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The geochronology of uraniferous minerals in the Witwatersrand Triad; an interpretation of new and existing U–Pb age data on rocks and minerals from the Dominion Reef, Witwatersrand and Ventersdorp Supergroups

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Uranium and lead analyses of rock samples from the Witwatersrand, Ventersdorp, and Transvaal supergroups give mainly discordant ages. Samples from the Upper Witwatersrand of the Orange Free State give $^{207}Pb/^{206}Pb$ ages of *ca.* 3000 Ma. These data when considered together with earlier total conglomerate U–Pb analyses from the Dominion Reef Supergroup lead to the conclusion that the uraniferous minerals of the Dominion Reef, Witwatersrand, Ventersdorp and Transvaal conglomerates are 3050 ± 50 Ma old. In the northern parts of the Witwatersrand Basin the parent uraniferous minerals experienced a major reworking at 2040 ± 100 Ma which brought about the partial or complete resetting of the original 3050 Ma age. Radiogenic lead released during this reworking crystallized as galena in veins and fissures which cut across the uraniferous conglomerate horizons. This reworking appears to have had little effect in the Orange Free State to the south. Its age and geographical extent suggest it was caused by thermal effects which accompanied the emplacement of the Bushveld Igneous Complex at 1950 ± 150 Ma.

Samples from the south, which were relatively unaffected by the *ca.* 2040 Ma reworking generally show the effects of recent uranium loss. In the northern part of the basin discordant age patterns characteristic of lead loss have been imposed on uranium–lead systems which were generally reset (partially or completely) by the *ca.* 2040 Ma event.

The presence of 3050 Ma old minerals in sedimentary sequences which are probably younger than *ca.* 2740 Ma suggests the simple interpretation that the uraniferous minerals are predominantly detrital.

1. Introduction

Auriferous and uraniferous quartz–pebble conglomerates occur in the Dominion Reef, Witwatersrand, Ventersdorp and Transvaal Supergroups of South Africa. These weakly metamorphosed sedimentary/volcanic successions rest with marked unconformity on a highly metamorphosed basement complex comprising the Older Granite and the Swaziland Sequence which together constitute an early Precambrian granite–greenstone terrain.

The three lower supergroups are closely related both areally and structurally, they appear to constitute a major geological cycle and have been named the Witwatersrand Triad (Hamilton & Cooke 1960). The maximum total thickness of the triad has been estimated to be about 15000 m of which nearly half is lava (Whiteside 1970).

Three main theories have been advanced to explain the presence of gold and uranium in the conglomerates. These are:

(1) The placer theory, which holds that the gold and uraninite are detrital. The theory was later modified to allow the gold to be dissolved and reprecipitated almost *in situ*.

(2) The hydrothermal theory, which considers that the gold, uraninite and certain other minerals, in particular sulphides, were introduced by hydrothermal solutions.

(3) The precipitation theory, which states that the gold and uranium were precipitated possibly together with pyrite during the deposition of the conglomerates or during subsequent diagenesis.

The main development of gold and uraninite bearing conglomerates occurs in the Upper Witwatersrand. Uraniferous conglomerates also occur in the Basal Sedimentary Formation of the Dominion Reef Supergroup. The Ventersdorp Contact Reef occurs at the base of the Ventersdorp Supergroup and is auriferous and uraniferous where it lies uncomformably on the eroded and bevelled beds of the Witwatersrand rocks. The gold and uraninite are here clearly derived from the underlying Witwatersrand sediments. The Black Reef which occurs in the overlying Transvaal Supergroup is locally auriferous and uraniferous. Here again it is accepted that the gold and other heavy minerals, including uraninite, were derived by the erosion of underlying Witwatersrand conglomerates.

		Area				
		1	2	3	4	5
Supergroup/ Group	Formation	Odendaalsrus–Welkom	Virginia	Klerksdorp	Carletonville	East Rand
Transvaal	Black Reef			212 Black Reef		
Ventersdorp	Contact Reef			216, 222 V.C.R.	217 V.C.R.	
Upper Witwatersrand	Elsburg	226 Elsburg Freddies		223 no. 5 Reef		
	Kimberley	229 'A' Reef		224 Dennys Conglomerate		233 Upper Kimberley
	Bird	228 Intermediate Reef, 231 Basal Reef	227 Leader Reef, 230 Basal Reef	221 Vaal Reef		
	Main				218, 220 Carbon Leader	232 Main Reef Leader

FIGURE 1. Stratigraphic correlations and sample location (1–5 as in figure 2).

The present investigation was made on conglomerates – bankets – from the main conglomerate horizons within the Witwatersrand, Ventersdorp and Transvaal Supergroups (see figures 1 and 2). Age determinations were made on 'total rock' specimens (cf. Nicolaysen, Burger & Liebenberg 1962). Rock for analysis was selected from the interstitial material between the quartz and other pebbles, avoiding as far as possible the inert pebbles. No claim is made that these total rock specimens reflect the bulk composition of the conglomerates. The method of sampling was adopted because microscopic examination shows that uraninite is frequently associated with exsolved, and presumably radiogenic galena, which occurs both as specks within the uraninites and as more completely exsolved rims. Normal mineral separation procedures bring about a mechanical separation of the uraninite and exsolved radiogenic galena (Nicolaysen *et al.* 1962) giving rise to discordant age patterns.

The analyses were made on a suite of conglomerates provided by Dr H. C. M. Whiteside and the analytical data are given in table 1. The specimens are all of typical 'banket' conglomerate composed of quartz and chert pebbles in a fine-grained matrix of quartz, phyllosilicates and opaques. Pyrite is the chief opaque mineral and occurs as allogenic, concretionary

authigenic, and reconstituted authigenic forms (Köppel & Saager 1974). Uraninite occurs as individual rounded grains lying within the quartzitic and sericitic matrix (the detrital form of Liebenberg 1958) and in some specimens is intimately associated with hydrocarbon. Secondary uraninite occurs as specks, patches and veinlets in the matrix and also in the hydrocarbon.

