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Geochemical and Morphological Characteristics of Gold Particles from Recent River Deposits and the Fossil Placers of the Witwatersrand

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A relationship has been established between morphological features and fineness of gold particles and the distance over which they have been transported in recent alluvial placer deposits, such as the rivers of the Barberton Mountain Land, South Africa and the river Rhine in Germany. It was possible to show that most gold particles from the Witwatersrand conglomerates retained their detrital morphology and by comparing them with particles from recent alluvial gold deposits it was possible to estimate the distance of transport for the Witwatersrand gold, which in most cases ranged from 10 km to 30 km. Gold particles in recent placers show a characteristic increase in fineness with increasing distance of transport because of the leaching of the silver from them. The Witwatersrand gold particles on the other hand, have retained their primary fineness, because leaching of silver in the oxygen-deficient Precambrian atmosphere was not feasible chemically.

INTRODUCTION

Gold placer deposits, including fossil placers such as the Witwatersrand deposits, are the result essentially of the weathering and erosion of primary deposits and the re-distribution and concentration of the gold by alluvial, eolian and mass-wasting processes. During transport the original crystalline gold is deformed and comminuted on the stream bed in the frequently agitated gravels on the bed and to a lesser extent in the load suspended in the water above the bed (Parker, 1974). Transport is most rapid and comminution is greatest where gravel layers on the bed are thin or where there is no permanent gravel layer on the bed.

The decrease in particle size is rapid until equilibrium is reached between the gold particles and the stream and its load. Further decrease in size is then very gradual. The comminution of gold particles ceases when the particles have moved in the gravel layer below the zone of transportation. Some fine gold, however, is transported at all stages.

Because of its pronounced malleability gold acquires characteristic morphological features during transportation. Gold particles become rounded or flaky from the hammering action of the pebbles, and some flaky gold may be rolled and sandwiched into more compact particles. As the properties relevant to the hydraulic transport of the gold flakes change with the increase in their surface area these particles can be transported over a longer distance than rounded gold particles of the same weight (Ramdohr, 1965). There is a functional relationship between the shape and especially the flatness, of a gold particle and its rate of settling in water (Shilo and Shumilov, 1970).

Studies of the abrasion rates for gold in a simulated high-energy environment (Yeend, 1975) showed that the velocity of the particle appears to be a more important factor in the abrasion of gold than the distance of travel - a fourfold increase in velocity produced a tenfold increase in rate of abrasion of the gold.

Gold particles recovered from the Witwatersrand conglomerates by hydrofluoric acid (HF) treatment (Neuerburg, 1975) often display plate and nugget-like shapes (Hallbauer and Joughin, 1973; Hallbauer, 1974), and as will be shown later, have a remarkably well preserved surface texture similar to that found in recent placer deposits contrary to the commonly expressed opinion that the gold is no longer present in its original form (Feather and Koen, 1975).

The samples were not moved during the four to six week period of HF-treatment and tests showed that brief screening and even concentration in the micropanner produced no further noticeable scratches or deformation.

As an estimate of the distance that gold particles of the Witwatersrand have been transported is of great interest, an investigation was undertaken to study the relationship between the transport distance and changes in the surface morphology as well as changes in the chemical composition of gold particles during transport. Several samples of gold-bearing river sand were, therefore, taken from the Kaap River and its tributaries in the Barberton area of South Africa. Sampling was carried out at various distances from known outcrops of gold-bearing rocks. A sample of alluvial gold from the Rhine river near Karlsruhe, Germany, as an example of prolonged transport was used for comparison in the study.

The gold particles were examined in a scanning electron microscope (Leitz - A M R 1000), and their fineness[1] (Fisher, 1945) determined with the energy dispersive X-ray micro-analyser attachment.

DESCRIPTION OF SAMPLING SITES

The primary gold deposits in the Barberton area occur almost exclusively in the Archean greenstone belt of the Swaziland Super Group. This area is considered to be a provenance model of the source area for the Witwatersrand deposits (Viljoen and Saager and Viljoen, 1970; Saager, 1973; Köppel and Saager, 1974) and not as often misunderstood the direct source of the Witwatersrand detritus. Gold mineralisation is prominent along faults in shear zones, and is considered to be of hydrothermal origin, or in some cases, is of a stratiform, volcanic-exhalative character (Viljoen and Viljoen, 1969; Anhaeusser, 1976). Samples used in this study originated from the Blue Rock Reef of the Fairview Gold Mine.

The frequent occurrence of platinoid minerals and chromite in the Witwatersrand conglomerates points to the presence of ultra-basic rocks with associated chromite-platinum deposits in the original source area. As gold is found frequently as a minor constituent in the platinum deposits of the Merensky reef proper (Cousins, 1964), a sample of gold from that deposit recovered by the HF method was included in this investigation as an example of primary gold with microcrystalline texture.

The particles of alluvial gold from the rivers of the Barberton area can be seen as the direct erosion products of various primary gold deposits which were transferred to and then transported by the rivers in their bed load.

The sampling sites shown in Fig. 1 are described in more detail in Table 1.

