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FISSION XENON DATING OF WITWATERSRAND
URANINITES: IMPLICATIONS FOR GEOLOGICAL
ACTIVITY IN THE CENTRAL KAAPVAAL CRATON
AROUND 1 Ga AGO

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INFORMATION CIRCULAR No. 284

UNIVERSITY OF THE WITWATERSRAND

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ABSTRACT

Four uraninite separates from different conglomerate reefs in the Central Rand Group (Upper Witwatersrand Supergroup) and Ventersdorp Supergroup rocks were obtained from gold mines in the East Rand, West Rand, and Welkom Goldfields of the Witwatersrand Basin in South Africa. These samples were dated by the Xe_sXe_n spectrum technique. All four age spectra have a typical "step-up" form with plateaux for high temperature Xe release. The plateau ages are 0.84 ± 0.04 Ga for the Carbon Leader Reef sample from Blyvooruitzicht Gold Mine, 1.035 ± 0.05 Ga for the Basal Reef sample from Free State Geduld Gold Mine, 1.12 ± 0.05 Ga for the Main Reef uraninite concentrate from Sub Nigel Gold Mine, and 1.26 ± 0.02 Ga for the Ventersdorp Contact Reef sample from Kloof Gold Mine. No single apparent ages in excess of about 1550 Ma were recorded.

These results clearly do not correspond to primary crystallization ages for the uraninites. Instead, they most likely represent ages of resetting events that affected the U-Xe isotopic system in these samples. In comparison with other recent chronological results obtained from rocks in and around the Witwatersrand Basin, it can be confidently concluded that the region was affected by about 1.3 to 1 Ga magmato-tectonic activity, the latter not only being localized along the southern margin of the Kaapvaal Craton, the so-called Namaqua-Natal Thrust Front, but also affecting other parts of the craton.

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INTRODUCTION

The Witwatersrand Basin (Fig. 1) in the central part of the Kaapvaal Craton in South Africa is well known for its exceptional gold mineralization. In addition, uranium was mined for several decades, particularly during the Seventies, from a number of the Witwatersrand conglomerate reef horizons. Uranium, mainly in the form of uraninite and its alteration products coffinite and brannerite, occurs together with gold in all exploitable reefs, but is particularly concentrated in some of the conglomerate reefs of the Central Rand Group.

