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The Witwatersrand pyrites and metamorphism

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ABSTRACT. Evidence obtained from morphological and extensive trace element studies, and from the examination of mineral and fluid inclusions in Witwatersrand pyrites, shows three major types of pyrite: (i) detrital pyrite (rounded pyrite crystals transported *into* the depositional environment); (ii) synsedimentary pyrite (round and rounded aggregates of fine-grained pyrite formed *within* the depositional environment); and (iii) authigenic pyrite (newly crystallized and/or recrystallized pyrite formed *after* deposition). The detrital grains contain mineral inclusions such as biotite, feldspar, apatite, zircon, sphene, and various ore minerals, and fluid inclusions with daughter minerals. Most of the inclusions are incompatible with an origin by sulphidization. Recrystallized authigenic pyrite occurs in large quantities but only in horizons or localities which have been subjected to higher temperatures during the intrusion or extrusion of younger volcanic rocks. Important additional findings are the often substantial amounts of pyrite and small amounts of particles of gold found in Archaean granites (Hallbauer, 1982) as possible source rocks for the Witwatersrand detritus. Large differences in Ag and Hg content between homogeneous single gold grains within a hand specimen indicate a lack of metamorphic homogenization. The influence of metamorphism on the Witwatersrand pyrites can therefore be described as only slight and generally negligible.

KNOWLEDGE of the gold distribution in Witwatersrand conglomerates is essential for effective mining operations. Its study requires an intimate knowledge of all mineral components in the reefs, in particular their genetic relationship to gold and their usefulness as markers of distribution patterns.

Of the ore minerals present in Witwatersrand reefs, pyrite is the most conspicuous, occurring typically in the matrix of the conglomerate, as layers on fore-sets within the conglomerate, and in the quartzitic rock of the footwall and hangingwall sequence. Rounded pebble pyrite (Ramdohr, 1958) constitutes the largest proportion of the pyrite present.

The commonly applied method of studying these pyrites is by means of polished sections in reflected light. Recently, however, the hydrofluoric acid (HF) leaching technique (Neuerburg, 1975) has been applied to the study of Witwatersrand pyrite (Hallbauer, 1977). In combination with scanning electron microscopy (SEM), it allows a more detailed and realistic division of Witwatersrand pyrite than before. Thus, a classification of the major types of Witwatersrand pyrite based on genesis as inferred from morphological characteristics (Hallbauer, 1982) has been suggested. The three main types are: (i) allogenic detrital pyrite; (ii) synsedimentary pyrite; and (iii) authigenic, post-sedimentary pyrite.

Recently, the question of the origin of the Witwatersrand pyrite has been revived, especially in the context of the evolution of the Earth's atmosphere. Pyritization of magnetite and ilmenite on a large scale has been suggested as the formation mechanism (Dimroth, 1979; Clemmey, 1981) so as to account for the apparent lack of black sand components in the Witwatersrand sediments; volcanic sulphur and the oxidation of detrital pyrite were proposed as sources of sulphur.

Ramdohr (1958) noted, however, that 'pyrite is present as indisputably abraded pebbles (in great abundance)' and that 'pure magnetite, hematite, and maghemite could not in all probability, produce relict structures. In fact, no forms have been observed which can unequivocally be traced back to them'. Our own results underline that pyritization of oxides can have had only a very minor effect on the amount of pyrite present.

Allogenic detrital pyrite. A study of hydrofluoric acid leach concentrates of Witwatersrand conglomerates shows that often more than 90% of the pyrite can be classified as abraded pyrite pebbles

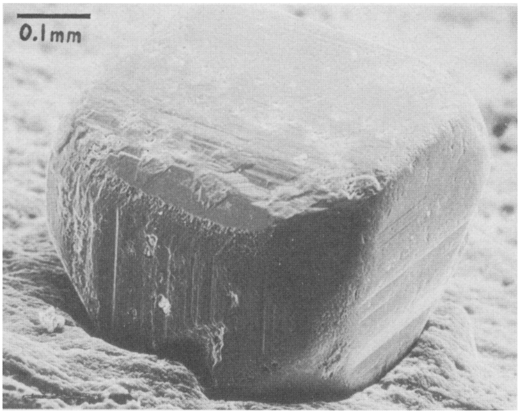

FIG. 1. Pyrite pebble, slightly rounded during fluvial transport. Ventersdorp Contact Reef (VCR), East Driefontein gold mine.

