The Witwatersrand Basin, South Africa: Geological framework and mineralization processes

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Received 19 December 1994; accepted 19 June 1995

Abstract

The Witwatersrand Basin formed over a period of 360 Ma between 3074 and 2714 Ma. Pulses of sedimentation within the sequence and its precursors were episodic, occurring between 3086–3074 Ma (Dominion Group), 2970–2914 Ma (West Rand Group) and 2894–2714 Ma (Central Rand Group). Detritus was derived from a mixed granite–greenstone source of two distinct ages; the first comprises Barberton-type greenstone belts and granitoids > 3100 Ma old, and the second consists of the greenstone belt-like Kraaipan Formation and associated granitoids ≤ 3100 Ma old. Subsequent granitoid plutonism was episodic and coincided with hiatuses in sediment deposition, but continued throughout the evolution of the basin. Many of the provenance granitoids are characterized by hydrothermal alteration, are geochemically anomalous with respect to Au and U, and may represent viable source rocks for palaeoplacer mineralization. Tectonically, the basin evolved in response to processes occurring within a Wilson cycle, associated with the encroachment and ultimate collision of the Zimbabwe and Kaapvaal cratons. Metamorphism of the Witwatersrand Basin occurred at ca. 2500, 2300 and 2000 Ma. The first two events coincided with the progressive loading of the basin by Ventersdorp and Transvaal cover sequences, whereas the last reflects intrusion of the Bushveld Complex and/or the Vredefort catastrophe.

Mineralization is concentrated in the conglomerates of the Central Rand Group and is represented by a complex paragenetic sequence initiated by early accumulation of detrital heavy minerals. This was followed by three stages of remobilization caused by metamorphic fluid circulation. An early event of authigenic pyrite formation at 2500 Ma was followed at 2300 Ma by maturation of organic material, fluxing of hydrocarbon bearing fluids through the basin and the radiolytic fixation of bitumen around detrital uraninite. This was followed at around 2000 Ma by peak metamorphism which resulted in the widespread redistribution of gold and the formation of a variety of secondary sulphides. Post-depositional fluid conditions were such that metal solubilities were low and precipitation mechanisms very effective, resulting in the superimposition of both primary and secondary mineralization.

1. Introduction

The major mineral deposits of the world form as a result of a fortuitous combination of events which is not often repeated in the course of geological history. Crustal processes occasionally interact at a particular point on the earth's crust in such a way as to form metal concentrations that are virtually unique, examples of which might include Sudbury, the Zambian copper belt, the Viburnum trend, the Bushveld Complex, etc. The Witwatersrand Basin is a classic example of where a variety of geological processes have coincided in time...
and space to produce the world's greatest gold province. In just over 100 years of mining, about 45 000 tons of gold (more than 35% of all the gold mined in the history of mankind; Handley, 1990) and over 150 000 tons of uranium have been produced from the Witwatersrand Basin.

The nature of the geological processes which contributed to this prodigious accumulation of metals has been much debated ever since the commencement of mining activities, in the area which was to become Johannesburg, in 1886. Controversies such as the timing of mineralization and the tectonic framework for basin deposition, as well as uncertainties in the age of sedimentation, the limits of the basin and the source of gold and uranium, have restricted geological understanding in the region. Over the past decade, however, a number of advances have contributed to resolving many of the problems of Witwatersrand metallogeny. The purpose of this review is to highlight some of the pertinent features relating to the geological environment and mineralization processes in the Witwatersrand Basin, drawing especially on research carried out in recent years.

2. Geological framework

2.1. The Kaapvaal Craton

The Kaapvaal Craton is the name given to the ancient segment of continental crust which formed in southern Africa between about 3.7 to 2.7 Ga. Much of this continental nucleus actually formed prior to 3.1 Ga by the formation of an extensive granitoid basement and amalgamation with arc-like oceanic terranes represented by mafic/ultramafic volcanics and associated sediments (De Wit et al., 1992). Subsequent growth of the craton was accomplished by further continental magmatic activity, possible Cordilleran-style accretion of composite terranes along the margins of the proto-continent, and the deposition of appropriate sedimentary basins. Although the question of when the Kaapvaal continent attained its cratonic rigidity is a semantic one, it is clear that by 2.7 Ga, or shortly thereafter, this segment of crust was not subjected to further major orogenesis. The Witwatersrand Basin is an approximately 7000 m thick terrigenous sequence comprising mainly arenaceous and argillaceous, together with minor rudaceous, lithologies deposited in a fluvo-deltaic environment in the centre of the Kaapvaal Craton (Fig. 1 and Fig. 3). Until recently it was considered to be early Proterozoic in age and, consequently, to have developed subsequent to the formation of the craton. As will become clear in this review the basin is now viewed as an integral part of the Kaapvaal Craton, having formed during the latter stages in the development of what is now known to be one of the most extensively mineralized segments of the earth's crust.

2.2. Age constraints

Even as recently as 1986 the Witwatersrand Basin was believed to have been deposited during the early Proterozoic era, between about 2.6 and 2.3 Ga (Allsopp and Welke, 1986). Age constraints depended largely on the limited availability of whole rock Rb-Sr and Pb-Pb isotope determinations carried out on ante- and supercedent rock formations, which have since proved, in most cases, to reflect a resetting event that has been superimposed on to the rock system and not the age of formation. Recent work has utilized precise, single zircon U-Pb dating techniques in order to accurately constrain the true age of Witwatersrand deposition; these results now show that the basin is Archaean in age and was deposited over an extended period of 360 Ma.

A summary of recently acquired U–Pb zircon age data (after Barton et al., 1989; Robb et al., 1990a, b, 1992; Armstrong et al., 1991) is presented in Fig. 1 and summarized below. Witwatersrand sediments overlie, either the Archaean granite–greenstone basement, or the volcano–sedimentary Dominion Group. The maximum age of deposition is, therefore, controlled by the ages of these units. Several basement granitoids which stratigraphically underlie Witwatersrand strata have now been dated and range in age between 3174 ± 9 and 3086 ± 3 Ma. Volcanics in the upper part of the Dominion Group yield an age of 374 ± 6 Ma and this figure represents the maximum available constraint for onset of Witwatersrand deposition. Dominion sediments were deposited over a relatively short interval, between 3086 ± 3 Ma (the age of the Westerdam granite, Fig. 2, on top of which Dominion sediments have been deposited) and 3074 ± 6 Ma, the age of overlying lavas. Detrital zircon and monazite grains extracted from Dominion sediments range in age between 3191 and 3105 Ma. The
Fig. 1. Age constraints for the Witwatersrand Supergroup on the basis of available U–Pb zircon ages (data compiled from Barton et al., 1989; Robb et al., 1990a, b, 1992; Armstrong et al., 1991). *D* = Dominion Group deposition; *WR* = West Rand Group deposition; *CR* = Central Rand Group deposition; *VCR* = Venterdorp Contact Reef deposition. For the sake of clarity error bars are not shown; errors for rock ages are quoted in the text; errors for individual detrital zircon ages are listed in the original source and are in all cases less than ±20 Ma, and in most cases less than ±10 Ma.
latter date provides an independent assessment of the maximum age of deposition for the Dominion Group.

The minimum age of Witwatersrand deposition is provided by the age of the Venterdorp lavas which immediately overlie the sequence over wide areas. The lower basaltic portion of these lavas has been dated at 2714 ± 8 Ma, whereas higher up in the succession quartz porphyries have been dated at 2709 ± 4 Ma (Fig. 1). The total time span available for Witwatersrand deposition is, therefore, in the order of 360 million years, between 3074 and 2714 Ma.

Placing constraints on the timing of the various pulses of sedimentation within the Witwatersrand sequence is difficult and requires dating of thin intercalated lava units and detrital grains, the latter providing maximum estimates for the ages of deposition. The Crown lava, which occurs just beneath the West Rand–Central Rand Group transition about half way up the sequence, has been provisionally dated at 2914 ± 8 Ma (Fig. 1), a figure which provides approximate maximum and minimum constraints on Central Rand and West Rand Group deposition, respectively. Detrital zircon grains from the Orange Grove quartzite and the Promise reef, both in the West Rand Group, range in age between 3330 ± 5 and 2970 ± 3 Ma; the latter date represents a maximum constraint on the depositional age of the West Rand Group, with the minimum age determined by the Crown lava at 2914 Ma (see stippled box marked WR, Fig. 1). It is reassuring to note that, of the 30 detrital grains independently analysed from the West Rand Group, none is younger than 2914 Ma, a feature which provides credibility to the age of the Crown lava.