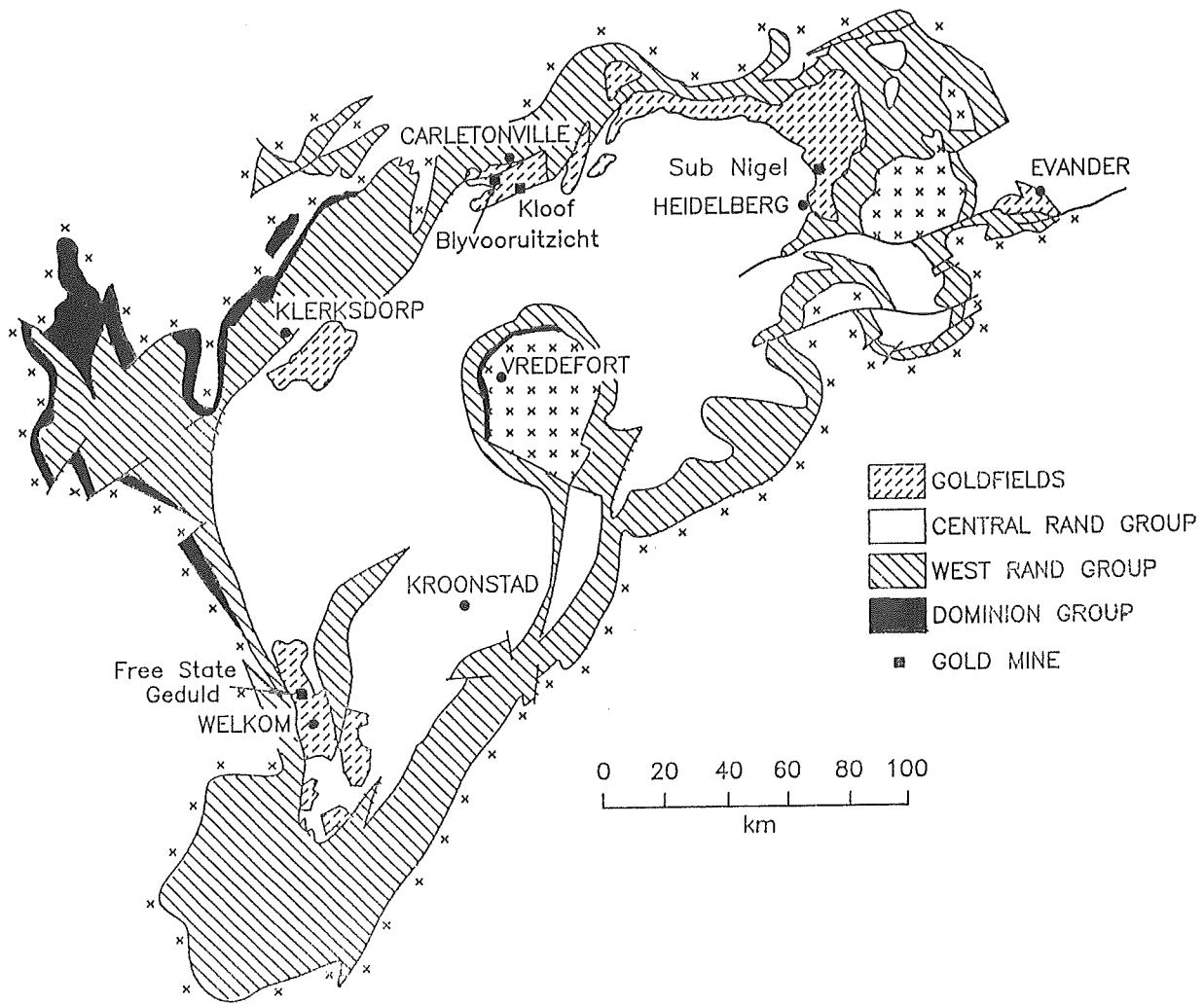


Figure 1: Schematic geology of the Witwatersrand Basin with the locations of the four gold mines (Sub Nigel, Kloof, Blyvooruitzicht, Free State Geduld), from which samples were obtained for this study.

