7 Mesoarchaean Gold Mineralisation in the Barberton Greenstone Belt: A Review

Andrea Agangi, Axel Hofmann, Benjamin Eickmann, and Johanna Marin-Carbonne

Abstract

The Barberton Greenstone Belt hosts abundant structurally controlled gold mineralisation of Mesoarchaean age. More than 300 gold occurrences have been reported, although most of the gold production so far (>350 tonnes Au) has come from a handful of deposits located along the northern margin of the greenstone belt. Most deposits are hosted by greenschist-facies metasedimentary and metamafic rocks, with the notable exception of the amphibolite-facies rocks at New Consort mine. Mineralisation is associated with quartz–carbonate veins that truncate major compressional structures at the greenstone belt scale. The age of mineralisation is loosely constrained at circa 3080–3030 Ma, based on U–Pb dating of hydrothermal rutile and titanite. In greenschist-facies deposits, the ore assemblage is dominated by pyrite and arsenopyrite, which contain up to thousands of ppm of 'invisible' gold, Ni–As–Sb sulphides and native gold. At New Consort mine, mineralisation includes massive replacement-style ore and vein-hosted or disseminated types. Both structural studies in the field and microstructural observation point to a multistage ore deposition process, which is reflected in the re-activation of brittle to ductile structures and the overprinting of sulphide assemblages. The presence of mass-independently

B. Eickmann

J. Marin-Carbonne

Laboratoire Magmas et Volcans, UCA, IRD, CNRS, Université Lyon—UJM, Saint Étienne, France

Present Address:

J. Marin-Carbonne

Faculté des géosciences et de l'environnement, Institut des sciences de la Terre, University of Lausanne, Lausanne, Switzerland

fractionated S isotopes (Δ^{33} S = –0.6 to +1.0‰) in pyrite from Sheba and Fairview mines suggests that hydrothermal fluids mobilised S from volcanic and sedimentary rocks of the greenstone belt and places constraints on the origin of the Au itself.

Keywords

Archaean • Gold • Mineralisation • Multiple S isotopes • Barberton

7.1 Introduction

The Palaeoarchaean Barberton Greenstone Belt is host to some of the earliest known Au mineralisation (Anhaeusser [1976](#page-10-0), [1986;](#page-10-0) de Ronde et al. [1991;](#page-11-0) Dziggel et al. [2010](#page-11-0)). The mineralisation is structurally controlled and occurs in strongly deformed greenschist- to amphibolite-facies metamorphic rocks. Deciphering mechanisms and timing of mineralisation in the Barberton Greenstone Belt and other Archaean terrains is important for understanding Archaean tectonic and hydrothermal processes (Anhaeusser [1986;](#page-10-0) Kolb et al. [2015;](#page-12-0) Sahoo and Venkatesh [2015;](#page-13-0) Hazarika et al. [2017](#page-11-0); Mishra et al. [2017\)](#page-12-0).

Mineralisation in the Barberton Greenstone Belt has significant economic relevance and, since the discovery of gold in this area in 1882, more than 350 tonnes of gold have been extracted (Anhaeusser [1976;](#page-10-0) Dirks et al. [2009;](#page-11-0) Pearton and Viljoen [2017\)](#page-12-0). In plan view, the distribution of the main deposits reveals a heterogeneous distribution of gold mineralisation. Despite the fact that more than 300 deposits and prospects are known in the greenstone belt, the bulk of the gold production comes from a handful of mines, namely Fairview, Sheba, New Consort, Agnes and Princeton mines, all located in the northern portion of the greenstone belt near the contact between the Barberton Greenstone Belt and the Kaap Valley and Stentor plutons (Anhaeusser [1976;](#page-10-0) Ward [1995](#page-13-0), [1999;](#page-13-0) Dirks et al. [2009](#page-11-0)) (Fig. [7.1](#page-2-0)). The fluids

A. Agangi (\boxtimes) · A. Hofmann Department of Geology, University of Johannesburg, Johannesburg, South Africa e-mail: aagangi@uj.ac.za

Isotopengeochemie, Universität Tübingen, Tübingen, Germany

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responsible for mineralisation are believed to have been low-salinity and H_2O-CO_2 -rich (de Ronde et al. [1992\)](#page-11-0) and, similar to other orogenic gold deposits, Au is interpreted to have been transported as complexes of reduced S such as Au $(HS)_2^-$ and AuHS (Pokrovski et al. [2014](#page-12-0)). Structural and microtextural evidence convincingly points at multiphase processes of mineralisation (Agangi et al. [2014](#page-10-0); Munyai et al. [2011](#page-12-0); Dziggel and Kisters [2019\)](#page-11-0).

Some aspects of the genesis of this mineralisation are still controversial, and some of the open questions include the origin of H_2O -CO₂ mineralising fluids and, by inference, the origin of Au itself. Possible sources of fluids and Au include supracrustal lithologies (volcanic and sedimentary rocks), felsic intrusions, and the mantle. The mineralisation is believed to have occurred between circa 3080 and 3030 Ma, based on rutile and titanite U-Pb ages, but precise timing of mineral deposition is also uncertain, given the scarcity of available data (de Ronde et al. [1992](#page-11-0); Dziggel et al. [2010](#page-11-0); Dirks et al. [2013\)](#page-11-0).