Detrital zircon grains from conglomerate reefs in the auriferous Central Rand Group become progressively younger upwards in the stratigraphy and the youngest grains occur in the Elsburg and Venterdorp Contact (VCR) reefs (2894 ± 10 and 2780 ± 5 Ma, respectively). The VCR must, therefore, have been laid down sometime after 2780 Ma ago, but before or at 2714 Ma which is the age of the Venterdorp lavas (Armstrong et al., 1991). Central Rand Group deposition is more difficult to constrain because the youngest detrital grains occur in rocks high up in the succession. If it is assumed that the igneous (granitic?) rock within which the zircon formed was emplaced prior to the pulse of erosion and sedimentation which followed exhumation of that rock, then the Central Rand Group may have been deposited subsequent to 2894 Ma (the youngest zircon in the Elsburg reef) and possibly before 2780 Ma (which is the maximum age constraint on VCR deposition). The latter limits are feasible only if the absence of the post-2894 Ma VCR zircons in the Elsburg reef reflects a geological reality and not a sampling bias. The maximum constraints on Central Rand Group deposition are shown in Fig. 1 as a sloped dashed line to reflect the uncertainties in placing this limit on the basis of detrital zircon ages.

In summary, the availability of precise, single U–Pb zircon ages in and around the Witwatersrand Basin has resulted in a reasonably well constrained set of age limits to the various pulses of sedimentation that formed the depository. Dominion sedimentation occurred over a relatively brief interval some time after 3086 Ma but before 3074 Ma ago. This was followed by what was probably an equally rapid pulse of bimodal volcanism at around and preceding 3074 Ma. West Rand Group deposition commenced subsequent to 2970 Ma and, consequently, a significant hiatus of some 100 million years appears to exist between the Dominion and West Rand Groups. The latter pulse of sedimentation was largely complete by 2914 Ma, when the Crown lava was extruded. The onset of Central Rand Group deposition commenced after 2914 Ma but may have started as late as 2894 Ma, the age of the youngest detrital zircon grain in the Elsburg reef (Fig. 1). The VCR was deposited after 2780 Ma but
before or around 2714 Ma. The extent of the hiatus between Turffontein Subgroup and VCR deposition is not known.

2.3. Nature of the Witwatersrand provenance

There are two ways of studying the provenance of a sedimentary basin; these are (a) the nature and distribution of detritus and the chemistry of shales within the depository, and (b) the eroded remnants of the source area from which the sediments are derived, which might also require, as in the case of the Witwatersrand Basin, study of borehole intersections of largely uneroded sub-surface portions of the provenance. In the following section an attempt is made to reconstruct the nature of the Witwatersrand provenance by referring to data from both sources.

Detritus and shale chemistry

Zircon and chromite are two of the most abundant heavy minerals to have been concentrated in the Witwatersrand conglomerate reefs. Their presence attests to the existence of both felsic and mafic/ultramafic rocks in the source area. Individual reefs are characterized by very different chromite:zircon ratios (Viljoen, 1963) and, assuming similar conditions of erosion and deposition, this suggests that the relative proportions of felsic and mafic source rocks differ from one portion of the provenance to another.

The distribution of immobile elements in shales from the Witwatersrand sediments confirm the above, and systematic changes in Cr/Zr, Ni/Co and Cr/V ratios upwards in the succession indicate that the proportions of felsic to mafic source material changed as the basin was progressively deposited (Wronkiewicz and Condie, 1987). The West Rand Group provenance appears to have been dominated by 40–60% tonalite and lesser proportions of granite and basalt, whereas the Central Rand Group was apparently derived from a source where granite and basalt were relatively abundant (40–50% each) but tonalite was a minor component (Wronkiewicz and Condie, 1987).

The age of the Witwatersrand provenance area can also be inferred from the distribution of ages obtained from the dating of individual zircon and monazite grains concentrated within the reefs. A histogram of all available detrital grain ages (Barton et al., 1989; Robb et al., 1990a, b) shows that the granitoid rocks, from which this detritus was largely derived, were formed between 3200 and 2900 Ma (Fig. 2). Two distinct peaks are, however, evident, one with a mean at around 3200 Ma and the other with a mean of about 3075 Ma. These two populations reflect an older source terrane of dominantly tonalitic material that was intruded at a later stage by more evolved granitic crustal components, a trend that is consistent with the inferences from shale geochemistry. The suggestion that the basaltic (greenstone) component of the source area also became more abundant as Witwatersrand deposition proceeded (Wronkiewicz and Condie, 1987) is perhaps linked to the extrusion of post-3200 Ma basalts such as parts of the Dominion Group at 3075 Ma and the Kraaipan Formation greenstones which are also tentatively dated at 3070 Ma (L.J. Robb, unpubl. data).

Eroded surface and sub-surface remnants of Witwatersrand provenance

The best exposed portion of the Archaean basement on the Kaapvaal Craton is the Barberton region and Swaziland to the east of the Witwatersrand Basin (Fig. 3), where detailed study over several decades has provided an extensive data base for the study of early crustal evolution. The region is now known to comprise a collage of amalgamated terranes each of which consists of tonalite–trondhjemite–granodiorite (TTG) gneisses and an associated assemblage of metavolcano–sedimentary supracrustal rocks (Lowe, 1994). These terranes were mainly formed at ca. 3480–3440 and 3250–3220 Ma (Armstrong et al., 1990; De Ronde and De Wit, 1994; Kamo and Davis, 1994), although isolated remnants of TTG material and associated metavolcanics in the southwestern portion of the belt are as old as 3550–3530 Ma (Kröner et al., 1991). The TTG basement of the Ancient Gneiss Complex of Swaziland contains minor relics which are as old as 3650–3500 Ma (Compston and Kröner, 1988) and, therefore, predate the Barberton greenstone belt.

Greenstone belt formation and TTG plutonism was followed in the Barberton area by the emplacement of voluminous, sheet-like granodiorite–adamellite (or quartz-monzonite) batholiths at 3106 ± 3 Ma (Kamo and Davis, 1994). This period was followed by an epicontinental style of sedimentation (the Pongola Supergroup) and inter-continental rifting which was accompanied, in the Barberton region, by at least three pulses of granite plutonism and possibly more else-
where on the craton. An early period of S-type granite intrusion occurred at ca. 3080 Ma and again at 2820–2860 Ma and this was followed by an I-type event at 2740–2690 Ma (Meyer et al., 1994a, b).

The sub-surface Archaean basement between Barberton and the Evander goldfield has been studied by referring to bore-hole intersections drilled through the Karoo Sequence (Ferraz, 1989). The area south and east of Evander is underlain by a significant proportion of greenstone material (Fig. 3) which is associated with granitoids of tonalitic composition. By inference, this terrane probably equates with the 3450–3230 Ma Barberton greenstone belt and surrounding TTG terrane (Fig. 3). The high proportion of greenstone in the Evander provenance is consistent with features such as the high platinum group element contents, high Cr/Zr ratios and diverse, chert-rich, polymictic pebble population which typify the principal economic conglomerate horizons in this goldfield.

To the north and west of the Witwatersrand Basin exposure is poor and discontinuous (Fig. 3) and the evolution of the Archaean basement is less well understood. An older generation of TTG gneiss is preserved in at least three areas, namely; (a) in part of the Johan-
nesburg dome (the 3170 ± 34 Ma Linden tonalite; Anhaeusser and Burger, 1982); (b) to the west of the Colesburg magnetic anomaly trend (Fig. 3) where TTG gneisses have been dated at ca. 3250 Ma (Drennan et al., 1990) and; (c) in the Kraaipan–Disaneng areas (Fig. 3) where the basement gneisses have not been dated (Anhaeusser, 1991). Granodiorites and adamellites dated at 3174 ± 9 and 3120 ± 5 Ma (Armstrong et al., 1991; Robb et al., 1992) occur in the Hartbeestfontein area (Fig. 3), while the Westerdam dome exposes eroded remnants of granodiorite dated at 3086 ± 3 Ma (Robb et al., 1992). Younger, more evolved, S-type granites, akin to those in the Barberton region, underlie the Coligny (3031 ± 11 Ma) and the Schweizer–Reneke (2880 ± 2 Ma) domes (Robb et al., 1992; Fig. 3). Unique on the Kaapvaal craton is the large rapakivi granite–anorthosite–rhyolite complex in Botswana represented by the Gaborone Granite Suite and Kanye Formation (Fig. 3). These two units have now been shown to be coeval and were emplaced at a high level in the crust at 2785 ± 2 Ma (Grobler and Walraven, 1993; Moore et al., 1993). The late I-type granites documented in the Barberton region are also recorded in the Witwatersrand provenance in the form of the Uitloop granite (2687 ± 2 Ma; De Wit et al., 1993) and a granite which was intersected in a borehole (number 1633, Fig. 3) beneath the Venterdorp lavas northwest of the Welkom goldfield, dated at 2727 ± 6 Ma (Robb et al., 1992).

