ std	0.11		540			295			2	1		0.6	5.3			0.06	
	147-2/4/Un	21.29	149	73905	3520	n.a.	6350	n.a.	<55	7	9	<2	6.5	113	<0.3	<0.3	88	0
	1σ std	0.38	21	1534	1060		325			3	3		1.8	27			17	0.48
	142/5/Un	21.66	155	6755	2430	<1000	7465	10670	169	<2500	3	<5	1.9	15	<0.4	<1	33	<0.2
	1σ std	1.49	23	4990	360		2340	990	33		1		1.0	1.3			31	

925 Table 3 –Pt concentration in individual fluid inclusion from LA-SF-ICPMS microanalysis.

926

FIA number	NaCleqv. wt %	Pt ppb	±Pt ppb	Host ppb
FIA-1	14.4	<27		<0.69
	13.4	12	4	<0.85
	13.5	155	35	<0.51
FIA-2	17.8	<460		0.26
FIA-3	7.6	<46		0.42
	7.8	<386		0.2
FIA-4	3.6	<1	<1	<0.32
	3.6	<1		0.51
	3.6	9	2	0.27
	3.6	<6		0.24
	3.6	<2		0.21
	3.6	31	3	<0.30
	3.6	14	2	<0.34
	3.6	<2		0
FIA-5	5.0	<28		<0.22
FIA-6	3.6	<5		0.22
	3.4	25	3	<0.50
	3.6	<3		<0.55
	3.6	<4		0.39
	3.6	<7		<0.37
	3.6	<6		<1.1
FIA-7	5.9	<20		<0.41
FIA-8	3.9	<4		0.24
	3.9	<15		<0.65
FIA-9	20.2	12	1	<0.75
FIA-10	3.6	16	1	0.54
	3.6	<350		<0.41