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Gold accumulation in the Archaean Witwatersrand Basin, South Africa — Evidence from concentrically laminated pyrite

A. Agangi [a,*], A. Hofmann [a], C. Rollion-Bard [b,1], J. Marin-Carbonne [c,2], B. Cavalazzi [d], R. Large [e], S. Meffre [e]

[a] Department of Geology, University of Johannesburg, Auckland Park 2006, Johannesburg, South Africa
[b] CRPG-CNRS, Université de Lorraine, 15 rue Notre Dame des Pauvres, 54500 Vandoeuvre-lès-Nancy, France
[c] Department of Earth and Space Sciences, UCLA, 595 Charles Young Drive East, Los Angeles, CA 90095, USA
[d] Dipartimento di Scienze Biologiche, Geologiche e Ambientali, Università di Bologna, Via Zamboni 67, 40126 Bologna, Italy
[e] CODES ARC Centre of Excellence in Ore Deposits, University of Tasmania, Hobart, Tasmania 7005, Australia

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ABSTRACT

Concentrically laminated pyrite is a relatively common, although volumetrically minor, component of auriferous conglomerates in the Archaean (ca. 3.0–2.7 Ga) Witwatersrand Basin of South Africa. This type of pyrite contains high amounts (several tens of ppm) of Au, but the origin of the pyrite is debated, and the timing of Au deposition in these grains is not known. In order to constrain the formation of pyrite, we have studied concentrically laminated pyrite and other coexisting types of pyrite (inclusion-rich, massive pyrite) by analysing the contents and distribution of Au and other trace elements by laser ablation ICP-MS, the S and Fe isotope composition by SIMS, and the mineral inclusions by scanning electron microscope and laser Raman spectroscopy. Trace element maps indicate that concentrically laminated pyrite is enriched in Sb, Mn, Au, Ag, Tl, Cu, Mo, Mn, and contains two types of gold: finely dispersed Au ("invisible gold", with Au/Ag ~0.1 and likely of primary origin) and Au inclusions with Au/Ag ~10 of secondary origin. The study of mineral inclusions revealed the presence of muscovite, chlorite, fine-grained carbonaceous matter, monazite, Ti-oxides, and quartz. Iron and multiple S isotopes suggest that concentrically laminated pyrite and inclusion-rich pyrite were formed from two separate pools of S and Fe with different isotope characteristics. Sulfur was derived from atmospheric S that had undergone mass-independent isotope fractionation to form SO_4^{2-} with negative $\Delta^{33}S$ that constituted concentrically laminated pyrite, and elemental S with positive $\Delta^{33}S$ that formed inclusion-rich pyrite. Iron pools were derived from partial oxidation of Fe^{2+}, so that concentrically laminated pyrite formed from a low-$\delta^{56}Fe$ residual Fe^{2+} (average +0.2‰) and inclusion-rich pyrite formed from a high-$\delta^{56}Fe$ Fe^{3+} pool (average +2.7‰). Biological activity may have been involved in the reduction of SO_4^{2-}, causing a wide spread of $\delta^{34}S$ values (~25‰, S reducing microorganisms), as well as in the partial oxidation of Fe^{2+} (anaerobic photosynthetic Fe reducers or photosynthetic O_2 producers), and in the formation of pyrite from Fe^{3+} (dissimilatory Fe reducers). We propose that concurrent biogenically-mediated pyrite formation and Au trapping suggest that microbial activity was responsible for the accumulation of Au and other trace elements (e.g. Sb, Mn, Ag, Tl, Cu, Mo, Mn) which are commonly enriched in organic matter-rich sediments.

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[*] Corresponding author at: Applied Geology Department, Curtin University, Kent St, Bentley 6102, WA, Australia Tel. + 61(0)892667074.
E-mail address: andrea.agangi@curtin.edu.au (A. Agangi).

[1] Present address: Institut de Physique du Globe - Sorbonne Paris Cité, Université Paris Diderot, CNRS, 1 rue Jussieu, 75238 Paris cedex 05, France.

[2] Present address: Laboratoire Magmas et Volcans – Transferts Lithosphériques UMR CNRS 6524 Université Jean Monnet 42023 Saint-Etienne, Cedex 2.