A vertical profile through the granitoid basement underlying the Witwatersrand Basin has been exposed in the core of the Vredefort structure (Fig. 3), which is interpreted, either as a meteorite impact site (Grieve et al., 1990; Leroux et al., 1994) formed at 2025 Ma (no error margin provided; Kamo et al., 1995), or a post-Transvaal push-up structure associated with a bend in a large strike-slip shear (Coward et al., 1995). The uplifted core of the structure now preserves a near vertical succession of granitoid crust which grades from ca. 3100 Ma upper crustal potassic gneisses, through a mid-crustal magnetic discontinuity and down into a metamorphosed TTG-greenstone segment that is around 3500 Ma old (Hart et al., 1991). The tectonic and metamorphic evolution of the Vredefort structure has important implications for the preservation, alteration and tectonic dismemberment of the Witwatersrand sediments.

A few, poorly exposed remnants of greenstone belts are preserved in the Witwatersrand provenance and, as with the granitic terrane, their geological evolution is not well understood. In the southern portion of the Johannesburg dome the Roodekrans and Muldersdrift ultramafic complexes intrude a sequence of komatiitic basalts which collectively resemble the lower portions of the Onverwacht Group in the Barberton greenstone belt (Anhaeusser, 1977; Fig. 3). The Kraaipan Formation in the western Transvaal is made up by a different lithological package, which comprises voluminous banded iron formation, magnetite quartzite, serpentinite and mafic to felsic schists (Anhaeusser, 1991). Felsic schists within this package have yielded a tentative U–Pb zircon age of ca. 3070 Ma (L.J. Robb, unpubl. data), suggesting that the latter are unrelated to the Barberton greenstone belt. The above characteristics suggest that the Kraaipan greenstones, which occur in north–south-trending linear belts, may represent a phase of platformal volcano–sedimentary activity that was coeval with Dominion volcanism.

The sub-surface Archaean basement southwest of the Welkom goldfield is typified by an apparent paucity of greenstone material and two quite different granitoid terranes which are separated by a shallow, easterly dipping geophysical feature known as the Colesburg magnetic anomaly trend (Drennan et al., 1990). West of this north–south orientated magnetic feature (Fig. 3), basement consists largely of ca. 3250 Ma old tonalitic gneisses and migmatites, while east of the trend granitoids are massive-textured, granodiorites and adamellites with distinctly more evolved chemical compositions. These differing terranes are considered to reflect a shallow-dipping crustal profile analogous to, but differing in certain details, from the vertical profile exposed in the core of the Vredefort structure (Drennan et al., 1990; Hart et al., 1991).

**Relationship between granitoid intrusion and basin development**

The development of the Kaapvaal craton occurred over a prolonged (> 800 million year) period of earth history and involved a large number of discrete tectono–magmatic events (De Wit et al., 1992). From the viewpoint of the Witwatersrand Basin, however, these events can be simplified into two periods (Fig. 3 and Fig. 4); those that preceded Witwatersrand deposition (> 3100 Ma) which in this review are termed Swazian,
and those associated with the development of the "Witwatersrand Triad" (i.e. the Dominion, Witwatersrand and Ventersdorp sequences deposited between 3100–2700 Ma) which are termed Randian. Among the Randian granites in the immediate basin provenance, several pulses of magmatism are recognized, namely at ca. 3090, 3030, 2880, 2780 and 2730 Ma (Fig. 4). In relation to episodes of sedimentation in the Witwatersrand Triad there is a suggestion that individual sediment pulses may have occurred subsequent, and therefore represent a response, to an event of granite magmatism. For example, the Westerdam granite (3086 ± 3 Ma) was emplaced just prior to Dominion sedimentation, the Coligny granite (3031 ± 11 Ma) was emplaced prior to West Rand Group deposition and the Schweizer–Reneke granite (2880 ± 2 Ma) may have preceded Central Rand Group sedimentation (Fig. 4). The Gaborone granite and Kanye rhyolites were emplaced at 2785 ± 2 Ma in an interlude which may have stimulated Turffontein Subgroup deposition (i.e. the upper portion of the auriferous Central Rand Group; Fig. 4), although no detrital zircons of this age have yet been found in the latter sequence. In the Welkom goldfield Central Rand Group sediments have been deformed, and in places overturned, prior to the deposition of the final pulse of Witwatersrand sedimentation, known in the Welkom goldfield as the Boulder Beds. This deformation is probably related to the emplacement of a granite (intersected in borehole 1633) at 2727 ± 6 Ma (Fig. 4) which occurs very close to the western edge of the Welkom goldfield (Fig. 3). If the Boulder Bed is approximately coeval with the Ventersdorp Contact Reef (VCR) then it follows that the granite to the west of Welkom may have been emplaced in the hiatus between Central Rand Group and VCR deposition (Fig. 4). The existence of several pulses of syn-sedimentary granitoid plutonism close to the margins of the basin (demonstrated schematically

Fig. 4. Magmatic and sedimentary events pertaining to the evolution of the Witwatersrand Basin and environs (data sources as for Fig. 1).
in Fig. 6) is a new concept in terms of Witwatersrand evolution and may also prove to be important in understanding the nature and origin of the contained mineralization.

**Fertility of the Witwatersrand provenance**

Syngenetic models of Witwatersrand mineralization require that the vast quantities of Au and U that are concentrated within the basin were ultimately derived from source rocks in the Witwatersrand provenance. Arriving at a viable mass balance for such a scenario implies that, either the source area was unusually well endowed in Au and U (Robb and Meyer, 1987, 1990), or placer deposits of this magnitude can be created by sedimentological concentration processes from normal Archaean basement (Loen, 1992). Analysis of Witwatersrand provenance rocks shows that significant enrichments of both Au and U occur, especially in the relatively uneroded granites that occur as suboutcrop in the provenance. These granites are often characterized by an overprint of hydrothermal alteration (Robb and Meyer, 1990; Robb et al., 1990a, b) and point to the possibility that the Witwatersrand source rocks may indeed be geochemically anomalous.

The sub-surface basement to the north and west of the Witwatersrand Basin appears to be underlain by an apparently high proportion of evolved Randian granites and the Kraaipan greenstones, which although of fairly limited exposure, may be more extensively developed beneath younger cover sequences. It is suggested that the combination of evolved granites emplaced episodically throughout the entire deposition of the Witwatersrand sequences, together with the younger greenstones, combine to provide a fertile source environment that was both Au and U specific. In particular it is observed that many of the granites in the region are hydrothermally mineralized, a feature which is manifest as pervasive phyllic and propylitic alteration, vein-related alteration (a dominantly quartz–chlorite–carbonate–sulphide paragenesis), hydrofracturing and, specifically in peraluminous granites, a form of greisen alteration comprising a quartz–muscovite–chlorite–albite–fluorite paragenesis (Robb and Meyer, 1990; Robb et al., 1990a, b). Although it is difficult to accurately date the age of hydrothermal alteration in these granites, precise U–Pb dating at two localities has shown that exogenous quartz–microcline–albite–pyrite–rutil veins are derived from the nearby 2880 Ma Schweizer–Reneke pluton, whereas greisen-related fluorites are broadly coeval with, and have similar Pb-loss histories to, the 3086 Ma Westerdam granite within which they are hosted (Robb et al., 1992). It is evident, therefore, that the paragenetically early hydrothermal alteration is related to fluid saturation within the granite intrusions themselves and that this was associated with both endo- and exo-granitic metal concentrations that occurred repeatedly with successive pulses of granite magma emplacement. Paragenetically later hydrothermal alteration, which post-dates Witwatersrand deposition, is also evident in some of the provenance granitoids; for example, bituminous nodules which coat grains of uraninite/uranotherite in peraluminous granites adjacent and beneath the basin are believed to be related to be the migrated products of Witwatersrand kerogen forming ca. 2300 Ma (Robb et al., 1994).

