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LEAD ISOTOPE EVIDENCE FOR THE
DETRITAL ORIGIN OF WITWATERSRAND PYRITES
AND ITS BEARING ON THE PROVENANCE
OF THE WITWATERSRAND GOLD

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ABSTRACT

The lead isotopic composition of sulphides from primary gold deposits of the early Precambrian greenstone belts of the eastern Transvaal, South Africa, is compared with the lead isotopic composition of detrital and authigenic pyrites from the placer gold-deposits of the Witwatersrand basin, to test the hypothesis that the detrital sulphides and the gold of the Witwatersrand sediments were derived from greenstone belts of the Swaziland Sequence.

The isotopic composition of the lead of some detrital pyrite samples from the placer deposits was found to be similar to that observed in sulphides from primary gold deposits. The lead of one of the detrital samples originating from the Orange Free State Goldfield is identical to the lead of galena from the Rosetta Mine, a primary gold deposit in the Barberton Mountain Land. The same lead also comprises the first stage lead of two other detrital pyrite samples obtained from the same locality in the Orange Free State Goldfield. These results provide conclusive evidence that the greenstone belts of the Swaziland Sequence and their numerous gold deposits contributed detritus to the Witwatersrand depository.

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1. INTRODUCTION

Since the discovery of the Witwatersrand goldfields the metallogenesis of the ores has been a controversial matter between the placerists and hydro-thermalists (Mellor, 1916; Young, 1917; Graton, 1930; Sharpe, 1949; Davidson, 1953, 1964; Antrobus, 1956).

Liebenberg (1955) and Ramdohr (1955), in their minerographic studies, independently demonstrated the placer nature of the gold and uraninite in the Dominion Reef and Witwatersrand sediments. These studies, as well as the geological and sedimentological data acquired by Brock and Pretorius (1964a), Winter (1964), Pretorius (1966) Armstrong (1968), and many others, led to the widespread recognition of the *modified placer theory* as a metallogenetic concept for the Witwatersrand ores.

The modified placer theory, which in principle had already been proposed by Mellor (1916), suggests that subsequent to their deposition some of the placer minerals - notably gold and some of the pyrite - were reconstituted during the low-grade metamorphic alteration of the embedding sediments and thereby lost their detrital characteristics.

Although the placer origin of the Witwatersrand ores is now generally accepted, the problem of the original source of the gold and the sulphides has never been investigated in detail. Brock and Pretorius (1964b) were among the few workers who gave some consideration to the problem of the provenance terrain of the Witwatersrand ores and sediments. Pointing to the structural and geological similarities which exist between the Rhodesian and the Transvaal-Orange Free State crustal fragments, they postulated that a goldfield analogous to the Rhodesian goldfield existed in the area now occupied by the Bushveld Igneous Complex to the north of the Witwatersrand. From this area, sedimentological processes would have transported the released gold, either directly, or in stages, into the Witwatersrand depository.

Saager (1970), synthesizing several studies on the Witwatersrand pyrite, concluded that the detrital pyrite originated from gold-quartz vein-deposits or massive sulphide ores; pyrite with detrital features may, however, also represent pseudomorphs after black sand constituents.

Using the recently published geological and mineralogical data on the Archaean granite-greenstone basement of the Rhodesian and Transvaal-Orange Free State crustal fragments and on the rocks of the Witwatersrand basin, Viljoen and others (1970) suggested that the primary source of the Witwatersrand gold would have to be sought in the greenstone belts of the Kaapvaal and Rhodesian craton. These greenstone belts are generally well-mineralized and constitute the most important source of gold in Southern Africa, excluding that recovered from the Witwatersrand goldfields. Using the greenstone terrane of the Barberton Mountain Land as a model source terrain for the Dominion Reef and Witwatersrand sediments, Viljoen and others (1970) demonstrated a high degree of correlation between expected placer minerals derived from successive erosional levels of the greenstone terrane and the actual placer minerals encountered in the Witwatersrand depository.

The present investigation was undertaken with the aim of testing the gold source hypothesis of Viljoen and others (1970). Ideally the test would have been accomplished by comparing the isotopic composition of lead in the gold from primary deposits with that in the gold from the Witwatersrand sediments in a manner similar to the study carried out by Antweiler and others (1972) who investigated the origin of the placer gold at Hahn's Peak, Colorado. However, the Witwatersrand gold has undergone recrystallization. This probably occurred during the sedimentational process or during the solution stage of the modified placer theory as is demonstrated by the low and uniform silver content (Saager, 1969) and by textural evidence (Liebenberg, 1955; Ramdohr, 1955). The lead in the gold can thus not be expected to be the same as in the gold from primary deposits. Furthermore the generally fine grain-size (< 0,5 mm) of the Witwatersrand gold and the immediate proximity of uranium bearing compounds renders an isotopic study of the lead in the gold impracticable. The test for the origin of the gold had therefore to be carried out by comparing the lead isotopic composition of pyrites from the Witwatersrand sediments with that of pyrites and lead-bearing sulphides associated with primary gold-

deposits in the Barberton Mountain Land. This study forms part of a more comprehensive mineragraphic and geochemical study of a large number of gold deposits in South African greenstone belts.

GEOLOGICAL SETTING

(a) The Granite-Greenstone Basement and its Associated Primary Gold-Deposits

The Kaapvaal and the Rhodesian cratons, forming a part of the ancient crystalline shield of southern Africa, consist of complex granite terranes, incorporating a number of tightly folded greenstone belts which, on the Kaapvaal craton, are collectively referred to as the Swaziland Sequence. The cratons have ages mostly in excess of 2 600 m.y. and are encircled by younger, often linear metamorphic belts (Limpopo, Mozambique, Natal-Namaqua belts) forming an unique pattern (Figure 1). In these mobile belts granite-greenstone material has repeatedly been metamorphosed between 2 000 m.y. and 500 m.y. (Anhaeusser and others, 1969; Saggerson and Owen, 1969).

Today most of the Kaapvaal craton is covered by younger, mostly sedimentary sequences. However, portions of the ancient granite-greenstone basement (Figure 1) occur fully exposed and preserved below the great escarpment in the Eastern Transvaal Lowveld, where the thick volcano-sedimentary assemblages or greenstone belts of the Swaziland Sequence form one of the most primitive rock systems developed on Earth (Engel, 1968; Anhaeusser, 1973).