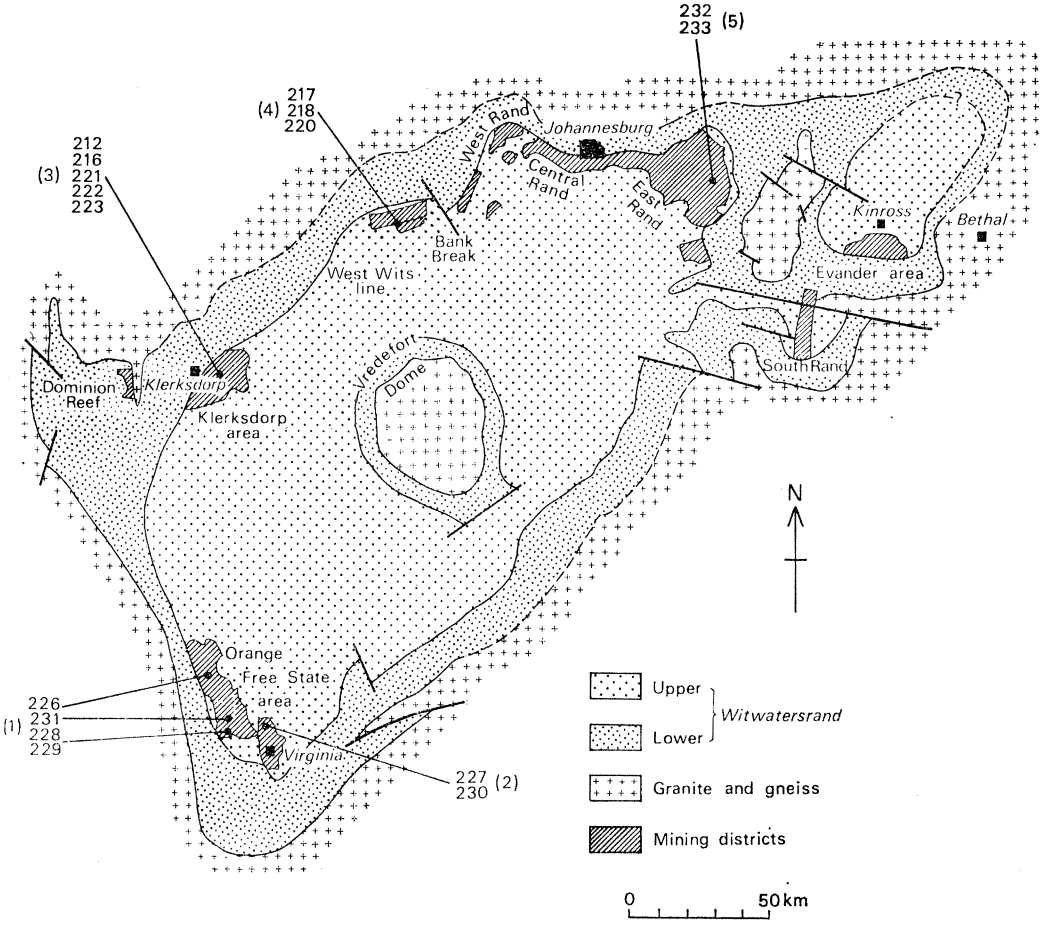

FIGURE 2. Simplified geological map of the Witwatersrand Basin beneath the Ventersdorp and younger cover. Sample locations 1–5 as in figure 1.

2. ANALYTICAL METHODS AND PRECISION

Portions of the matrix, avoiding as far as possible the pebbles, were cut in the form of cubes about 2 cm in size, fragmented in a percussion mortar and finely ground in a Tema swing mill (grain size < 75 μm). The approximate concentrations of U and Pb were then determined by X-ray fluorescence analysis. Samples for analysis were decomposed in a mixture of hydrofluoric and perchloric acids (Aristar grade) and after evaporation to dryness the residues were dissolved and made up to 100 ml in 1.2 M hydrochloric acid. Minute amounts of residue sometimes proved insoluble and consisted of unattacked sample – chromite, rutile and traces of zircon – and crystalline perchlorates which spectrographic analysis showed to be free of uranium and lead.

Appropriate aliquots of the rock solution were taken for lead isotope analysis and for isotope dilution analysis of Pb and U using ^{208}Pb and ^{235}U enriched tracers respectively. Uranium and

lead were separated using ion exchange columns of Dowex 1 × 8 resin, lead was further purified by dithizone extraction, and uranium by precipitation as hydroxide with ammonia gas. The precipitated hydroxides were dissolved and repassed through the ion exchange column and finally iron was extracted with di-isopropyl ether.

Mass spectrometric analyses were made using a Thomson C S F THN 206, 60°, 30 cm radius mass spectrometer. Samples were loaded on a rhenium filament together with silica gel and phosphoric acid. Ion beam intensities were measured from a chart recorder for samples 212, 218, 221, 226, 228, 229, 230 and 231. Subsequently a digital recording system using voltage to frequency conversion became operative.

Lead isotope ratios were monitored throughout the investigation by analysis of the NBS lead isotope standard SRM. 981. The following results were obtained: (1) by chart measuring, $^{206}Pb/^{204}Pb = 16.914 \pm 0.014$, $^{207}Pb/^{204}Pb = 15.445 \pm 0.012$; (2) by digital output, $^{206}Pb/^{204}Pb = 16.946 \pm 0.007$, $^{207}Pb/^{204}Pb = 15.498 \pm 0.008$. Catanzaro, Murphy, Shields & Garner (1968) give the following ratios for this standard: $^{206}Pb/^{204}Pb = 16.937$, $^{207}Pb/^{204}Pb = 15.491$.

Estimates of precision in the determination of uranium, lead and lead isotope ratios have been obtained from replicate data. Pooled standard deviations have been determined as follows:

(1) Uranium: concentration range 39–87 parts/10^6, pooled standard deviation ± 1 part/10^6; concentration range 100–215 parts/10^6, pooled standard deviation ± 2.2 parts/10^6; concentration range 1000–1500 parts/10^6, pooled standard deviation ± 22.2 parts/10^6.

(2) Lead: concentration range 18–28 parts/10^6, pooled standard deviation ± 0.35 part/10^6; concentration range 190–350 parts/10^6, pooled standard deviation ± 1.5 parts/10^6; concentration range 750–1100 parts/10^6, pooled standard deviation ± 4.2 parts/10^6.

The precision is thus generally better than ± 2 % for uranium and ± 1.5 % for lead. Applying these uncertainties to the data, and reducing them by a factor of $N^{-\frac{1}{2}}$ when N replicates have been made, the standard errors on the U/Pb ratios are less than ± 2.2 % with the exception of 223 (± 3.3 %), 217 (± 4.2 % and 224 ± 6 %). For sample 230 which has an extremely low uranium content of 2.78 parts/10^6 the standard error has been calculated using duplicates of 2.77 and 2.79.

Replicate determinations of lead isotope ratios on these samples gave pooled standard deviations as follows: $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb < 30$, ± 0.27; $^{206}Pb/^{204}Pb$, and $^{207}Pb/^{204}Pb > 30$, < 60, ± 0.17; $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb > 60$, < 300, ± 0.37. These precision estimates are relatively poor compared with those obtained on the NBS standard galena. They presumably reflect additional uncertainty introduced by sampling, chemical processing and blank correction.

Errors in the $^{207}Pb^*/^{235}U$ and $^{206}Pb^*/^{238}U$ ratios have been calculated by combining in the appropriate way the above precision estimates. The common lead correction results in an error enhancement on the lead isotope ratios by a factor of $M/(M-C)$ where M is the measured ratio and C the assumed common lead ratio. Samples 217 and 212 are most strongly effected by this error magnification, and assuming the Rosetta common lead correction the $^{206}Pb/^{204}Pb$ errors are enhanced by a factor of 2.8 and 1.6 respectively and the $^{207}Pb/^{204}Pb$ errors by a factor of 9 and 4.7.

The percentage errors on the ages are less than those on the appropriate D/P ratios by a factor $\dfrac{(D/P)}{(1+D/P)\ln(1+D/P)}$. For a 2000 Ma sample this factor is 0.43 for the $^{207}Pb/^{235}U$ age and 0.86 for the $^{206}Pb/^{238}U$ age. For a 3000 Ma sample the factors are 0.32 and 0.8 respectively.