Several reviews have dealt with gold mineralisation in the Barberton Greenstone Belt and have mostly focussed on the meso- to mega-scale structural and metamorphic aspects of various mines (Anhaeusser [1986;](#page-10-0) Dziggel et al. [2007](#page-11-0); Pearton and Viljoen [2017;](#page-12-0) Dziggel and Kisters [2019\)](#page-11-0). Here, we review some of the main characteristics of gold deposits in the Barberton Greenstone Belt with an emphasis on geochemical and microstructural characteristics. We refer to the mentioned papers for more complete information on the control of structures and deformation on mineralisation. We distinguish mineralisation hosted in mostly greenschistfacies rocks from mineralisation hosted in amphibolite-facies rocks. We then evaluate different models of genetic mechanisms of mineralisation.

7.2 Geological Setting of the Barberton Greenstone Belt

The Palaeoarchaean Barberton Greenstone Belt is situated in the eastern part of the Kaapvaal craton, southern Africa (Fig. [7.1\)](#page-2-0). Its ca. 3550–3220 Ma volcano-sedimentary succession, which forms the Barberton Supergroup (formerly Swaziland Supergroup), is preserved in a southwest-northeast-trending belt surrounded by granitoid rocks belonging to the trondhjemite-tonalite-granodiorite (TTG) and granite-granodiorite-syenogranite (GMS) series (Viljoen and Viljoen [1969](#page-13-0); Lowe and Byerly [2007](#page-12-0); de Wit et al. [2019\)](#page-13-0). The supracrustal succession has been subdivided into three main lithostratigraphic units: the Onverwacht Group, the Fig Tree Group, and the Moodies Group, in ascending order (SACS [1980](#page-13-0)).

The Inyoka–Saddleback Fault System, a southwest to northeast-trending structure interpreted by some as a suture

zone, separates a northern and a southern terrane of different age and geochemical characteristics (Kamo and Davis [1994;](#page-12-0) Kisters et al. [2003](#page-12-0); Lowe and Byerly [2007](#page-12-0)). The Onverwacht Group consists mostly of komatiite, komatiitic basalt and basalt, with minor felsic volcanic rocks, and has been dated at circa 3550–3300 Ma in the southern terrane (Kröner et al. [1996](#page-12-0), [2016](#page-12-0)). North of the Inyoka-Saddleback fault, the Onverwacht Group is composed of the Weltevreden Formation, which contains mafic-ultramafic volcanic rocks and numerous layered ultramafic complexes (Anhaeusser [2001;](#page-11-0) Stiegler et al. [2012](#page-13-0)). The Fig Tree Group is a largely marine, northwards deepening succession dominated by turbiditic greywackes, shales, banded iron formation and cherts (Lowe [1999](#page-12-0); Hofmann [2005\)](#page-12-0). The Moodies Group is a shallow marine to continental succession characterised by coarse-grained clastic sedimentary rocks, mainly sandstones and conglomerates, and only minor shale with a minimum depositional age of 3219 Ma (SACS [1980;](#page-13-0) Heubeck et al. [2013](#page-12-0), [2016;](#page-12-0) Drabon et al. [2017](#page-11-0)). The metamorphic grade of the Barberton Greenstone Belt is generally low but increases towards the contacts with the surrounding gneiss domes (Dziggel et al. [2002](#page-11-0), [2005](#page-11-0); Diener et al. [2005\)](#page-11-0). The structure of the greenstone belt is rather complex and is dominated by steeply plunging synforms separated either by thrust faults or narrow anticlines (Lowe et al. [2012\)](#page-12-0).

The tectonic evolution of the Barberton Greenstone Belt is complex, and the multiple phases of deformation affecting these rocks have been described elsewhere (de Ronde and de Wit [1994](#page-11-0); Kamo and Davis [1994](#page-12-0); Lowe and Byerly [1999;](#page-12-0) de Ronde and Kamo [2000;](#page-11-0) Lana et al. [2010\)](#page-12-0). According to de Ronde and de Wit [\(1994](#page-11-0)) and de Ronde and Kamo ([2000\)](#page-11-0), four main tectono-metamorphic events affected the greenstone belt. The first of these events (D1) remains to some extent enigmatic. D1 occurred between circa 3445 and 3416 Ma and was restricted to Onverwacht Group rocks in the southern part of the greenstone belt (de Ronde and de Wit [1994\)](#page-11-0). D1 was coeval with the intrusion of TTGs along the southern margin of the greenstone belt and with an early phase of low-pressure amphibolite-facies metamorphism, with estimated peak conditions of \sim 550 °C and 4.5 kbar (Cutts et al. [2014](#page-11-0)).