In the Barberton Mountain Land, the largest and best documented of all the South African greenstone belts, three different divisions have been recognized. The lowermost, or Onverwacht Group, forming the base of the Swaziland Sequence has been described by Viljoen and Viljoen (1969a, c). It consists, in the lower portion, of mafic and ultramafic rocks mainly of subaqueous extrusive origin. The upper portion is characterized by a succession of mafic to acid volcanic cycles (Anhaeusser, 1971; Viljoen and Viljoen, 1969c). From a quartz porphyry in the upper division of the Onverwacht Group van Niekerk and Burger (1969) obtained a minimum U-Pb age of 3360 ± 100 m.y. on a total zircon sample.

The Onverwacht Group is overlain by the Fig Tree and Moodies Groups which consist mainly of argillaceous and arenaceous sediments.

The volcano-sedimentary remnants of the Swaziland Sequence are surrounded by a variety of granites which deformed the assemblages and produced the low-grade regional metamorphism that exists throughout the greenstone belt terranes. Based on U-Pb ages of sphene, apatite and zircon and on Rb-Sr whole rock ages, three major granitic episodes ranging in time from 3310 ± 40 m.y. (U-Pb) to 2550 ± 70 m.y. (Rb-Sr) can be distinguished (Allsopp and others, 1962, 1968; de Gasparis, 1967; Oosthuyzen, 1970). Each of these granitic episodes possesses its own distinctive tectonic style, mode of emplacement and geochemistry (Anhaeusser, 1969; Viljoen and Viljoen, 1969b).

The numerous mineralized gold occurrences in the Barberton Mountain Land and in other greenstone belts are restricted to the volcanic and sedimentary rock assemblages, the surrounding granites being devoid of any significant gold and sulphide mineralization. Gold has been found in gold-quartz veins, stratabound massive sulphide deposits and, more rarely, in disseminated complex sulphide-deposits in porphyry bodies. The processes which led to the formation of some of these gold-deposits have been discussed by Viljoen and others (1969) and Saager (1973).

(b) The Witwatersrand Basin

The fossil gold and uranium placers forming the Witwatersrand deposits are contained in conglomerate horizons in the lower portion of the Dominion Reef Sequence and in conglomerate and carbon reefs within the upper portion of the Witwatersrand Sequence. These two argillaceous to arenaceous rock sequences, together with the arenaceous and volcanic rocks of the Ventersdorp Sequence, are commonly referred to as the Witwatersrand Triad (Haughton, 1969).

From the whole rock samples of an underlying granite Allsopp (1964) obtained a maximum Rb-Sr age of 2820 ± 55 m.y. for strata of the Dominion Reef Sequence. The Ventersdorp Sequence at the top of the Witwatersrand Triad has been dated at 2300 ± 100 m.y. by the U-Pb method using zircons

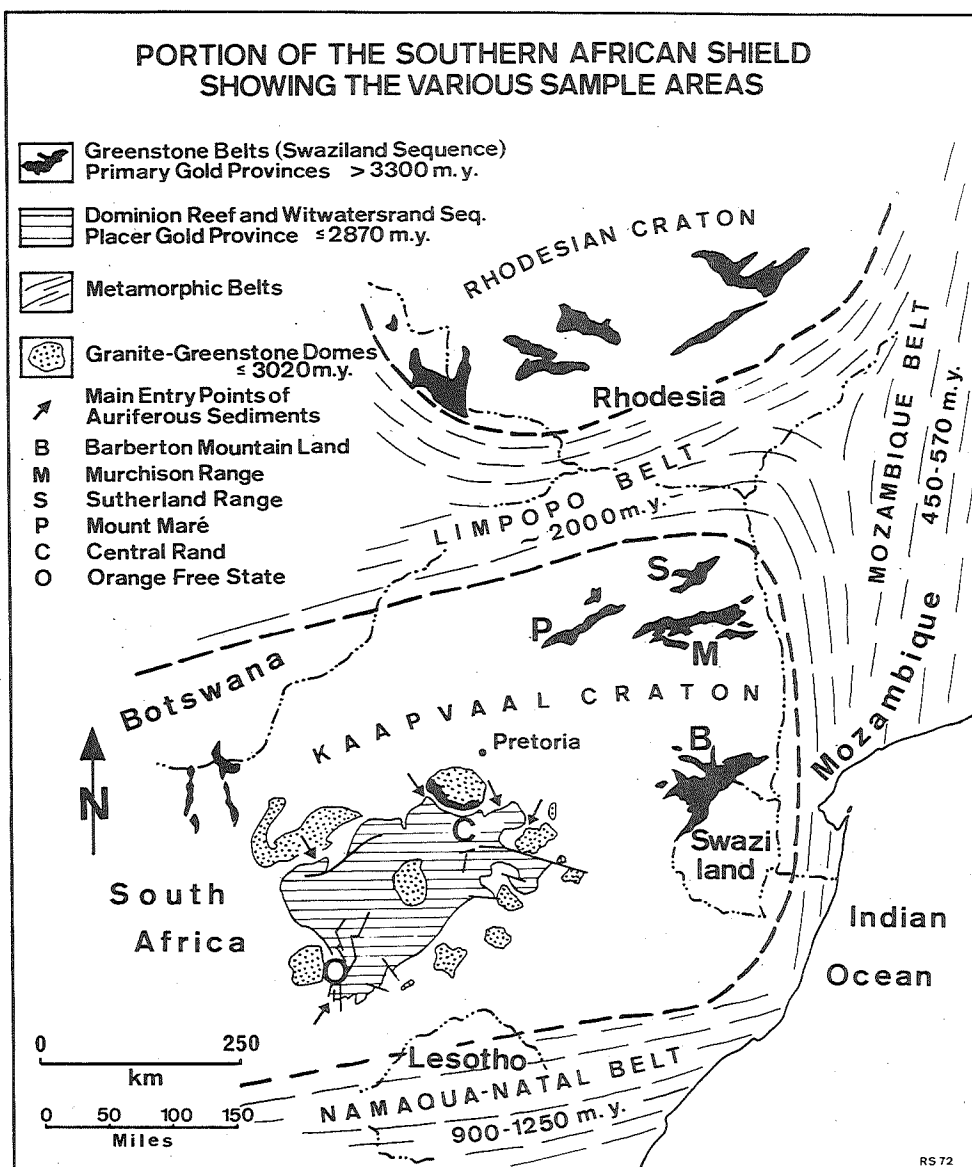


Figure 1 : Sketch map of the Kaapvaal craton showing the greenstone belts and the Witwatersrand basin.

from an interbedded quartz porphyry lava (van Niekerk and Burger, 1964). The age of the Witwatersrand Sequence, which is approximately 7600 m thick, lies somewhere between the two abovementioned values.