TABLE 1. NEW U–Pb DATA, TOTAL CONGLOMERATE SAMPLES FROM THE UPPER WITWATERSRAND GROUP, VENTERSDORP CONTACT REEF, AND THE BLACK REEF (TRANSVAAL SUPERGROUP)

	concentration parts/10^6		Pb isotope ratios			calculated ages Ma		
sample	U	Pb	206/204	207/204	208/204	$t^{206}\mathrm{Pb}/^{238}\mathrm{U}$	$t^{207}\mathrm{Pb}/^{235}\mathrm{U}$	$t^{207}\mathrm{Pb}/^{206}\mathrm{Pb}$
212	85.73	82.49	34.64	18.35	40.49	1493 ± 30	2089 ± 40	2738 ± 105
216	246.3	182.5	116.4	32.62	38.03	2506 ± 25	2577 ± 15	2632 ± 25
217	25.48	98.34	22.29	16.45	34.28	2985 ± 105	3046 ± 65	3084 ± 215
218	60.75	25.77	574.4	82.80	56.75	2112 ± 24	2051 ± 15	1998 ± 10
220	513.9	168.0	2686	364.3	96.26	1804 ± 20	1952 ± 15	2110 ± 10
221	1486	778.7	139.9	33.27	40.76	1985 ± 15	2168 ± 10	2344 ± 20
222	319.7	179.8	330.9	74.83	46.82	2434 ± 25	2609 ± 10	2746 ± 25
223	772.4	345.7	274.9	57.61	48.52	1968 ± 55	2250 ± 30	2513 ± 10
224	12.41	16.83	91.82	26.83	64.71	3330 ± 150	2810 ± 60	2453 ± 45
226	39.50	23.00	60.99	21.41	41.73	1497 ± 30	1885 ± 30	2341 ± 65
227	403.3	485.9	61.53	24.33	32.29	2927 ± 25	2904 ± 16	2887 ± 45
228	102.3	187.7	77.80	29.05	49.54	4077 ± 50	3411 ± 20	3038 ± 35
229	216.5	348.7	56.90	23.26	36.32	3441 ± 30	3090 ± 20	2867 ± 50
230	2.78	17.00	52.58	22.94	35.78	8127 ± 20	4416 ± 25	2974 ± 35
231	1051	1070	119.7	37.13	38.37	3211 ± 25	3046 ± 15	2937 ± 35
232	234.8	115.8	539.8	98.35	49.71	2345 ± 30	2403 ± 15	2451 ± 10
233	1063	300.0	948.4	243.2	74.99	1398 ± 25	2251 ± 20	3150 ± 10

Decay constants: $\lambda^{235}\mathrm{U} = 0.98485 \times 10^{-9}\ \mathrm{a}^{-1}$, $\lambda^{238}\mathrm{U} = 0.15525 \times 10^{-9}\ \mathrm{a}^{-1}$.
Common lead correction ratios: $^{206}\mathrm{Pb}/^{204}\mathrm{Pb} = 12.52$, $^{207}\mathrm{Pb}/^{204}\mathrm{Pb} = 14.13$, $^{208}\mathrm{Pb}/^{204}\mathrm{Pb} = 32.35$.

212 Black Reef, Western Reefs mine.
216 Ventersdorp Contact Reef, Western Reefs mine.
217 Ventersdorp Contact Reef, Western Deep Levels mine.
218 Carbon Leader, Western Deep Levels mine, 100–69 2E.
220 Carbon Leader, Western Deep Levels mine, 97/60 Pillar Line (Reef Drive).
221 Vaal Reef, Western Reefs mine.
222 Ventersdorp Contact Reef, Western Reefs mine.
223 Elsburg No. 5 Reef, Western Reefs mine.
224 Dennys Conglomerate, Western Reefs mine.
226 Elsburg Series, Freddies Consolidated mine.
227 Leader Reef, Free State Saaiplaas mine.
228 Intermediate Reef, President Steyn mine.
229 'A' Reef, President Steyn mine.
230 Basal Reef, Free State Saaiplaas mine.
231 Basal Reef, President Brand mine.
232 Main Reef Leader, East Rand area, E. Geduld mine.
233 Kimberley Reef, East Rand area, E. Geduld mine.

3. AGE LIMITS TO THE WITWATERSRAND SUPERGROUP

Early attempts to assign age limits to the Witwatersrand Supergroup have been summarized by Hales (1960). At that time it was clear that some granitic members of the basement complex underlying the Witwatersrand Triad were at least 3000 Ma old. Hales further reported that Rb–Sr whole-rock studies by Schreiner on Dominion Reef acid volcanics indicated an age of about 3000 Ma. Mendelssohn, Burger, Nicolaysen & de Villiers (1958) had previously reported a lead–lead age of 2160 ± 100 Ma on an authigenic monazite from a vein cutting Witwatersrand sediments which was generally accepted as setting a younger limit to the age of the Supergroup.

Nicolaysen *et al.* (1962) made U–Pb determinations on a detrital monazite and total conglomerate from the Dominion Reef conglomerate. Their data are given in table 2. Ages have been recalculated using the most recently determined uranium decay constants (Jaffey *et al.* 1971).

Although the monazite showed a discordant U–Pb age pattern indicative of uranium loss, the lead–lead age of 3120 ± 100 Ma† was accepted as its age of formation, and since the

† See comments by Nicolaysen *et al.* (1962, footnote, p. 18) concerning the interpretation of this age.

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monazite is unquestionably detrital it set an older limit to the Dominion Reef Supergroup. This was further confirmed by results on three total conglomerate specimens which contain small, oval uraninites, considered to be detrital in origin, together with various other detrital heavy minerals and subhedral secondary uraninite. One sample gives a virtually concordant age of 3060 ± 70 Ma (B153) while the other two, KCG1 and KCG4, although showing a lead loss discordant age pattern, give $^{207}Pb/^{206}Pb$ ages of 3048 ± 100 Ma and 3018 ± 100 Ma respectively. Although the presence of subhedral uraninites suggests some degree of recrystallization, it is clear that total conglomerate B153 has remained a closed chemical system with respect to both U and Pb. An age of about 3060 ± 70 Ma can thus be accepted as dating an episode or uraninite mineralization in the pre-Dominion Reef basement and sets an older limit to the Dominion Reef and younger deposits.

TABLE 2. U–Pb DATA, DOMINION REEF CONGLOMERATES
(Nicolaysen *et al.* 1962)

	Concentration		Pb isotope ratios			calculated ages/Ma		
sample	% U	% Pb	206/204	207/204	208/204	$t^{206}Pb/^{238}U$	$t^{207}Pb/^{235}U$	$t^{207}Pb/^{206}Pb$
KCG Mon	0.332	0.981	195	58	596	3640 ± 120	3322 ± 120	3120 ± 100
B 153	2.46	1.77	571	142	52.6	3075 ± 100	3059 ± 100	3050 ± 100
KCG 1	0.201	0.112	249	68.3	62.6	2182 ± 110	2654 ± 120	3048 ± 100
KCG 4	0.520	0.350	326	84.7	69.1	2648 ± 110	2862 ± 120	3018 ± 100

KCG 1 Monazite concentrate, Dominion Reef conglomerate, Klerksdorp Consolidated Mines Ltd.
B 153 Total < conglomerate, Upper Reef in the Bramley 6, N3 section of the Dominion Reefs Mine.
KCG 1, KCG 4 Total conglomerate reef from the upper zone, showing supergene alteration, of the Klerksdorp Consolidated Mines Ltd mine.
Ages recalculated according to revised decay constants (Jaffey *et al.* 1971):
$\lambda^{235}U = 0.98485 \times 10^{-9}\ a^{-1}$, $\lambda^{238}U = 0.15525 \times 10^{-9}\ a^{-1}$.
Common lead correction ratios: $^{206}Pb/^{204}Pb = 12.52$, $^{207}Pb/^{204}Pb = 14.13$, $^{208}Pb/^{204}Pb = 32.35$.