The origin of the Au and U specificity in the Witwatersrand Basin source is an intriguing and contentious issue. In the Barberton area, for example, U content increases progressively in successively younger and chemically more evolved granites (Meyer et al., 1986; Fig. 5b). However, in order for a granite to be a viable source for a placer uranium deposit, it should be U specific (U/Th>0.5) such that it can contain primary uraninite or uranotherite. In Fig. 5b it is apparent that, although the youngest granitoids in the Barberton region have the highest U contents, they are not U specific and are, therefore, unlikely to represent a viable source of uraninite. Only the 3080 Ma S-type Sinceni granite in Swaziland (Fig. 3), however, represents a deviation from the normal U and U/Th trends in Fig. 5b and might contain accessory uranium minerals (Trumbull, 1993); it may, therefore, represent a viable source rock for subsequent placer concentrations. Granites from north and west of the Witwatersrand Basin are, by contrast with the Barberton area, often U specific and contain primary uraninite (Robb et al., 1990a, b). In Fig. 5b it is apparent that granites emplaced at around 3100 Ma, which are peraluminous and may be S-type granites (e.g., GH1 and Westerdam), are U specific (i.e. have high U/Th ratios) and could have acted as a source of detrital uraninite for both the Dominion and Witwatersrand sequences. Another viable source rock is represented by the 3174 Ma greisenized granites around Hartebeestfontein (Fig. 3) which have very high U/Th ratios by virtue of their low Th contents (Fig. 5b) and abundant ura-
ninite. The peraluminous, 2880 Ma old Schweizer–Reneke granite is also U specific, having been emplaced not long before the main pulse of S-type granitoid plutonism in the Barberton region, and could, therefore, represent a source of uraninite for the Central Rand Group conglomerates.

Au source is far more difficult to account for in the Witwatersrand provenance because of the enormous
quantities of the metal that have to be reconciled, and the requirement that a source of particulate gold is required if deposition is to be explained by placer processes. Small, mesothermal gold deposits proliferate in greenstones of both the Barberton and Kraaipan types, and this source could account for some detrital gold in the Witwatersrand conglomerates. It is generally held, though, that greenstone-hosted gold is volumetrically inadequate to account for the prodigious accumulations present in the basin. Particulate gold has, however, also been detected in hydrothermally altered granites of the western Transvaal (Hallbauer, 1975, 1984). If, as suggested above, metal enrichment in these hydrothermally altered granites either pre-dates or is coeval with, deposition of the Witwatersrand Basin, then a more ubiquitous Au source than that represented by the greenstones may exist (Robb and Meyer, 1990). It has also recently been shown that the 2785 Ma Kanye volcanics in southeastern Botswana (Fig. 3) represent an enormous extrusive rhyolite province within which there is perhaps potential for the development of epithermal precious metal mineralization (Moore et al., 1993). Accurate analyses of hydrothermally altered granites from throughout the provenance have shown that many of these rock types are enriched in Au (Robb and Meyer, 1990). Fig. 5a shows Au contents for Barberton granitoids, eroded surface remnants of Witwatersrand provenance granitoids and sub-surface hydrothermally altered granitoids, and clearly shows that the latter exhibit wider ranges in Au content and are characterized by mean values well in excess of the Clarke value. There is no doubt that many of the granitoids in the Witwatersrand Basin source area are both geochemically anomalous and potentially prospective with respect to Au, but whether these rocks represent an adequate source for all the Au in the depository remains a contentious issue.

Another option for Au source in Witwatersrand ores is the suggestion that gold and pyrite were concentrated along the basin edge by shallow marine discharge vents that circulated hydrothermal solutions along growth faults during sedimentation (Hutchinson and Viljoen, 1988). As with other hypotheses this exhalative model remains contentious, especially given that the association of putative auriferous discharge vents with tholeiitic volcanics and chemical sediments appears to be restricted to the gold-poor West Rand Group and that such associations are apparently absent in the gold-bearing upper sequences of the basin. If indeed a primary source of Au cannot be found or inferred, then consideration must be given to the possibility that a substantial contribution of metal was made epigenetically, during post-depositional fluid circulation. This topic is discussed further in a later section.

2.4. Tectonic environments during Witwatersrand deposition

Plate tectonic theory has resulted in a dramatic change in the perception of the tectonic environment for Witwatersrand deposition over the past decade. Originally thought to have been laid down on a stable continental basement in an early-Proterozoic extensional half-graben environment (Pretorius, 1976), the basin is now thought to represent a complex response to a variety of plate tectonic linked processes in the late-Archaean.

The age constraints outlined above clearly indicate that the Witwatersrand Basin represents one component in a long-lived sequence of events that gave rise to an early continental nucleus known as the Kaapvaal craton. The latter stages of the development of the craton are thought to be closely related to the progressive encroachment of the Zimbabwe and Kaapvaal cratons and the formation of the Limpopo orogeny between the two at around 2700 Ma (De Wit et al., 1992). Early dynamic models, therefore, tended to regard the Witwatersrand depository as a foreland basin (Burke et al., 1986; Winter, 1987). Clendenin et al. (1988) subsequently envisaged the concept of “successor basins” in the region, whereby the Dominion-Witwatersrand-Ventersdorp-Transvaal depositional systems formed in response to alternating compressional and extensional forces localized along the sites of pre-existing tectono-thermal weakness. Ultimately, Stanistreet and McCarthy (1991) recognized the existence of a Wilson Cycle, suggesting that five major stages can be recognized in the evolution of the Witwatersrand Triad. These are:

1. Development of a structural grain on the craton formed through the distribution and structural evolution of greenstone belts;
2. Deposition of the Dominion sequence in a continental rift environment (extensional), with the outpouring of a thick bimodal volcanic pile;
3. Thermal collapse and subsequent initiation of foreland basin tectonics with the progressive encroachment of the Zimbabwe craton, giving rise to the widespread epicontinental, subtidal sedimentary sequence of the West Rand Group;
4. Collision of the two cratons, and development of foreland basin tectonics, and the deposition of the Central Rand Group; and
5. Extrusion of the lower Ventersdorp (Klipriviersburg) tholeiitic flood basalts and the beginning of renewed extensional tectonics (impactogenal rifting) to form the mid-Ventersdorp (Platberg) graben-fill sequences.

Many of these notions have been incorporated into a much broader model for the construction of the entire Kaapvaal shield and craton by De Wit et al. (1992). In this model the craton is considered to be made up of a collage of sub-terranes each with distinct geological attributes. Pertinent is the suggestion that the Witwatersrand hinterland comprises two distinct terranes, one to the north and northwest of the basin made up of > 3000 Ma crust and the other to the west (the Amalia terrain) comprising rocks < 3000 Ma.

In Fig. 6 an attempt has been made to summarize the major tectonic scenarios associated with the development of the Witwatersrand Triad in the context of a time-constrained depositional framework (Robb et al., 1991). In this model an attempt is made to incorporate the various pulses of magmatic activity, especially granitoid plutonism, which are often ignored in this type of reconstruction. During the Dominion extensional and thermal collapse phase (3100–3010 Ma) plutons such as Westerdam and Coligny were intruded, together with plutons hidden beneath younger cover for which evidence is recorded in the detrital zircon population. The early foreland basin stage which gave rise to West Rand and possibly portions of the Central Rand Group sedimentation was accompanied elsewhere on the craton by the emplacement of the S-type granites which are exposed in the Barberton region.

The continent–continent collisional stage between 2840–2720 Ma was accompanied by emplacement of the voluminous Gaborone–Kanye event, the Schweizer–Reneke granite and the hydrothermally altered granite (borehole 1633) northwest of the Welkom goldfield (Fig. 3). Finally, the largely extensional Ventersdorp event was also coeval with widespread I-type granitic activity particularly in the northern and eastern portions of the craton.