The rocks of the Witwatersrand Triad, together with still younger strata in the central portion of the Kaapvaal craton are flanked by a number of small dome-like inliers of granite-greenstone basement. These diapiric domes, at present either exposed or under cover, form part of the elliptical framework defining the depositional basin of the Dominion Reef and Witwatersrand sediments (Figure 1).

The sediments, despite their great age, show only the effects of low-grade regional metamorphism and mild deformation caused by epeirogenic movements (Brock and Pretorius, 1964b). Argillaceous sediments have been converted to compact shales and slates, while arenaceous sediments show effects of secondary silicification and pyrophyllitization (Young, 1907; Reinecke, 1927; Fuller, 1958). In general, the auriferous conglomerates are composed of pebbles of water-worn vein quartz, quartzite and chert, all of which are firmly cemented in a fine-grained matrix of quartz, sericite, phyllosilicates and opaques. Partial metamorphic reconstitution of certain sedimentary minerals can frequently be observed (Liebenberg, 1955; Ramdohr, 1955).

The area around the northwest flank of the Witwatersrand basin appears to have been a more active sedimentary source than the southeastern one. This is manifested by the location of almost all the payable gold and uranium along the northwestern arc of the basin where the mineralization occurs in fan-like areas or deltas. A number of sedimentological features led to the recognition of five main entry points of the sedimentary material (Figure 1) into the depositional basin along its northern, northwestern and western margin (Brock and Pretorius, 1964a, b). High energy environments close to the entry points of the deltas gave rise to typical conglomerate reefs whereas, in the distal facies regions of the deltas, low energy environments led to the formation of carbon-seam reef horizons (Pretorius, 1966). Generally, the overall coarseness of the sediments varies sympathetically with the grade of mineralization and the pay streaks (i.e. better developed conglomerates with higher gold values) radiate outwards from the entry points. These features indicate that the conglomerate constituents, including gold, uraninite and detrital pyrite, entered the basin at the same points.

In addition to the sedimentary criteria, the placer origin of most of the Witwatersrand ore-minerals has also been confirmed by numerous mineralogical observations. Some of these include : (i) the sympathetic correlation of gold and uranium (Liebenberg, 1955; Saager and Esselaar, 1969) (ii) the concentration of ore-minerals along the footwall contacts of the reef (Young, 1917) (iii) the abraded form of many pyrites (Becker, 1896; Ramdohr, 1955) (iv) the uniform silver content of the gold (Saager, 1969) (v) the occurrence of typical primary inclusions in abraded sulphides (Ramdohr, 1955) and (vi) the pressure solution effects on rounded pyrite grains (Schidlowski and Trurnit, 1966).

The structural development of the Witwatersrand basin, the situation of the main gold mineralization and the age relationship between the various early Precambrian sequences of the Kaapvaal craton, allow the conclusion to be drawn that the Witwatersrand sediments and the gold derived from the granite-greenstone basement immediately to the north and northwest of the Witwatersrand basin (Brock and Pretorius, 1964b). The exact provenance terrain is now virtually impossible to pinpoint precisely because of the ubiquitous cover of younger strata in these regions, coupled with the added possibility that the original gold-bearing source rocks may have been entirely eroded away. For these reasons Viljoen and others (1970), in their attempt to explain their gold source concept, focused attention on the Eastern Transvaal Lowveld where the granite-greenstone basement is fully exposed and well-preserved. They consequently used the Barberton Mountain Land, situated more than 300 km to the east of the basin rim, as a model provenance terrain for the sediments of the Witwatersrand Triad. It must, however, be emphasized that the actual mother-lode of the Witwatersrand gold, as it is the case with so many other placer-deposits, is still unknown.

3. MINERALOGY

In the gold-vein deposits of the Barberton Mountain Land pyrite is generally the most abundant ore-mineral and, particularly in the massive sulphide ores, it is the most important gold carrier. In all deposits pyrite forms up to several millimetre sized euhedral grains and is associated with varying amounts of pyrrhotite, arsenopyrite and magnetite. Chalcopyrite, sphalerite, tetrahedrite and galena occur almost exclusively as small inclusions in the pyrite and arsenopyrite. Uranium-bearing minerals have not been observed in any of the samples investigated.

The complex sulphide ore, in contrast to the vein and stratiform ore-types, contains high amounts of base metal sulphides and relatively minor amounts of pyrite (Plate 1H). More detailed mineralogical descriptions of the Barberton Mountain Land gold ores are available in reports by de Villiers (1957), Schweigart and Liebenberg (1966), and Viljoen and others (1969).

In the Witwatersrand conglomerates, pyrite constitutes, in many cases, over 90 per cent of the opaque minerals. Ramdohr (1955) and Saager (1970) attempted to classify the different pyrite varieties encountered in the Witwatersrand ores and were able to distinguish between the following types :

(i) Allogenic pyrite, which always has rounded, abraded, outlines is completely fresh and is generally less than 0,5 mm in diameter (Plate 1A, B, D). Besides the water-worn shapes, the presence of pressure indentations and the occurrence of primary gold inclusions possessing higher silver contents than the reconstituted gold in the conglomerate matrix provide additional evidence supporting an allogenic origin of this pyrite. Further criteria suggesting a detrital mode of deposition for the rounded pyrite have been presented by Ramdohr (1955), Viljoen (1964), and others. The primary inclusions in the allogenic pyrite grains are essentially the same as those in the pyrite of the primary mineral deposits of the Swaziland Sequence Greenstone belts.

(ii) Concretionary, authigenic pyrite with loose, skeletal structures and rounded outlines have diameters several times larger than those of the allogenic pyrite variety. Saager and Mihálik (1967) found that most concretionary pyrite aggregates consist of a complex intergrowth of at least two chemically different pyrite varieties. Since the extremely delicate nature of the concretions exclude any possible transport over long distances, these authors proposed an *in situ* formation "from Fe-rich silica gels by the addition of H_2S ". Numerous inclusions of gold and base metal sulphides indicate that the loose concretionary pyrite acted as a trap for the metamorphically mobilized ore minerals which infiltrated and filled the pores of the marginal areas of the concretions (Plate 1C, G).

(iii) Reconstituted authigenic pyrite occurs as euhedral grains in the conglomerate matrix or forms encrustations on detrital components and is, in rare cases, closely associated with enrichments of chalcopyrite, sphalerite and/or galena (Plate 1F). This pyrite variety was formed by reconstitution and remobilization during the metamorphic period. In certain places, dykes intrusive into the Witwatersrand rocks have influenced the content of authigenic minerals, but such effects are very local and do not account for the bulk of reconstituted sulphides in the Witwatersrand ores (Plate 1E).