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1. Introduction

The origin of Au–U deposits in the Archaean Witwatersrand Basin of South Africa is the subject of longstanding discussion. Because of the close spatial association of pyrite with Au and U in the mineralised conglomerates, the origin of pyrite is a central aspect in all genetic hypotheses of Witwatersrand mineralisation, in both primary "placer" models (Robb and Meyer, 1995a,b; Frimmel, 2005), and post-sedimentary, "hydrothermal" models (Barnicoat et al., 1997; Phillips and Law, 2000). The two models have broad implications, given the constraint that the stability of detrital sulfides in a subaerial sedimentary environment would give on the composition of the Archaean atmosphere (Krupp et al., 1994; England et al., 2002a; Frimmel, 2005). Further, high concentrations of Au and U are in some cases associated with thin but laterally continuous layers of carbonaceous matter. Although the carbonaceous matter is ultimately of biogenic origin, as indicated by carbon isotope ratios, the process of its formation is unclear, either representing bitumen derived from migrated hydrocarbons (Spangenberg and Frimmel, 2001; England et al., 2002b) or microbial mat remains (Mossman et al., 2008). Other independent evidence for the former presence of life exists in the Witwatersrand basin, including microbially-induced sedimentary structures (Noffke et al., 2006), and possible sulfidised microfossils (Schidlowski, 1965).

In this paper, new evidence for the association of biological activity and the accumulation of gold in the Witwatersrand conglomerates is provided by the volumetrically minor, but relatively widespread concentrically laminated pyrite (CLP). This type of pyrite, reported in the Elsburg Reef, Basal Reef and, most commonly, in the Ventersdorp Contact Reef (Schweigart and von Rahden, 1965; Hallbauer, 1986; England et al., 2002a) has been previously described as "oolitic pyrite", a term with a certain genetic implication. It locally occurs in association with other types of round pyrite ("buckshot" pyrite), such as "mudball" pyrite (e.g. Hallbauer, 1986). Mudball pyrite, characterised by roughly globular shape and polygonal cracks on the surface, is common in Witwatersrand reefs and locally forms 2–10 mm thick-layers (Hallbauer, 1986; Barton and Hallbauer, 1996). Both types of pyrite are known to contain appreciable amounts of gold (up to several tens of ppm of Au; Hallbauer, 1986; Guy et al., 2010; Koglin et al., 2010; Agangi et al., 2013). Preservation of delicate textures of CLP suggests short-distance transport (Saager, 1970; Large et al., 2013). Thus, the study of this pyrite type can potentially give information on the conditions at, or near, the site of final deposition of the conglomerates.

The analysis of multiple S isotope ratios in pyrite has proven to be a powerful tool in the study of ancient rock successions, especially since it was discovered that sedimentary rocks older than c. 2.3 Ga show mass-independent S isotope fractionation, which is interpreted to be the result of photochemical reactions distinctively occurring in the early atmosphere (Thiemens, 1999; Farquhar et al., 2000; Farquhar and Wing, 2003; Whitehouse et al., 2005; Johnston, 2011). Further, recent advances in analytical techniques have demonstrated that Fe isotopes can be applied to Archaean and Phanerozoic iron suphides to discriminate biological and abiotic processes for their origin (Beard et al., 2003a; Yamaguchi et al., 2005; Whitehouse and Fedo, 2007). Finally, the combined study of Fe and multiple S isotopes is emerging as a promising approach in addressing these issues (Archer and Vance, 2006; Hofmann et al., 2009; Marin-Carbonne et al., 2014).

We have studied the chemical and Fe, S isotope compositions of CLP grains and other types of pyrite from the Ventersdorp Contact Reef (VCR), Witwatersrand Basin in order to gain information on their origin. Abundant elemental and some isotopic compositional data are available in the literature, which have shown that significant differences in trace element and isotopic composition exist between different conglomerates, and different generations of pyrite (e.g. Utter, 1978; Meyer et al., 1990; Barton and Hallbauer, 1996; Zhao et al., 2006; Guy et al., 2012; Large et al., 2013). However, most of the available chemical and isotopic analyses were made on whole grains. Because Witwatersrand pyrite is commonly zoned (Foya et al., 1999; Reimold et al., 2004; Agangi et al., 2013), these whole-grain analyses must be considered as "average compositions" of different zones. Inclusions of sulfide and silicate minerals can also severely affect the results. Bearing this in mind, we have chosen an in situ analytical approach, including ion and electron microprobes, and laser ablation ICP-MS, to be able to analyse small-scale textures,

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