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### Paragenesis and composition of ore minerals in the Randalls BIF-hosted gold deposits, Yilgarn Craton, Western Australia: Implications for the timing of deposit formation and constraints on gold sources

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#### ABSTRACT

The Randalls district comprises three individual gold deposits – Cock-Eyed Bob, Maxwells, and Santa-Craze – hosted in the same unit of banded iron-formation (BIF) in the southern Eastern Goldfields Superterrane, Yilgarn Craton, Western Australia. The iron formation is a silicate/oxide-facies unit with overprinting sulfides, and has undergone metamorphism (upper-greenschist facies) and deformation (two generations of folds). The gold deposits are hosted in two structural locations: hinge zones of anticlinal folds (e.g., Santa-Craze and Maxwells) and overturned, steeply dipping limbs of anticlinal folds (e.g., Cock-Eyed Bob). Gold dominantly occurs as inclusions of native gold and/or electrum within or around pyrrhotite, magnetite, and arsenopyrite.

The earliest mineral assemblage preserved in the banded iron formation at Randalls is banded magnetite–quartz. Magnetite in all forms has elevated Mg, Al, Ti, V, Mn, Cr, Zn, and W, but very low Ni compared to other BIFs globally. Pyrrhotite, the most abundant sulfide, is restricted to ore zones and occurs as laminae, "blebs", and veinlets, all of which have replaced magnetite. Some pyrrhotite in the hydrothermally altered BIFs (especially in near-surface samples) has been re-sulfidized to pyrite. Trace elements associated with pyrrhotite include Co, Ni, Ag, Sb, and Pb. Large, coarse-grained arsenopyrite overgrows the pyrrhotite-defined foliation; it is enriched in Co, Ni, Sb, and Te, but contains very little 'invisible' gold (<0.01 ppm Au), unlike arsenopyrite in many orogenic Au systems globally (commonly  $\geq$ 0.1 ppm Au).

Petrographic studies of several mineralized BIFs from Randalls reveal that pyrrhotite and arsenopyrite are associated with Fe(-Ca) amphibole, which grew during contact metamorphism caused by granite plutons. This spatial sulfide-amphibole relationship indicates that gold mineralization likely occurred during granite intrusion, consistent with prior studies on Randalls. The trace element characteristics of the sulfides show that they were not inherited from replaced magnetite. Further, these characteristics suggest that the fluid from which sulfides crystallized was not indigenous to the BIFs, and that this Srich fluid either contained S in abundance originally, or interacted with a lithology/lithologies that were rich in S before reaching the BIFs. Black shale containing diagenetic nodular pyrite occurs in the upper Black Flag Group, which unconformably underlies the Belches Supersequence at depth ( $\sim$ 3 km). The trace element composition of nodular pyrite from this unit fits well with the trace element characteristics of the BIF pyrrhotite and arsenopyrite, and the nodular pyrite contains an average of 0.5 ppm Au. However, the nodular pyrites have an average  $\delta^{34}$ S value of 5.6‰ (*n* = 13), which contrasts markedly with the  $\delta^{34}$ S average of pyrrhotite, pyrite and arsenopyrite in mineralized BIFs (0.8%, n = 17). Thus, a definitive source of gold, arsenic, silver, and tellurium in the Randalls system remains elusive, although we would contend that the upper Black Flag Group should be considered as a possible metal and S source in areas of the Yilgarn where it forms a thick substrate to ore camps.

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#### 1. Introduction

The majority of Precambrian gold deposits may be divided into two groups – Witwatersrand-type and orogenic (i.e., structurally

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controlled)-type – which, combined, account for much of Earth's accessible gold endowment. The orogenic category is further subdivided into sediment-hosted and greenstone (altered mafic volcanics)-hosted. A variant of the sediment-hosted subclass is the banded iron-formation (BIF)-hosted gold deposit group, members of which are commonly small and subeconomic. One exception to this general rule is the Homestake BIF deposit (South Dakota, USA), which contained over 40 million ounces (Moz) of gold (Caddey et al., 1991).

The Randalls orogenic (structurally controlled) Au district, Western Australia (~0.6 Moz total contained Au, Integra Mining Pty Ltd., 2011; Figs. 1 and 2), is one of the few gold camps globally where banded iron-formation (BIF) is the primary ore host. Other orogenic gold districts (both sediment- and greenstone-hosted) have BIF in the regional stratigraphic sequence but it is not the principal host to the ore (e.g., Sunrise Dam, Western Australia). The banded iron-formation at Randalls includes oxide (magnetite) and silicate (chlorite) facies, and a replacement sulfide 'facies' (pyrrhotite and arsenopyrite) is localized in or near the individual gold deposits. The latter lithology contains the majority of the gold budget.

BIF-hosted gold deposits, although small in size and number, have been the subject of much research and a protracted debate for nearly a century. For example, the origin of Homestake has been reviewed numerous times since the early 1900s (Paige, 1923; Gustafson, 1933; Noble, 1950; Rye and Rye, 1974; Caddey et al., 1991). Several papers published in the 1970s (e.g., Fripp, 1976; Anhaeusser, 1976; Hutchinson, 1976) argued for a syngenetic origin of all BIF-hosted gold deposits worldwide, drawing analogies between banded iron-formations and seafloor exhalites. The following decade marked a departure from this mode of thinking toward a metamorphic-hydrothermal, or 'epigenetic', formation model, in which the iron formations were simply efficient chemical traps for migrating metal- and H<sub>2</sub>S-rich fluids (Phillips et al., 1984; Groves et al., 1987). This model has remained the paradigm in academic and exploration literature.