In 1964 Allsopp reported whole-rock Rb–Sr determinations on various basement granites from the Western Transvaal. An intensive study was made of the Schweizer Reneke granite which intrudes a complex belt of schists some 320 km southwest of Johannesburg. The granite is overlain unconformably by rocks of the Ventersdorp Supergroup. Allsopp's data gave an isochron corresponding to an age of 2700 ± 55 Ma ($\lambda = 1.39\ 10^{-11}\ a^{-1}$) with an initial $^{87}Sr/^{86}Sr$ ratio of 0.704 ± 0.005. Recalculation of these data, allowing for errors (as quoted by Allsopp) in both coordinates, by the method of Williamson (1968) gives a slightly different result of 2740 ± 20 Ma with an initial $^{87}Sr/^{86}Sr$ ratio of 0.703 ± 0.001. Allsopp also reported apparent ages on a suite of granite samples, some from boreholes and others from small isolated exposures. Ages were calculated assuming an initial $^{87}Sr/^{86}Sr$ ratio of 0.705 and gave results agreeing within experimental error either with the age of the Johannesburg–Pretoria granite (3200 ± 65 Ma, Allsopp 1961, 1964) or the Schweizer Reneke granite. One of the granite samples which gave an apparent age of 2720 ± 100 Ma was overlain by Lower Witwatersrand rocks (borehole sample An952), another sample (borehole RSPIC) gave an apparent age of 2820 ± 55 Ma and was overlain by sediments of the Dominion Reef. However, Allsopp considered it likely that sample RSPIC had suffered chemical alteration, since it occurred only 0.9 m beneath the Dominion Reef contact. Although not absolutely conclusive, these data strongly indicate that the Witwatersrand Triad is younger than *ca.* 2740 Ma. The Ventersdorp rocks certainly are,

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and there is no reason to believe that the episode of granite intrusion dated at 2740 ± 20 Ma occurred after the deposition of the Witwatersrand sediments and before the formation of the Ventersdorp rocks. The apparent age of An 952 (2720 ± 100 Ma) accords with this view but it must be admitted that an age calculated assuming an initial $^{87}Sr/^{86}Sr$ ratio must be viewed with caution.

More recently van Niekerk & Burger (1964, 1969*a*, *b*) have reported lead–lead and uranium lead ages on lavas from the Dominion Reef and Ventersdorp Supergroups so providing both lower and upper limits to the time of deposition of the Witwatersrand Supergroup. They quoted ages of 2800 ± 109 Ma and 2300 ± 80 Ma respectively. Only lead–lead data are available for the Dominion Reef lavas; regression of these data weighted according to the quoted errors, and calculation of the age using the most recently determined uranium decay constants, gives a somewhat younger age of 2725 ± 75 Ma. Similar treatment of the Ventersdorp lavas gives an age of 2290 ± 85 Ma. Two zircons from these lavas give discordant age patterns indicative of uranium loss and yield a concordia intersection age of 2285 ± 50 Ma, identical within error limits with the aforementioned lead–lead age.

The age of the Schweizer Reneke granite and the Dominion Reef lavas cannot be resolved, suggesting that the unroofing of the granite and subsequent deposition of the Dominion Reef sediments and lavas occurred in a relatively short time. This is compatible with the observations that the Dominion Reef rocks were deposited on an uneven floor suggesting that there was insufficient time for a peneplain surface to develop.

The younger limit to the age of the Witwatersrand deposits is thus set by the Ventersdorp age determination of 2290 ± 85 Ma. For the older limit we suggest it is appropriate to adopt the weighted mean age of the Schweizer Reneke granite and the Dominion Reef lavas, namely 2740 ± 19 Ma.

4. Previous research on the U–Pb geochronology of the Witwatersrand

The results of previous U–Pb age determination studies on uraninite and thucholites mainly from the Witwatersrand sediments by Louw (1954) and Horne & Davidson (1955) have been summarized and discussed by Holmes & Cahen (1957). The data, plotted on a concordia diagram are shown in figure 3. The concordia diagram has been constructed, and ages calculated using the recently revised uranium decay constants. Rosetta lead has been used for the common lead correction. However, if it is assumed that the ages reflect a major rejuvenation at about 2040 Ma (see below) then a more radiogenic common lead correction will be appropriate. Any lead incorporated in the rejuvenated uraninites would, at that time, have an isotopic composition comparable to that of the extracted lead. This lead has a variable isotopic composition and defines a secondary isochron reflecting mixing of entirely radiogenic lead with original *ca.* 3000 Ma old common lead (Burger, Nicolaysen & de Villiers 1962). We have accordingly adopted one of the most radiogenic of these leads, namely NPRL No. 163, $^{206}Pb/^{204}Pb = 23.32$, $^{207}Pb/^{204}Pb = 18.21$, for the alternative common lead correction and the appropriate data points are indicated by vertical crosses (see comments below, §5).

The calculated ages are in the main discordant and in some cases affected by uncertainties concerning the common lead correction. However, Holmes & Cahen (1957) and later Burger *et al.* (1962) considered that three samples yielded concordant or nearly concordant ages which

were not too dependent on assumption concerning the common lead correction and which 'appear to have started their history 2040 ± 100 million years ago' (Burger *et al.* 1962). The greatest age was given by a sample from the Dominion Reef ($^{207}Pb/^{206}Pb$ age = 2760 Ma) with the remaining samples giving $^{207}Pb/^{206}Pb$ ages in the range *ca.* 2000 Ma to 2540 Ma. Holmes & Cahen (1957) proposed two hypotheses to explain the age patterns. The first implied that the uraninites were of two distinct generations, the oldest having an age in excess of 2760 Ma and the youngest being about 2000 Ma old. The second considered that all the uraniferous minerals were of the same age as the Dominion Reef uraninite and hence older than 2760 Ma. Concerning the cause of the discordant ages Holmes & Cahen were particularly impressed with the number of $^{206}Pb/^{238}U$ ages in the 1200 Ma to 1400 Ma range and suggested an important reworking causing lead loss at this time.

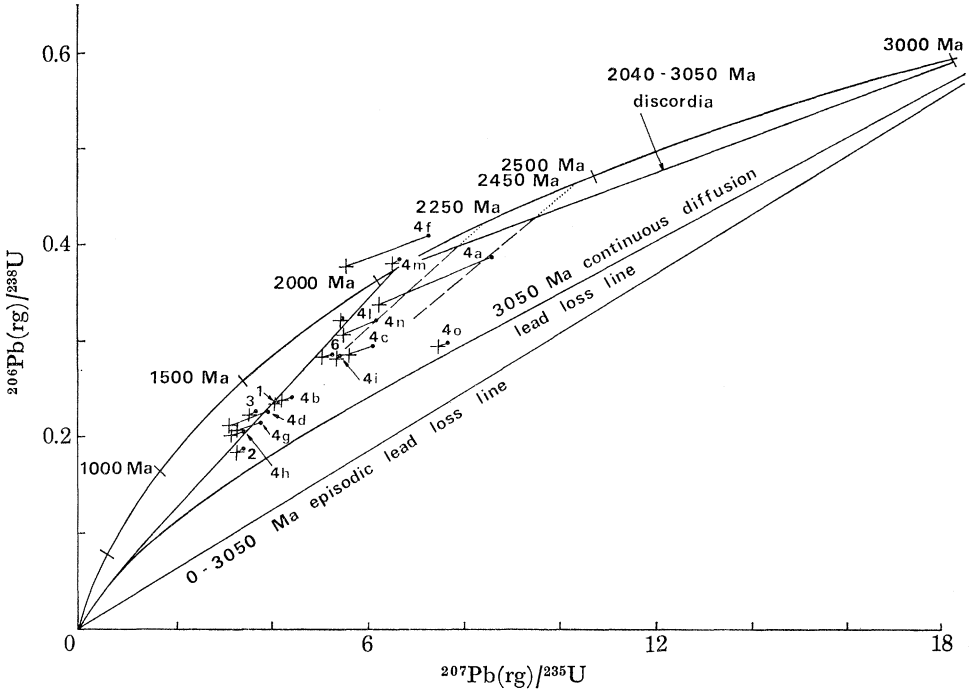

FIGURE 3. Concordia diagram showing data points for previous uraninite and thucholite U–Pb age determinations. Sample numbers refer to Holmes & Cahen (1957), Table 1, suite V11 (sample V11, 1 uses lead isotope data H. 1).