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[1] $\text{fineness} = \frac{Au}{Au + Ag} \times 1000$

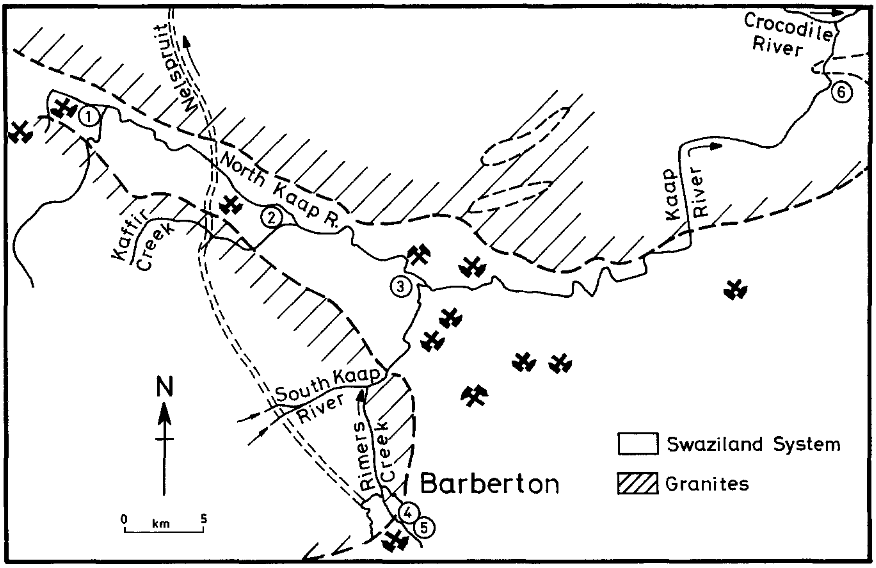

Fig. 1. Location of sampling sites in the vicinity of Barberton

The gold particles from the Rhine river were recovered from gravel deposits in the vicinity of Karlsruhe by panning. A small quantity of this gold was made available for this study by the courtesy of Prof. P. Ramdohr (Mineralogical Institute, University of Heidelberg, Germany).

The present concentration of gold in the younger unconsolidated sediments of the Rhine is the result of reworking the older river terraces. A relatively long distance of transport can be assumed. The primary deposits for most of the alluvial gold in the Rhine gravel are thought to be located in the upper regions of the Rhine, more than 150 km from the present locations of the alluvial deposits (Kirchheimer, 1965; Ramdohr, 1965).

MORPHOLOGY OF GOLD PARTICLES

Gold particles extracted by the HF method from primary gold ore in the Barberton area generally exhibited crystalline features. Although the overall shapes of the particles were jagged and irregular the surfaces showed the outlines of distorted octahedrons, often contorted and rounded. Twins after (111) were observed frequently, often arranged in irregular groups or distorted to form sheet-like aggregates. A typical particle of primary gold from the Barberton area is shown in Fig. 2.1 and 2.2. The overall diameters of these particles vary from a few micrometres to several hundred micrometres with an occasional large grain of up to five millimetres.

Gold in the Merensky reef occurs as relatively large grains (average 238 μm) which suggested early crystallisation (Vermaak and Hendriks, 1976). According to their morphology these gold particles can be divided into crystalline particles of the type described above and irregularly formed, particles with a spongy structure shown in Fig. 2.3 and 2.4. The latter are generally smaller than the crystalline particles. It appears that the morphological differences, as well as the greatly varying fineness of 680 to 940 point to two genetically different types of gold. This question, however, should be the subject of a separate investigation.

The preservation of at least part of the crystalline morphology is characteristic