4. SAMPLE PREPARATION

Samples were collected in different mines from various economic auriferous reef horizons of the upper Witwatersrand Sequence (for Sample locations see Table 3 and Figure 1). As several pyrite varieties usually occur irregularly distributed within the same sample, pure concentrates of one pyrite-type had to be obtained in the following manner :

Each hand specimen was cut into two sections, one of which was polished for microscopic investigation and the other put aside in case further sample material might be required. Samples displaying unusual concentrations of either detrital or concretionary pyrites were crushed, sieved and cleaned in a water stream. Thereafter, the pyrite concentrates were obtained by handpicking grains according to morphological criteria. The concentrates were treated with HCl and cleaned ultrasonically in distilled water until free of visible turbidity.

Polished sections were made of all concentrates and their purity examined under the ore-microscope (Plate 1B). Because of their small sizes difficulty was experienced in obtaining good samples of detrital pyrites and this placed constraints on the number of samples analysed.

The preparation of the pyrite samples collected from primary ore-deposits of the Barberton Mountain Land and other greenstone belts was essentially similar to that of the Witwatersrand samples. The larger grain-size and the presence of only one pyrite-type greatly facilitated the sample preparation permitting a far greater number of samples to be investigated.

Analytical Procedure

The pyrite concentrates were gently crushed in an agate mortar and leached in 4.5 n HCl at about 80°C for 20 minutes. The sample was then cleaned in distilled water for about one hour. Between 50 and 150 mg of pyrite were dissolved in 3 to 5 ml of 6 n HNO_3 . Thereafter, the solution

was dried and collected in 12 ml of 1.5 n HBr and 1 ml of 2 n HCl. This solution was passed through an ion exchange column holding 2 ml of Dowex 1x8 AG resin that was cleaned with 20 ml of 9 n HCl and 20 ml of H₂O, and prepared with 6 ml of 1.0 n HBr. The column was rinsed with 8 ml of 1.0 n HBr and 8 ml of 2 n HCl. The lead was eluted with 6 ml of 9 n HCl. The solution was brought to dryness and the evaporate redissolved in a few drops of 6 n HNO₃ and again brought to dryness.

Both the HCl and the HNO₃ were infrared distilled starting with suprapure reagents. The HBr was of suprapure grade. In the last stage of purification the water was also infrared distilled.

The lead contributed from the reagents was of the order of 1 to 2 nanograms. Total blanks ranged from 10 to 15 nanograms indicating a considerable contribution from the air. The blanks, however, were negligible compared with the amount of lead in the pyrite which, by means of atomic absorption analyses, were found to contain not less than 500 ppm Pb (i.e. larger than 1 microgram in the analysed sample).

The lead was analysed using the silica-gel method in a CH5-type mass spectrometer with two analysing tubes in tandem configuration and automatic peak switching. The signal was collected by a Cary 401 amplifier and recorded by a digital voltmeter that fed the data directly into a computer.

One analysis consisted of 3 to 4 sets of measurements taken at different temperatures between 1200°C and 1300°C, the temperature being controlled with the aid of an optical pyrometer. One set of measurements consisted of three sequences, each of which comprised ten measurements in the order : ²⁰⁸Pb-²⁰⁷Pb-²⁰⁶Pb, ²⁰⁶Pb-²⁰⁴Pb-²⁰⁴Pb, and again ²⁰⁸Pb-²⁰⁷Pb-²⁰⁶Pb.

Table 1 lists the raw ratios of the NBS common lead standard SRM 981 and equal atom lead standard SRM 982. The values for samples loaded directly onto the filament do not differ from those passed through the ion exchange column; the reproducibility is estimated to be ± 0.2 per cent. These measurements yielded the factors necessary to correct the isotopic ratios of the samples to absolute ratios of the NBS standards.

5. PREVIOUS ISOTOPIC AND GEOCHRONOLOGICAL INVESTIGATIONS

Before discussing the present results it may be useful to consider the isotopic evidence given by other authors regarding the age and history of minerals from the Witwatersrand Triad.

Nicolaysen and others (1962) reported U-Pb ages of detrital uraninites and monazites as well as of "total conglomerate" samples from the Dominion Reef Sequence. From a concordant uraninite age and from the linear array of discordant data points on a U-Pb evolution (concordia) diagram the authors concluded that the uranium minerals are 3080 ± 100 m.y. old. Burger and others (1962) reported lead isotopic compositions of Dominion Reef galenas which, on a ²⁰⁷Pb/²⁰⁴Pb versus ²⁰⁶Pb/²⁰⁴Pb plot, define a secondary isochron with a slope of 0.38 ± .01 indicating thereby a lead loss of the uranium minerals 2040 ± 60 m.y. ago. The period of alteration of the uranium minerals coincided in time with the formation of the Bushveld granite at Houtbeck, as determined by the U-Pb method on monazites (Nicolaysen and others, 1958).

Most of the uranium minerals of the Witwatersrand Sequence yielded discordant ages, some, however, being approximately concordant around 2000 to 2300 m.y. (Louw, 1954; Horne and Davidson, 1955). Wetherill (1956) concluded that most of the uranium minerals were formed 2040 ± 50 m.y. ago and that they suffered a recent lead loss. Burger and others (1962) also reported lead isotopic compositions of galenas from the Witwatersrand Sequence. Many of the data points again define a secondary isochron with a slope of 0.37 ± .01. The remaining points all lie below the secondary isochron. Assuming the 2040 m.y. event to signify a period of reconstitution of most of the uranium minerals - which also implies a period of lead loss - the slope of the secondary isochron indicates that the uranium minerals of the Witwatersrand Sequence are 2980 ± 100 m.y. old, i.e. equal in age to the uranium minerals of the Dominion Reef Sequence. The galena data points which lie below the best fit of the secondary isochron can be explained either by a later period of galena formation after a further lead loss of the uranium minerals, or by later additions of radiogenic lead with a ²⁰⁷Pb/²⁰⁶Pb ratio lower than existing galenas.

