



Nature of ore-forming fluid and formation conditions of BIF-hosted gold mineralization in the Archean Amalia Greenstone Belt, South Africa: Constraints from fluid inclusion and stable isotope studies



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ABSTRACT

Orogenic gold mineralization in the Amalia greenstone belt is hosted by oxide facies banded iron-formation (BIF). Hydrothermal alteration of the BIF layers is characterized by chloritization, carbonatization, hematization and pyritization, and quartz-carbonate veins that cut across the layers. The alteration mineral assemblages consist of ankerite-ferroan dolomite minerals, siderite, chlorite, hematite, pyrite and subordinate amounts of arsenopyrite and chalcopyrite. Information on the physico-chemical properties of the ore-forming fluids and ambient conditions that promoted gold mineralization at Amalia were deduced from sulfur, oxygen and carbon isotopic ratios, and fluid inclusions from quartz-carbonate samples associated with the gold mineralization.

Microthermometric and laser Raman analyses indicated that the ore-forming fluid was composed of low salinity H₂O-CO₂ composition (~3 wt% NaCl equiv.). The combination of microthermometric data and arsenopyrite-pyrite geothermometry suggest that quartz-carbonate vein formation, gold mineralization and associated alteration of the proximal BIF wall rock occurred at temperature-pressure conditions of 300 ± 30 °C and ~2 kbar. Thermodynamic calculations at 300 °C suggest an increase in *f*O₂ (10⁻³²–10⁻³⁰ bars) and corresponding decrease in total sulfur concentration (0.002–0.001 m) that overlapped the pyrite-hematite-magnetite boundary during gold mineralization. Although hematite in the alteration assemblage indicate oxidizing conditions at the deposit site, the calculated low *f*O₂ values are consistent with previously determined high Fe/Fe + Mg ratios (>0.7) in associated chlorite, absence of sulfates and restricted positive δ³⁴S values in associated pyrite. Based on the fluid composition, metal association and physico-chemical conditions reported in the current study, it is confirmed that gold in the Amalia fluid was transported as reduced bisulfide complexes (e.g., Au(HS)₂⁻). At Amalia, gold deposition was most likely a combined effect of increase in *f*O₂ corresponding to the magnetite-hematite buffer, and reduction in total sulfur contents due to sulfide precipitation during progressive fluid-rock interaction.

The epigenetic features coupled with the isotopic compositions of the ore-forming fluid (δ³⁴S_{SS} = +1.8 to +2.3‰, δ¹⁸O_{H2O} = +6.6 to +7.9‰, and δ¹³C_{ΣC} = -6.0 to -7.7‰ at 300–330 °C) are consistent with an externally deep-sourced fluid of igneous signature or/and prograde metamorphism of mantle-derived rocks.

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1. Introduction

Archean orogenic (or so-called mesothermal) gold deposits share similar structural complexities that extend deep along crustal-scale faults and are widely considered to form from similar hydrothermal fluids; notably H₂O-CO₂ ± CH₄ ± N₂-NaCl fluids of relatively low to moderate salinities that were tapped from deep

magmatic/mantle- or metamorphic-related processes (Groves et al., 1998). Gold mineralization in the deposits commonly occurs in quartz ± carbonate veins or as disseminations in wall rocks close to these veins. On the other hand, minor differences in the deposits have been linked to host rock composition, redox conditions of gold formation and/or other physico-chemical parameters at the individual deposit. In terms of redox conditions, both reduced and oxidized end members of the Archean gold deposits are widely documented in the literature; each exhibiting distinctive mineral and metal associations with gold. The classification of gold

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deposits by Phillips and Powell (1993, 2015) into the 'gold-only' and 'gold-plus' scheme identified the chemistry of gold in relation to metal complexing, salinity of ore-forming fluids and redox state. Geological, mineralogical, geochemical, fluid inclusion and stable isotope studies have been useful in understanding the redox control in these deposits. Processes relating to the formation of reduced gold deposits occur at relatively low oxygen fugacity (fO_2) conditions and are relatively easy to comprehend. On the other hand, various processes have been implied for the formation of the oxidized end members. Mass balance considerations based on oxidized-reduced sulfur species suggest that the fO_2 value of the mineralizing fluids was high (e.g., Cameron and Hattori, 1987; Phillips et al., 1996). Evidence for this is the occurrence of hematite in altered wall rocks, sulfate minerals and sulfides of varied isotopic compositions (Cameron and Hattori, 1987). Unlike in the Archean Eon, oxidized hydrothermal fluids are common in the post-Archean Eon. However, in the Archean, the largest individual primary gold deposits are of oxidized character e.g., the Hemlo and Hollinger-McIntyre camps in Canada and Golden Mile (Kalgoorlie) in Australia (Phillips et al., 1996; Groves et al., 2015). Therefore, it is quite evident that in spite of the common acceptance of a generally reduced environment in the Archean, hydrothermal fluids that are characterized by high oxygen fugacity are also important in the formation of gold deposits in that Eon.