From consideration of the distribution of the data points on a concordia diagram, Wetherill (1956) suggested that the Witwatersrand uraninites and thucholites 'were formed 2040 ± 50 million years ago and suffered loss of lead because of some alteration or leaching process 200 ± 150 million years ago'. Wetherill further noted certain minor regularities which, if real, could be interpreted as two additional groups of minerals having true ages of 2250 Ma and 2450 Ma which had also experienced lead loss at *ca.* 200 Ma. Wetherill had interpreted the data solely on the basis of episodic lead loss. Nicolaysen (1957), however, had previously shown that the characteristic lead-loss pattern of discordant ages could equally arise from continuous loss of lead by solid diffusion. When plotted on concordia the lead loss by diffusion trajectory for a *ca.* 2040 Ma sample hardly differs over much of its length from the discordia

chord resulting from episodic lead loss at 200 Ma (Tilton 1960). In the present case the distinction is somewhat irrelevant, lead loss is clearly indicated and could equally reflect continuous diffusion or episodic loss related to the thermal disturbances which may have been associated with the widespread Karroo volcanism. Before further consideration of this earlier U:Pb data it is appropriate to review the findings of Burger *et al.* (1962) concerning the isotopic composition of lead from galenas from the Witwatersrand and Orange Free State, which led to the formulation of their hypothesis concerning the evolution of the Witwatersrand uraninites, and also to consider the new data obtained during this investigation.

Burger *et al.* noted not only the strongly radiogenic nature of the lead in galenas occurring in veins cutting the uraniferous conglomerates of the Witwatersrand and Dominion Reef, but also the fact that many of them displayed regular linear relations when plotted on a $^{207}Pb/^{204}Pb$ against $^{206}Pb/^{204}Pb$ diagram, and that the lines so defined passed through or close to the isotopic composition of lead from the Rosetta Mine galena. This is one of the least radiogenic terrestrial leads and occurs in rocks equivalent to those underlying the Witwatersrand deposits. The linear relations (metachrons or secondary isochrons) of the Witwatersrand, Dominion Reef and Rosetta galenas imply that they have evolved by the mixing of Rosetta type lead with radiogenic lead resulting from uranium decay. Burger *et al.* summarize the evolution of these galenas as follows:

(1) At some time in the past, an original or 'parent' uraninite was present in a particular region. The time of crystallization of the parent uraninite is denoted t_s.

(2) The uraninite was associated with varying amounts of a primary lead. Isotopically the Rosetta lead is, as indicated above, an appropriate primary lead.

(3) In parts of this region uraninite crystals were subjected to a sudden chemical alteration at time t_m. Some lead was separated from the parent uraninite ores. This lead would consist of radiogenic lead formed between t_s and t_m, together with some primary or common lead originally present in the uraniferous minerals.

Burger *et al.* show that if the alteration at time t_m affects a series of uraninite ores with different U/Pb ratios the separated lead will have different but linearly related isotopic ratios, and will define a metachron or secondary isochron.

The major postulate made by Burger *et al.* was to identify the parent uraninite with the oval and spherical uraninite grains present in the Witwatersrand conglomerates and considered by many works to be of detrital origin. They showed that these uraninites were at least 2550 million years old (this limit becomes 2602 Ma using new decay constants and the recalculated metachron slope; see below) and noted the apparent discrepancy between this minimum age and the conclusion reached by other workers, namely that certain of the Witwatersrand uraninites appeared to have started their present chemical history about 2040 Ma ago.

Faced with this dilemma they proposed that a parent uraninite may undergo a solid-state chemical alteration such that lead is selectively removed from the crystal, and that the oval and spherical Witwatersrand uraninites had undergone such a rejuvenation about 2040 million years ago. They noted Liebenberg's observation (1958) that lead occurs as small veinlets and specks of galena in the uraninite while other uraninites are surrounded by thin rims of more completely exsolved galena, and suggested that with complete mobilization this exsolved lead is deposited as galena in veins cross-cutting the uraniferous conglomerate horizons. Thus at about 2040 Ma parts of the Witwatersrand Basin experienced a thermal event of sufficient intensity to locally mobilize lead present both as a trace component in the

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Witwatersrand detrital sulphide, and that generated over the preceding 1000 Ma in the uraninites and thucholites. This lead was mixed and deposited as galena in minor cross-cutting quartz veins. The great majority of these veins are entirely barren of uranium, gold or sulphides, and only carry sulphides, including galena, where they cut or lie within the mineralized conglomerates.

In more recent time further mobilization of both uranium and lead has occurred, as is testified by the presence of secondary uranium minerals in mine workings, and three-stage galenas (Burger *et al.* 1962, section VI (*b*)).

We would note that the new uranium decay constants, some new analyses of galenas from cross-cutting quartz veins (Köppel & Saager 1974) and refinements in the method of calculating regression lines necessitate slight modifications to the original treatment of the metachrons. Following Köppel & Saager (1974) the original galena isotopic data given by Burger *et al.* have been corrected on the basis of the measurements of the U.S. Geological Survey lead standard reported by them, and by Catanzaro *et al.* (1968). These modified data have been regressed following the method of Williamson (1968). Samples 33, 34 and 35 of Burger *et al.* (1962) have been excluded from the regression of the Witwatersrand galenas since we agree with Köppel & Saager (1974) that they may represent three-stage leads. Two additional galena samples analysed by Köppel & Saager (1974) have been included. The Witwatersrand main trend and the Dominion Reef galenas give metachrons or secondary isochrons of slopes 0.386 ± 0.003 and 0.413 ± 016 respectively. At the two-sigma level there is no significant difference between these metachrons. Their weighted mean, however, is strongly influenced by the high precision of the Witwatersrand metachron which largely reflects the high precision of Köppel & Saager's measurements on two extremely radiogenic samples. We accordingly think it more realistic to take an unweighted mean of the two slopes, namely 0.399 ± 0.019. Adopting 2040 Ma as the age of rejuvenation the metachron equation gives the age of the parent uraniferous mineral phases as 3065 ± 100 Ma, somewhat older than the age of 2980 ± 100 Ma ($= 2911$ Ma using revised decay constants) originally proposed by Burger *et al.* 1962).†

5. The significance of new total rock U–Pb analyses

The new analytical data are given in table 1. Ages have been calculated using the isotopic composition of lead from the Rosetta galena for the common lead correction.