TABLE 1

Raw Data of NBS Lead Standards

1. NBS Equal Atom Standard, SRM 982

Date	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$
3.7.72	$0,9971 \pm ,0009$	$0,4664 \pm ,0006$	$36,59 \pm ,07$
3.7.72	$0,9992 \pm ,0007$	$0,4673 \pm ,0002$	$36,58 \pm ,21$
3.7.72	$0,9984 \pm ,0002$	$0,4668 \pm ,0001$	$36,64 \pm ,02$
8.8.72	$0,9975 \pm ,0001$	$0,4666 \pm ,00002$	$36,47 \pm ,13$
24.1.72	$0,9988 \pm ,0006$	$0,4666 \pm ,0006$	$36,66 \pm ,01$
Average	$0,9982 \pm ,0009$	$0,4667_5 \pm ,0003$	$36,59 \pm ,07$
Catanzaro and others (1968)	1,00016	0,46707	36,739

2. NBS Common Lead Standard, SRM 981

Date	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
15. 3.72	$16,870 \pm ,014$	$15,407 \pm ,010$	$36,447 \pm ,027$
15. 3.72	$16,878 \pm ,022$	$15,424 \pm ,018$	$36,485 \pm ,032$
22. 3.72	$16,885 \pm ,022$	$15,435 \pm ,017$	$36,541 \pm ,045$
3. 7.72 *	$16,895 \pm ,011$	$15,438 \pm ,015$	$36,504 \pm ,035$
3. 7.72 *	$16,879 \pm ,011$	$15,423 \pm ,016$	$36,477 \pm ,005$
24.11.72 *	$16,893 \pm ,006$	$15,431 \pm ,008$	$36,515 \pm ,030$
Average	$16,883 \pm ,009$	$15,426 \pm ,011$	$36,495 \pm ,032$
Catanzaro and others (1968)	16,937	15,491	36,724

Errors are 1 sigma errors.

* Sample passed through the ion exchange column

The galenas of the Witwatersrand Sequence having $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios plotting on the secondary isochron with a slope of 0.37 may be treated, for the purpose of this paper, as two-stage leads, although one is not justified in assuming that a sedimentary environment contains a single-stage lead at the time of its formation. However, quasi single-stage leads may result from mixing processes during the sedimentation cycle. Like Burger and others (1962) the present authors consider lead isotopic compositions plotting below the secondary isochron as representing three-stage leads.

From the foregoing observations it is possible to denote in a $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram an area encompassing the lead isotopic compositions of the authigenic minerals of the Witwatersrand Sequence. The secondary isochron, due to a first lead loss of uranium minerals 2040 m.y. ago, is the upper boundary. The lower boundary, due to further lead loss, starts at the lower intercept of the secondary isochron with the lead growth curve and has a slope of $0.124 \pm .004$ which corresponds to the present-day $^{207}\text{Pb}/^{206}\text{Pb}$ ratio of 2040 ± 50 m.y. old uranium minerals (Figure 2). The slope of the lower boundary, therefore, implies that after 2040 m.y. the uranium minerals only once lost lead, or in the case of a continuous lead loss, that the authigenic minerals of the Witwatersrand Sequence received only one addition of radiogenic lead.

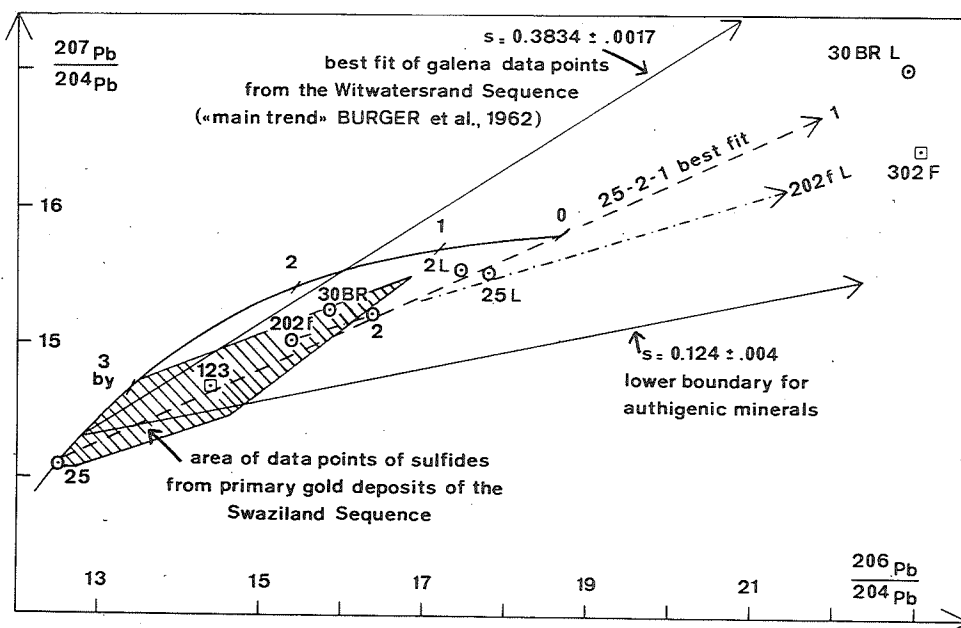


Figure 2 : $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram

The following parameters were used for calculating the growth curve :

$$(^{206}\text{Pb}/^{204}\text{Pb})_0 = 9.56 \quad (^{207}\text{Pb}/^{204}\text{Pb})_0 = 10.42$$

$$T_0 = 4.55 \text{ by} \quad ^{238}\text{U}/^{204}\text{Pb} = 9.0 \quad \lambda_{238} = 1.537 \times 10^{-10} \text{y}^{-1}$$

$$\lambda_{235} = 9.722 \times 10^{-10} \text{y}^{-1}$$

Circles denote detrital pyrites, squares denote authigenic pyrites from the Witwatersrand Sequence. "L" denotes the data point of the HCl leach of the respective sample. For further explanation see text.

6. RESULTS

The results are listed in Table 2 and plotted in Figures 2 and 3. The hatched area in Figure 2 contains the data points of sulphides from primary gold deposits in the major greenstone belts of the Swaziland Sequence. The data will be discussed in a separate paper (Köppel and Saager, in preparation.)

