Metallogeny of precious and base metal mineralization in the Murchison Greenstone Belt, South Africa: indications from U-Pb and Pb-Pb geochronology

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Abstract

The 3.09-2.97 Ga Murchison Greenstone Belt is an important metallotect in the northern Kaapvaal Craton (South Africa), hosting several precious and base metal deposits. Central to the metallotect is the Antimony Line, striking ENE for over 35 km, which hosts a series of structurally-controlled Sb-Au deposits. To the north of the Antimony Line, hosted within felsic volcanic rocks, is the Copper-Zinc Line where a series of small, ca 2.97 Ga Cu-Zn VMS-type deposits occur. New data are provided for the Malati Pump gold mine, located at the eastern end of the Antimony Line. Crystallization of a granodiorite in the Malati Pump mine and of the Baderoukwe granodiorite are dated at 2964 ± 7 Ma and 2970 ± 7 Ma, respectively (zircon U-Pb), while pyrite associated with gold mineralization yielded a Pb-Pb age of 2967 ± 48 Ma. Therefore, granodiorite emplacement, sulfide mineral deposition and gold mineralization all happened at ca. 2.97 Ga. It is, thus, suggested that the major styles of orogenic Au-Sb and the Cu-Zn VMS mineralization in the Murchison Greenstone Belt are contemporaneous and that the formation of meso- to epithermal Au-Sb mineralization at fairly shallow levels was accompanied by submarine extrusion of felsic volcanic rocks to form associated Cu-Zn VMS mineralization.

Keywords

Gold mineralization, VMS deposit, Antimony Line, Kaapvaal Craton, Murchison range, South Africa.

Introduction

The 3.09-2.97 Ga Murchison Greenstone Belt (MGB; Poujol et al. 1996) represents one of a number of Archaean volcano-sedimentary belts within the Kaapvaal Craton and is located in the northeastern portion of the craton (Fig. 1a), approximately 200 km north of the Barberton Greenstone Belt (BGB).

The MGB is well known for its numerous precious and base metal deposits, including: (1) Sb and Au mineralization along a central structural lineament, the *Antimony Line* (AL); (2) massive sulfide–style Cu-Zn mineralization associated with acid volcanic rocks along the northern margin of the belt; and (3) beryl-emerald mineralization associated with granitoid intrusions along the southern margin.

The present study focuses on the Malati Pump mine (also referred to as the Malati Store), which is a small granodiorite-hosted gold deposit along the AL and aims to: (1) date the Au mineralization by U-Pb and Pb-Pb isotopic determinations on zircon and pyrite, respectively; and (2) assess the role that granitoids played in that system. The dating provides additional insights into the metallogenic system at the scale of the MGB.

Geological setting

The east-northeast-trending MGB comprises folded (e.g., Graham 1974; Vearncombe et al. 1992), complexly deformed metavolcanic and metasedimentary rocks intruded by diverse Archaean granitic gneisses. Intense deformation and lack of definitive relationships have, in the past, prevented the recognition of a volcano-sedimentary stratigraphy (Vearncombe 1988), although, more recently, based on available geochronological data, Poujol (2001) proposed a stratigraphic column for the MGB successions. The undated Leydsdorp and Mulati Formations are a mafic to ultra-mafic succession along the southern flank of the MGB. In the centre of the belt, the ca 3.09 Ga Weigel Formation (Poujol et al. 1996) comprises mafic to felsic volcanic rocks and volcaniclastic sedimentary rocks and hosts the AL. Felsic sedimentary rocks (MacKop Formation) have a minimum age of 3076 ± 4 Ma (Poujol et al. 1996). Intermediate to felsic lavas, pyroclastic rocks, quartz-feldspar porphyries and the mineralized "Copper-Zinc Line" constitute the Rubbervale Formation, deposited between 2974 and 2963 Ma ago along the northern flank of the MGB (Brandl et al. 1996; Poujol et al. 1996; Poujol 2001; Schwarz-Schampera et al. 2010).

The Baderoukwe granodioritic gneiss was emplaced syn-tectonically along the eastern side of the MGB (Minnitt and Anhaeusser 1992). The Discovery granite located along the southern contact of the MGB was dated at 2969 ± 14 Ma (Poujol 2001). The Maranda granodiorite, at the western extremity of the AL, was emplaced at a minimum age of 2901 ± 20 Ma (Poujol et al. 1996). The peraluminous Lekkersmaak granite intruded the southern margin of the MGB at 2795 ± 8 Ma (Zeh et al. 2009). The final magmatic event in this region is represented by the Mashishimale pluton, emplaced to the south of the belt and dated at ca 2.67 Ga (Poujol 2001; Zeh et al. 2009).