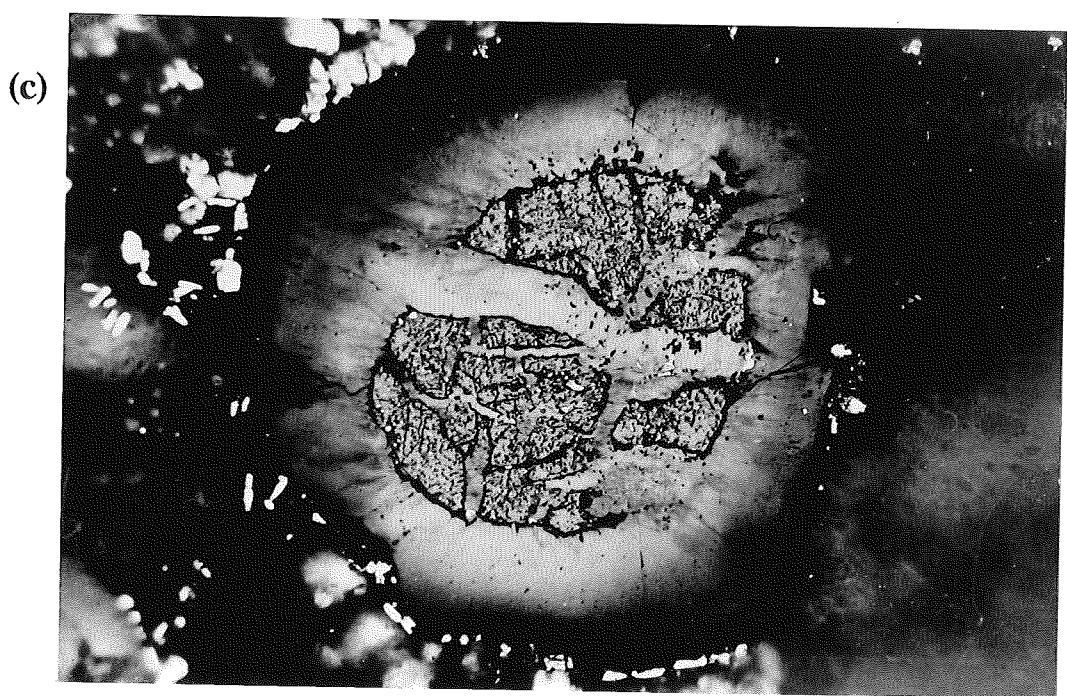
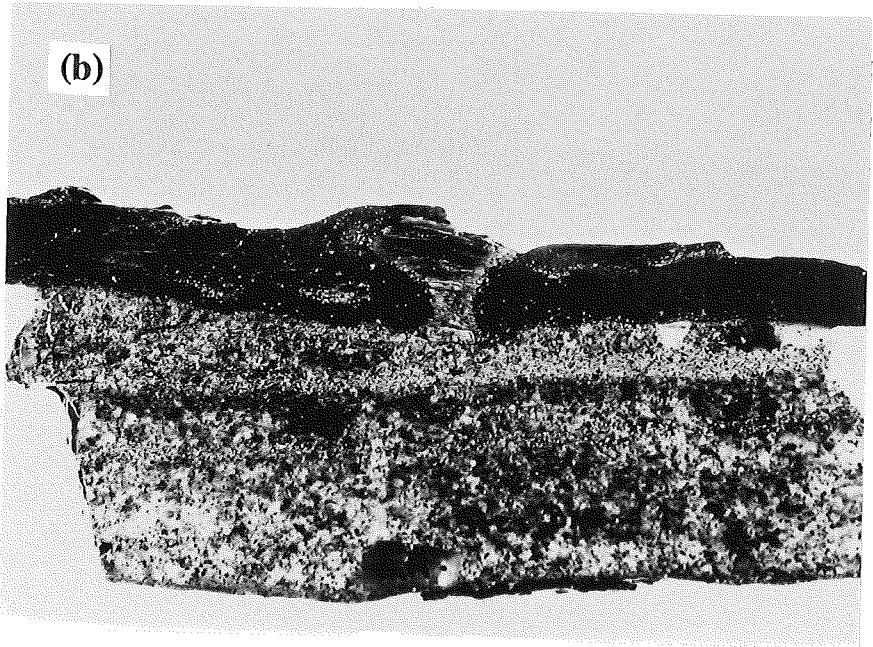
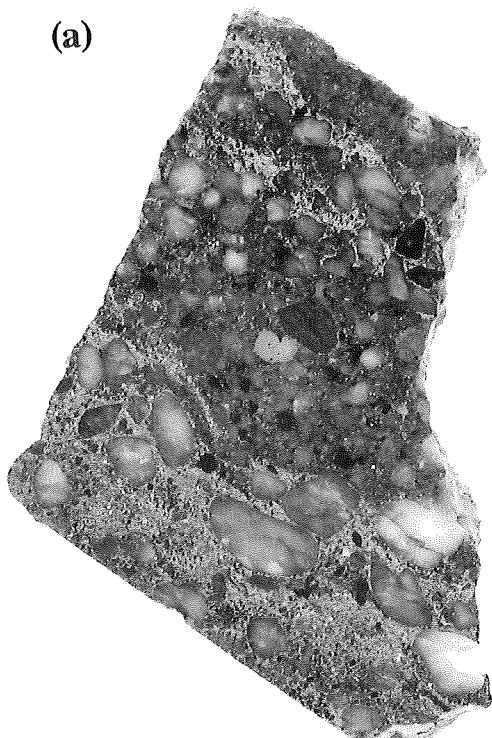
Over the years several theories have been proposed for the origin of the gold and uraninite in the Witwatersrand Basin (see e.g., reviews by Pretorius, 1976, 1981; Frimmel et al., 1993; Frimmel, 1994). Various schools of thought periodically supported either a purely detrital (e.g., Hallbauer, 1986; Minter et al., 1993) or a purely hydrothermal (e.g., Graton, 1930; Davidson, 1957; Phillips and Myers, 1989) origin for the Witwatersrand ores. In addition, a large number of workers have subscribed to the so-called 'modified placer hypothesis' (first discussed by Mellor, 1916, and repeatedly invoked in recent years: e.g., Feather and Koen, 1975; Schidlowski, 1981; Pretorius, 1976; Robb and Meyer, 1991), which assumes a detrital origin for the primary gold and uraninite (probably derived from granite-greenstone terrane sources in the hinterland to the basin - e.g., Barton et al., 1989; Drennan et al., 1990; Robb et al., 1990a,b), but also acknowledges later post-burial stages of in situ remobilization and redeposition of gold and uraninite. These processes may have affected the size and morphology of gold particles, but not those of uraninite (Koen, 1961). A large portion of the uraninite was entrapped by primitive algal communities during early deposition (e.g., Smits, 1984, 1992).