The 3229–3227 Ma D2 event was responsible for the main regional strain and affected the entire greenstone belt (de Ronde and Kamo [2000](#page-11-0); Schoene et al. [2008](#page-13-0)). Event D2 coincided with emplacement of TTG intrusions such as the 3227 Ma Kaap Valley Tonalite, the 3290–3230 Ma Badplaas pluton and the 3236 Ma Nelshoogte pluton (Kamo and Davis [1994](#page-12-0); Kisters et al. [2010;](#page-12-0) Matsumura [2014\)](#page-12-0). During this event, the rocks of the lowermost units of the Onverwacht Group, the Theespruit and Sandspruit Formations, experienced high-pressure amphibolite-facies metamorphism (Dziggel et al. [2002](#page-11-0), [2005](#page-11-0); Diener et al. [2005;](#page-11-0) Moyen et al. [2006\)](#page-12-0). D2 marked the switch from sedimentation of the

Fig. 7.1 Geological map of the Barberton Greenstone Belt and distribution of the main gold deposits (modified from de Ronde et al. [1992\)](#page-11-0). Dashed line indicates position of Fig. [7.2](#page-4-0)

3260–3230 Ma Fig Tree Group in a relatively deep marine environment to the \sim 3225 Ma Moodies Group in a shallow marine to continental environment (de Ronde and de Wit [1994;](#page-11-0) Kamo and Davis [1994;](#page-12-0) de Ronde and Kamo [2000](#page-11-0); Kisters et al. [2010;](#page-12-0) Heubeck et al. [2013](#page-12-0)). The structures associated with D2 are truncated by potassic intrusions such as the 3203 Ma Dalmein pluton in the southern part (Lana et al. [2010](#page-12-0)), which places constraints on the end of this phase of deformation. The tectonic circumstances that gave rise to the D2 event are controversial and directly mirror the controversy relating to the nature of Archaean tectonics and the origin of TTG magmas. Some authors interpreted D2 as the consequence of compression, subduction and accretion of the southern terrane along the Inyoka-Saddleback Fault during northwards-directed subduction (Moyen et al. [2006](#page-12-0); Kisters et al. [2010\)](#page-12-0). In this interpretation, the syntectonic granitoids to the north of the greenstone belt represent the roots of a volcanic arc. D2 was synchronous with, or immediately followed by, a period of syn-orogenic extension and solid-state doming that eventually resulted in the steepening of fabrics during the orogenic collapse of the belt (Kisters et al. [2003\)](#page-12-0). Others interpret this deformation as a result of crustal overturn due to density inversions between buoyant (partially molten) TTG intrusions and cooler, denser greenstones (Anhaeusser [1984](#page-10-0); Van Kranendonk et al. [2009\)](#page-13-0). These authors argue that the relatively high-pressure conditions and the low temperature/pressure gradients can be achieved during sagging of the supracrustal succession.

The later, post-D2 events are less clear as geochronological constraints are scarce, and deformation gave rise to reactivation of earlier shear zones. The D3 tectonic event appears to have been related to NW-SE directed compression and orogen-parallel stretching accommodated by strike-slip shear zones (de Ronde and de Wit [1994\)](#page-11-0). D3 structural elements thus parallel those of D2 (de Ronde and de Wit [1994\)](#page-11-0). The timing of D3 has been constrained at <3126 Ma (de Ronde and Kamo [2000](#page-11-0)), although much looser time constraints (circa 3220–3080 Ma) were inferred for this event by other authors (de Ronde and de Wit [1994](#page-11-0); Kamo and Davis [1994;](#page-12-0) Schoene et al. [2008](#page-13-0)). The D4 tectonic event was characterised by a switch to transtensional deformation (de Ronde and de Wit [1994\)](#page-11-0). Geochronological constraints on this event are few and suggest that D4 occurred at around \sim 3080 Ma, based on U-Pb ages on hydrothermal rutile from Fairview mine (de Ronde et al. [1991](#page-11-0); de Ronde and de Wit [1994](#page-11-0)).

Gold mineralisation is interpreted to be associated with D3 and/or D4 deformation stages. The age of \sim 3080 Ma (de Ronde et al. [1991\)](#page-11-0), estimated on rutile in an altered, although not mineralised, sample offers a good indication of the timing of hydrothermal alteration at Fairview mine. However, much younger ages (3027 Ma) have been obtained from titanite associated with sulphide mineralisation at New Consort mine (Dziggel et al. [2010](#page-11-0)), and a maximum age of \sim 3015 Ma was estimated based on the zircon age of a felsic dyke interpreted to be syn- minerali-sation at Fairview mine (Dirks et al. [2013\)](#page-11-0).

7.3 Greenschist-Facies Gold Deposits

Gold deposits associated with host rocks at greenschistfacies grade are the most abundant in the Barberton Greenstone Belt and include the well-studied Sheba and Fairview mines. The greenschist-facies deposits all share distinctive alteration characteristics and structural style. Mineralisation is spatially associated with major D2 compressional structures (Figs. [7.1](#page-2-0) and [7.2](#page-4-0)), although mineralising fluids are interpreted to have moved along later, extensional faults that crosscut D2 compressional structures (de Ronde et al. [1991](#page-11-0); Dirks et al. [2009](#page-11-0), [2013\)](#page-11-0).