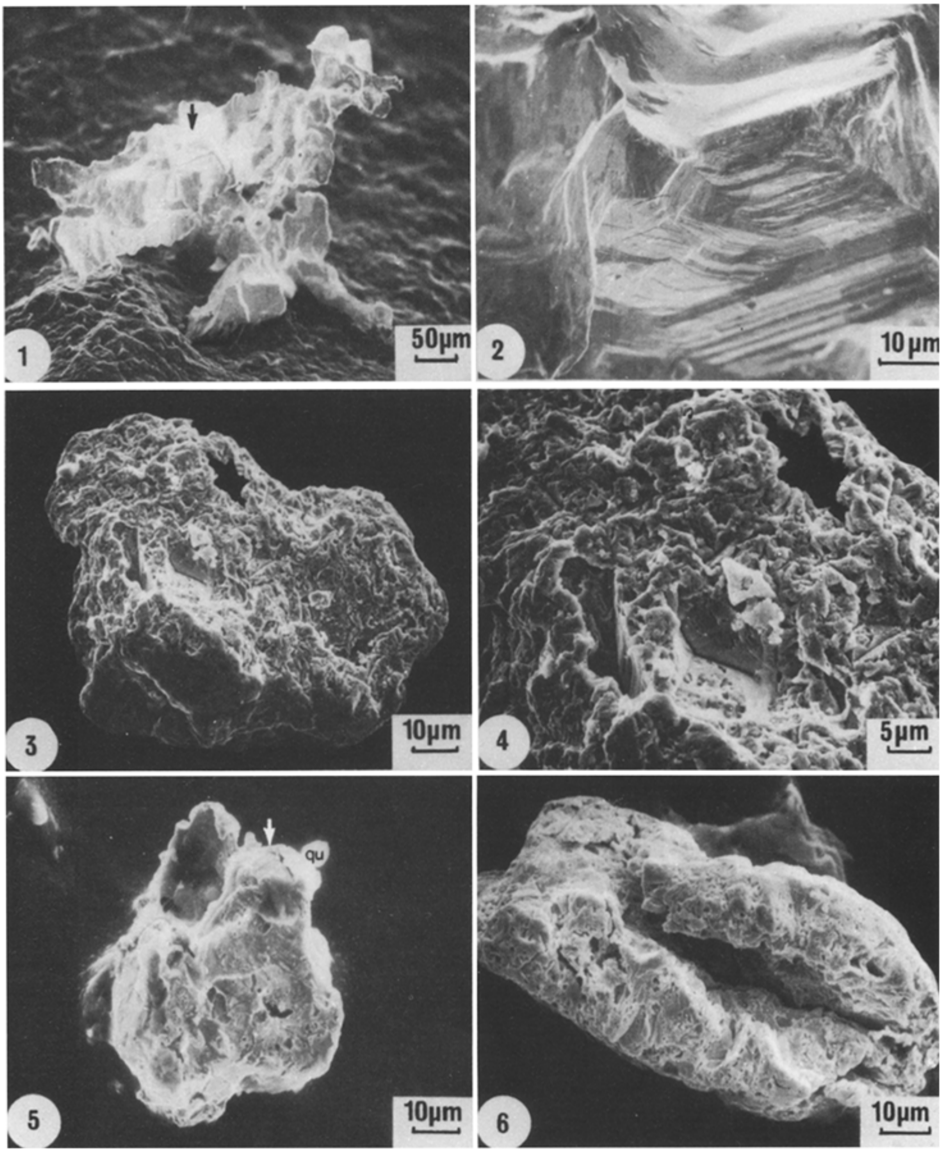

Fig. 2. Scanning electron photomicrographs of gold particles.
2.1 Primary hydrothermal gold particle from a sample of Blue Rock Reef, Fairview Gold Mines, Barberton.
2.2 Surface details of the above particle (indicated by an arrow in Figure 2.1) showing the crystalline nature.
2.3 Gold particle from the Merensky Reef, Rustenburg.
2.4 Surface detail of the above particle demonstrating the cryptocrystalline nature and spongy texture.
2.5 Detrital gold particle from a creek near an old gold mine in the Barberton area showing a "glazed" surface with remnants of the crystalline surface (arrow). Attached quartz grain indicated by "qu".
2.6 Sausage shaped, rolled gold particle with a spongy micro texture from locality 4

Table 1. Description of sampling sites in the Barberton area

Sample No.	Location	Probable origin and transport distance of gold
1.	Farm Waterfall 125, about 4km N of the Barberton-Kaapsehoop road, in a creek coming from NE and joining the North Kaap River	The spring of this creek is located about 4km NE of the sampling site. The old gold mines Great Britain and Condor are near its spring. Transport distance of gold is about 5km
2.	South bank of the North Kaap River, near Kaffir Creek	The gold may originate from any site within the catchment area of the North Kaap River including the vicinity of sampling site No. 1
3.	South Bank of the North Kaap River at the bridge of the Noordkaap-Consort Mine road	As for sampling site No. 2, but here a transport distance of a few km to 25km is possible
4. and 5.	Taken from a sluice in the Rimers Creek south of Barberton; No. 4 from the north sluice, No. 5 at the sluice	About 1km south of the sampling site the Rimers Creek crosses the gold occurrences mined Castalian Mine. Transport distance: 1km - 5km
6.	West-bank of the Kaap River NW of Greenstone station	These gold particles could originate from the primary gold-occurrences all along the greenstone belt southeast of the sampling site. Maximum transport distance: 80km

of alluvial gold particles transported for short distances in the rivers of the Barberton area. This ranges from undeformed crystalline features (Fig. 2. 5) to barely recognisable octahedral morphology (Fig. 3. 1). The overall shape of the particles is flaky and jagged with occasional particles becoming elongated and sausage shaped. A peculiar glazed surface with numerous irregular protuberances a few micrometres in size (Fig. 2. 5) is very common on gold particles transported for short distances. Similar characteristics were described for gold particles from rivers and sediments of the alpine molasse (Schmid, 1973). Possible explanations for this phenomenon include chemical reactions on the surface, leaching of silver and the associated local dissolution and the immediate precipitation of gold as described for alluvial particles from New Guinea (Fisher, 1945). The crystalline portion of the "glazed" surface of the grains shown in Fig. 2. 5. were analysed using the X-ray micro-analyser attachment of the scanning electron microscope (SEM). The fineness determined as 964 and 996, respectively, which supports the theory of a silver depletion during alluvial transport. Slightly deformed, spongy particles as shown in Fig. 2. 6 may originate from primary particles such as those shown in Fig. 2. 3.