Evidence for the detrital origin of Witwatersrand uraninite (see Fig. 2c) and gold is provided by their specific concentration at the very base of conglomerate reefs, where they were deposited together with other heavy minerals, such as pyrite, arsenopyrite, chromite, and zircon, before the pebble fraction of the conglomerate itself (e.g., Liebenberg, 1955, 1960; Koen, 1961; Schidlowski, 1966, 1970). Detailed petrographical and chemical studies of conglomerate reef transects have, however, shown that significant amounts of gold and uraninite were remobilized throughout the entire reef zone (from lower parts of the hanging-wall through the reef itself into the footwall rocks) (e.g., Liebenberg, 1955; Ramdohr, 1955; Hiemstra, 1968; Reimold and Boer, unpubl. data). Locally, primary detrital gold and uraninite may no longer be present in the ores, and petrographic analysis shows that hydrothermal processes have pervasively overprinted such reef zones.

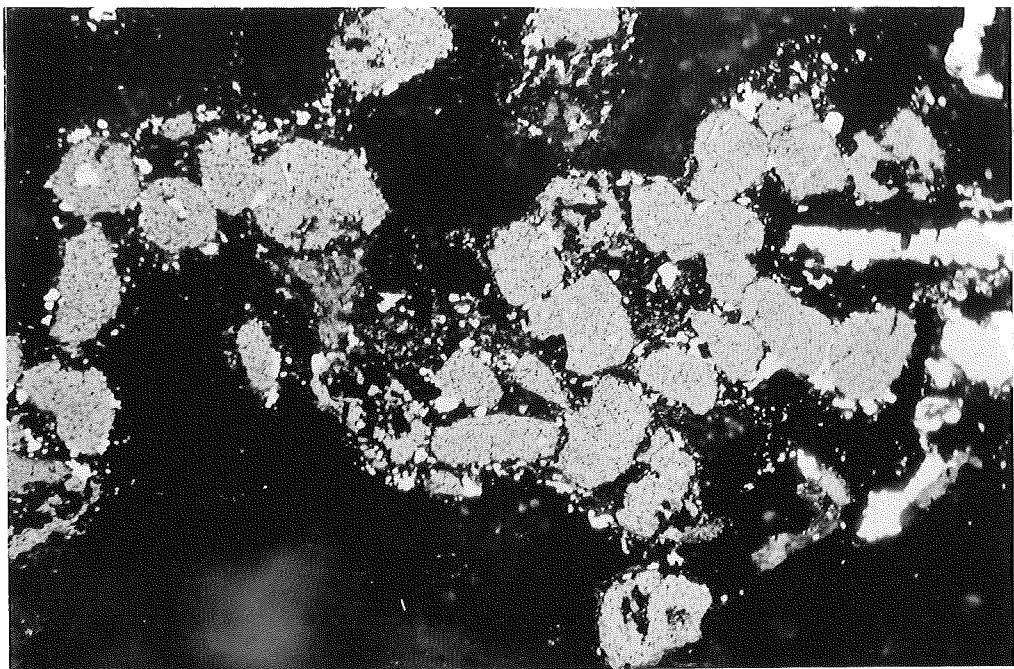
Microscopic studies indicate that detrital gold fills interfilamentous spaces in kerogen and, hence, was entrapped mechanically. Uraninite, in contrast, was at least partially trapped by biological processes (e.g., Smits, 1984; 1992; Feather and Glatthaar, 1987). The texture of kerogen, the metamorphic product of organic matter, strongly suggests that primitive organisms grew onto detrital uraninite (Fig. 2d) and also penetrated these grains along crystallographic planes or fractures, thus fragmenting some grains into smaller subgrains (Fig. 2e). Unfragmented uraninite grains encapsulated by kerogen are similar in size and shape to the detrital uraninite which is not enclosed by kerogen (Smits, 1984). In both modes of occurrence, the uraninite remained unaffected and nowhere underwent oxidation or recrystallization.