The upper boundary of the field in which the lead isotopic ratios of authigenic minerals from the Witwatersrand Sequence should plot has been recalculated using the isotopic ratios of Burger and others (1962), after correcting them on the basis of the measurements of the USGS lead standard reported by Catanzaro (1968). Samples 33, 34 and 35 of Burger and others (1962), i.e. the samples with the lowest $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios were not used for the calculation because of the possibility that they represent three-stage leads. These data points would shift the lower portion of the best fit line towards the right and thereby increase its slope. On the other hand, the galena samples 42 and 202 reported in this paper were included in the calculation, which was carried out by the method proposed by York (1966). The slope obtained for the secondary isochron, i.e. the upper boundary of the authigenic Witwatersrand sulphides is $0.3834 \pm .0017$ which is slightly higher than the

TABLE 2

Isotopic Ratios of Galenas and Pyrites from the
Witwatersrand Sequence and of the Rosetta Galena from the
Barberton Mountain Land

<u>Sample</u>	<u>$^{206}\text{Pb}/^{204}\text{Pb}$ *</u>	<u>$^{207}\text{Pb}/^{204}\text{Pb}$ *</u>	<u>$^{208}\text{Pb}/^{204}\text{Pb}$ *</u>
42, galena	26,478 \pm ,014	19,560 \pm ,007	34,82 \pm ,03
202, galena	41,16 \pm ,07	25,38 \pm ,04	34,97 \pm ,06
25, detrital pyrite	12,503 \pm ,010	14,095 \pm ,013	32,30 \pm ,02
25, leach	17,80 \pm ,08	15,52 \pm ,07	37,48 \pm ,17
1, detrital pyrite	37,43 \pm ,05	20,92 \pm ,05	34,23 \pm ,05
1, leach	33,21 \pm ,04	19,02 \pm ,04	35,44 \pm ,05
2, detrital pyrite	16,369 \pm ,025	15,216 \pm ,021	36,02 \pm ,07
2, leach	17,453 \pm ,019	15,546 \pm ,015	37,27 \pm ,04
202f, detrital pyrite	15,38 \pm ,07	15,02 \pm ,05	34,93 \pm ,19
202f, leach	38,528 \pm ,019	19,743 \pm ,002	36,42 \pm ,01
30BR, detrital pyrite	15,853 \pm ,013	15,246 \pm ,030	33,98 \pm ,09
30BR, leach	22,875 \pm ,004	17,014 \pm ,005	36,63 \pm ,02
302F, authigenic pyrite	23,042 \pm ,020	16,424 \pm ,020	33,91 \pm ,03
302F, leach	159,32 \pm ,25	33,990 \pm ,010	43,69 \pm ,01
123, authigenic pyrite	14,396 \pm ,007	14,684 \pm ,007	32,82 \pm ,02
3.1, authigenic pyrite	27,339 \pm ,015	19,062 \pm ,015	34,34 \pm ,04
3.2, authigenic pyrite	29,81 \pm ,03	19,43 \pm ,05	34,77 \pm ,07
3.4, authigenic pyrite	26,27 \pm ,04	18,96 \pm ,03	34,31 \pm ,05
3.4, leach	49,13 \pm ,09	21,63 \pm ,02	35,54 \pm ,04
3.6, authigenic pyrite	28,52 \pm ,04	19,21 \pm ,03	34,44 \pm ,06
Galena, Rosetta	12,522 \pm ,012	14,126 \pm ,018	32,35 \pm ,05

* Corrected ratios. Errors are 1 sigma errors

value of $0.37 \pm .01$ obtained by Burger and others (1962). However, the conclusions reached by these authors are not invalidated. Assuming a time of alteration of 2040 m.y. for the uranium minerals of the Witwatersrand Sequence, their age would be 3040 ± 50 m.y., which is identical with the age of 3080 ± 100 m.y. for uranium minerals from the Dominion Reef Sequence reported by Nicolaysen and others (1962).

7. DISCUSSION

(a) Authigenic Pyrites

In the $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram all data points of authigenic pyrites (including the leaches) plot in the area predicted for authigenic minerals of the Witwatersrand Sequence (Figures 2 and 3).

Samples 3.1, 3.2, 3.4 and 3.6 were single concretionary pyrites (so-called buckshot pyrites, Saager, 1970) collected from one hand specimen (Plate 1G). The four data points and the one from the leach of sample 3.4 yielded a linear array with a slope of $0.117 \pm .001$ (Figure 3). The slope of the line is the $^{207}\text{Pb}/^{206}\text{Pb}$ ratio of the three-stage lead which is most likely due to the *in situ* decay of uranium present in inclusions in amounts varying between 15 and 70 ppm after leaching. According to the $^{207}\text{Pb}/^{206}\text{Pb}$ ratio of the three-stage lead and assuming closed system behaviour of the uranium-bearing phases, their age is 1945 ± 25 m.y. which is still in agreement with the period of reconstitution of the uranium minerals in the Witwatersrand Sequence 2040 ± 50 m.y. ago and discussed earlier. The buckshot pyrites possess high lead contents ranging from 650 to 1130 ppm and comparatively low uranium contents of 15 to 70 ppm. It is thus impossible to shift the data points of the leached pyrites along the best fit line back to intercept with the upper boundary of the authigenic Witwatersrand minerals by correcting for the three-stage lead on the assumption of a closed uranium-lead system.

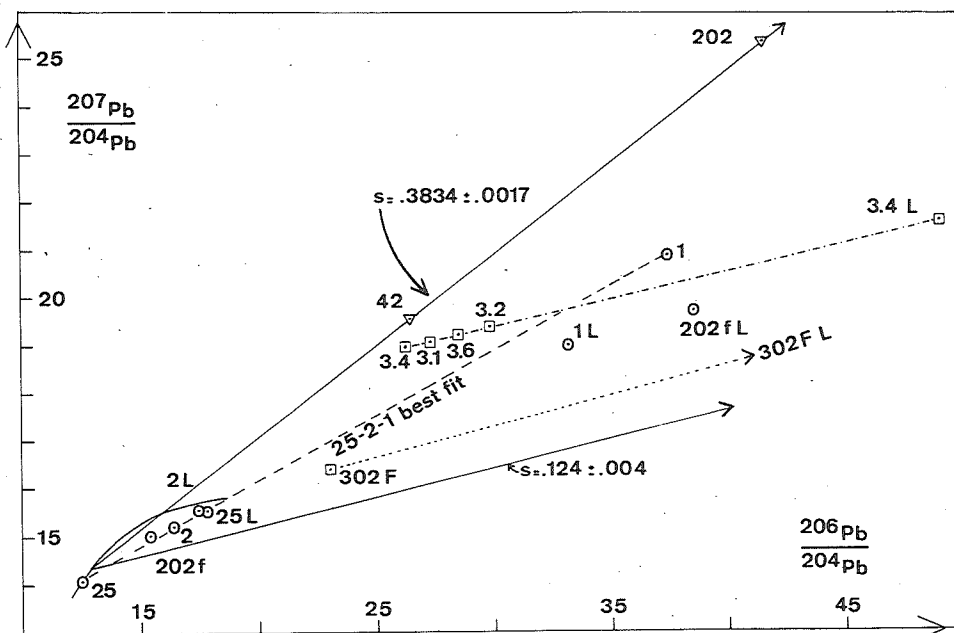


Figure 3 : $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ diagram showing the data points of galenas (triangles), detrital pyrites (circles) and authigenic pyrites (squares) with highly radiogenic lead from the Witwatersrand Sequence. The lines with slopes of 0.3834, i.e. the best fit of galena data points from the Witwatersrand Sequence (Burger and others, 1962), and of 0.124 are the upper and lower boundaries respectively for data points of authigenic minerals from the Witwatersrand Sequence. "L" denotes the HCl leaches of the respective sample. For further explanation see text.