(fig. 1). There must, therefore, have existed a large source of primary pyrite, and a recent search has revealed the remnants of large bodies of Archaean metasomatically altered granite close to the Witwatersrand Basin. These typically contain up to 0.2% pyrite, no magnetite, little ilmenite, zircon, rutile and molybdenite, and occasionally sphalerite and traces of gold particles and uranium minerals. This discovery apparently solves 'one of the greatest difficulties of the placer hypothesis' (Clemmey, 1981), especially as it points to large volume sources devoid of magnetite. Details of the types of granite and their associated mineralization will be published in the near future (Hallbauer, in prep.). At this stage it suffices to report that a great many similarities exist between 'reef pyrite' and this primary pyrite in granite especially with regard to mineral inclusions, fluid inclusions and geochemistry (fig. 2).

The detrital pyrites in Witwatersrand sediments can be recognized as primary by a number of mineralogical and geochemical characteristics in addition to their abraded morphology which at the same time indicate either no, or a very limited, effect of metamorphism. Among the most striking characteristics of the detrital pyrites are their fluid and mineral inclusions.

Detrital pyrite grains from several localities across the Witwatersrand and from Archaean granites were selected for a study of fluid and mineral inclusions. From each sampling point, about twenty pyrite grains were taken from the hydrofluoric acid leach residue and then split and prepared for SEM observation (Hallbauer, in press). Several hundred inclusions were examined from each locality.

The minerals most frequently observed in polished sections as inclusions in pyrite include pyrrhotine, pentlandite, chalcopyrite, galena, ilmenite, rutile, and quartz. An examination of the fresh fracture surfaces of pyrite grains by SEM provides information on the morphology of the inclusions, their composition, and the composition of the condensed fluid from fluid inclusions (Hallbauer, in press).

The crystalline components observed with the SEM on fresh fracture surfaces can be divided into three groups;

1. Daughter minerals (Metzger *et al.*, 1977) in fluid inclusions. These are crystal phases that formed at temperatures lower than those at which the fluids were originally trapped, from the often dense fluids or later products of reactions between the pyrite mother crystal and the fluid (fig. 3*a*).
2. Crystals or crystalline aggregates of salts resulting from the drying of fluids (fig. 3*b*).
3. Mineral inclusions that are not visibly connected with fluid inclusions and can be classified as primary inclusions (fig. 3*c* shows ilmenite inclusions, clear evidence against 'pyritization' of oxides).

The results available so far from inclusion and trace element studies indicate that different types of allogenic detrital pyrite occur in Witwatersrand conglomerates. This finding supports the view that there were several primary sources that supplied detritus to the basin. The results indicate that the sources of the gold probably included gold–quartz veins, pegmatites, massive sulphide deposits, and, as bulk sources, mineralized granitic rocks.

Of special interest with regard to the survival of primary pyrite in Witwatersrand sediments during metamorphism are fluid inclusions of a high salinity; these would be vulnerable under conditions of high temperatures and pressures. Crushing and leaching of detrital pyrite from a locality in the Ventersdorp Contact Reef showed NaCl contents of up to 0.5% (of *total pyrite* by weight) indicating not only the high salinity of the fluid inclusions but also that this pyrite was not a product of pyritization.