Migmatites and orthogneisses of Tonalite-Trondhjemite-Granodiorite (TTG) affinity occur both to the north (Groot-Letaba gneiss) and to the south (Makhutswi gneiss) of the MGB. Rocks from the Groot-Letaba gneiss have been dated at 3171 ± 6 Ma (Brandl and Kröner 1993), 3170.5 ± 0.3 Ma (Kröner et al. 2000), 2839 ± 8 Ma, (Zeh et al. 2009) and 3063 ± 12 Ma for the Makhutswi gneiss (Poujol and Robb 1999). Finally to the south of the MGB, the Harmony granite yielded an age of 3091 ± 5 Ma, contemporaneous with the age of the Weigel Formation (Poujol and Robb 1999).

The MGB represents an important metallotect because it hosts several styles of mineralization; Sb-Au (+ As, W, Hg) are the most frequent associations found within hydrothermal mineralized systems in the AL (Vearncombe et al. 1992; Viljoen et al. 1978). In addition, Cu-Zn mineralization in the Rubbervale Formation occurs in volcanogenic massive sulfide systems (Schwarz-Schampera et al. 2010 and references therein). Finally, emerald mineralization occurs to the south of the MGB (Groat et al. 2008).

Cu-Zn massive sulfide mineralization of the Copper-Zinc Line

The massive base metal sulfide mineralization of the Copper-Zinc Line is located in the southern part of the Rubbervale Formation, along a zone between tuffaceous rhyolite and overlying pelitic metasedimentary rocks (Terblanche and Lewis 1995). The mineralization is of a felsic VMS-type of syngenetic origin (Taylor 1981; Terblanche and Lewis 1995; Schwarz-Shampera et al. 2010), like those observed in many Archaean terranes. The 12 known deposits are typically small deformed lenses (500 to 1000 m long, 500 m wide), that are Zn-rich (up to 27%) with subordinate Cu (0.4%), and variable Pb, Au and Sb. They are closely associated with felsic volcanic centres (Schwarz-Shampera et al. 2010). The deposits are hosted in dacitic to rhyolitic volcanic rocks and were dated by U-Pb evaporation technique on zircon grains, yielding ages between

2974.8±3.6 and 2963.2±6.4 Ma (Schwarz-Shampera et al. 2010).

Antimony mineralization along the Antimony Line

Very significant Sb mineralization occurs within the MGB. The production of Sb from the Consolidated Murchison mine reached 25 000 t (15 000 metal) in 1951 and, by 1986, total Sb production in the MGB represented 18% of the world production (Pearton and Viljoen 1986; Ward 1998). The MGB is estimated to have an indicated resource of 7.4 million tons at 2.47 % for Sb (Metorex Limited 2011).

Antimony mineralization occurs in the form of stibnite and berthierite associated with pyrite and arsenopyrite, mainly in quartz-carbonate veins. Mineralization occurred intermittently along the entire length of the 35 km long, 250 m wide Antimony Line, the latter representing an upper-crustal shear zone (Vearncombe et al. 1988; Jaguin et al. 2012); the ores are characterized by strong metamorphic remobilization but, although broadly orogenic in character, their detailed metallogenesis is still poorly understood. Archaean Sb deposits are a rare phenomenon, which implies that either this style of mineralization is characterized by an unusual set of processes and/or that they were poorly preserved. Au is also commonly associated with the Sb mineralization, which suggests that both elements were enriched during the same mineralization processes (Pearton and Viljoen 1986).

Gold mineralization

Gold in the MGB has been mined from 89 deposits over the past century (Fig 1), most of which have now been worked out. Some 27 of these deposits show a strong association with Sb: 32 t of Au were recovered from the MGB, with two-thirds as a by-product of Sb production (Ward and Wilson 1998).