The ages fall into two groups, one composed of samples yielding $^{207}Pb/^{206}Pb$ ages of *ca.* 3000 Ma, namely 217, 227, 228, 229, 230, 231 and 233, while the second gives younger ages. Two of the *ca.* 3000 Ma samples, 217 and 227, give concordant ages of 3029 ± 55 Ma 2921 ± 13 Ma respectively. Of the remaining samples in this group, 228, 229, 230 and 231 show a typical uranium loss pattern of ages while sample 233 shows a lead loss pattern. We would suggest that a first approximation to the age of the *ca.* 3000 Ma samples is given by regression of their 207/204 against 206/204 ratios (figure 4). The regression applies the appropriate pooled standard deviation to each ratio, and yields a best-fit line of 2945 ± 40 Ma. Sample 233 was excluded

† Slope of metachron or secondary isochron

$$= \frac{\exp(\lambda 235 t_s) - \exp(\lambda 235 t_m)}{137.8\,[\exp(\lambda 238 t_s) - \exp(\lambda 238 t_m)]}$$

where $\lambda 235$ and $\lambda 238$ are the decay constants for ^{235}U and ^{238}U respectively, and t_s and t_m are as defined in the text.

from the regression because its extremely high ratios dominate the calculation. The Rosetta lead fits this line within limits of error so providing independent justification for the use of this lead for the common lead correction. The mean square of weighted deviates (hereafter m.s.w.d., Brooks, Hart & Wendt 1972) is high (about 4), indicating that the scatter of data points about the best-fit line is greater than can be accounted for by experimental error alone. The inclusion of 233 in the regression gives a significantly greater age of 3090 ± 30 Ma with no improvement of the fit of data points about the line.

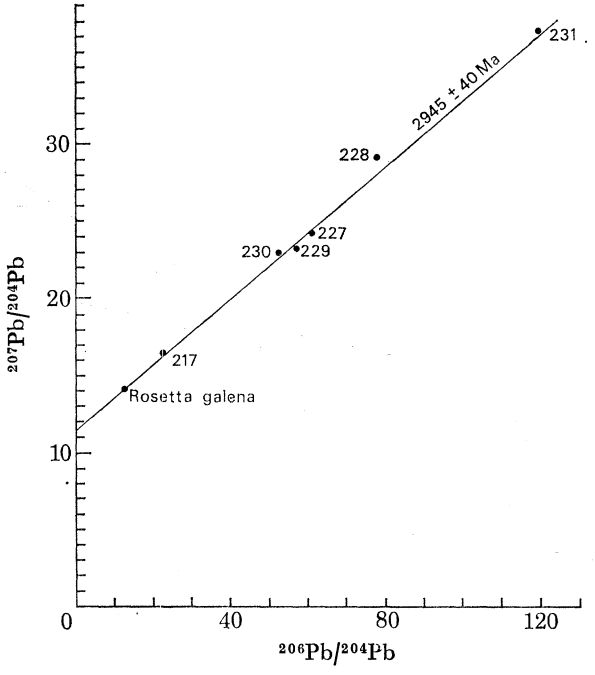

FIGURE 4. $^{207}Pb/^{204}Pb$ against $^{206}Pb/^{204}Pb$ plot for Upper Witwatersrand and Ventersdorp Contact Reef samples giving lead/lead ages of *ca.* 3000 Ma.

This *ca.* 3000 Ma-old suite of samples includes total conglomerates from both the Upper Witwatersrand and Ventersdorp Contact Reef. The age immediately suggests an affinity with the uraniferous minerals of the Dominion Reef and it is pertinent to plot the Dominion Reef total conglomerate and monazite data (Nicolaysen *et al.* 1962) on concordia together with the *ca.* 3000 Ma Witwatersrand and Ventersdorp samples (figure 5). It will be seen that total conglomerate samples 233, KCG1, KCG4, 217, B153, 228, 230, and detrital monazite KCG M give a good linear array. The best-fit line through these points gives a slope of 0.0329 ± 0.0007, and an intercept of −0.0262 ± 0.0111. The m.s.w.d. is only 0.4, indicating that all scatter of points about the line can be accounted for by experimental error alone and the line intersects concordia at 3050 ± 50 Ma. Thus all these samples can be regarded as having been derived from parent rocks containing 3050 ± 50 Ma uranium bearing minerals, some have experienced recent lead loss (233, KCG1 and KCG4), others recent uranium loss (KCG M, 228 and 230), while others have remained closed systems with respect to U and Pb and yield concordant ages not significantly different from 3050 ± 50 Ma (217, 3029 ± 55 Ma and B153, 3061 ± 25 Ma).

It might be argued that the inclusion of sample 230, which has extremely high 206*/238 and 207*/235 ratios, has a dominant effect on the regression calculation. However, it can be

shown that exclusion of this sample has no significant effect on the calculation and results in a 3060 ± 50 Ma intersection of the regression line with concordia, an effectively zero intercept, and an identical m.s.w.d.

Samples 227, 231 and 229 lie appreciably to the left of this line and their inclusion in the regression calculation, although giving a similar chord/concordia intersection of 2975 ± 60 Ma results in a significantly higher m.s.w.d. of 3.4. Thus scatter of these data points about the best-fit line is greater than can be accounted for by experimental errors alone, indicating that samples 227, 231 and 229 have experienced geochemical disturbance in addition to recent lead or uranium loss.

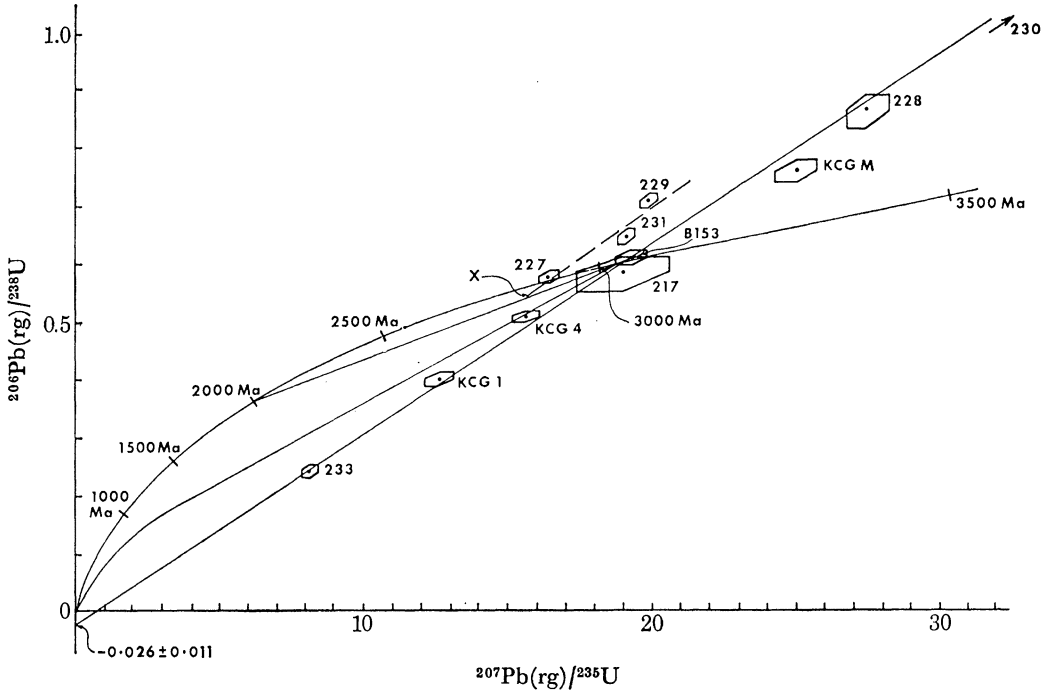

FIGURE 5. Concordia plot of *ca.* 3000 Ma-old samples from the Dominion Reef, Witwatersrand and Ventersdorp Supergroups. Sample 230 cannot be shown.