From the buckshot pyrites data one is tempted to conclude that they were formed simultaneously with the major part of the galena 2040 m.y. ago. According to Ramdohr (1955) and Saager (1970), however, textural evidence points to a formation of these pyrites before, and/or during, the diagenesis of the sediments, i.e. prior to 2040 m.y. One therefore would have to expect these pyrites to contain three different types of lead : a first type corresponding to a hypothetical single-stage lead incorporated during their formation; the second type corresponding to the two-stage lead which is observed in many of the galenas and which would have infiltrated the pyrites 2040 m.y. ago and, the third type, due to the *in situ* decay of uranium that infiltrated the pyrites also 2040 m.y. ago. One may assume that leaching preferentially removes the third as well as the second type of lead and consequently one would expect to find a greater

slope than the one observed or, more likely, a scatter of the data points. Based on the normal range of 50 to 200 ppm of lead in pyrites (Saager, in preparation) and on the uranium contents of the leached samples, one has therefore to conclude that the two-stage lead forms the major part of the high lead content and that a possible scatter of the data points may be masked by the high proportion of the two-stage lead. A significant scatter may only be observed if, for example, the ratio of the two-stage to the single-stage lead is less than 30, and if the ratio of the two-stage to the single-stage lead, removed by the HCl treatment, is higher than 5. Furthermore, the lead isotopic composition of the single-stage lead has to correspond with that given by the lower intercept of the best fit through the galena data points with the growth curve, i.e. $^{206}\text{Pb}/^{204}\text{Pb} = 12.90$ and $^{207}\text{Pb}/^{204}\text{Pb} = 14.30$. The present isotopic data, therefore, does not provide an answer to the question of the time of formation of the buckshot pyrites.

The high lead content of the buckshot pyrites suggests that lead infiltration took place after their formation. Microscopic examination of the analysed pyrites revealed only few galena inclusions with diameters of the order of a few microns. Examination with the scanning electron microscope confirmed the paucity of galena inclusions at the 0.1 micron level. A Pb-As-S profile recorded with an electron microprobe revealed a very inhomogeneous Pb-As distribution within the range of a few microns varying from several hundred to more than 4000 ppm Pb and As, the latter varying sympathetically with Pb. The buckshot pyrites also contain portions very low in As and other minor elements (Saager and Mihálik, 1967), but where As was below the detection limit of approximately 300 ppm, the Pb content was still high, varying from a few hundred to about 2000 ppm. These examinations therefore did not yield conclusive evidence for a later Pb diffusion into pyrites.

Sample 302F and its leach show a pattern similar to that of the buckshot pyrites (Figure 3). The slope of the tie line is $0.1289 \pm .0008$. Extrapolating the line back to the upper limit for the authigenic minerals, one enters the field of "normal" leads, but not the area in which are plotted the data of primary pyrites from the greenstone belts.

Sample 123 contained only small amounts of additional radiogenic lead. It lies close to the data points of the least radiogenic leads found by Burger and others (1962). It also lies in the field of sulphides from primary gold deposits of the Swaziland Sequence (Figure 2).

(b) Detrital Pyrites

The isotopic composition of the lead in sample 25 is identical with the lead in the galena from Rosetta, a vein-type gold deposit located in the lower part of the Onverwacht Group in the Barberton Mountain Land. This sample, therefore, provides proof that the detrital pyrites and most likely, also the gold of the Witwatersrand sediments, were derived from greenstone belts of the Swaziland Sequence type.

The data points of the samples 1, 2 and 25, originating from the same locality, as well as the leach of sample 2, exhibit a linear array (Figures 2 and 3). The residuals of the coordinates of the data points vary between 0.3 and 0.7 per mil and are, in all cases, within the reproducibility of ± 2 per mil. They all contain the same first stage lead indicative of an origin in the greenstone belts. Sample 1 and 2 contain, after the HCl treatment, an additional radiogenic component with a $^{207}\text{Pb}/^{206}\text{Pb}$ ratio of $0.271 \pm .003$. A simple explanation for this is that these detrital pyrites, since their formation, contained some uranium - probably as minute inclusions - that contributed radiogenic lead. The slope of this lead corresponds to a $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3360 ± 20 m.y. of the uranium system in the pyrites. It is interesting to note that this age is identical to the lead-lead age of a zircon sample from the Barberton Mountain Land (van Niekerk and Burger, 1969) which is a minimum age for the upper part of the Onverwacht Group of the Swaziland Sequence.

The lead leached from sample 1 is noticeable, because it is less radiogenic than the remaining lead and appears itself to be a three-stage lead similar to some of the galenas reported by Burger and others (1962).

The remaining detrital pyrite samples 202f and 30BR yielded lead isotopic compositions similar to those obtained from sulphides of primary gold deposits in the greenstone belts of the Swaziland Sequence. It is apparently significant that the slopes of the tie-lines to the respective leaches are steeper than the analogous slopes of the authigenic pyrites. The hypothetical first stage lead of the detrital pyrites resembles those of galenas from primary deposits in the greenstone belts (Figures 2 and 3; see also Ulrych and others, 1967).