Similarly, inclusions in pyrite of minerals which are not represented in the conglomerate matrix, such as biotite, orthoclase, albite, carbonates, and some ore minerals, point to an inheritance from primary sources. Another strong point in favour of the primary nature of one type of pyrite is the small but often numerous inclusions of rutile oriented parallel to (100) of the pyrite host. This type of inclusion was observed in hydrothermal pyrite from the Fairview Gold Mine in the Barberton Mountain land.

Geochemical features of the abraded detrital

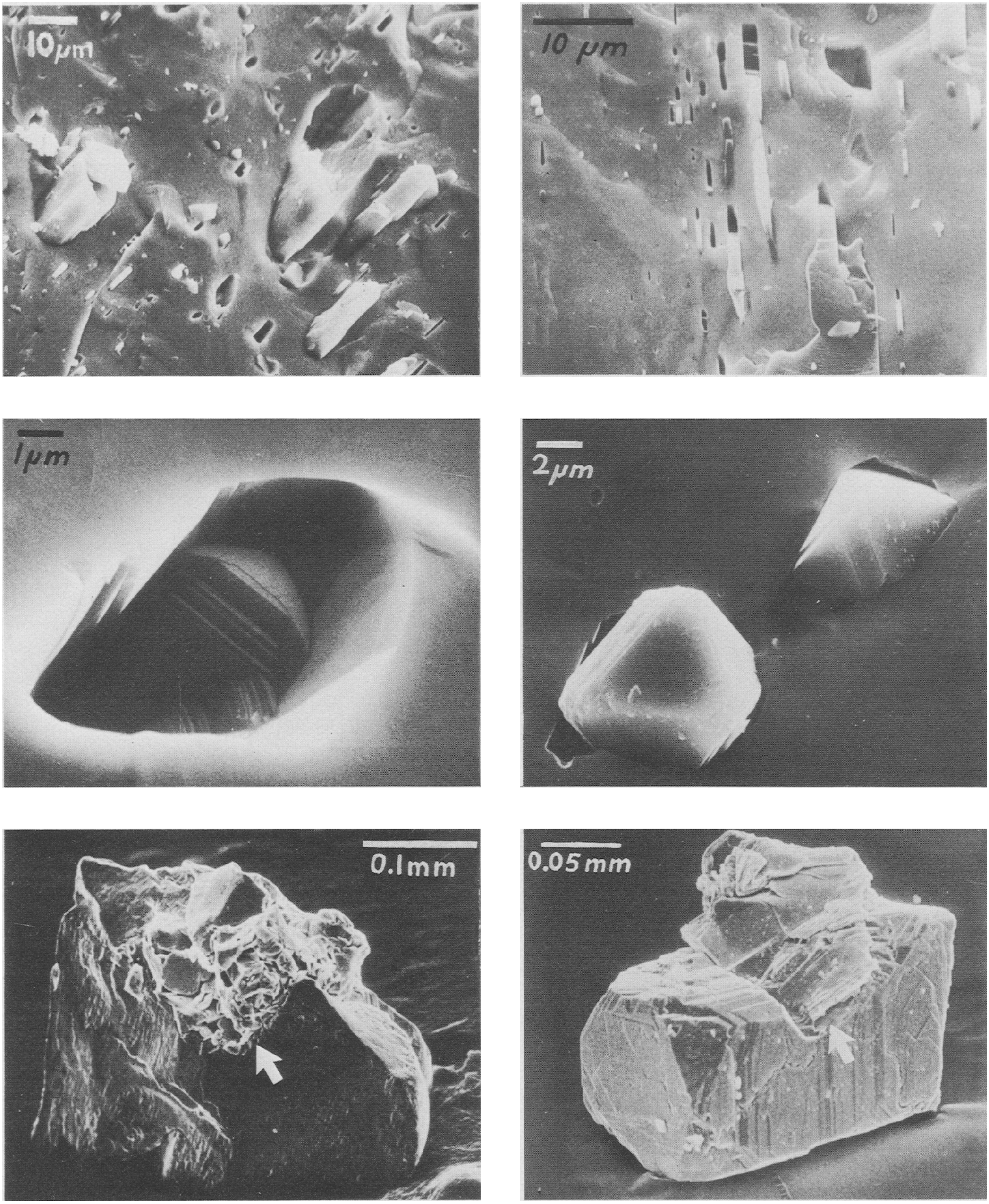

FIG. 2. Mineral inclusions in detrital pyrite from Witwatersrand conglomerates (left) and in pyrite separated from Archaean granites (right). Top—Inclusions of albite parallel to (100) of pyrite; Centre—Magnetite inclusions; Bottom—Pyrite intergrown with molybdenite (arrowed).