We suggest that this disturbance can be correlated with the 2040 Ma reworking proposed by Burger *et al.* (1962). Lead loss from rock samples occurring at this time would cause their data points to migrate along a chord between 3050 Ma and 2040 Ma. Two samples analysed in the course of this investigation (216 and 232, figure 6) in fact plot on the 3050 Ma/2040 Ma chord within limits of error and it will be noted that the use of a more radiogenic common lead correction merely displaces the data points along the chord. This effect is to be expected since the radiogenic common lead is one of the galenas from the Witwatersrand metachron. Burger *et al.* (1962) postulated that this lead was extracted at *ca.* 2040 Ma from a *ca.* 3000 Ma parent uraninite. If this postulate is correct then the use of the isotopic composition of any galena which plots on the Witwatersrand metachron as a 'radiogenic' common lead correction, is bound to move the respective data points along a trajectory parallel to the 2040 Ma/3050 Ma discordia. Sample 218 (figure 6) is virtually concordant irrespective of the common lead correction with a weighted mean of ^{206}Pb/^{238}U and ^{207}Pb/^{235}U ages of 2065 ± 13 Ma. This does not differ

significantly from the reworking age of 2040 ± 100 Ma, proposed by Burger *et al.* and it is clear that sample 218 experienced rejuvenation and virtually complete lead loss at this time.

Samples 227, 229 and 231 which we have shown to lie significantly to the left of the extension of the zero to 3050 Ma chord, thus show the effect of recent uranium loss imposed on slight lead loss at 2040 Ma, i.e. lead loss at 2040 Ma moved the data points a little way along the 3050 Ma/2040 Ma chord to about a point *X*, figure 5, and recent uranium loss caused the data points to move off the 2040 Ma/3050 Ma chord along trajectories away from zero and across concordia. Such a trajectory is indicated by a dashed line in figure 5. It will be observed that recent uranium loss from sample 227 was just sufficient for the datum point to come to rest on concordia and the sample provides a possibly unique case of accidental concordance.

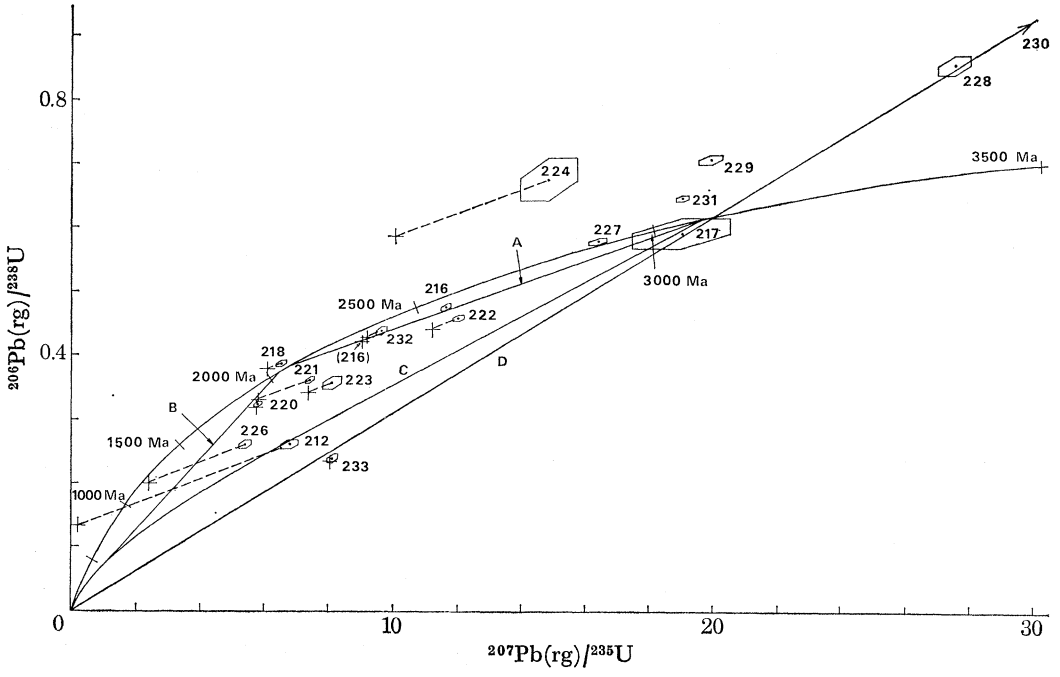

FIGURE 6. Concordia plot of 'total conglomerates' from the Witwatersrand, Ventersdorp and Transvaal Supergroups. Line A, *ca.* 3050 Ma/2050 Ma discordia; line B, *ca.* 2050 Ma lead loss by continuous diffusion trajectory; line C, *ca.* 3050 Ma lead loss by continuous diffusion trajectory; line D, recent episodic lead loss trajectory from *ca.* 3050 Ma sample.

These results provide a striking confirmation of the hypothesis first advanced by Burger *et al.* (1962) concerning the evolution of uraniferous minerals in the Witwatersrand Supergroup which predicts an age of the parent uranium minerals of 3065 ± 100 Ma, virtually identical with the estimate of 3050 ± 50 Ma derived above. The results carry the implication that the uraniferous conglomerates of the Dominion Reef, Witwatersrand and Ventersdorp Supergroups contain a common suite of parent uraniferous minerals. The age of 3050 ± 50 Ma for this parental suite is significantly older than the older limit to the deposition of the Witwatersrand sediments (2740 ± 19 Ma) and implies that the uraniferous minerals are detrital in origin.

With the exception of sample 224 the remaining samples (212, 220, 221, 222, 223 and 226) when plotted on the concordia diagram (figure 6) all fall within the triangular area bounded by the 2040 Ma/3050 Ma, zero/2040 Ma, and zero/3050 Ma chords. We suggest that these samples originally contained 3050 Ma-old uranium minerals which suffered variable lead loss

in response to the 2040 Ma reworking such that their data points were distributed along the 2040 Ma/3050 Ma chord. Subsequent lead loss, either episodic or by continuous diffusion, would then cause the data points to migrate into the triangular region defined above. It is not possible to decide which lead loss process was operative. Thus sample 220 must have experienced virtually complete resetting at 2040 Ma and now plots on the 2040 Ma lead loss by continuous diffusion trajectory. However, over most of its course this trajectory does not differ from the 200 Ma/2040 Ma episodic lead loss chord. Similarly, although the discordant age pattern of sample 212 is completely explicable in terms of lead loss by continuous diffusion from a 3050 Ma parent (it plots on the 3050 Ma continuous diffusion trajectory), it could equally reflect partial resetting in response to the 2040 Ma reworking involving about 50 % lead loss at that time, with further lead loss in response to a more recent (*ca.* 200 Ma) disturbance.

Sample 224 plots above concordia and gives a uranium loss discordant age pattern. We consider that this reflects a similar history to that experienced by samples 227, 229 and 231. In this case considerable lead loss ($\approx$ 50 %) from a 3050 Ma parent occurred in response to the 2040 Ma reworking moving the data point about half way down the 2040 Ma/3050 Ma chord. Recent uranium loss then caused the point to move away from the origin, and across concordia to its present position.

It is now appropriate to reconsider the early uraninite/thucholite U–Pb age determinations in the light of the above considerations. Wetherill (1956) had observed that most of the determinations when plotted on concordia fell on or close to a 200 Ma/2040 Ma chord indicating episodic lead loss at 200 ± 150 Ma from a 2040 Ma parent (see figure 3). Over most of its length the 200 Ma/2040 Ma chord hardly differs from the 2040 Ma lead loss by continuous diffusion trajectory so that the discordant age patterns of samples 1, 2, 3, 4*b*, 4*g*, 4*h* and 4*l* can be equally well explained in terms of episodic lead loss at 200 Ma or lead loss by continuous diffusion since 2040 Ma. Sample 4*f* plots above concordia and its age pattern can be explained in terms of recent uranium loss effecting a 2040 Ma parent. All these samples could have evolved from 3050 Ma old parent uraninites which experienced virtually complete resetting at 2040 Ma.