Gold mineralisation is hosted by different lithologies, ranging from metamafic–ultramafic rocks of the uppermost Onverwacht Group to metasedimentary rocks (greywacke, shale) of the Fig Tree and Moodies Groups. The Fairview and Sheba mines are located largely south of the Sheba Fault, although both mines exploit ore bodies on the northern side of the fault as well. The Sheba Fault has been interpreted as a north-west verging thrust fault that structurally superposed Fig Tree Group (meta)greywacke and shale of the Ulundi syncline onto Moodies Group arenites of the Eureka syncline (Anhaeusser [1976](#page-10-0); Ward [1999;](#page-13-0) de Ronde and Kamo [2000](#page-11-0)) (Fig. [7.2](#page-4-0)). Minor metamafic rocks of the Onverwacht Group are preserved between the Ulundi and Eureka synclines as tight antiformal structures made of talc-carbonate schist of the Zwartkoppie Formation¹ (Schouwstra and De Villiers [1998;](#page-13-0) Schouwstra [1995](#page-13-0)).

Mineralisation occurs as auriferous quartz-carbonate \pm rutile veins with associated semi-massive replacement sulphide bodies developing as part of wall-rock alteration (Schouwstra [1995\)](#page-13-0). Alteration zones associated with mineralisation in mafic lithologies include, from distal to proximal, talc-carbonate, quartz-carbonate, fuchsite-quartzcarbonate \pm sulphides, and fuchsite-quartz-sulphides \pm carbonate (Schouwstra [1995](#page-13-0)). Sericite is present instead of fuchsite as the K-mica in the neighbouring greywacke and shale. Mineralised fractures in the Sheba–Fairview area extend for tens of metres, trending northeast to eastnortheast, and are arranged in hundreds of metre-scale fracture zones (Dirks et al. [2009](#page-11-0)). In microtextural observations, veins directly associated with mineralisation appear as dilatant, mostly brittle structures, as demonstrated by euhedral prismatic quartz growing from the walls inwards (Agangi et al. [2014](#page-10-0)) (Fig. [7.3](#page-5-0)a).

The mineralisation occurs within or around quartz-carbonate veins that truncate the foliation (Fig. [7.3](#page-5-0)b, c). The mineralisation hosted in metapelite and greywacke at Fairview mine is dominated by arsenopyrite and pyrite, with

minor chalcopyrite, gersdorffite, galena and Sb-sulphides. Mineralisation hosted in metamafic–ultramafic rocks at Sheba mine is typically dominated by pyrite and also contains As–Ni-sulphides, chalcopyrite, sphalerite and minor galena (Fig. [7.3d](#page-5-0)). Gold occurs in a variety of forms, as 'invisible' (or refractory) gold in sulphides, as free gold grains in quartz-carbonate veins, and as sulphide-hosted inclusions of native gold (Cabri et al. [1989](#page-11-0); de Ronde et al. [1992](#page-11-0)).

At Sheba and Fairview mines, gold is mostly hosted in arsenopyrite and pyrite, both as micro-inclusions (Fig. [7.3e](#page-5-0)) and as finely dispersed, sub-microscopic Au (refractory gold). The concentrations and distribution of invisible Au and other trace elements in sulphides have been studied by different methods, including electron microprobe (EPMA), secondary ion mass spectrometry (SIMS) and protoninduced X-ray emission (PIXE) (Cabri et al. [1989;](#page-11-0) Agangi et al. [2014;](#page-10-0) Altigani et al. [2016\)](#page-10-0). These studies found that Au concentrations are extremely heterogeneous and vary from <300 ppm (the EPMA detection limit in Cabri et al. [1989](#page-11-0) study) to 4400 ppm Au even in a single arsenopyrite grain from Sheba mine, and up to 1020 ppm have been measured in pyrite at Sheba mine. In the Au versus As diagram (Fig. [7.4\)](#page-6-0), pyrite compositions from mineralised samples at Sheba and Fairview mines plot both above and below the Au-saturation line of Reich et al. ([2005](#page-13-0)). This is compatible with the presence of Au in different oxidation states and crystallographic positions, namely Au⁺ included in the crystal lattice and Au^0 in gold inclusions. Mapping of trace element distributions at the micrometre scale has highlighted complex intragranular textures that indicate multiple phases of growth, resorption and recrystallisation of sulphide minerals (Fig. [7.5;](#page-7-0) Cabri et al. [1989](#page-11-0); Agangi et al. [2014](#page-10-0), [2015](#page-10-0)).

The largely refractory nature of gold (below the zone of surface oxidation) in these deposits requires the sulphide ore minerals to be oxidised during the process of extraction in order to liberate the gold. In the past, this was achieved by roasting, with consequent production of large amounts of SO2 gas and volatile As oxides, a process that posed a considerable environmental hazard. Oxidation is now carried out through biogenic means using sulphide-oxidising bacteria, such as Thiobacillus ferrooxidans (Barberton Mines [2010](#page-11-0)).

7.4 Amphibolite-Facies Gold Deposits—The New Consort Mine

The New Consort mine (Figs. [7.1](#page-2-0) and [7.2](#page-4-0)) represents an exception in the Barberton Greenstone Belt in being one of the few gold deposits hosted in amphibolite-facies rocks. It is located north of Sheba and Fairview mines and occurs in

¹The Zwartkoppie Formation is not recognised by SACS and its use is informal.