The characteristics of particles transported up to 30 km include numerous scratches on the surface, bent and hammered edges (Fig. 3. 2) which sometimes enclose other mineral particles and occasionally exhibit remnants of the original crystalline structure (Fig. 3. 1). Flaky and rounded particles (nugget-shaped) are common. During prolonged transport the gold particles are deformed increasingly and acquire a dough-like microtexture with numerous scratches and abrasions on their surfaces (Fig. 3. 2 to 3. 4). Repeated folding of previously flattened particles occurs and scratches on earlier surfaces can be observed on the inner side of the folded edges. Where the overall shapes of the particles from rivers in the Barberton area are rounded and flaky, the grains are assumed to have been transported for 80 to 100 km.

Gold particles from the Rhine river can be taken as having travelled in a fluviatile environment for more than 150 km. These particles have been hammered characteristically, into thin sheets, often less than 10 μm in thickness, by the river pebbles (Fig. 3. 5). Repeated folding and bending produced thin gold particles consisting of several thin layers, an arrangement described as "sandwich-structure" (Ramdohr, 1965). The micromorphology is characterised by thin scales about 5 μm in diameter, crater-like indentations, and scratches in random directions (Fig. 3. 6). It can be assumed that the comminution which took place was caused by repeating thinning and folding.

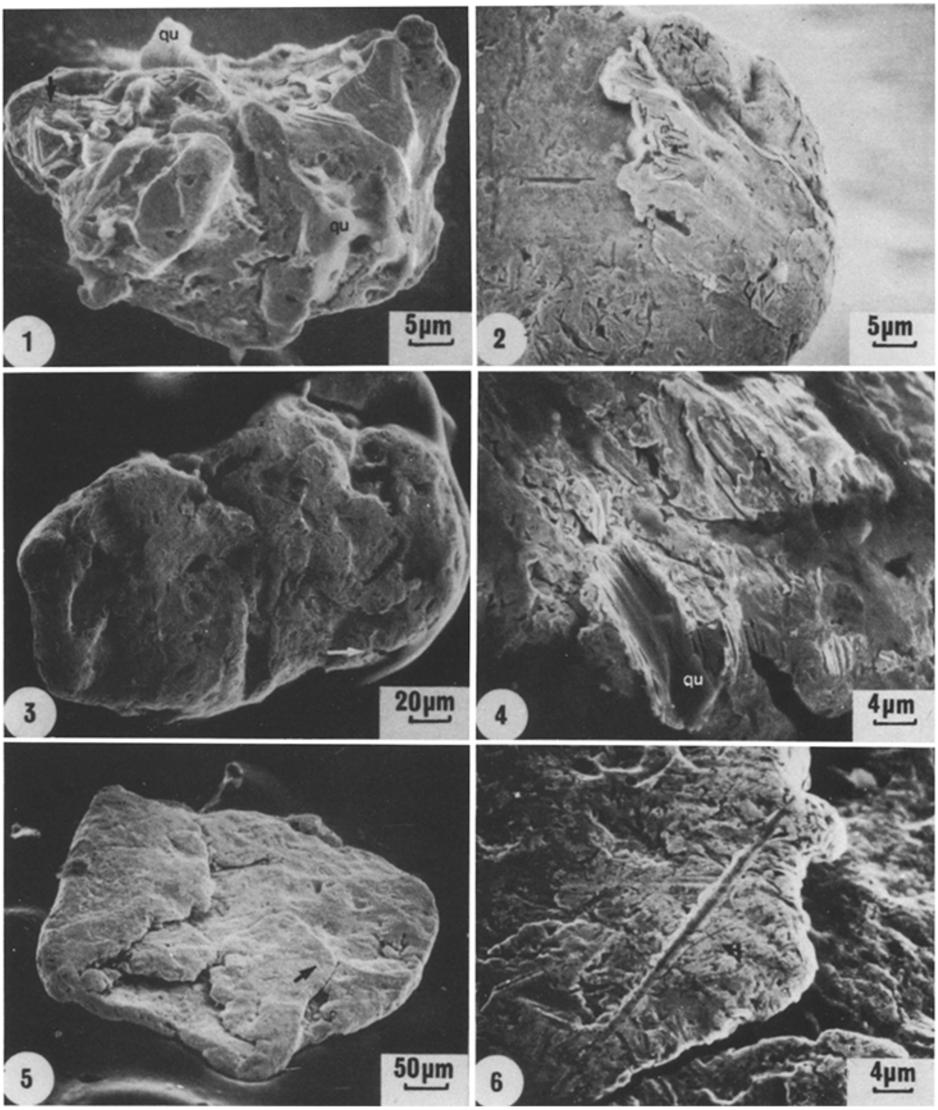

Fig. 3. Scanning electron photomicrographs of detrital gold particles.
3.1 Detrital gold particle from locality 2 with remnant crystalline features (arrow) but otherwise heavily distorted. Attached quartz grains indicated by "qu".
3.2 Gold particle from the Rimers Creek (location 5) demonstrating a dough-like micro texture after prolonged deformation and transport.
3.3 Gold particle from the Kaap River after more than 50 km transport.
3.4 The magnified surface of the particle in Fig. 3.3 shows the extensive gouging of the gold by quartz grains and the formation of a laminated texture. Attached quartz grain indicated by "qu".
3.5 Flaky gold particle from the Rhine river, Germany showing repeated folding of the flattened particles resulting in a sandwich-structure.
3.6 At higher magnification the scaly surface texture and the fine, randomly oriented scratches are evident