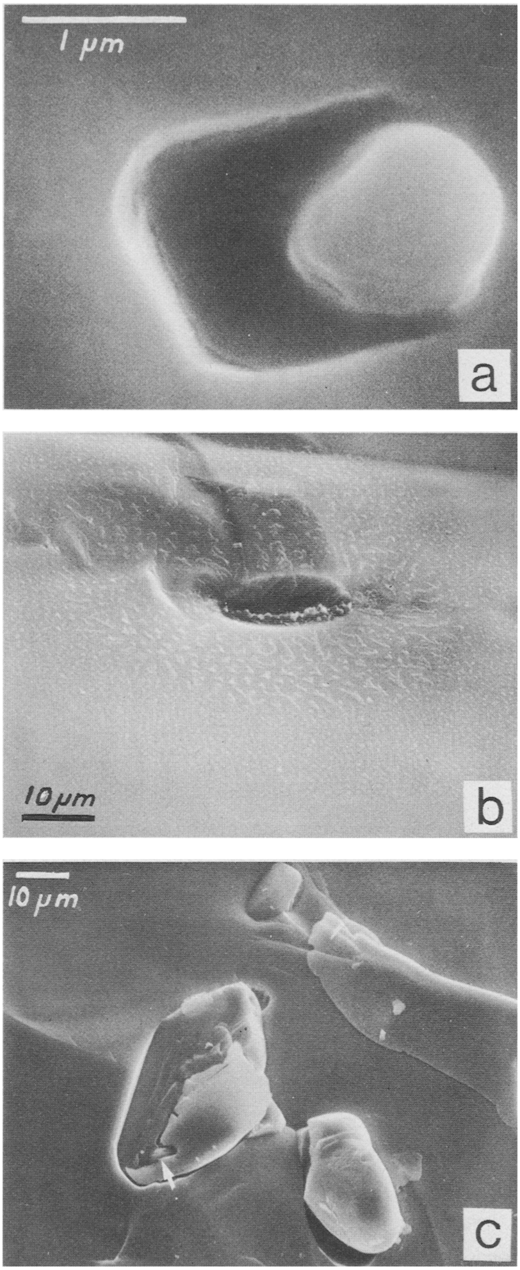

FIG. 3(*a*). Sodium chloride daughter crystal in the fluid inclusion void of a detrital pyrite. Carbon Leader Reef, Blyvooruitzicht gold mine. (*b*) Crust of dried salt solution around an inclusion void in a detrital pyrite from the Ventersdorp Contact Reef, East Driefontein gold mine. (*c*) An inclusion of chlorite (top centre) and two inclusions of ilmenite, one with biotite (arrowed), in a detrital pyrite from the Ventersdorp Contact Reef, East Driefontein gold mine.

pyrite which are neither compatible with formation during metamorphism nor origin by sulphidization are their trace element contents (Hallbauer, in press). The rather high tellurium contents (up to 200 ppm) in some detrital pyrites are, however, compatible with those found in pyrite from Archaean granites.