Redox conditions are important for understanding the physico-chemical conditions for gold transport and deposition in mineral deposit systems. In general, three chemical processes that effectively promote gold deposition are: (1) the decrease of solution fO_2 , ideally into the reduced carbon field; (2) the decrease in total sulfur in solution; (3) the increase of solution fO_2 into the hematite field with generation of sulfate. The last two processes bring about gold deposition by lowering the concentration of reduced sulfur. However, Phillips and Powell (2010) and Cameron and Hattori (1987) noted that evaluating the importance of the process of fluid oxidation to the hematite field that is applicable to gold-only deposits is not an easy task. Based on studies conducted on the Kalgoorlie gold deposits (e.g., Phillips and Gibb, 1993; Phillips et al., 1986), Phillips and Powell (2010) suggested that the fO_2 of auriferous fluids would not shift well into the hematite field, where SO_4^{2-} would greatly exceed reduced sulfur (i.e. $H_2S + HS$). This is reflected in the fact that the expected full range up to 20‰ $\delta^{34}S$ is not attained in the deposits (Lambert et al., 1984; Evans et al., 2006; Hodkiewicz et al., 2009). As such, Phillips and Powell (2010) concluded that it is possible to form hematite with no sulfate in orogenic gold-only deposits, which in turn, may result in precipitation of sulfides with narrow positive $\delta^{34}S$ values (cf. gold solubility experiments of Gibert et al., 1998). It is worth noting that most of the gold solubility experiments for high temperature environments were conducted under saturation vapor-pressure conditions that are typical for epithermal gold deposits (Ohmoto and Rye, 1979). These conditions are not necessarily reflective of the high lithostatic pressure gradients that are documented for the mesozonal-hypozonal classes of Archean orogenic gold deposits. Therefore, a possibility that is often overlooked in the literature is formation of oxidized gold deposits under high pressure-low fO_2 conditions that could yield sulfur with restricted narrow positive $\delta^{34}S$ values and no sulfates. These considerations formed the basis for presenting this paper on the Amalia banded iron formation (BIF)-hosted gold deposit (also known as the Blue Dot deposit; New Dawn Mining Corp. press release, 2009). The cut-off grade in the deposit is 2.0 g/t at USD 750/oz gold price (New Dawn Mining Corp. press release, 2009). The Amalia deposit is an example of a relatively oxidized Archean gold deposit that contains hematite, pyrite with narrow positive $\delta^{34}S$ values and no sulfate (Adomako-Ansah et al., 2013).

In spite of intermittent gold mining activities since 1903, the ore-forming physico-chemical conditions on the genesis of the Amalia gold deposit have not been fully established yet. Previous works on the deposit (e.g., Vearncombe, 1986; Hunt, 1996; Cheshire, 1996; Kiefer, 2004; Adomako-Ansah et al., 2013) concentrated on the geological, mineralogical, whole rock geochemistry and structural controls on gold mineralization. Orogenic gold mineralization in the BIF at Amalia is intimately associated with quartz-carbonate veins that cut across the BIF layers (Vearncombe, 1986; Kiefer, 2004; Adomako-Ansah et al., 2013). Gold is not found in the quartz-carbonate veins themselves but is hosted within sulfidized-alteration haloes and selvages of the proximal BIF units that are in contact with these veins. Field observation and textural evidence suggest that the gold-associated sulfides in the Amalia BIF occurred only around the contacts between the quartz-carbonate veins and the BIFs. Sulfidation of BIF layers is not observed in other sections of the BIF units where quartz-carbonate veins are absent. Geochemical data have recorded high gold concentrations in sulfidized BIF that surround the cross-cutting quartz-carbonate veins, but register no gold showings in the non-sulfidized BIF (e.g., Kiefer, 2004; Adomako-Ansah et al., 2013). Therefore, on the basis of the geological, textural and geochemical features, fluid-rock interaction between an externally-derived auriferous fluid and the Amalia BIF is considered to be responsible for the gold mineralization at Amalia (Vearncombe, 1986; Kiefer, 2004; Adomako-Ansah et al., 2013).

Determination of the composition of paleo-fluids trapped within minerals such as quartz and carbonate that crystallized directly from the ore-forming fluid and reasonably inferred to be related to the gold mineralization can provide valuable information on the gold transport mechanism as well as estimation of the pressure (P)-temperature (T)-depth (d) conditions of gold deposition. Based on textural relationships and mineral-chemical data on chlorite, carbonate and arsenopyrite-pyrite assemblage, Adomako-Ansah et al. (2013) documented that an increase in fO_2 and decrease in total sulfur contents were predominant factors that facilitated gold deposition at Amalia. However, fluid inclusion studies remain the fundamental analytical tool used to directly obtain both qualitative and quantitative data on such fluid properties, as well as other behavioral characteristics of the fluid components that may have implication(s) for the nature of the ore-forming processes (e.g., phase separation, fluid immiscibility or mixing). In addition, independent estimates of T (or P), such as those based on mineral thermometry or oxygen isotope geothermometry, make a study of fluid inclusions a powerful technique to constrain the physical conditions of gold deposition. In order to better constrain the physico-chemical conditions of gold transport and deposition in the Amalia gold deposit, this paper presents results from fluid inclusion study and stable isotopic compositions of hydrothermal quartz and carbonates from the deposit that were integrated with data from chlorite and arsenopyrite compositions and $\delta^{34}S$ values of hydrothermal pyrite from the deposit (Adomako-Ansah et al., 2013). The conclusions from this study have implication for the nature of gold transport and deposition by reduced gold-sulfide complexes in relatively oxidized hydrothermal fluids in Archean orogenic gold deposits.

2. Geological setting, alteration, quartz-carbonate veins and gold mineralization

The BIF-hosted Amalia gold deposit is located in the Amalia greenstone belt, about 325 km WSW of Johannesburg, South Africa. The Amalia greenstone belt forms part of the Amalia-Kraaipan greenstone-granitoid terrane in the Kimberley Block of the Kaapvaal craton (Fig. 1). The poorly exposed Amalia-Kraaipan

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