The model established for gold deposits of the BGB has been applied to the MGB with a classification into three groups (Saager and Köppel 1976):

(1) massive stratabound deposits that could be of exhalative origin, such as Letaba, Gravelotte, Monarch and United Jack mines;

(2) disseminated sulfide ores in veinlets of Na-rich porphyry; they could either be secretions from the country rock at the time of granite emplacement (Viljoen et al. 1969 1970) or subvolcanic equivalents of the

VMS with gold being concentrated during differentiation (Saager 1973, 1974); and

(3) gold-rich quartz veins, such as the Old Star mine, that would have formed from late volatile emanations of granites or metamorphic mobilization from the country rocks (Saager 1973 1974; Viljoen et al. 1969; 1970).

Saager and Köppel (1976) suggested that granites played an important role in the mineralization in the BGB, but that they could not be considered as the ultimate source of the gold. More recently, Vearncombe et al. (1992) classified the gold mineralization from the MGB into 7 different types: (1) mineralization associated with stibnite and berthierite in carbonaceous rocks (AL); (2) mineralization associated with arsenopyrite and pyrite in ferruginous cherts and banded iron formation; (3) disseminated sulfides in chlorite, amphibolitic or talcose schists; (4) Au-bearing pyrite and other sulfides in shear zones; (5) quartz veins spatially related to shear zones; (6) zones with minor quartz veining and disseminated pyrite; and (7) disseminated auriferous pyrite within albitized granodiorite intrusions.

Several small, now albitized, TTG intrusions were emplaced along the AL into the Weigel Formation. Ward and Wilson (1998) listed similar albitized bodies associated with gold mineralization in other parts of the MGB (Discovery Shaft, Minerva, Sutherland mines, type 7 of Vearncombe's classification). Moreover, they indicated that 25 of the gold deposits are spatially associated with granitoid intrusives or granite-gneiss contacts. Kedda (1992) studied gold mineralization associated with albitized felsic intrusions, that he related to deuteric and post-magmatic fluids. The mineralization is characterized by an atypical Au-Mo-W-Be-B-Sb-Hg paragenesis associated with a mesothermal (250-350°C) temperature regime. Stable isotopes, as well as fluid inclusion data, indicate an intimate relationship between Au-Sb and the granites as well as a fluid homogeneity on a regional scale (Kedda et al. 1990; Kedda 1992).

Characteristics of the mineralization at Malati Pump mine

The mineralization at Malati Pump is essentially Au-rich with only minor Sb. Gold (up to 3 g/t) is found in quartz-tourmaline-pyrite veins near the apex of the granodiorite pluton and is related to fine disseminated pyrite in the wall rock and as visible and microscopic inclusions in the pyrite (Kedda 1992). The exposed intrusion is a cupola zone of an underlying granodioritic body intruded along the AL (Fig. 2, Kedda 1992; de Beer et al. 1984). The granodiorite intruded quartz-chlorite schists of the Weigel Formation and was, in turn, intruded by a late (Palaeoproterozoic or Mesozoic) dolerite dyke (Kedda et al. 1990). It is an S-shaped body, 50 m in diameter (Vearncombe et al. 1992). The cupola has undergone albitization, carbonation, sulfidation, tourmalinization and silicification, at about 250°C (Kedda 1992). The hosting schists underwent silicification, sericitization, carbonation and sulfidation and display quartz veins, some carrying gold. Quartz vein stockworks have been reported in the granodioritic body (Kedda 1992), which may represent the equivalent of the quartz-veined zones associated with oxidized sulfides of Vearncombe et al. (1992).

Results: Petrography and Age Determinations

Petro-geochronology of the intrusions: U-Pb dating of the Malati Pump and Baderoukwe granodiorites

Zircon grains were separated using standard techniques. Handpicked zircon grains were casted in epoxy mounts, imaged and analyzed by LA-ICP-MS in the Magmas et Volcans Laboratory (Clermont-Ferrand, France). Additional information on the analytical procedure can be found in Poilvet et al. (2011).

The Baderoukwe gneiss (Fig. 1b, samples 5 and 82) is a coarse-grained biotite-trondhjemite, altered in some places (with secondary albite, epidote, titanite, white mica). The samples provided stubby to elongated zircon grains, with luminescent core and visible oscillatory zoning (Fig. 3a, inset). They have low Pb contents (24-136 ppm, Table 1) and variable U contents (34-243 ppm). Four concordant analyses provide a Concordia date (Ludwig 1998) of 2961.9 ± 9.4 Ma (MSWD = 0.019). Twenty analyses out of seventeen grains give a similar upper intercept date of 2967.3 ± 6.7 Ma with a lower intercept at 76 ± 140 Ma (MSWD = 10.1).