Fig. 7.2 a Map of the Eureka and Ulundi synclines and location of gold deposits (modified from Dirks et al. [2009](#page-11-0)), b cross sections of Fairview and Sheba mines (modified from Barberton gold mines [2014\)](#page-11-0)

the hanging wall of the contact between greenstone lithologies and the Stentor Pluton. Mineralisation is hosted by upper greenschist- to upper amphibolite-facies rocks and is associated with the Consort Bar, a mylonitic shear zone separating (ultra)mafic rocks and chert of the Onverwacht Group from clastic sedimentary rocks of the Fig Tree Group (Otto et al. [2007;](#page-12-0) Munyai et al. [2011](#page-12-0)). Mineralisation is found in three bodies, the Seven Shaft Shoot, Isaura Shoot, and Prince Consort Shoot. The style of mineralisation in underground workings includes massive replacement-style ore and vein-hosted or disseminated mineralisation (Otto et al. [2007](#page-12-0)).

Two phases of mineralisation have been identified: the early phase in the footwall, characterised by löellingite-pyrrhotite ore assemblages and a high-temperature alteration calc-silicate assemblage composed of garnet, clinopyroxene, hornblende, K-feldspar, quartz, calcite and biotite (Dziggel et al. [2007](#page-11-0); Otto et al. [2007](#page-12-0); Dziggel and Kisters [2019](#page-11-0)). Pressure-temperature estimates for first-phase alteration assemblages in metamafic and intercalated metasedimentary rocks are circa 600–700 °C and 6–8 kbar (Otto et al. [2007\)](#page-12-0).

The second, and main, phase of mineralisation has been attributed to oblique shear zones crosscutting the Consort Bar and was associated with the development of Cr-muscovite-K-feldspar-plagioclase-quartz or Cr-muscovite-tourmalineplagioclase-rutile as typical alteration assemblages proximal to ore. This second mineralisation event occurred at temperatures ranging from 520 to 600 °C and pressures between 1 and 3 kbar; the ore assemblages vary from arsenopyritepyrrhotite to arsenopyrite-pyrrhotite-chalcopyrite-löellingite with increasing depth and temperature (Dziggel et al. [2006,](#page-11-0) [2010](#page-11-0); Otto et al. [2007](#page-12-0)). Shearing was accompanied by emplacement of pegmatite that has been dated at

Fig. 7.3 Ore microtextures of samples from Sheba and Fairview mines. a Quartz-carbonate vein in chert, Fairview mine. Pyrite and arsenopyrite are present in the host rock. Note quartz crystals growing perpendicular to the vein walls, indicating dilatant behaviour of the cracks. b, c Carbonate-quartz veins in metagreywacke at Fairview mine. Arsenopyrite and pyrite are visible in both the vein and the alteration halo. Foliation is indicated by dashed lines. d Pyrite,

 3040 ± 84 Ma (Rb–Sr whole-rock; Harris et al. [1995](#page-11-0)). Overall, textural relations and thermometric estimates indicate a clock-wise P-T path with a near-isobaric decompression to 520–600 °C and 1–3 kbar followed by near-isobaric cooling (Dziggel et al. [2006](#page-11-0)).

Munyai et al. [\(2011](#page-12-0)) and Dirks et al. ([2013\)](#page-11-0) focussed their observations on gold-associated fractures on surface exposure at, and in the vicinity of, New Consort mine. In

sphalerite and chalcopyrite in metamafic schist at Sheba mine. e Pyrite-arsenopyrite ore with micro-inclusions of native gold and chalcopyrite, Fairview mine. All images transmitted polarised light, except for a and c (crossed polarisers in lower half), d (reflected light in right-hand half) and e (reflected light). Abbreviations: Asp arsenopyrite, cb carbonate, Ccp chalcopyrite, Ms K-mica, Py pyrite, Qtz quartz, Sp sphalerite

stark contrast with what has been described earlier, they described gold mineralisation associated with lowtemperature alteration assemblages and brittle deformation that developed during late-stage extensional tectonics. Thus, mineralisation at New Consort mine and surrounding areas is multiphase and may have formed at very different temperature conditions at different times (Dziggel and Kisters [2019](#page-11-0)).

Fig. 7.4 Plot of Au versus As content of pyrite and arsenopyrite grains from Fairview and Sheba mines (data from Agangi et al. [2014](#page-10-0) and Cabri et al. [1989](#page-11-0)). Gold saturation line in arsenian pyrite from Reich et al. ([2005](#page-13-0))

7.5 Fluid Inclusion and Mineral Stable Isotope Compositions

Fluid inclusion studies from the major deposits indicate low-salinity (NaCl eq = $5-6$ wt%), H₂O-CO₂-rich fluids, and homogenisation temperatures in the T = 290 to 310 $^{\circ}$ C range (de Ronde et al. [1992\)](#page-11-0). As-in-arsenopyrite geothermometry at Fairview mine indicates temperatures between $\langle 300 \rangle$ and $400 \degree$ C (Agangi et al. [2014\)](#page-10-0). These temperature estimates are in broad agreement with the greenschist-facies alteration assemblages. Based on O, H and C isotope analyses of mineralisation-related quartz and carbonate, the ore fluid would have had narrow ranges of δ^{18} O (+4.7 to + 5.8‰), δ^{13} C (−4.5 to −2‰) and δD (−35 to −41‰), recalculated based on fluid-mineral equilibria at 300 °C (de Ronde et al. [1992](#page-11-0)). Hydrothermal sulphides have slightly positive δ^{34} S values (+1.2 to +3.9‰) for pyrite and arsenopyrite (Kakegawa and Ohmoto [1999\)](#page-12-0). Overall, the fluid inclusion compositions and mineral stable isotope values indicate distinct homogeneity of mineralising fluids at the greenstone belt scale.