According to the modified placer theory (Ramdohr, 1958; Hiemstra, 1968; Liebenberg, 1973; Saager, 1973; Köppel and Saager, 1974; Pretorius, 1975), gold in the Witwatersrand conglomerates recrystallized during the slight metamorphism to which the rocks were subjected. Treatment with hydrofluoric acid of whole conglomerate specimens and subsequent microscopic examination of the liberated gold particles using the SEM showed, however, that most of the gold preserved its detrital morphology. Deformation caused by diagenetic compaction of the sediments are evident as minute indentations. It appears, however, that the framework of pebbles forming the reef protected the gold particles occuring in the interstices sufficiently well to prevent major deformation. Recrystallized gold particles have been observed occasionally but they have a different morphology (Hallbauer, in prep.). Gold contained within the carbonaceous matter often displays structures which strongly point to a mode of concentration by biological processes (Hallbauer and van Warmelo, 1974; Hallbauer, 1975). Detrital gold that was trapped by the original plant mat can, however, be observed frequently around and covering the columnar carbonaceous matter (Hallbauer, 1975).

The gold in Witwatersrand conglomerates can be divided into groups according to the degree of deformation sustained during transport as is the case with particles from recent alluvial deposits.

It is shown in the following section that leaching of silver can be excluded for gold particles transported in a Precambrian oxygen-deficient environment. The crystalline texture of the detrital gold particles in the Witwatersrand conglomerates is therefore not concealed by secondary surface formations but can be observed up to the stage of its destruction by mechanical deformation during transport.

Detrital gold particles carried in the bed load for short distances (perhaps up to 5 km) under these conditions are characterized by numerous randomly oriented scratches on the crystalline surfaces as the result of collisions with sharp-edged debris (Fig. 4.1 and 4.2). Protruding parts show plastic deformation caused by hammering and frictional movement against accompanying pebbles and bed rock. The gradual rounding and smoothing of the bed load constituents during continuous transport will result in a progressive decrease in gouging and scratching of the gold particles. As shown for gold particles from recent alluvial deposits, plastic deformation caused by hammering between pebbles and repeated folding are the major processes which influence the morphology of gold particles during prolonged transport in a fluviatile environment.

The early stages of such deformation can be observed on gold particles of the Vaal Reef taken near the entry point of the hypothetical river into the Klerksdorp fluvial fan (Fig. 5) (Minter, 1976). Although part of the crystalline texture is still visible (Figs. 4.3. and 4.4) the overall shape of the particles has been moulded by the hammering action of moving pebbles thus creating a dough-like texture and scaly surface. Gold particles equivalent to the flattened and completely deformed gold particles typical of prolonged transport in recent alluvial deposits were observed frequently in samples of Witwatersrand conglomerates (Figs. 4.5 and 4.6). They could be seen as indicators of a distant fan position or of prolonged transport if found near an entry point to the basin.

The examples shown in Figures 4.5 and 4.6 originate from the Basal Reef of the St. Helena Gold Mine near Welkom in the Orange Free State goldfield (Fig. 5), an area thought to be near the apex of the fluvial fan. These gold particles are accompanied by particles of the less deformed type shown in Fig. 4.3 and 4.4 as well as by detrital molybdenite, sphalerite, chalcopyrite, various types of pyrite (Saager, 1973; Hallbauer, 1974) and other heavy minerals. This simultaneous occurrence of two types of gold particles