Current Witwatersrand researchers are in agreement that relatively low-temperature ($\sim 270 - 400$ °C) metamorphism affected the entire basin in post-depositional times (Frimmel, 1994 and references therein). Evidence for localised intense fluid-related alteration is known from all reefs and all goldfields (Phillips, 1987; Phillips and Myers, 1989; Phillips and Law, 1993; Frimmel, 1994; Boer et al., 1993a,b). As a consequence of these secondary events gold, uranium, and other base metals were, basin-wide and on a grand scale, remobilized and redeposited as hydrothermal assemblages (e.g., Feather, 1981; Schidlowski, 1981). It has been widely speculated whether these effects were the result of

Figure 2: Examples of typical Ventersdorp Contact Reef and Carbon Leader Reef conglomerate. (a) VCR conglomerate from East Driefontein Gold Mine (located close to Kloof Gold Mine) with sulphide-rich bands at the bottom and upper parts of the specimen. Most pebbles shown consist of vein quartz, but several dark shale pebbles are seen in the upper part. Maximum diameter: circa 15 cm. (b) Basal Reef from President Brand Gold Mine. Thickness of kerogen seam is 12 mm. (c) Kerogen nodule (diameter 0.2 mm) enclosing fragmented uraninite; from Basal Reef on President Brand Gold Mine.



(d)



(e)



Figure 2
(contd)

(d) Group of detrital uraninite grains (average diameter 70 - 100 μm) from the Monarch Reef, West Rand Consolidated Gold Mine. (e) Fragments of a uraninite grain (original size: 100 μm) locked in a kerogen seam (Vaal Reef from Vaal Reef East Gold Mine).

deposition of the Ventersdorp extrusives on top of the Witwatersrand Supergroup (Fig. 3) around 2.7 Ga ago, or whether they took place at later times (during the approximately 2.5–2.25 Ga Transvaal, 2.05 Ga Bushveld, or even post-Bushveld times; discussed for example by Frimmel, 1994). Evidence for several successive alteration events has been recorded (e.g., Zhao et al., 1993; Reimold and Boer, 1993), particularly during studies of the Ventersdorp Contact Reef (VCR) at the unconformity between Ventersdorp and Witwatersrand Supergroups (Fig. 3). The basin-wide low-temperature metamorphism has often been related to the 2.054 Ga (Walraven and Hattingh, 1993) emplacement of the Bushveld Complex to the northeast of the basin and/or to the formation of the Vredefort Dome in the centre of the Witwatersrand Basin (Frimmel, 1994; Reimold, 1994). Reimold (1994) also discussed petrographic, chemical, and chronological results obtained on the VCR in the Central and West Rand Goldfields, which suggest a direct link between a circa 2.017 Ga tectonic and alteration event that affected the northern and northwestern parts of the Witwatersrand Basin and the formation of the Vredefort Dome at approximately that time (viz. at 2.024 Ga ago – some 30 Ma later than the Bushveld event – Kamo et al., 1995; cf. also Trieloff et al., 1994; Spray et al., 1995).