Of the remaining samples, 4*a*, 4*c*, 4*i*, 4*n* and 6 are considered to have had an original age of 3050 Ma but experienced only partial lead loss in response to the 2040 Ma resetting event such that they plotted on the 2040 Ma/3050 Ma chord. Subsequent lead loss, either in response to a more recent resetting event (200 ± 150 Ma?), or by continuous diffusion, caused the data points to migrate into the triangular area bounded by the 2040 Ma/3050 Ma, zero/2040 Ma, and zero/3050 Ma chords along trajectories indicated by dashed lines in figure 3. It is the extension of these trajectories across the 2040 Ma/3050 Ma chord on to concordia (indicated by dotted lines in figure 3) which led Wetherill (1956) to suggest the existence of *ca.* 2250 Ma and *ca.* 2450 Ma parent uraninites. There is clearly no need to postulate the existence of uraninites of these ages.

Sample 4*o* is from the Dominion Reef Supergroup and plots within error limits on the 3050 Ma lead loss by continuous diffusion trajectory. Nicolaysen *et al.* (1962) had previously noted that the discordant age pattern of this particular specimen could be explained solely in terms of such lead loss and showed that the true age of the uraninite would be about 3040 Ma.

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6. Some conclusions

Total rock U–Pb analyses on samples from the Witwatersrand Ventersdorp and Transvaal Supergroups, together with earlier data reported by Nicolaysen *et al.* (1962) from the Dominion Reef Supergroup show that the uraniferous minerals in the conglomerates constitute a 3050 ± 50 Ma-old system. This system experienced a major reworking at 2040 ± 100 Ma which varied in intensity, but locally resulted in a complete resetting of U:Pb ages to *ca.* 2040 Ma in both uraninites and total rock. Radiogenic lead released during this reworking crystallized as galena in veins and fissures which cut across the uraniferous conglomerates. Isotopically these leads define a metachron or secondary isochron the slope of which is fully consistent with a *ca.* 2040 Ma reworking extracting lead from a *ca.* 3050 Ma parent uraniferous mineral.

Subsequent loss of lead or uranium has produced a plethora of discordant ages. In some cases it appears most likely that the lead and uranium loss occurred in very recent time and that the lead loss was episodic rather than by continuous diffusion. In other cases lead loss by continuous diffusion is possible, but there remains no objective way of distinguishing between lead loss by continuous diffusion from a 3050 Ma uraniferous parent mineral, and lead loss by either continuous diffusion or episodic loss from a 3050 Ma parent mineral which had previously undergone partial resetting in response to the *ca.* 2040 Ma reworking. Many of the separated uraninites analysed by the earlier workers show evidence of virtually complete resetting at *ca.* 2040 Ma. Their lead loss age patterns, however, can be interpreted either in terms of an episodic loss at 200 ± 150 Ma, a resetting event that can be correlated with the Karroo volcanism, or by continuous diffusion since 2040 Ma.

There is considerable indication of an areal control to the discordant age patterns. Thus samples from the Upper Witwatersrand to the south in the Orange Free State (regions 1 and 2 of figure 2, the Odendaalsrus–Welkom–Virginia area) show little effects of the 2040 Ma reworking, evidence of recent uranium loss, and no evidence of recent lead loss (see figure 5).

Total conglomerates from the Dominion Reef in the Klerksdorp area analysed by Nicolaysen *et al.* (1962) are either concordant (B153) or show evidence of an episodic recent lead loss, but, like the samples from the Orange Free State area, show little if any effects of the 2040 Ma reworking. A separated uraninite from the Dominion Reef gives a discordant age pattern which can be interpreted solely in terms of continuous lead loss from a *ca.* 3050 Ma parent. A separated detrital monazite shows evidence of recent uranium loss (see figure 5).

With the exception of sample 233 (E. Rand, E. Geduld Mine) and possible 212 (Basal Reef, Western Reefs Mine) the remaining total conglomerates analysed in this work all show very strong effects of the *ca.* 2040 Ma reworking (figure 6). These samples are all from the Klerksdorp, Carletonville, and E. Rand areas. Sample 233 gives a discordant age pattern unaffected by the *ca.* 2040 Ma reworking. Sample 212 comes from the Black Reef in the Transvaal System and plots on the 3025 Ma lead loss by continuous diffusion trajectory. As pointed out above, however, the discordant age pattern could equally well be due to recent episodic lead loss from a 3050 Ma parent which had previously been partially reset at 2040 Ma.

Sample 224, a strongly sheared rock from the Dennys conglomerate in the Western Reefs mine shows the effects of recent uranium loss following the 2040 Ma reworking. Samples 218, 216, and 232 have remained closed chemical systems with respect to uranium and lead since the 2040 Ma reworking, while the remaining samples, namely 220 221, 222, 223 and 226 have all experienced lead loss since the 2040 Ma reworking.

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The problem of the correlation of the 2040 Ma old reworking of the Witwatersrand uraninites with a specific geological thermal event was fully discussed by Nicolaysen *et al.* (1962). They were unable to come to a definite conclusion, however, because of the uncertainty concerning the age of the Ventersdorp volcanism. It is now clear that the reworking of the uraninites at *ca.* 2040 Ma occurred after the Ventersdorp volcanism. We suggest that the occurrence of galenas, rich in radiogenic lead which plot on the Witwatersrand metachron, in cross-cutting veins in the Black Reef implies that the *ca.* 2040 Ma reworking is of late or post Transvaal age. This thermal event could thus be correlated with volcanic manifestations in the Transvaal Supergroup and the emplacement of the Bushveld Complex dated at 1950 ± 150 Ma, as suggested by van Niekerk & Burger (1964). In this connection we would note that all the analysed total rocks which show the effect of *ca.* 2040 Ma reworking come from the northern parts of the Witwatersrand Basin, i.e. the nearest to the Bushveld Complex.

The presence of 3050 ± 50 Ma-old uraniferous minerals in the Dominion Reef and Witwatersrand sediments which we have argued must have been deposited after about 2740 ± 19 Ma, carries with it the simple implication that the uraniferous minerals (originally mainly uraninite) are of detrital origin.

It might be argued that some of the uranium in the Witwatersrand sediments was deposited from solution either at the time of sedimentation or during diagenesis. Such uranium–lead systems would plot on concordia at about 2550 Ma but would mix with the older detrital component to generate a chord between 2550 Ma and 3050 Ma. We see no evidence of such a chord. It must be remembered, however, that most of the analysed samples have been affected to some extent by the 2040 Ma rejuvenation, and by subsequent loss of lead or recent loss of uranium. It would be virtually impossible to distinguish between disturbed detrital systems and disturbed systems with both detrital and authigenic components. The data presented here can be interpreted entirely in terms of a 3050 Ma old U–Pb system; the earlier work on separated uraninites and thucholites and on galenas fully supports this interpretation. We would suggest that authigenic mineralization has made at the most only a minor contribution to the total uranium in the Witwatersrand Basin.

We are indebted to our colleagues for their help and encouragement. We would in particular thank Dr S. H. U. Bowie at whose suggestion the work was undertaken, Dr I. G. Swainbank for much advice and help with the analytical work, and Mr R. D. Beckinsale for stimulating discussion which contributed greatly to the interpretation of the discordant ages advanced here. The analysed samples were generously provided by Dr H. C. M. Whiteside.

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