Synsedimentary pyrite. A large number of trace element determinations of the major pyrite types and their multivariate statistical analysis has shown a distinct geochemical difference between detrital and synsedimentary pyrite, which includes porous pyrite, oolitic pyrite and pyrite mud balls (Hallbauer and Utter, 1977). While detrital pyrite and authigenic pyrite show a slight overlap of their respective plots in the multivariate Mahalanobis diagram (fig. 4), synsedimentary pyrites plot in an area significantly removed from the others. This clearly indicates a different genesis for synsedimentary pyrite.

The apparent lack of hydraulic equivalence amongst pyrite grains of different types is not really an argument against the placer hypothesis (Clemmey, 1981) but a misunderstanding of the concept of hydraulic equivalence. This concept applies to static conditions in a settling tube but not to the dynamic conditions of fluvial transport. Although these conditions are not completely understood at present, they apparently allow different sizes to be deposited at the same locality due to variations in flow velocity, depth of water, and certain other parameters. Some synsedimentary pyrite (or 'buckshot pyrite' as it is commonly called) can be traced back to fine-grained muds. There is sufficient evidence to show that such pyrite muds were formed on the floor of abandoned channels (M. Nami, pers. comm.; Tucker, 1980). X-ray radiographs of rock slabs occasionally show mud

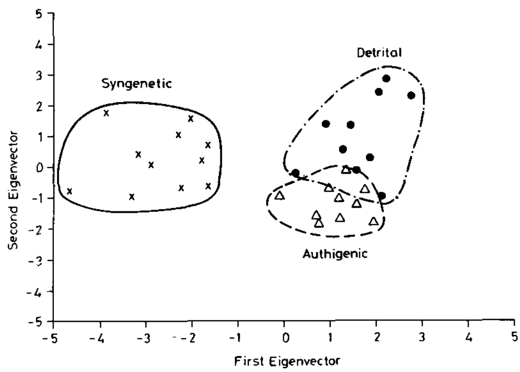

FIG. 4. Mahalanobis diagram showing the fields for the three major types of pyrite in Witwatersrand conglomerates.

flake structures in fine-grained pyrite layers (Hallbauer, 1974). It is conceivable that these heavy muds were dislocated during flash floods and that the mud balls (fig. 5*a*) subsequently formed (Karcz, 1969) were deposited after being transported for a short distance.

Due to their nature as heavy, fine-grained deposits, the pyrite muds contain other heavy minerals in fine-grained form including zircon, rutile, chromite and gold, and quartz and phyllosilicates. This explains the difference in trace element content between detrital and porous synsedimentary pyrite as discussed above. Arguments against the formation of the mud-ball-like pyrites by sulphidization of ferric clays (Clemmey, 1981) are their co-existence with clay clasts and the absence of only partially sulphidized clasts.

A synsedimentary origin can also be assumed for oolitic pyrite nodules and those of radiatingly structured marcasite appearance (fig. 5*b*). X-ray powder patterns of the latter only show the presence of pyrite. This could be taken as an indication of temperatures rising above the inversion temperature of marcasite (157 °C, after Rising, 1973) during metamorphism.

Apart from the general presence of large amounts of detrital pyrite in Witwatersrand sediments, the presence of pyrite mud, and mud balls particularly, implies an oxygen-deficient atmosphere during transport and deposition.

Post-sedimentary pyrite. Metamorphism and local heating events may produce, locally, growth of new pyrite. The complete absence of overgrowths in some deeply buried parts of the Witwatersrand, however, emphasizes the limited metamorphism in many parts of the basin. Almost complete recrystallization, as is very common in parts of the Ventersdorp Contact Reef, usually cannot be attributed to regional metamorphism but to local heat sources such as the overlying Ventersdorp Lava or intruding dykes. In places of extreme heating, an alteration of pyrite to pyrrhotine can be observed. Overgrowths on synsedimentary pyrite are randomly oriented according to the random nature of the aggregates, while those on detrital pyrite generally occur oriented crystallographically.