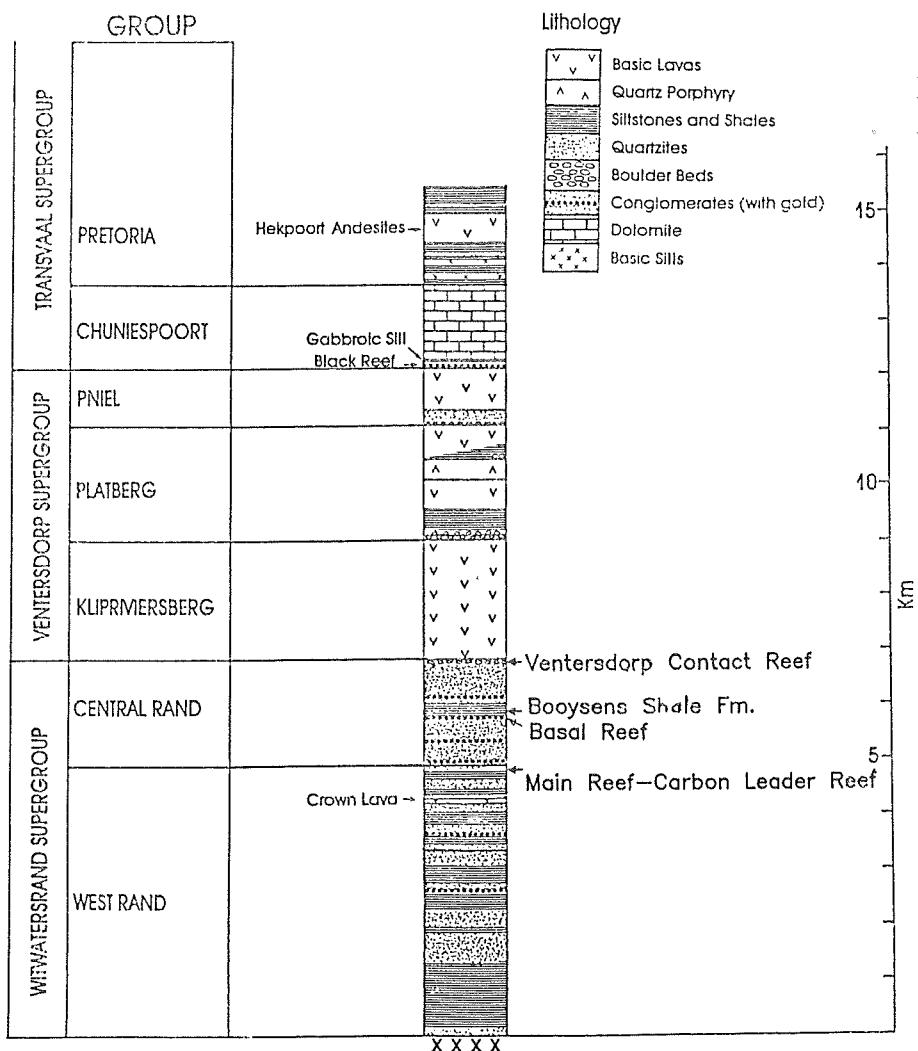


Figure 3: Schematic stratigraphy for the region of the Witwatersrand Basin (modified after Fletcher and Reimold, 1989). Indicated are the stratigraphic positions of the Main Reef, Carbon Leader Reef, Basal Reef, and Ventersdorp Contact Reef that yielded samples for this study.