Sample MUR 09-111 was collected from the Malati Pump mine, (Fig. 1b, locality 111). It is an albitized granodiorite comprising coarse-grained albite (An 0-2) and quartz. Minor phases include Fe-Mg-Ca-carbonates, rutile, tourmaline, white mica, sulfides (mostly pyrite, pyrrhotite, arsenopyrite). The sample provided homogeneous, weakly luminescent, elongated zircon grains, with low Pb contents (10-227 ppm) and variable U contents (12-982 ppm). Ten analyses out of eight grains give an upper intercept date of 2963.8 ± 6.6

Ma (MSWD = 5.3) if the lower intercept is anchored to 0 ± 100 Ma (Fig. 3b).

Mineralization: Pb-Pb dating of pyrite

Pyrite grains were extracted from the Malati Pump mine granodiorite and from the host quartz-chlorite schists of the Weigel Formation. The sample provided two types of minerals. Type 1 is typical, cream-colored, euhedral pyrite, whereas type 2 corresponds to reddish euhedral pseudomorphs of haematite after pyrite. The fractions were carefully selected under a binocular microscope, washed in acetone and dissolved in a Savilex beaker. Lead was separated and purified on ion exchange resin and the isotopic ratios were measured on a VG Sector mass spectrometer (University of Montpellier II, France). Additional information on the analytical procedure can be found in Poujol et al. (1999).

On a $^{206}Pb/^{204}Pb$-$^{207}Pb/^{204}Pb$ diagram (Fig. 3c), the two types of minerals plot in different positions. Type 1 pyrite defines a restricted range of values while type 2 hematite displays more radiogenic values. Type 1 pyrite, together with Vearncombe's (1992) data, as well as pyrite from the intruded felsic schist of the Weigel Formation, define a Pb-Pb secondary isochron which yields a date of 2967 ± 48 Ma (MSWD=2) with a $\mu 1=7.84$ (Fig. 3d). The more radiogenic type 2 haematite does not provide any meaningful age.

Discussion and conclusions

The Baderoukwe Batholith

The U-Pb dates obtained for the Malati Pump (2964 ± 7 Ma) and the Baderoukwe (2970 ± 7 Ma) granodiorite plutons are identical within error (Fig. 3a and b). They are interpreted as dating the emplacement age of the granodiorite. Therefore these two granodiorite bodies are regarded as part of the same large-scale batholith, now referred to as the Baderoukwe Batholith. Following geophysical evidence (de Beer et al. 1984), this batholith is likely to be present all along the AL (Fig. 1c). Moreover, the 2.97 Ga Discovery granite (Poujol 2001) demonstrates that the Baderoukwe Batholith could possibly be extended to the south of the MGB. The Maranda granodiorite to the west of the AL has a minimum age of 2.90 Ga and could therefore eventually represent the western extremity of the Baderoukwe Batholith. In addition, the ca 2.97 Ga volcanic rocks of the

Rubbervale Formation are identical in age with the emplacement of the Baderoukwe Batholith, suggesting that they might represent its extrusive equivalent. This is further confirmed by the fact that they share similar calc-alkaline affinities (Jaguin et al., in prep.).

Age of the mineralization

The Pb-Pb date of 2967±48 Ma is interpreted as the age of the pyrite crystallization. This sulfidation is therefore comparable in age (within error) to the emplacement of the Baderoukwe Batholith. As shown in Fig 3c, pyrite includes, or is associated with, various other Sb-As-Cu-Co-Zn-sulfides. Moreover, Kedda (1992) demonstrated a direct genetic link between pyrite and the Au-Sb mineralization, because gold is found within hydrothermal pyrite. Therefore granodiorite emplacement, sulfide mineral deposition and gold mineralization are considered contemporaneous.

Some pyrite grains (type 2) were altered to hematite suggesting that an oxidizing fluid circulated within the system. The U content in the type 2 hematite is higher than in the type 1 pyrite (1.4 ppm versus 600 ppb) and its Pb-Pb signature is, consequently, more radiogenic. This demonstrates that the fluid responsible for the oxidation consistently carried more U and/or radiogenic Pb. Unfortunately this oxidation event was not datable (Fig. 3c, Table 2) and could be either close to, or much younger than 2.97 Ga. This distinct fluid may eventually have partially remobilized Au.