These models represent a reasonable first order approximation of the tectonic evolution of the region, but lack resolution in terms of detailed scrutiny. A recent attempt has been made to chart the evolution of thrust systems in and around the basin using seismic sections, and relate this to the broader craton-wide tectonic scenario (Coward et al., 1995). Major thrust systems are shown to have been responsible for the development of topographic expression to the northwest of the depository, thereby controlling the main avenues of sediment input. These southeast directed thrusts are also shown to migrate and become younger to the southeast, reflecting the development of coeval tectonic activity and accretion along the northwestern edge of the Kaapvaal Craton (Coward et al., 1995). Details of processes along the craton edges are still, however, poorly constrained at this stage. The 2785 Ma Gaborone–Kanye rapakivi granite–anorthosite–rhyolite complex, for example, is chemically anorogenic in character and its significance in terms of existing tectonic scenarios is not clear. Furthermore, recent sequence stratigraphic correlations, together with new age determinations in the northern Limpopo belt, suggest that the orogeny was longer-lived than previously thought, and that actual continent–continent collision post-dated 2500 Ma and may have been as recent as 2000 Ma (Cheney, 1995; Jaeckel et al., 1995). A considerable amount of work, especially on the sequence and timing of events along the margins of the craton, is required before the significance of the Limpopo orogeny with respect to Witwatersrand Basin evolution can be fully evaluated.

2.5. Metamorphism

Within 500 million years of its formation the Witwatersrand Basin had been progressively buried by more than 7 km of overlying sedimentary and volcanic cover, while at ca. 2050–2025 Ma the region was affected by a major thermal anomaly in the form of the Bushveld Igneous Complex and its associated satellite intrusions. In addition at around this time the Witwatersrand Basin was affected by a catastrophic tectonic event represented by the Vredefort dome, whose origin is contentious but which is now commonly held to be a meteorite impact site (Grieve et al., 1990; Leroux et
Fig. 6. Schematic representation illustrating the stages of tectonic evolution in the formation of the Witwatersrand Triad (after Robb et al., 1991). Sections do not have a specific geographic locality.
POST-DEPOSITIONAL EVENTS AFFECTING THE WITWATERSRAND BASIN


al., 1994). It follows that the Witwatersrand Basin will have been affected by both burial metamorphism as well as thermal perturbations and enhanced fluid flow. A good understanding of these events is critical to an assessment of the timing and extent to which primary Au–U mineralization in the basin has been modified by metamorphism.

A number of studies have recently been able to constrain the pressure–temperature conditions of metamorphism in the basin (Phillips, 1987; Wallmach and Meyer, 1990; Wallmach et al., 1990). It is generally agreed that peak metamorphism regionally attained conditions of about 350°C and 2.5 kb. What is less clear is whether the metamorphic climax coincides with burial or a thermal anomaly (or both), and what the timing of this peak condition was. Clues regarding post-depositional alteration and metamorphism in the Witwatersrand Basin are provided by the sequence of geological events that superceded basin formation and radiometric age determinations which reflect isotopic re-setting during a given event. A summary of post depositional events and age determinations pertaining to the Witwatersrand Basin is provided in Fig. 7. Witwatersrand deposition was terminated at 2714 Ma by extrusion of the Ventersdorp flood basalts, an event which was followed at around 2560 Ma by the-onset of lower Transvaal Supergroup deposition (Fig. 7). This was followed at about 2250 Ma with the deposition of the upper Transvaal sequence. The Bushveld Igneous Complex was then emplaced at 2050 Ma and this was closely followed by the Vredefort ‘event’ which is now accurately dated at ca. 2025 Ma (Fig. 7). A perusal of available age data from within the basin and its immediate environs indicates that, although errors are generally large, isotope systems have been reset at ca. 2500, 2300 and 2000 Ma. Authigenic pyrite and
rutile in the basin yields ages of 2500 and 2578 Ma, respectively (Giusti et al., 1986; Giusti, 1988; Robb et al., 1990a, b) whereas granites along the West Rand anticline appear to have had their Rb–Sr isotope systems reset at 2525 Ma (Barton et al., 1986). A second generation of pyrite and also the bitumen in the basin and surrounding granites yield ages of 2260 and 2320 Ma (Allsopp et al., 1986; Giusti et al., 1986; Robb et al., 1994). Pb–Pb and Rb–Sr isotope systems in the Vastersdorp lavas and tonalites of the Johannesburg dome have also been reset at 2230 and 2264 Ma, respectively (Van Niekerk and Burger, 1964; Barton et al., 1986; Walraven et al., 1990). Finally, uraninite in the basin has been reset at about 2040 Ma (Rundle and Snelling, 1977) and Witwatersrand shales have yielded K–Ar whole rock ages of around 1950 Ma (Layer et al., 1988). Clay mineral separates from the Vestersdorp Contact Reef have yielded K–Ar ages of 1888 and 2021 Ma (Zhao et al., 1995; Fig. 7).

The above ages either date authigenic mineral formation or record isotope resetting events and it is likely, therefore, that they also reflect metamorphism in and around the basin. If this is the case then at least three events can be recognized. The 2500 and 2300 Ma events appear to be related to progressive burial metamorphism caused by successive loading of the lower and upper portions of the Transvaal sequences. The 2000 Ma event probably is probably related, either to thermal perturbations caused by intrusion of the Bushveld Igneous Complex or, the catastrophic tectonism associated with the Vredefort impact event, or both. The correlation that exists between the reset ages and chronostratigraphic-dates for the region provides a framework within which assessments of the post-depositional redistribution of ores in the Witwatersrand Basin can be made. The significance of these “events” at ca. 2500, 2300 and 2000 Ma in terms of mineralization processes is discussed more fully in the following sections.

3. Mineralization processes

The economically important Witwatersrand reefs are contained within conglomerates of the fluvially dominated Central Rand Group (Fig. 1). This sequence comprises numerous unconformity or disconformity bounded stratigraphic units which accumulated in response to periodic tectono-magmatically induced uplift in the basin hinterland. The sedimentary packages are composed mainly of arenites, with lesser rudites, and were deposited in alluvial fan and alluvial braid plain environments. Ore bodies are associated with gravel facies and occur mainly in the form of very mature scour-based pebble lag and gravel bar deposits. The gravel facies is generally located on degradation surfaces, either on the basal unconformity of a genetic sedimentary package or on retrenched degradation surfaces within such packages (Minter, 1978, 1981). It is clear, however, that Witwatersrand conglomerates show a highly varied set of characteristics. A wide range of combinations of sand and pebble layers is present, extending from single layers of scattered pebble to well-packed thick conglomerates which tend to contain internal partings of pebbly pyritic arenite. In general, sedimentary features of Witwatersrand conglomerates can be expressed in terms of common lithologies in modern braided stream systems (Minter, 1978).

Most models of the origin of the Witwatersrand mineralization focus on host rock–ore relations and on the source, genesis and mode of occurrence of the most important ore constituents, namely gold, uraninite, carbonaceous matter and pyrite. The development of metallogenic models for Witwatersrand-type mineralization has been a polemic and continually contentious affair over the past century, and has recently been reviewed in detail by Pretorius (1991). Recent debate still revolves around whether ore constituents were introduced syngenetically with later modification and remobilization of ores (i.e. the “modified placer theory”; Robb and Meyer, 1991), or whether the bulk of the mineralization is entirely epigenetic with metals having been introduced by post-depositional fluids (Phillips and Myers, 1989).

The modified placer theory was substantiated by the detailed microtextural analyses of early petrographers (Liebenberg, 1955; Ramdohr, 1955), who postulated that sedimentological processes during basin deposition, as well as post-depositional modification, had played major roles in localizing ore constituents. Although many of the major ore minerals, including gold, pyrite and uraninite, may be recognizably allo-genic (Schidlowski, 1981; Hallbauer, 1986; Minter et al., 1993), about half of the more than 70 ore minerals observed display textural relationships which are
inconsistent with a detrital origin and indicative of precipitation from a hydrothermal fluid. Genetically, the observed diversity of ore textures has been explained in terms of a three stage paragenetic sequence, comprising initial deposition of detrital minerals, followed by authigenic pyrite formation and a final stage of gold remobilization and secondary sulphide mineralization (Feather and Koen, 1976). It is this approach which is described and expanded upon in the following sections.

3.1. Syn-sedimentary mineralization

Numerous quantitative sedimentological studies have demonstrated a relationship between sedimentary characteristics and the presence of mineralization, and widespread use is made on the gold mines of quantitative sedimentology as a tool in the prediction of the economic potential of conglomerates (Minter, 1978, 1981; Smith and Minter, 1980; Buck and Minter, 1985; Els, 1990). A correspondence exists between mineralization and sedimentary features such as unconformities, fluvial channelways, cross-bed foreset laminae and deflation surfaces. The highest potential for the concentration of detrital heavy minerals (including gold and uraninite) is in those conglomerates which have the best packing, most resistant clasts, and the highest abundance of pyritic foresets. Sedimentary processes can, in many but not all cases, also explain the widely observed gold–uranium correlation in conglomerates and associated arenites and the fact that uranium concentrations are highest in the relatively distal parts of the braided fluvial system. The physical processes responsible for the concentration of gold and uranium were more complex than those predicted by simple Stokesian settling behaviour, and the heavy and light detrital minerals are interpreted to have been deposited by complex interactions that probably included entrainment, saltation and other hydrodynamic dispersion processes.