The mobilization of pyrite and its subsequent redeposition in small veins and veinlets can usually be directly related to a local rise in temperature caused by an intrusion of sills and dykes or to tectonic movements. Often this pyrite is accompanied by galena, sphalerite, and chalcopyrite, but rarely by gold. The detrital and synsedimentary pyrite in those parts of the strata which are a few metres away from 'vein' pyrite are generally not affected. An overriding influence of regional metamorphism on the formation of pyrite in veinlets can be ruled out.

Banded chert pebbles from parts of the Leader Reef (Welkom gold field) as well as in the Kimberley Reef of the Evander gold field (Hirdes, 1979), have been found to contain layers of pyrite. Hirdes also suggested pyritization of iron oxides in pebbles of banded iron formation during metamorphism in the Witwatersrand Basin.

It should, however, be kept in mind that some Archaean gold deposits, such as those from the Amalia greenstone belt in the Western Transvaal, are known to contain primary pyrite layers in banded iron formation. Such a deposit could have contributed detritus to the Witwatersrand.

Evidence from the composition of gold particles.

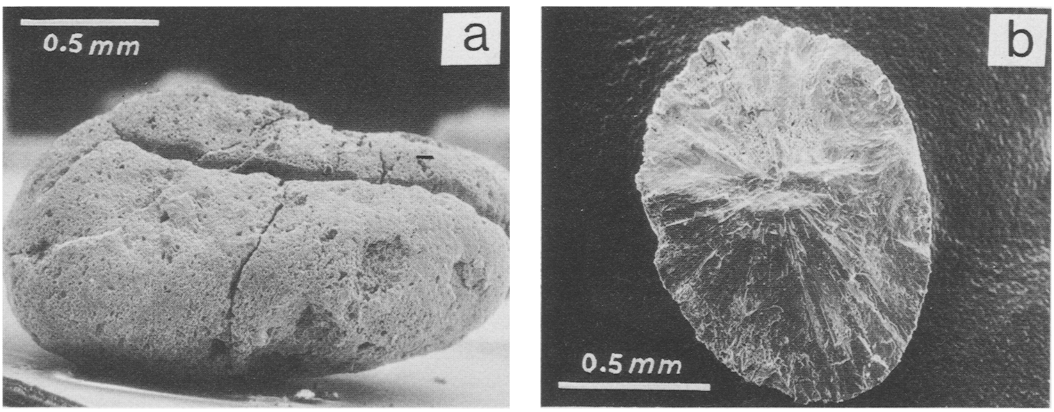

FIG. 5(*a*). Round aggregate of fine-grained pyrite with the morphology of 'rolled dough' as an example of a mud ball pyrite. Kimberley Reef, Vredefort area. (*b*) Cross section of a round pyrite pebble showing radiating lath-like crystals of marcasite morphology. Elsburg A12 reef, Loraine gold mine.

Ag and Hg contents of gold particles were determined in cross sections of single gold grains in concentrates from various Witwatersrand reefs, and Archaean hydrothermal samples from the Barberton area, using an electron microprobe. The results show that Ag and Hg are distributed homogeneously within individual gold grains but show distinct differences between populations of grains which may even occur within a single concentrate made from 1 to 2 kg of ore (fig. 6). This information strongly suggests that these element contents in Witwatersrand gold grains represent geochemical signatures inherited from their primary sources. It further implies that:

1. an oxygen deficiency existed during the transportation and in the depositional environment, as Ag would have been rapidly removed at least from the outer parts of gold grains under present-day conditions;