Model for the Au mineralization and implications regarding other deposits of the MGB

This study demonstrates for the first time that several different styles of mineralization in the MGB are contiguous and related to a period of magmatism that witnessed, at 2.97 Ga, coeval emplacement of granodioritic (TTG) intrusions and calc-alkaline type magma extrusion. Central to this study has been the Malati Pump intrusion with its Au mineralization and associated Sb-As-Hg metal suite that is perhaps more reminiscent of a high-level epithermal setting than of a mesothermal origin (e.g. Nesbitt and Muehlenbachs 1989). In addition to the Malati Pump intrusion, the entire AL is decorated with numerous small TTG type intrusions (Pearton and Viljoen 1986; Kedda 1992), geophysical evidence for which suggests a spatial link at depth and the possible existence of a larger batholith (de Beer et al. 1984). A continuum of Au-Sb mineralization, as described for example in Groves et al. (2003) and Nesbitt and Muehlenbachs (1989), is a feature of the AL, and in this regard it seems likely that this system can be related in its entirety to a single system where heat input and fluid flow were at least in part directly related to TTG emplacement at 2.97Ga.

This study also shows that the Rubbervale Formation and its syngenetic VMS-style Cu-Zn mineralization (Scharwz-Shampera et al. 2010) are contemporaneous and directly related to the emplacement of the Baderoukwe Batholith, again at circa 2.97Ga. Consequently, the calc-alkaline rocks of the Rubbervale Formation are likely the extrusive equivalents of a major TTG type intrusive event, here termed the Baderoukwe Batholith. It is, therefore, suggested (1) that the major styles of orogenic Au-Sb and the VMS style Cu-Zn mineralization in the MGB are contemporaneous and (2) that the formation of meso- to epithermal Sb-Au mineralization at fairly shallow intrusive levels was accompanied by extrusion of felsic volcanic rocks in a subaqueous shallow marine environment to form associated Cu-Zn VMS mineralization, all at circa 2.97 Ga. A close spatial relation between on one side VMS deposits plus their extrusive host rocks, and on the other side intrusive magmatic-hydrothermal system, has not often been described in Archaean settings (Franklin et al. 2005). It should also be noted that the beryl-emerald mineralization occurring along the southern margins of the MGB and spatially associated with the intrusion of the Discovery granite, might have also occurred at 2.97 Ga, the age of the latter intrusion. In the light of these data, we recognize the MGB as a major metallotect that has been under-explored for precious and base metal mineralization. Our data suggest that a new exploration strategy should be employed in the region, recognizing the Baderoukwe Batholith as the central geological feature that is spatially and secularly related to substantial and varied styles of mineralization. In particular, the structures that have played a key role along and into which phases of the Baderoukwe Batholith have been emplaced should remain the object of concerted exploration targeting in this region.

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Figures and Tables caption

Fig. 1 Inset (a) shows the location of the Murchison Greenstone Belt in the Kaapvaal Craton. (b) Simplified geological map of the MGB (modified after SACS 1980 and Vearncombe et al. 1992; Gold deposits from Ward and Wilson 1998). Inset (c) shows cross sections from the geophysical survey undertaken by de Beer et al. (1984), with available ages (Ga)

Fig. 2 Quarry face in the Malati Pump mine showing the geometry of the different rock types (man to the right for scale)

Fig. 3 Geochronological diagrams (a) Concordia diagram for the Baderoukwe granodiorite. Inset: CL image of a zircon grain from sample MUR 09-5, with oscillatory zoning (bar scale 100µm). (b) Concordia diagram for the Malati Pump granodiorite. (c) Pb-Pb diagrams for pyrite and haematite grains from the Malati Pump mine and Weigel Formation displaying two populations with distinct isotopic Pb signature. Inset: SEM image of the Malati Pump granodiorite showing the association between pyrite (Pyr) with mingled chalcopyrite (Ccp) and ullmanite (Ull, bar scale 100µm). (d) Isochron for the pyrite population (Type 1). Isoplot software, Ludwig (2000)

Fig. 4 Sketch for the MGB metallogenic system as proposed in this study

Table 1 LA-ICP-MS U-Pb isotope data for zircon

Table 2 ID-TIMS Pb-Pb isotope data for pyrite and haematite.