In addition to the primary sedimentary dispersion patterns, other indications for a detrital origin of many Witwatersrand ore constituents are provided by micro-morphological and chemical studies which demonstrate that grain morphologies (including those of certain gold particles, uraninite and certain pyrites) are consistent with the shape of micro-nuggets which have been affected by abrasion during sediment transport and compaction during diagenesis (Hallbauer and Utter, 1977; Utter, 1979, 1980; Schidlowski, 1981; Minter et al., 1993). Detrital gold grains typically display flattened and edge-overturned (or peaned) morphologies and may be overgrown by authigenic pyrite (Fig. 8a). Uraninite is often concentrated at the basal contacts of conglomerates, or occurs as small subhedral grains in carbonaceous matter. In the first case, the abraded muffin-shape of individual grains is regarded as clear evidence for the allogenic nature of the mineral (Schidlowski, 1981). Radiometric dating of uraninite indicates ages in excess of 3 000 Ma (Rundle and Snelling, 1977; Saager, 1981). Mineral-chemical analyses reveal highly variable ThO₂/UO₂ ratios, suggesting that uraninite originated from granitic and pegmatitic source rocks, and could not have formed within the basin by low-temperature hydrothermal fluids, as the Th contents are too high (Feather and Glatthaar, 1987). Round compact pyrite and arsenopyrite is often oscillatory zoned in terms of its As content and these growth zones are truncated at grain boundaries indicating mechanical abrasion during transport (McLean and Fleet, 1990). In addition, radiometric ages of round pyrite, sphalerite and osmiridium grains are older than the maximum age of the basin, suggesting that they originated as discrete particles from an external source (Saager, 1981; Giusti et al., 1986; Hart and Kinloch, 1989). A typical allogenic assemblage comprising rounded, compact, pyrite, zircon and chromite is displayed in Fig. 8b and also schematically illustrated in the summary paragenetic sequence in Fig. 12a.

Finally, brief mention is made of the nature of the heavy mineral assemblage in Witwatersrand conglomerates compared to younger quartz-pebble conglomerates. The significance of the Witwatersrand assemblage, comprising pyrite–uraninite–rutile–low fineness gold, compared to the magnetite–hematite–ilmenite–high fineness gold assemblage in the younger Tarkwaian conglomerates, for example, is interpreted differently depending on whether ore genesis is viewed as epigenetic or syngenetic. Phillips and Myers (1989) have suggested that the dominance of sulphide minerals, especially pyrite, in the Witwatersrand conglomerates is the result of extensive post-depositional sulphidation of an original “black sand” assemblage comprising mainly Fe- and Fe–Ti-oxides, as at Tarkwa. Although evidence for sulphidation of detrital components does exist and has been reported by many workers, it cannot be used as an explanation for all the pyrite
Fig. 8. Photomicrographs in reflected light of mineral assemblages and textures in Witwatersrand conglomerate reefs. (a) Detrital gold grains (Au) showing ovoid, flattened and edge-overturned morphologies. One gold grain (upper left) is partially overgrown by authigenic pyrite (py). Width of field (WOF) = 0.2 mm. (b) Dominantly allogetic assemblage comprising pyrite, zircon and chromite. WOF = 2 mm. (c) Authigenic sulphide assemblage comprising euhedral pyrite (py), chalcopyrite (cpy) and pyrrhotite (po). WOF = 1 mm. (d) Gold particles (Au) adhering to the edge of a rounded, As-zoned, detrital pyrite grain. WOF = 0.2 mm. (e) Three grains of rounded, detrital uraninite (light grey) in association with rounded, detrital pyrite. Uraninite grains are partially overgrown and/or replaced by bitumen (dark grey). WOF = 0.2 mm. (f) Close-up view within a bitumen seam showing a thin filament of gold cutting through the latter. WOF = 0.1 mm.
types in the basin, and the major proportion of compact, round pyrite is regarded as detrital in origin. Indeed, the differences in the nature of detrital heavy mineral suite from one sedimentary sequence to the next, should be viewed in terms of the prevailing conditions at various times during the Archaean and early-Proterozoic evolution of the surface of the earth. In a recent reassessment Krupp et al. (1994) have suggested that the particular mineral assemblage of the Witwatersrand conglomerates (as well as other ore-deposit types) is a function of both atmospheric and hydrospheric conditions at the time of deposition. It is considered that the pre-2350 Ma atmosphere was essentially oxygen free \((P_{O_2} < 10^{-10} \text{ bars})\) and comprised mainly CO\(_2\) \((P_{CO_2} < 1 \text{ bar})\) and N\(_2\), with minor H\(_2\)S \((P_{H_2S} = 10^{-7.5} \text{ bars})\) and H\(_2\); rainfall and surface runoff under these conditions was acidic \((pH < 4)\), giving rise to pronounced chemical weathering in the continental domain. During sediment deposition under these conditions, river waters were within the pyrite stability field but outside the stable region for Fe-Ti oxides, alluvial gold would retain its high silver content, and uraninite solubility would also be very low in the extremely anoxic surface environment (Krupp et al., 1994). Subsequent to 2350 Ma, however, the production rate of photosynthetic O\(_2\) would begin to exceed that of volcanic-derived H\(_2\)S such that the atmosphere would progressively have been purged of the latter; at higher \(f_{O_2} ( > 10^{-30} \text{ bars})\) Fe-Ti oxides would have been stable and the Ag content of Au-Ag solid solutions would be reduced by complexation with chloride under these moderately oxidizing conditions. The heavy mineral assemblages of pre- and post-oxyatmospheric inversion quartz-pebble conglomerates would, therefore, appear to be consistent with atmosphere-hydrosphere evolution and derivation of part of the ore suite by weathering, release and preservation of primary particles from a suitable source area. In this light it would appear to be unnecessary to have to invoke different ore genetic concepts to explain the placer component of Witwatersrand- and Tarkwa-like quartz pebble conglomerates.

3.2. Post-depositional mineralization

Detailed mineragraphic and mineral-chemical analysis has shown that many of the constituents of the Witwatersrand reefs, including gold, pyrite, sphalerite, rutile and cobaltite appear as both allogenic and authigenic phases. Other mineral associations such as cobaltite–arsenopyrite–gersdorffite, ilmenite–rutile–leucoxene and uraninite–brannerite–uraniferous leucoxene represent paragenetic continua which span syn-genetic and epigenetic processes (Robb and Meyer, 1990). A number of sulphide phases, including chalcopyrite, galena, pyrrhotite, cubanite, mackinawite, millerite, proustite and tennantite, occur only as authigenic minerals and these display easily recognizable textural characteristics (Fig. 8c). Although detrital gold micro-nuggets are preserved in many reefs (for example the Basal Reef; Minter et al., 1993), it is nevertheless recognized that a significant proportion, perhaps even the majority, of gold grains display authigenic textures, probably as a result of post-burial dissolution and reprecipitation. It is difficult to evaluate the extent and magnitude of gold remobilization, but many workers have suggested that dissolution and reprecipitation took place virtually in situ (Liebenberg, 1955; Ramdroh, 1955; Schidowski, 1970; Frimmel et al., 1993; Meyer et al., 1994b). Remobilized gold is preferentially associated with paragenetically-late phases such as authigenic sulphides, kerogen and secondary chlorite, but also commonly occurs as fracture-fillings in, and as overgrowths on, allogenic pyrite which only rarely contains primary gold inclusions (Meyer et al., 1990a, b). There is also abundant textural evidence for the partial replacement of compact pyrite by secondary gold, a feature which is almost certainly the result of fluid related gold mobilization and reprecipitation.

3.3. Gold–pyrite association

Recent investigations have shown that zonation of pyrite with respect to As, or Ni and Co (which substitute for S and Fe, respectively) results in mixed, n–p type semiconducting properties (Möller and Kersten, 1994). In such pyrites, the As-rich zones represent p-type semiconductors (cathodes) whereas the As-poor (or Co- and Ni-rich) zones become n-type semiconductors (anodes). Experimental results have shown that at p–n junctions in a pyrite crystal, differences in electrical potential are sufficient to induce the electro-chemical precipitation of gold, even from very dilute solutions. The fact that round detrital Witwatersrand pyrite is often oscillatory zoned with respect to As
Fig. 9. Model showing the mechanics of electrochemical reduction of the Au(HS)\(^-\)\(_2\) complex at the surface of a mixed np-type pyrite. Modified after Möller and Kersten, 1994.