TABLE 3

Description of Analysed Samples

Sample Number	Location and Stratigraphic Position	Mineral Analysed	Geological and Mineralogical Data
42	Durban Roodepoort Deep Central Rand Main Reef horizon	galena	Galena-rich sample from a quartz veinlet associated with a fault zone offsetting the Main Reef. The normal type fault is of pre-Transvaal age. Small ($\geq 100 \mu$) euhedral pyrite in an unusually large probably reconstituted galena nodule (Plate 1F).
202	Free State Geduld Mines Leader Quartzite horizon	galena	Galena inclusions in a large (15 mm) pyrite cube from a quartz vein associated with a fault cutting the Leader Quartzite. Other inclusions in the pyrite cube are chalcopyrite and pyrrhotite.
25	Free State Geduld Mines	pyrite,	Sample taken relatively close to the Basal Reef suboutcrop to the west of the mine. Large (0.6-2 mm) rounded detrital pyrite grains. With the exception of pyrrhotite, no other sulphide inclusions were observed in these pyrites.
1	Orange Free State Goldfield	detrital	
2	Basal Reef horizon		
202f	Welkom Gold Mining Co. Orange Free State Goldfield Western part of mine Leader Quartzite horizon	pyrite, detrital	Detrital pyrite grains from the Leader Quartzite overlying directly the Upper Foot-wall Formation. This sample contains small (≥ 0.5 mm), but unusually numerous, detrital pyrite grains in a matrix of authigenic pyrite (Plate 1A, B).
30BR	Stilfontein Gold Mining Co. Klerksdorp Goldfield Vaal Reef horizon	pyrite, detrital	Rounded pyrite grains from the Vaal Reef. Other detrital constituents present in the sample, which comes from the lower portion of the reef, are uraninite and chromite.
302F	Free State Geduld Mine Orange Free State Goldfield Basal Reef horizon	pyrite, authigenic	Sample collected approximately 10 cm from the contact of a Ventersdorp dyke intersecting the Basal Reef. In addition to authigenic euhedral pyrite grains (0.01-2 mm), rutile, shattered zircon and rounded chromite were observed in this sample (Plate 1E).
123	Free State Geduld Mine Orange Free State Goldfield 2/49E Level, 13 Raise Basal Reef horizon	pyrite, authigenic	Euhedral pyrite (≥ 2 mm) from a thin quartz vein associated with a Karroo (?) dyke cutting the Basal Reef. All pyrite in this sample seems to be reconstituted by the dyke intrusion, no rounded pyrite grains were observed.
3.1	Virginia Gold Mining Co.	pyrite,	Large pyrite concretions, so-called buckshot pyrites (3-12 mm), which occur at the foot-wall contact of the B-Reef (Plate 1G). Other minerals present are detrital pyrite, uraninite, gold, galena, chalcopyrite. The four different samples were obtained from four different buckshot pyrites in the same hand specimen.
3.2	Orange Free State Goldfield	authigenic	
3.4			
3.6	B-Reef, Kimberley Stage		
	Rosetta Mine Barberton Mountain Land Onverwacht Group, Swaziland Sequence	galena/ tetrahedrite	Complex disseminated galena, tetrahedrite, gold ore in strongly sheared acid schists within the Lower Ultramafic Unit of the Onverwacht Group (Plate 1H).

Finally, one may draw attention to the fact that the hypothetical first stage lead of the authigenic minerals from the Witwatersrand Sequence is identical to some of the leads found in galenas from the greenstone belts. This may be a coincidental mixing effect or the result of similar leads present in the younger granite-migmatite areas being contributed as detritus to the Witwatersrand basin. One has to bear in mind, however, that on the Kaapvaal craton little evidence is available for the existence of any terrane older than the greenstone belts. Therefore, the first stage of the Witwatersrand lead can hardly be explained as a mixture of an old and young component. As unusually high lead concentrations ranging from 5 to 40 ppm have been reported for the Onverwacht lavas (Viljoen and others, 1969; Sinha, 1972), the greenstone belts have probably contributed a significant amount of lead to sedimentary basins.

8. SUMMARY AND CONCLUSIONS

The lead isotopic composition of some detrital pyrites from the Witwatersrand Sequence provides proof that they derive from the ancient greenstone belts like that of the Barberton Mountain Land in the Eastern Transvaal Lowveld. The primary gold deposits of the greenstone belts therefore constitute the most probable source of the gold and other detrital minerals in the Witwatersrand basin as proposed by Viljoen and others (1970).

Although only a few samples have been analysed it may tentatively be concluded that systematic differences exist in the pattern of the lead isotopic composition of leach and sample between detrital and authigenic pyrites. Despite textural evidence indicating formation of the authigenic pyrites before and/or during the diagenesis of the Witwatersrand sediments, the possibility cannot be ruled out that the analysed authigenic pyrites were formed 2040 m.y. ago. Further investigations are required to ascertain which of the two lines of evidence - the textural or the isotopic - must be reinterpreted.

The present study demonstrates, furthermore, the usefulness of investigating the lead isotopic composition of pyrites in different samples collected from one hand specimen or from a very restricted area where it has been assumed that the constituents experienced qualitatively the same history.

* * * * *

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Key to Plate

- Plate 1 : A : Welkom Gold Mine. Ore sample from the Leader Quartzite horizon. Detrital, in part shattered, rounded pyrite grains (white) in matrix of authigenic pyrite and gangue (dark grey). In places the authigenic pyrite clearly moulds around the detrital pyrite. Note also the two detrital grains of chromite (grey). Reflected light, x36.
- B : Welkom Gold Mine. Concentrate of detrital pyrites from the Leader Quartzite horizon (see also A). Sample 202f. Reflected light, x36.
- C : Free State Geduld Mine. Ore sample from the Basal Reef horizon. Large inhomogeneous and loose pyrite concretions, so-called buckshot pyrite (dull white) together with small compact detrital pyrite grains (white). Gold (bright white) in the centre of the photograph has marginally infiltrated the concretions. Gangue (black). Reflected light, x36.
- D : Free State Geduld Mine. Ore sample from the Basal Reef horizon. Detrital grains of pyrite (white) in typical association with chromite (dark grey) and uraninite (grey) having numerous inclusions and stringers of galena (white). Brannerite aggregates (dark grey) with galena inclusions show columnar forms. Gangue (black). Reflected light, oil immersion, x250.
- E : Free State Geduld Mine. Ore sample from the contact between a Ventersdorp dyke and the Basal Reef horizon. Authigenic, euhedral pyrite grains (white) with an inclusion of a shattered detrital chromite (grey). Gangue (black). Sample 302F. Reflected light, x36.
- F : Durban Roodepoort Deep Mine. Ore sample from the Main Reef horizon. Euhedral pyrite grains (white), partially embedded in a large irregular galena aggregate (dark white). Gangue (black). Sample 42. Reflected light, oil immersion, x250.
- G : Virginia Gold Mine. Photograph of a polished slab showing the footwall contact of the conglomeratic B-Reef horizon with underlying shales. Note the concentration of concretionary buckshot pyrites along the contact. Taken from the same hand specimen as samples 3.1, 3.2, 3.4, 3.6. x2.
- H : Rosetta Mine, Barberton Mountain Land. Ore sample showing typical intergrowth of tetrahedrite (grey), galena (white, scratched) and sphalerite (dark grey) with chalcopryite exsolution specks (white). Rhombic crystal in the upper right hand corner is arsenopyrite (white). Gangue (black). Reflected light, oil immersion, x250.

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PLATE 1