Despite the complexity of sulphide textures and trace element compositions, δ^{34} S values are confined to a narrow range of moderately positive values (de Ronde et al. [1992](#page-11-0); Kakegawa and Ohmoto [1999](#page-12-0)) which limits the use of S isotopes as tracers of the fluid source. As a further complication, $\delta^{34}S$ values are affected by a complex range of

processes during S transport and precipitation and associated redox reactions that can have overlapping effects. However, recent developments in S isotope studies have revealed that sulphate and sulphide minerals in the Archaean and early Palaeoproterozoic bear mass-independently fractionated S isotopes (MIF-S, indicated by the notation $\Delta^{33}S = \delta^{33}S$ $-0.515 \delta^{34}$ S) (Farquhar et al. [2000;](#page-11-0) Ono et al. [2003](#page-12-0); Johnston [2011\)](#page-12-0). MIF-S signals are interpreted to be the footprint of photolytic reactions caused by UV irradiation of S gases (such as SO_2) in an O_2 -free Archaean atmosphere, which would produce sulphide with $\Delta^{33}S > 0$ and sulphate with Δ^{33} S < 0. Once produced, MIF-S is very robust and only slightly affected by most mass-dependent fractionationinducing abiogenic and biogenic processes so that it can be used as a tracer of S processed in the atmosphere.

In situ multiple S isotope analyses of pyrite obtained by ion microprobe (SIMS) from samples of Fairview and Sheba mines revealed MIF-S with Δ^{33} S deviating significantly towards both positive and negative values (Δ^{33} S = -0.6 to +1.0‰) (Agangi et al. [2016](#page-10-0)). In the Δ^{33} S versus δ^{34} S plot, these values match the distribution peak of S compositions of pyrite and whole-rock analyses of the entire volcano-sedimentary succession and suggest derivation of S from the Barberton Supergroup (Fig. [7.6](#page-8-0)). Given the variability in Δ^{33} S, S may have been scavenged from different rock types and then been transported by hydrothermal fluids responsible for Au mineralisation. More specifically, negative Δ^{33} S values have been measured in sulphides from volcanic-hosted massive sulphide deposits (VHMS) of the Bien Venue deposit of the Fig Tree Group and hydrothermally altered mafic and ultramafic rocks of the Komati Formation, Onverwacht Group (Montinaro et al. [2015\)](#page-12-0). These rocks are interpreted to have derived their S isotopic signature from interaction with heated seawater and to have inherited the typically negative Δ^{33} S values of Archaean seawater sulphate (Bao et al. [2007;](#page-11-0) Montinaro et al. [2015\)](#page-12-0). They are thus possible sources of some S (and Au) for the sulphides at Sheba and Fairview mines. Positive Δ^{33} S and δ^{34} S values were measured in pyrite from chert, conglomerate and dolomite from the Onverwacht and Fig Tree Groups, as well as from bulk shales from the Fig Tree Group (Grosch and McLoughlin [2013;](#page-11-0) Roerdink et al. [2013;](#page-13-0) Montinaro et al. [2015\)](#page-12-0). In particular, some shale samples and pyrite from barite-free samples show a steep negative $\Delta^{33}S/\delta^{34}S$ slope that resembles the distribution of pyrite analyses at Sheba-Fairview. Irrespective of the origin of this negative trend, which may result from mixing of different S components or represent a primary photolytic signal (Roerdink et al. [2013](#page-13-0)), the similarity to the values observed for hydrothermal pyrite at Sheba-Fairview mines implies that similar rocks may be a good source of S in the Au deposits (Fig. [7.6](#page-8-0)b).

7.6 Discussion and Conclusions

7.6.1 Sources of S, Fluids, and Gold: Internal or External to the Greenstone Belt?

The origin of mineralising fluids and Au in Archaean greenstone belts has proved to be elusive, and the various hypotheses proposed reflect the difficulty in the identification of such sources in structurally-controlled Au deposits, both in Precambrian and Phanerozoic ages (Hronsky et al. [2012](#page-12-0); Gaboury [2013](#page-11-0); Goldfarb and Groves [2015\)](#page-11-0). Possible sources of sulphur in Archaean orogenic gold deposits include supracrustal rocks (Groves et al. [2003](#page-11-0); Phillips and Powell [2009;](#page-12-0) Tomkins [2013\)](#page-13-0), hydrothermal systems related to felsic magmatism (Salier et al. [2005](#page-13-0); Doublier et al. [2014](#page-11-0)), the lower crust, and the mantle (Hronsky et al. [2012](#page-12-0); Fu and Touret [2014\)](#page-11-0).