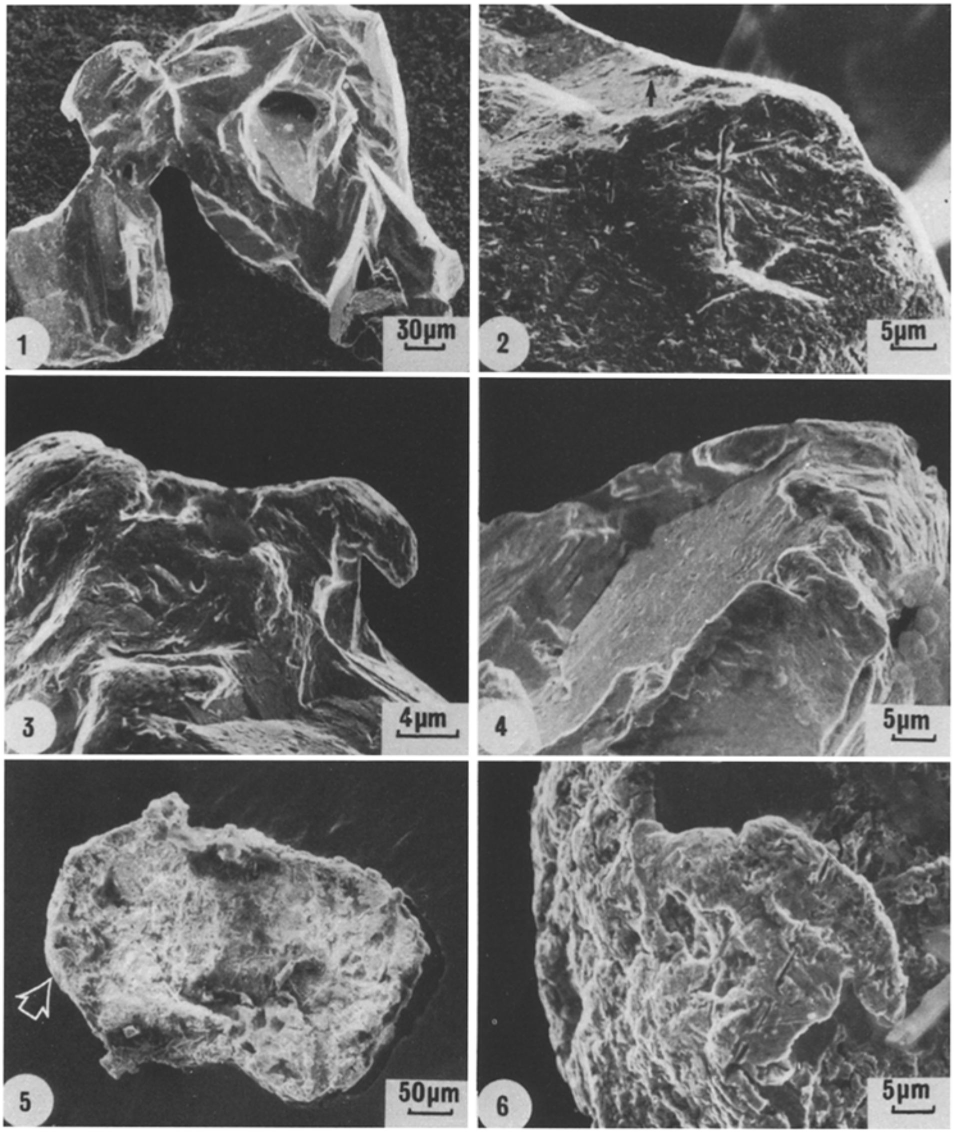

Fig. 4. Scanning electron photomicrographs of detrital gold particles from Witwatersrand reefs.
4. 1 Gold particle from the B-Reef, Lorane Gold Mine, Orange Free State, showing an overall crystalline structure.
4. 2 At higher magnification the particle of Fig. 4. 1 shows numerous scratches, and typical plastic deformation (arrow) indicating short transport in a fluviatile environment.
4. 3 Plastic deformation and remnants of crystalline structure on a gold particle from the Vaal Reef, Stilfontein Gold Mine.
4. 4 Plastic deformation on the protruding part of a crystalline gold particle from the Vaal Reef, Stilfontein Gold Mine.

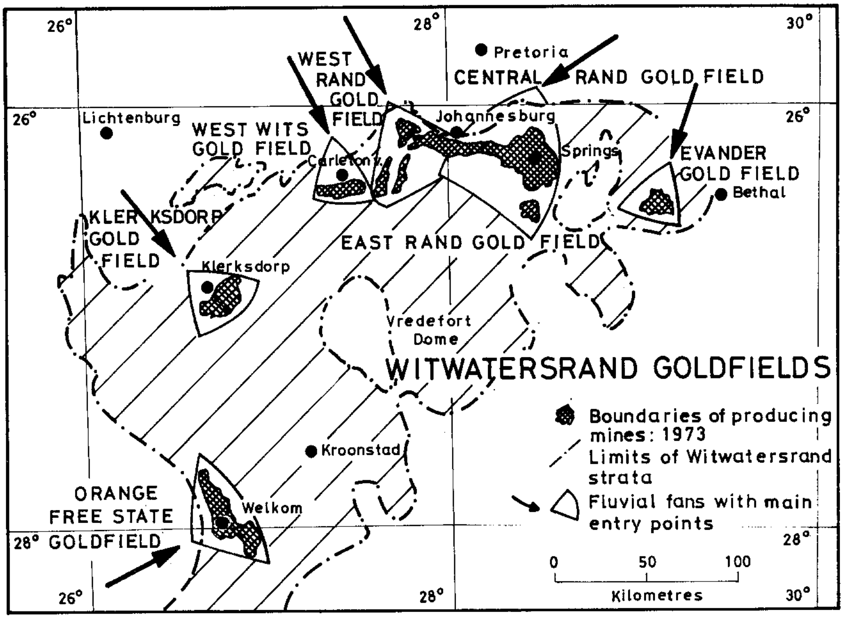

Fig. 5. Sketch map of the Witwatersrand Gold Fields showing the hypothetical fluvial fans and their entry points into the basin (Pretorius, 1975)

could indicate to different supply areas, near and far from the apex.

Gold particles of the type shown in Fig. 4.1 and 4.2 exhibiting only slight deformation were found to occur in abundance in the Elsburg reefs of the Loraine Gold Mine at the northwestern border of the Orange Free State goldfield. They are accompanied by pyrite "mudballs" (Hallbauer, 1974) shown in Figure 6 which, judging from the state of preservation of their delicate primary structures, can be taken as indicative of an arid or semi-arid environment (ephemeral streams) and the absence of major re-crystallisation processes after deposition. The location of such gold particles, which lack signs of extended transport, on the distal side of the hypothetical Orange Free State fan (Fig. 5) could be explained by a separate inflow into the basin at that point.