Syngenesis has been revived and revised in recent years for certain sediment-hosted orogenic gold resources (e.g., Large et al., 2007, 2009, 2011; Thomas et al., 2011). In the deposits studied by these authors, gold and other associated trace elements (e.g., As, Te, Bi, Ag, and Pb) were found to be incorporated into synsedimentary/early-diagenetic pyrite in fine-grained, carbonaceous sediments (i.e., black shale). Later generations of pyrite, interpreted to be late diagenetic/early metamorphic, had ordersof-magnitude smaller concentrations of gold in the pyrite structure, but often contained inclusions of native gold and/or precious-metal tellurides. These textures were interpreted to reflect a multistage process of gold enrichment, from pre-concentration in the early pyrite, to release and upgrading in structurally favorable zones during metamorphism and/or deformation, facilitated by the pyrite-to-pyrrhotite conversion at deeper levels in the basin.

This paper discusses the syngenesis versus epigenesis origin of the Randalls deposits, but the main aim is to evaluate the source of metals and sulfur from the textures and chemistry of the oxide-sulfide assemblages of BIF within the ore envelope, using various techniques. Constraints on the depositional age of the sequence hosting the BIFs are also addressed, for comparison with previous detrital zircon geochronology studies that focused on the southern (stratigraphically lower) portion of the Belches Basin (e.g., Krapež et al., 2000). Sulfur isotope analyses on both BIF-hosted sulfides and pyritic black shale from the Lucky Bay gold prospect (~15 km west of the Randalls district; Steadman et al., 2013), are used to evaluate sulfur sources. The Lucky Bay area was chosen because it does not have the same metamorphic history as the Belches Basin; rather, the maximum P-T conditions there were somewhat lower (lower-middle greenschist facies; Goscombe et al., 2009) compared to the Belches Basin. Furthermore, Steadman et al. (2013) maintained that the Lucky Bay black shale horizon correlates with the Black Flag Group shale/greywacke sequence at the Golden Mile and St. Ives gold camps, which means it is slightly older (~10 Ma; Squire et al., 2010) than the Belches Supersequence. These authors also postulated that distal correlates of the Lucky Bay shale underlie the Belches basin at depth, in keeping with the tectonic reconstruction of Czarnota et al. (2010), providing a potential source of S, CO<sub>2</sub>, and metals to the Randalls deposits.

#### 2. Regional geology

Swager (1997), Krapež et al. (2008), Czarnota et al. (2010), and Pawley et al. (2012) provide detailed accounts of the geologic framework of the Eastern Goldfields Superterrane. A brief summary of their papers is given below.

The Eastern Goldfields Superterrane is an amalgamation of five ancient volcanic arcs (now referred to as terranes; Fig. 1) that were sutured together to form one giant superterrane (Eastern Goldfields) at about 2660–2650 Ma. The Superterrane has a protracted tectonic history involving multiple cycles of magmatism, uplift, erosion, and sedimentation, the products of which have largely escaped preservation or are concealed beneath younger (but still Archean) rocks. The exception is the package of rocks formed during the youngest cycle, which makes up the overwhelming majority of lithologic units (including the Black Flag Group (BFG)) exposed in the Eastern Goldfields.

The Black Flag Group is a thick (~4000 m; Krapež and Hand, 2008; Krapež and Pickard, 2010) sequence of volcaniclastic siltstones, sandstones, conglomerates, and shales (this latter lithology comprises  $\sim$ 15% of the overall Group; Krapež and Pickard, 2010) that overlie all other lithologies in the Eastern Goldfields, with the exception of the late-stage basins (see below). The BFG is separated into an Early and Late sequence (Squire et al., 2010). Maximum depositional age of the Early BFG, as determined by detrital zircon U–Pb geochronology, is ~2690 Ma (Squire et al., 2010; Krapež and Pickard, 2010). Late Black Flag deposition is assumed to have commenced later than dolerite intrusion in the Kalgoorlie and St. Ives areas (i.e., after 2680 Ma; Squire et al., 2010). Steadman et al. (2013) analyzed detrital zircon from coarse grained sandstone interbedded with carbonaceous and pyritic black shale at the Lucky Bay gold prospect (Fig. 1c), and obtained an age of  $2679 \pm 11 \text{ Ma}(2\sigma)$ , similar to that of the Late Black Flag Group and the Golden Mile/Condenser Dolerite.

The Belches Basin is one of several so-called 'Late-Stage' basins that formed after the cessation of volcanic activity in the Eastern Goldfields Superterrane ca. 2660 Ma, where it adjoins the Kurnalpi and Gindalbie Terranes (Fig. 1). Tectonostratigraphic reconstruction of the Eastern Goldfields Superterrane by Czarnota et al. (2010) shows that the Belches Supersequence (Krapež et al., 2008) most likely deposited on top of Black Flag Group rocks, which formed the floors of graben blocks during D<sub>3</sub> extension. The >3600 m-thick Belches Supersequence is dominated by sandstone, which grades into mudstone over intervals of varying thickness. Krapež et al. (2008) identified two successions of sandstone, separated by a horizon of banded iron formation (maximum thickness 130 m; Fig. 2b), which is the focus of this paper. Each succession is ~1500 m thick, and sedimentary structures are generally well-preserved.

The first geochronologic study of the Belches Basin was by Krapež et al. (2000), who collected a lower-succession sandstone sample from an outcrop on Mt. Belches,  $\sim 10 \text{ km}$  south of the Randalls district. Their SHRIMP U–Pb detrital zircon age of  $2664 \pm 4 \text{ Ma}$  represents a maximum depositional age for at least the lower succession of the Belches Supersequence. Later sequence stratigraphy by Krapež et al. (2008) suggested that, despite the differences in sedimentary facies between the upper and lower successions, and

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