The hypothesis of a magmatic origin for mineralising fluids has been proposed in several cases of Archaean structurally-controlled Au deposits, such as in Neoarchaean Au deposits of Western Australia, based on the presence of coeval magmatism (Wang et al. [1993;](#page-13-0) Doublier et al. [2014\)](#page-11-0), trace element signature of accessory minerals (Bath et al. [2013](#page-11-0)), and Pb and noble gas isotope studies (Qiu and McNaughton [1999](#page-13-0); Kendrick et al. [2011\)](#page-12-0). A causal relationship between plutonism and Au mineralisation in the Barberton Greenstone Belt has been proposed mostly based on spatial distribution (Anhaeusser [1976,](#page-10-0) [1986](#page-10-0)). In the Barberton Greenstone Belt, mineralisation post-dates the main phase of potassic granite plutonism, which caused the emplacement of extensive batholiths, such as the

Fig. 7.6 Comparison of S isotope analyses of pyrite associated with gold mineralisation at Sheba and Fairview mines, and pyrite and barite from different units of the Barberton Greenstone Belt.
a Δ^{33} S versus δ^{34} S plot of pyrite and barite from different units of the Barberton Greenstone Belt $(n = 1680)$ and density distribution of pyrite data. **b** Δ^{33} S versus δ^{34} S plot of pyrite from Au-mineralised samples from Sheba and Fairview mines and some representative rocks of the Barberton Greenstone Belt. c Histogram of Δ^{33} S of pyrite from Sheba and Fairview mines compared with Barberton Greenstone Belt pyrite and barite. Data from: (1) Philippot et al. ([2012\)](#page-12-0), (2) Montinaro et al. ([2015\)](#page-12-0), (3) Roerdink et al. [\(2012\)](#page-13-0), (4) Bao et al. [\(2007](#page-11-0)), (5) Roerdink et al. [\(2013](#page-13-0)), (6) Grosch and McLoughlin ([2013\)](#page-11-0), (7) Agangi et al. ([2016](#page-10-0))

3160–3090 Ma Mpuluzi Batholith (Kamo and Davis [1994](#page-12-0); Murphy [2015\)](#page-12-0) and the circa 3067 Ma Stentor pluton (Kamo and Davis [1994\)](#page-12-0), and was accompanied by emplacement of small granitic dykes (porphyries) observed in different gold mines. However, the role of these dykes in the mineralising process is not clear. A magmatic source is expected to have $0 \pm 0.2\%$ Δ^{33} S (Farquhar et al. [2000\)](#page-11-0) so that the MIF-S measured at Sheba and Fairview mines requires the involvement of atmospherically processed S (Fig. [7.6b](#page-8-0)).

A mantle derivation of $CO₂$ -bearing mineralising fluids has been proposed in recent models that aimed at linking the presence of various types of gold deposits with the presence of ''fertile' metasomatised lithospheric mantle (Hronsky et al. [2012;](#page-12-0) Fu and Touret [2014](#page-11-0)). This is particularly applicable in the presence of mafic mantle magmas coeval with mineralisation (de Boorder [2012\)](#page-11-0). However, in the Barberton Greenstone Belt magmas coeval with mineralisation are felsic, and the marked MIF-S also argues against such a deep origin of fluids.

Homogeneity of fluid inclusion salinity and elemental compositions (H_2O, CO_2, CH_4) , stable isotope compositions of minerals and temperature estimates (in greenschist-facies deposits) suggest a homogeneous source of mineralising fluids at the greenstone belt scale. This source has been identified as external to the Barberton Greenstone Belt (de Ronde et al. [1992](#page-11-0)). This homogeneity of major element and isotope compositions does not imply constant content of metals, as indicated by zoning of sulphide minerals (Fig. [7.5\)](#page-7-0). Otto et al. ([2007\)](#page-12-0) estimated that mineralisation at New Consort mine occurred at peak conditions in the Fig Tree sedimentary rocks forming the hanging wall of the Consort Bar, whereas the Onverwacht Group rocks had already experienced metamorphic peak and were on their retrograde path. They concluded that, as a consequence, the host rocks are unlikely sources of fluids (also see Dziggel and Kisters [2019\)](#page-11-0).

In apparent contrast, multiple S isotope analyses with non-zero MIF-S values imply an atmospheric source, and heterogeneous Δ^{33} S values suggest that a variety of rocks with different Δ^{33} S acted as S sources for mineralising fluids. Similar results have been obtained from multiple S isotope analyses of pyrite and pyrrhotite of various Neoarchaean gold deposits of the Yilgarn Craton of Western Australia (Selvaraja et al. [2017\)](#page-13-0). These authors underlined the importance of shales as sources of gold in Archaean structurally controlled gold deposits. A sedimentary rather than igneous source for Barberton gold would be in agreement with low Au contents of komatiites from the Barberton Supergroup (Hofmann et al. [2017\)](#page-12-0).