The overall picture that emerges after studying the surface morphology of a number of gold particles from different locations within the Witwatersrand basin is that the transport distances of consistently more than 100 km, assumed in several publications (Stumpfl, 1974; Feather and Koen, 1975), cannot be substantiated. On the contrary, short trans-

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4.5 Flaky gold particle from the Basal Reef, St. Helena Gold Mine, Orange Free State. The surface is partially obscured by fine pyrophyllite flakes and rutile needles. See Figure 4.6 for detail indicated by arrow.
4.6 Detail from Figure 4.5 showing the folded rim of the particle with the scaly surface texture and random scratches typical for prolonged transport (See Figure 3.6)

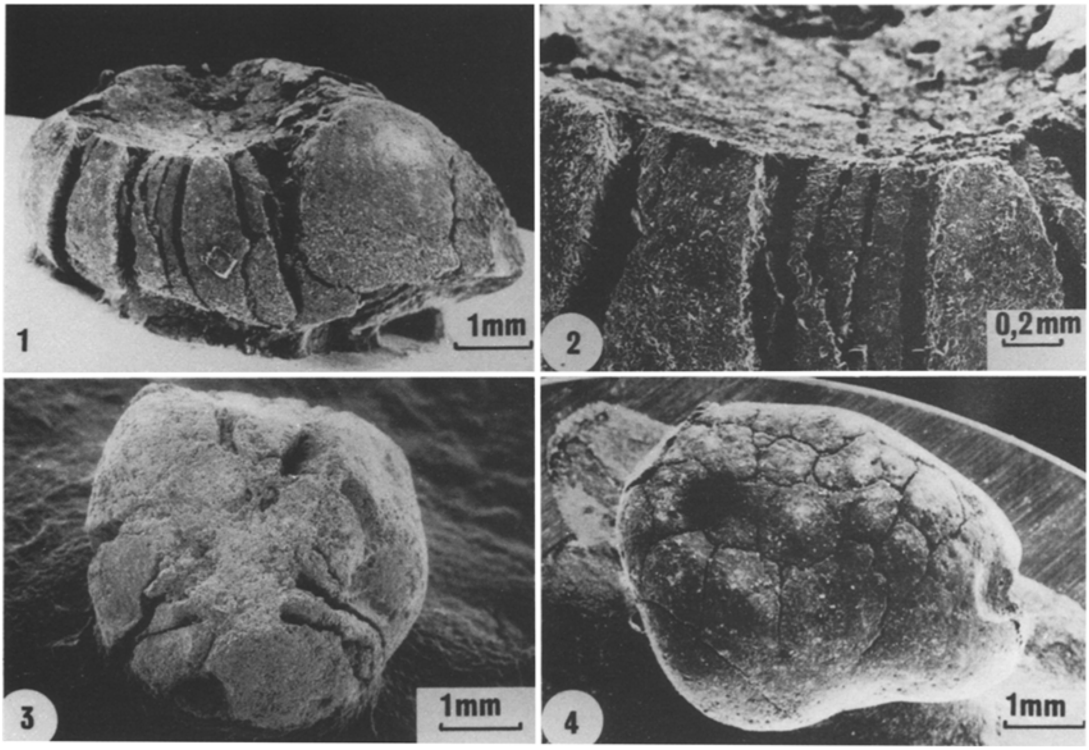

Fig. 6. Scanning electron photomicrographs of pyrite particles.
6.1 "Mudball" pyrite showing plastic deformation by pressure from an overlaying pebble.
6.2 Detail of Fig. 6.1 showing the preservation of delicate cracks caused by plastic deformation and cryptocrystalline texture of particle.
6.3 "Mudball" pyrite from the Vaal Reef, Stilfontein Gold Mine.
6.4 Polygonal cracks on one side of a "mudball" pyrite particle indicating a formation in an arid or semiarid environment

port distances of the order of 10 to 30 km are considered reasonable in a number of cases.

GEOCHEMICAL CONSIDERATIONS

The Precambrian atmosphere is regarded as having been deficient in free oxygen (Rubey, 1951; Abelson, 1966; Schidlowski and Junge, 1973; Cloud, 1976; Shimizu, 1976). In order for the uraninite present in Witwatersrand conglomerates to have survived as a detrital mineral the oxygen pressure in the Precambrian atmosphere must have been less than 10^{-2} of the present level (Grandstaff, 1974). The presence of photosynthesizing organisms as shown by the occurrence of fossilized plants (Hallbauer, 1975) points to an atmospheric oxygen pressure above the "Urey Level" at about 2.10^{-3} of the present one (Schidlowski and Junge, 1973). Judging from the stability of detrital pyrite and pyrite mud (Hallbauer, 1974; Utter, 1977) in the Witwatersrand paleo environment the oxygen fugacity in the surface water should be taken as $fO_2 = 10^{-70}$ (Drever, 1974). Under these conditions the leaching of silver from detrital gold particles as hypothesised (Saager, 1969; Saager, 1973) is not possible.

It is apparent from Table 2 that the only stable silver ion is Ag^+. As the Ag^{2+} ion is a stronger oxidizing agent than water it cannot occur in aqueous solutions.