Au\(^+\) + 2HS\(^-\) + S\(_2\)O\(_2\)H\(^-\) → 2H\(^+\) + 2Au(HS)\(^-\)\(_2\)

(McLean and Fleet, 1990) and is also one of the preferred substrates for secondary gold deposition, suggests that gold precipitation may have been effectively enhanced by the semiconductive properties of pyrite (Meyer et al., 1994b). An example of gold particles adhering to the surface of a rounded, As-zoned, detrital pyrite grain is shown in Fig. 8d; an explanation of the mechanism whereby gold is electrochemically precipitated on to this type of pyrite is provided in Fig. 9. The close, primary spatial association of detrital gold nuggets with round pyrite in the conglomerates, combined with a mechanism for the effective electrochemical precipitation of gold which is subsequently taken up into solution, provides circumstantial evidence for a scenario where gold remobilization is restricted and is largely confined to within the conglomerates themselves.

**Uraninite–bitumen–gold association**

In addition to the gold–pyrite association, the intimate textural relationship between uranium, carbonaceous matter and gold (Fig. 8e and f) is also considered to be a result of hydrothermal ore modification. Uraninite occurs as round compact grains, of detrital origin, which can occasionally be seen as isolated particles within the quartz-pebble matrix, but which are more commonly enveloped and partially replaced by carbonaceous matter (Fig. 8e). The latter is referred to as bitumen since it is regarded largely as organic material that was mobile as a viscous liquid and has since solidified (Robb et al., 1994). The bitumen is present as seams which generally run along the basal contact of the conglomerate layer, but can anastomose through the pebble band and even bifurcate around pebbles. These seams invariably contain innumerable partially replaced and fragmented uraninite particles. Bitumen also occurs as isolated round nodules within which an association with uranium is less obvious. Gold is very often intimately associated with the kerogen seams both along the edges of and within the hydrocarbon (Fig. 8f). Texturally, the paragenetic sequence observed in the association comprises detrital uraninite, followed by post-depositional hydrocarbon fixation and uraninite replacement (Robb and Meyer, 1990; Robb et al., 1994), with subsequent gold mobilization and reprecipitation around the kerogen.

Evidence for the mechanism of hydrocarbon fixation is provided by Raman and Fourier transform infra-red (FTIR) microspectroscopy. Witwatersrand bitumen is characterized by spectra which show systematic peak variations that are proportional to both the uranium content of kerogen and the proximity of the point of analysis to a uraninite grain (Landais et al., 1989; Rochdi and Landais, 1991). Many other organic parameters, such as the C/H atomic ratio and the methyl/methylene ratio, also vary as a function of the radiation dose that the bitumen has received. This indicates that the organizational structure (degree of polymerization) and composition of the hydrocarbon is a function of the intensity of the contained radiation. The highly pleochroic halos that demarcate the alpha-radiation envelope around individual uraninite grains within the bitumen suggest that changes occur within the organic matter as a function of radiation damage. FTIR analysis of Witwatersrand bitumen along a traverse approaching a uraninite grain (Fig. 10) reveals a progressive loss of aliphatic and aromatic CH and a relative increase in concentration of C = C and C = O functions (Rochdi and Landais, 1991). This indicates that the organic material is heterogeneous and suggests that its characteristics, and hence its formation, were controlled by radiolysis. Stable isotope evidence nevertheless indicates that the bitumen is of biogenic derivation, originating from decomposed organic remains probably buried in the Witwatersrand sediments (Schidlowski, 1981).
Bitumen nodules which are morphologically similar to nodules in the Witwatersrand conglomerates, are observed in association with high-U phases from Archaean peraluminous granites adjoining the basin (Klemd and Hallbauer, 1987; Robb et al., 1994). Available U–Pb isotopic analyses of bitumen, sepa-
rated from both Witwatersrand reefs and adjoining granites, are indistinguishable, yielding imprecise upper intercept concordia ages of ca. 2330–2380 Ma (Robb et al., 1994). It is suggested, therefore, that the sediment- and granite-hosted hydrocarbons are coeval and related to an event of oil production at around 2350 Ma. At this time light hydrocarbons migrated through the sediments as well as into the underlying/adjacent basement granites where polymerization and hydrocarbon fixation took place in response to radiation accompanying the presence of high-uranium phases.

It is now suggested that much of the organic matter in the Witwatersrand Basin is bituminous, although some kerogen will have been preserved in the basin. The precipitation of gold around the bitumen must have occurred after hydrocarbon fixation, indicating that redistribution of metals also took place after the event of oil production at ca. 2350 Ma (see later). It is suggested that interaction of gold-bearing solutions with bituminous nodules and seams would have resulted in a change of the prevailing redox state to more reducing conditions, thereby facilitating the deposition of gold (Fig. 11c). This reaction not only explains the gold–bitumen association, but, because of the genetic link between uraninite and bitumen, also explains the regional correlation between Au and U. It is clear, however, given the original detrital association between gold and uraninite and the radiolytic accumulation of bitumen around uraninite, that post-depositional ore modification did not result in major segregation of the ore components. In other words, despite pervasive, hydrothermal dissolution and reprecipitation of ore components, the primary, sedimentary distribution of Au and U was not significantly disturbed.

Fig. 11. (A) \( \text{FO}_2 \)-pH diagram showing solubility contours for the \( \text{Au(HS)}^- \), \( \text{HAu(HS)_2}^- \), and \( \text{AuCI}_2^- \) complexes. The hatched field indicates Witwatersrand fluid conditions (modified after Robb and Meyer, 1991). (B) Gold solubility versus temperature diagram for the \( \text{Au(HS)}^- \), \( \text{HAu(HS)}_2^- \), and \( \text{AuCI}_2^- \) complexes calculated for pyrite–pyrrhotite (py, po) and pyrophyllite–muscovite (pyr, musc) controlled fluid \( \text{FO}_2 \) and pH, respectively. The curve marked \( \Sigma \text{Au} \) represents total gold solubility. Typical fluid conditions in Archaean lode gold deposits are shown for comparison (after Neall and Phillips, 1987). (C) \( \text{FO}_2 \)-pH diagram showing solubility contours for the dominant gold complexes (\( \text{HAu(HS)}_2^- \) and \( \text{AuCI}_2^- \)) at pH below neutrality. The arrow indicates decreasing gold solubility with decreasing \( \text{FO}_2 \) (more reducing conditions). Decreases in fluid S and Cl content would also result in lower solubilities.
3.4. Conditions of fluid related ore modification

The nature of authigenic sulphides and silicates, as well as gold, in the conglomerate reefs is important in assessing the role that post-depositional fluids have played in ore modification. In this regard, one of the pertinent questions to be asked is whether mobilization was due to liquid-state transport or solid-state creep. Quartz is not normally amenable to solid state mobilization, and the abundance of quartz veining and of secondary overgrowth of quartz on detrital grains further suggests that the dominant mobilization mechanism was liquid-state transport (e.g. Marshall and Gilligan, 1987). The involvement of fluids in metal transport raises the additional question of whether hydrothermal processes involved only components already present in the host rock, or whether introduction of additional ore components from outside the basin was involved. At this stage only preliminary statements can be made with respect to aspects such as the origin, nature, and physiochemical conditions of the fluid system, and of the solubility of metal in such fluids.

Fluid inclusion microthermometry and other temperature determinations based on stable mineral assemblages, mineral thermometry, magnetic blocking temperatures and ESR analyses of carbonaceous matter (Phillips, 1987; Layer et al., 1988; Wallmach and Meyer, 1990; Wallmach et al., 1990; Ebert et al., 1990; Meyer et al., 1990b, 1991; Robb and Meyer, 1991; Frimmel et al., 1993) provide a temperature range from < 300 to about 400°C, for both peak metamorphic conditions and fluid temperatures. Palaeofluids trapped in quartz veins cross-cutting conglomerase reefs comprise two distinct types. One is H_2O-rich and is in places saturated with respect to salts, and the other is CO_2-rich and contains variable amounts (up to 90%) of CH_4, N_2, and heavier hydrocarbons such as ethane (Robb et al., 1989; G. Drennan, pers. commun., 1994). Evaluation of fluid conditions and Au and U solubilities was carried out utilizing information from fluid inclusion microthermometry and stable ore and silicate parageneses, to constrain fluid pH and oxygen fugacities as well as S, K, and Cl contents (Robb and Meyer, 1991). Post-depositional aqueous fluids in the Witwatersrand Basin were typically acidic (pH 4–5 at 350°C) with oxygen fugacities pertaining to the field of pyrite–arsenopyrite–pyrrhotite stability (Fig. 11a). It is pertinent to note that, under these conditions, magnetite was not stable, a feature which explains the absence of this mineral, either in allogenic or authigenic form, in the Witwatersrand paragenesis.