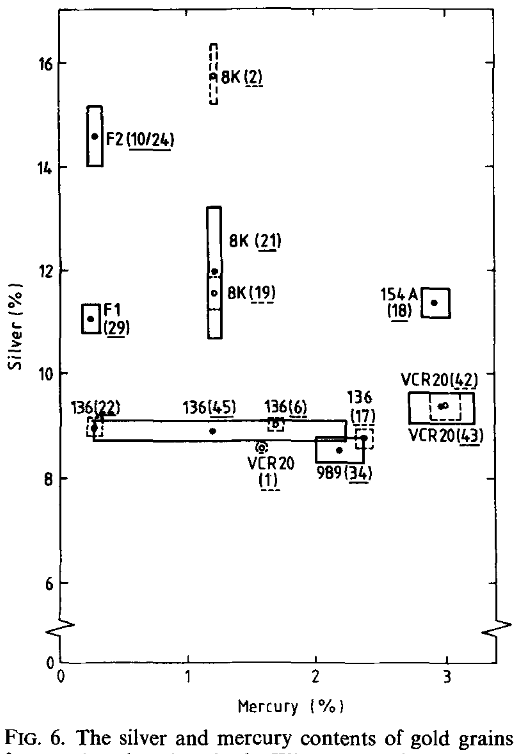

FIG. 6. The silver and mercury contents of gold grains from various locations in the Witwatersrand Basin (8K-VCR, Kloof gold mine; 136-B reef, Loraine gold mine; 154A-Basal Reef, St. Helena gold mine; VCR 20-VCR, Western Deep Levels gold mine; 989-Carbon Leader Reef, Blyvooruitzicht gold mine) and from Archaean primary deposits in the Barberton Mountain land, (F1 and F2-Fairview mine, Barberton). Circles and rectangles—means and standard deviations; figures in brackets—numbers of analyses. After von Gehlen (in press).

2. gold was transported and deposited in particulate form as silver and mercury would have been lost during any assumed solution stage;
3. the temperature during metamorphism cannot have reached the temperature at which mercury is released from gold (about 350 to 400 °C; Erasmus *et al.*, 1982); and
4. no major redistribution of gold took place either after deposition or during metamorphism.

Type of metamorphism. The mineralogical and petrological evidence points to the Witwatersrand metamorphism being essentially isochemical, that is without the supply of external material. A knowledge of the texture and composition of some mineral components in the reefs, and the measurement of fluid inclusion parameters, allow an estimate to be made of the temperature during metamorphism.

Although marcasite textures can be observed, all marcasite was found to have inverted to pyrite. The maximum temperatures during metamorphism should therefore have been between the inversion temperature of marcasite (Rising, 1973) and the decrepitation temperatures of the secondary fluid inclusions in Witwatersrand quartz pebble (Hallbauer and Kable, 1979) which is a range from 157 °C to 250 °C.

The assumption of an upper temperature limit of 250 °C is further supported by the presence of up to 17% volatiles in carbonaceous matter because these are expelled at above 250 °C from carbonaceous matter and coal (Karweil, 1956).

Conclusions. From the evidence presented it can be concluded that a division of Witwatersrand pyrite into different genetic types based on mineralogical and geochemical criteria is justified. It follows that these differences could not have been the result of a general sulphidization of 'black sand' components, and that the influence of metamorphism was confined to a comparatively minor recrystallization of the original detrital allogenic and synsedimentary pyrite.

The discovery, along the boundary of the Witwatersrand Basin, of remnants of Archaean granites containing disseminated pyrite mineralization and some particulate gold allows a comparison between detrital minerals in reefs and their possible primary counterparts. Apart from similarities between detrital pyrite in conglomerates and pyrite in granites, a strong geochemical relationship exists between the 'reef gold' and 'granitic gold'. Both types of gold have a similar high Hg content of about 1% or more, an almost identical range in Ag content, and are intergrown with pyrite, chalcopyrite and cobaltite-gersdorffite.

The uniform distribution of Ag and Hg in individual gold particles, and the variations observed between gold particles from the same locality within a reef, support the concept of non-homogenization during fluvial transport and metamorphism. The maximum temperature during metamorphism has been estimated as of the order of 250 °C.

Local heat sources, resulting from lava extrusions and intrusions, have had only limited, but locally and economically important, effects on the recrystallization and redistribution of gold and pyrite in Witwatersrand conglomerates.

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