Table 2. Standard electrode potentials for some reactions involving silver and water (Laist, 1954; Martell and Sillen, 1964)

Reactions	Potential (in volts)
$Ag^0 \rightleftharpoons Ag^+ + e^-$	+ 0,799
$Ag^+ \rightleftharpoons Ag^{2+} + e^-$	+ 1,98
$Ag^{2+} + H_2O \rightleftharpoons AgO^+ + 2H^+ + e^-$	+ 2,1
$2H_2O \rightleftharpoons O_2 + 4H^+ + 4e-$	+ 1,229

From the stability relations among various iron compounds, gold and silver in water (Tischendorf and Ungethüm, 1965) shown in Fig. 7, it is apparent that the stability field for Ag^+ is far removed from the stability field for pyrite and the "Sulphate-Sulphide Fence" (Krumbein and Garrels, 1952). The approximate localization of the Precambrian environment below this limit is supported by the occurrence of detrital gersdorffite, intergrowths of detrital gold and gersdorffite, and detrital chalcopyrite and cobaltite which were observed by the authors in a number of reefs. As shown elsewhere (Ling Ong and Swansen, 1974) the relationship in Fig. 7 demonstrates further that significant amounts of gold cannot be transported in solution in the oxygen-deficient Witwatersrand fluvial system as occasionally claimed (Pretorius, 1975; Reimer, 1975). The possibility of transporting gold in solution as AuS^- is confined to a strongly reducing environment containing HS^- concentrations of $> 10^{-6}$ moles per litre (Garrels and Christ, 1965) which could have been in existence in local stagnant pools but not in the general fluvial system.

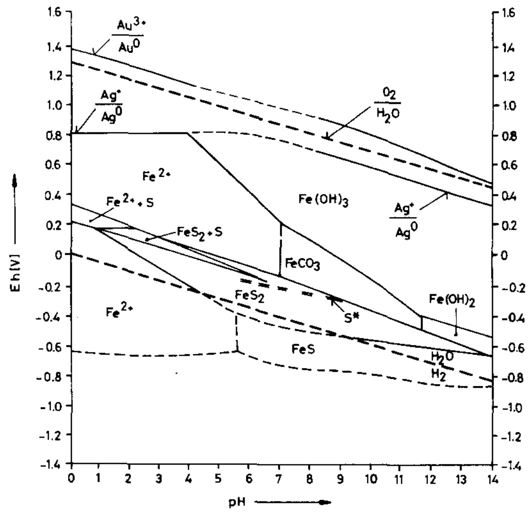

Fig. 7. Stability relations of some iron minerals, gold and silver in water at 25°C and 1 atmosphere total pressure. Total activity for iron minerals: $\Sigma Fe=10^{-6}$, $\Sigma S=10^{-2}$, $\Sigma [H_2CO_3]+[HCO_3^-]+[CO_3^{2-}] = 10^{-1}$ (Tischendorf and Ungethüm, 1965), S* = Sulphide - Sulphate - Fence (Krumbein and Garrels, 1952)

The transport of silver as AgCN [Ag (CN) $]^-$ or [$Ag(NH_3)_2$ $]^-$ can be ruled out as the presence of HCN, (CN_2) and NH_3 in the Precambrian atmosphere in significant amounts is unsubstantiated (Schopf, 1972). These geochemical considerations therefore show that the fineness of the detrital gold particles in Witwatersrand conglomerates which normally ranges from 870 to 960 can be taken as the original fineness.

The fineness of gold particles in the recent river sands from the Barberton area, however, was found to decrease with increasing distance from the known primary deposits as shown in Table 3. This is in accordance with earlier observations (Fisher, 1945) and the stability relations given in Fig. 7. The well aired river water has a high oxygen fugacity and provides the oxidizing environment necessary to dissolve silver.

Table 3. Change of gold fineness with increasing alluvial transport for samples from the Barberton area, South Africa, and for a sample of alluvial gold from the Rhine river near Karlsruhe, Germany

Locality	Approx. distance of transport in km.	Fineness of gold particles.	No. of Measurements.
New Consort Mine near locality 3 on Figure 1.	-	934	15
Fairview Mine	-	955	9
Nos. 4 & 5 on Figure 1.	1 - 5	921	9
No. 1 on Figure 1.	5	951	6
No. 3 on Figure 1.	25	962	4
No. 6 on Figure 1.	80	996	4
Rhine river at Karlsruhe, Germany.	>100	998	10

CONCLUSIONS

By comparing the Precambrian Witwatersrand gold particles with gold particles from alluvial placers it has been possible to show that the Witwatersrand gold has retained most of its original detrital morphology thus confirming the theory of placer origin of the Witwatersrand gold and ruling out large-scale recrystallisation and mobilisation.

As gold particles from recent alluvial deposits show characteristic changes in their morphology with increasing distance of transport, which can be related to the morphology of Witwatersrand gold, it is possible to estimate the distance that the Witwatersrand gold particles were transported of being in the range of 10-30 km.

Gold particles from recent alluvial deposits show a relationship between fineness and distance of transport due to the leaching of silver during stream transport. It has been shown, that this leaching of silver can only take place when sufficient free oxygen is present. Because of the absence of free oxygen in the Precambrian atmosphere it can be assumed that the Witwatersrand gold particles retained their original fineness during transport.

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