These fluid parameters, together with solubility data for the $\text{AuCl}_2$, Au(HS)$^-$, HAu(HS)$_2$ and U(OH)$_4$ complexes can be used for the calculation of Au and U solubilities (Shenberger and Barnes, 1989; Wilde et al., 1989; Hayashi and Ohmoto, 1991). For gold it is not possible to decide whether sulphur or chlorine anions dominated in the complexing ligand, and in the discussion below the total solubility estimate is based on the assumption that both may have contributed. The results presented in Fig. 11a and b show that the total Au solubility, at a temperature of 350°C, was unlikely to have been more than 10–30 ppb. The calculated U solubility was around 20 ppb.

The estimate of Au solubility obtained is more than an order of magnitude less than typical values from Archaean lode gold deposits (Neall and Phillips, 1987; Fig. 11b). The relatively low solubility argues against an introduction of Au into the basin from an external source because such fluids would be unable to account for the enormous concentration of gold without invoking excessively high fluid fluxes. Although suggestions have been made that a fluid dominated system did apply to the Witwatersrand Basin (Phillips, 1988; Harris and Watkins, 1990), the widespread preservation of sedimentary features and the generally limited degree of pervasive alteration indicate that the post-depositional environment as a whole, with the possible exception of faults, was probably rock dominated. The widespread development of authigenic gold in the conglomerates, coupled with low estimated Au solubility in hydrothermal solutions, implies that fluid-related mobilization was restricted to environments where primary gold concentration had already taken place. Gold dissolution by hydrothermal solutions and subsequent precipitation requires fundamental changes in the prevailing thermodynamic conditions, such as S and Cl activities, $T$, pH, and redox state. The effect that a lowering of oxygen fugacity, or changes in S and Cl activities, has on the solubility of Au is shown in Fig. 11c. Precipitation of gold is likely to have been promoted by reduction and/or decreases in the activities of S and Cl, in addition to the electrochemical mechanisms discussed previously.
3.5. Paragenetic sequence of mineralization

Mineralization in the Witwatersrand Basin appears to have developed in response to four distinct paragenetic stages which are schematically illustrated in Fig. 12. Initial concentration of heavy minerals was controlled by sedimentological processes resulting in the formation of placer deposits (Fig. 12a). Fluvially deposited coarse clastic sediments are the preferred hosts to the significant economic concentrations of gold and uranium mineralization. There is evidence that the heavy mineral load of the original placer deposits comprised significant accumulations of gold, uraninite and pyrite, in addition to zircon, chromite and several other detrital phases.

The Witwatersrand sediments were subsequently blanketed by a succession of cover sequences in the form of the Ventersdorp and Transvaal Supergroups. This resulted in a progressive burial of the contained mineralization and its remobilization by at least three stages of authigenic mineral formation. The first, at around 2500 Ma (Fig. 12b), accompanied deposition of the lower portions of the Transvaal Supergroup, and is represented by widespread formation of authigenic pyrite (Fig. 8a and c). This was followed by a second event, at about 2300 Ma, accompanying deposition of the upper portions of the Transvaal Supergroup, which involved catagenic maturation of organic material in the basin, mobilization of an aquo-carbonic fluid through the basin and into the adjacent granitoid basement, and the radiolytic fixation of hydrocarbons to form bituminous seams and nodules around concentrations of detrital uraninite (Fig. 12c). Some metal redistribution will also have accompanied fluid circulation associated with this event.

The attainment of peak metamorphic conditions in the Witwatersrand Basin (350°C and 2.5 kb) is believed to have been associated with intrusion of the Bushveld Igneous Complex at 2050 Ma and the Vredefort catastrophism at 2025 Ma. These events resulted in further hydrothermal fluid flow through the basin and the formation of a variety of late authigenic sulphides (Fig. 8c) as well as the widespread remobilization of gold (Fig. 12d). Much of this gold appears to have been efficiently re-precipitated on to As-zoned pyrite or bitumen which formed during earlier stages of the paragenetic sequence.

During all the above episodes of post-depositional alteration, fluid conditions were such that the solubility of gold and uranium species was low but mechanisms for the re-precipitation of these metals were very efficient. Thus, a substantial input of metals derived from hydrothermal fluids originating outside the basin is neither necessary, nor feasible, as this would require enormously high fluid–rock ratios, for which there is limited evidence. Consequently, metal redistribution occurred more or less in situ resulting in the superimposition of both primary detrital and secondary authigenic mineralization.

4. Conclusions

The Witwatersrand Au–U deposits formed as a result of a number of coincident geological events which all contributed to creating a unique mineral province. Important amongst these were: (a) the primitive stage of development of the earth’s surface, where the atmosphere and hydrosphere were less evolved than at present; (b) a prolonged (360 Ma) period of episodic sedimentation punctuated by tectono–magmatic activity which introduced progressively more evolved granitoids into the source area; (c) the presence of a fertile and hydrothermally enriched source area capable of providing pyrite, uraninite and particulate gold into the adjacent depository; and (d) several phases of fluid related alteration subsequent to deposition which further concentrated authigenic mineralization within the conglomerates.

Deposition of the Witwatersrand Triad commenced with the outpouring of the Dominion volcanics onto pre-3100 Ma old Swazian granite–greenstone basement, during a period of extensional tectonism. Subsequent thermal collapse of this proto-basin and the onset of a compressional tectonic environment resulted in deposition of the West Rand Group. Much of the Witwatersrand sequence appears to have formed in a basin which formed in response to encroachment and eventual collision of the Zimbabwe and Kaapvaal cratons. During this time the continent was intruded by a succession of compositionally evolved (Randian) granites whose emplacement punctuated sedimentary deposition and stimulated further uplift and erosion. The Randian granites may have been geochemically anomalous with respect to Au and U and their intrusion,
Fig. 12. Schematic diagram illustrating the paragenetic sequence of mineralization in the Witwatersrand conglomerate reefs. (A) Primary accumulation of heavy detrital minerals by sedimentary processes to form fluviodeltaic placer deposits. (B) Development of early authigenic pyrite in response to sedimentary loading at ca. 2500 Ma. (C) Catagenic stage of organic maturation and circulation of hydrocarbon-bearing aqueous fluids. Subsequent radiolytic fixation of bitumen around detrital accumulations of uraninite at ca. 2300 Ma. (D) Circulation of (peak metamorphic?) hydrothermal fluids and precipitation of various authigenic sulphides and gold related to the Bushveld and Vredefort events at ca. 2000 Ma.
often at a moderate-to-high crustal level, stimulated further hydrothermal concentration of Au and U in the source area. The secular tectono-magmatic evolution of a fertile hinterland at the same time as basin deposition may be one of the most compelling explanations why the Witwatersrand palaeoplacers are concentrated in the upper half of the basin while the lower sequences are virtually unmineralized. Witwatersrand deposition was terminated by the voluminous Ventersdorp volcanism which covered the basin and preserved it from further erosion. Progressive burial and thermal/tectonic perturbations at ca. 2500, 2300 and 2000 Ma resulted in the development of a complex paragenetic sequence of authigenic mineralization which was itself largely restricted to the conglomerates within which the original detrital accumulation of ore constituents had taken place. Although this review provides circumstantial evidence, and favours the view, that the fluids and metals involved with authigenic mineralization were derived from within the basin, this remains to be demonstrated unequivocally and quantified. As long as the source of the prodigious accumulations of Witwatersrand gold remain enigmatic, the possibility of epigenetic processes, however tenuous, will exist as an option in metallogenic considerations.

Acknowledgements

Parts of this paper appeared in the proceedings of the XVth Congress of Mining and Metallurgical Institutions and we are grateful to the South African Institute of Mining and Metallurgy for agreeing to their incorporation into the present publication. We thank David Groves and an anonymous reviewer for considerably improving an earlier version of the text.

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