The microscale heterogeneity of Δ^{33} S values implies that the ore-forming fluid was isotopically heterogeneous, and variable MIF-S values were not completely homogenised during hydrothermal fluid flow. The isotopic heterogeneity is

matched by trace element heterogeneity, which appears as growth zones, recrystallisation domains and veinlets, a characteristic also observed in several other orogenic gold deposits of Archaean and Palaeoproterozoic age (Morey et al. [2008;](#page-12-0) Fougerouse et al. [2016\)](#page-11-0). This evidence is compatible with a pulsating fluid flow rather than a single large event and suggests that single fluid pulses had a very localised effect in terms of ore deposition. This is similar to what is observed in orogenic gold deposits and typically attributed to periodic fluid pressure build-up and earthquake-related release, also known as fault-valve mechanisms (Sibson et al. [1988](#page-13-0)).

7.6.2 Timing of Gold Mineralisation in the Barberton Greenstone Belt and Tectonic Context

The age of mineralisation in the Barberton Greenstone Belt is still rather poorly constrained, namely because of a dearth of available data (Fig. [7.7\)](#page-10-0). At Fairview and Sheba mines, porphyry dykes crosscutting mineralisation give good estimates of minimum mineralisation ages. One of these dykes was dated at 3126 Ma (U-Pb on zircon; de Ronde et al. [1991](#page-11-0)). A rutile separate extracted from a pre-mineralisation, altered dyke at Fairview was dated at 3084 Ma (de Ronde et al. [1992\)](#page-11-0), which may be a good estimate for mineralisation. However, recent U-Pb zircon dating of two mineralised dykes at Golden Quarry, near Sheba mine, has been interpreted to indicate mineralisation at circa 3015 Ma (Dirks et al. [2013\)](#page-11-0). At New Consort mine, a 3027 ± 7 Ma U-Pb age on titanite (Dziggel et al. [2006\)](#page-11-0) associated with main-stage mineralisation and a whole-rock Rb-Sr age of 3040 ± 84 Ma on a syn-mineralisation dyke (Harris et al. [1995](#page-11-0)) may indicate that mineralisation was caused by a protracted or episodic tectonic activity that lasted for several tens of million years. The multiphase nature of mineralisation at New Consort mine seems to support such a conclusion (Dziggel et al. [2007,](#page-11-0) [2010;](#page-11-0) Dziggel and Kisters [2019\)](#page-11-0).

The tectonic regime in operation in the Kaapvaal craton at the time of Au mineralisation are inferred from a fragmentary record, and a complete picture has still to emerge. At the craton scale, mineralisation occurred shortly after the final consolidation of the Kaapvaal craton and at the beginning of an extension phase that led to bimodal volcanism at circa 3074 Ma (Dominion Group, Armstrong et al. [1991\)](#page-11-0) in the central part of the craton and at 2980 Ma to the southeast (Nsuze Group, Hegner et al. [1984](#page-12-0)).

In the eastern part of the craton, felsic magmatism is indicated by emplacement of the Sinceni granite in Swaziland at circa 3070 Ma (Maphalala and Kröner [1993\)](#page-12-0). A continent-wide felsic magmatic event at circa 3070 Ma is indicated by the abundance of detrital zircons of this age in

Fig. 7.7 Age constraints on Au mineralisation in the Barberton Greenstone Belt

the Witwatersrand and Pongola Supergroups (Kositcin and Krapez [2004;](#page-12-0) Wilson and Zeh [2019](#page-13-0)). At the same time in the north of the Kaapvaal craton, volcanic rocks and sedimentary rocks of the Murchison Greenstone Belt, such as the Weigel Formation and MacKop conglomerate, were deposited, possibly starting from circa 3090 Ma in what is interpreted as a convergent setting (Poujol et al. [1996](#page-13-0)). Following this magmatic event, volcanic rocks of the Rubbervale Formation in the Murchison Greenstone Belt and Rooiwater granitoids were emplaced at circa 2970 Ma. The emplacement of these volcanic rocks is taken as evidence of crust formation in an arc environment related to the accretion of the northern part of the Kaapvaal craton, the Pietersburg block, to the Witwatersrand block and collision at that time (Zeh et al. [2013](#page-13-0)).

Gold mineralisation in the Barberton Greenstone Belt is coeval with the tectono-magmatic event related to an extension that shortly followed the final phase of stabilisation of the central Kaapvaal craton. Greenstone gold mineralisation is frequently associated with the latest stage of stabilisation of cratons and can be related to the switch between compression-collision and extensions, when high-angle extensional faults allow deep fluids to flow across the crust (Goldfarb and Groves [2015](#page-11-0); Groves et al. [2000](#page-11-0)). Intriguingly, the available ages for mineralisation partly overlap with Re–Os age determinations of gold and detrital pyrite grains from the Vaal Reef of the Witwatersrand basin (circa 3030 ± 20 Ma, Kirk et al. 2002), the largest gold deposit in the world (Frimmel this volume). Based on this age and an unradiogenic initial $^{187}Os/^{188}Os$ ratio of 0.109, Kirk et al. [\(2002\)](#page-12-0) concluded that the Witwatersrand gold was eroded from a mantle-derived mafic-ultramafic source rock, although the specific origin of this detrital gold could not be identified. Therefore, gold mineralisation preserved in the

Barberton Greenstone Belt may have formed as part of this major crust- and Au-forming event, of which it would represent but a remnant.

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