In this study the fission xenon dating method is applied for the first time in an attempt to constrain the formation ages of uraninite grains in a series of samples collected from different Witwatersrand conglomerate reefs (stratigraphic positions indicated in Fig. 3). Samples were obtained from gold mines in the East Rand (Sub Nigel Gold Mine), West Rand (Kloof and Blyvooruitzicht Gold Mines), and Welkom (Free State Geduld Gold Mine) Goldfields (Fig. 1). The rationale for studying samples from these different goldfields was to investigate uraninite from an arc around the Vredefort structure, the latter having provided a possible source of thermal energy and fluids for the remobilization of metals in its environs at the time of its formation. In addition, samples from different reefs would be used to test the possibility that the Witwatersrand Basin may have been affected by igneous activity linked to the 2.054 Ga Bushveld Complex northeast of the basin.

GEOCHRONOLOGY

Earliest U-Pb dating of galena and uraninite from various reefs in the Witwatersrand and Ventersdorp Supergroups (approximately 3.07 to 2.70 Ga - Armstrong et al., 1991; Barton et al., 1989; Robb et al., 1990a) indicated an age dichotomy. Clear evidence for the formation of detrital phases prior to 3 Ga has been presented, but a major resetting event at about 1900-2200 Ma was also indicated (e.g., Köppel and Saager, 1974; Rundle and Snelling, 1977 and references therein). Ion microprobe dating of uraninite by Feather (1981) also yielded a relatively young age of 1770 ± 60 Ma. Köppel and Saager (1974) analysed pyrite from several Witwatersrand conglomerate reefs using the U-Pb technique and arrived at the conclusion that the formation of pyrite took place prior to 3 Ga ago, but that 'rejuvenation' occurred around 1950 Ma. Giusti (1988) deduced, from U-Pb isotopic analysis of pyrites from the hydrothermally altered Varkenskraal Granite in the northern hinterland to the Witwatersrand Basin, that these pyrites were formed in post-Ventersdorp times, probably not much later than 2200 Ma ago.

Much of the early chronological work was focussed on the Ventersdorp Supergroup, one reason being that the volcanic rocks of this sequence were believed to be suitable for determination of a minimum age for the metasedimentary packages of the Witwatersrand Supergroup. Earliest dating of zircons from the Makwassie Quartz Porphyry (Van Niekerk and Burger, 1964) indicated an age of 2.3 ± 0.1 Ga, and, while some ages by Van Niekerk and Burger (1978) suggested a Ventersdorp formation age of 2643 ± 80 Ma, Allsopp and Welke (1986) still entertained the possibility that the Ventersdorp Supergroup could be as "young" as 2300 Ma. Walraven et al. (1987) and Armstrong (1987) reported Pb-Pb whole rock ages of $2352 + 160/-184$ Ma and $2371 + 102/-106$ Ma for Makwassie quartz porphyry, respectively, but also a U-Pb zircon age of $2693 + 60/-59$ Ma (Walraven et al., 1987). Rb-Sr data for Klipriviersberg Lava (Ventersdorp Supergroup) suggested an age of 1960 Ma (Burger and Walraven, 1980). Pb-Pb whole rock analyses of two suites of Klipriviersberg Lava samples by Armstrong et al. (1986) yielded $2360 + 320/-350$ Ma and 2370 ± 70 Ma ages. A whole rock Rb-Sr isochron for Makwassie samples by these authors suggested an age of 2140 ± 260 Ma, whereas their first SHRIMP zircon data constrained the Makwassie age to 2699 ± 16 Ma. The Ventersdorp age was finally resolved, when Armstrong et al. (1991) obtained comprehensive SHRIMP results for felsic (Makwassie) and mafic (Alberton) Ventersdorp volcanics of 2709 ± 4 and 2714 ± 8 Ma, respectively. These results demonstrated that later disturbances of the U-Pb and Rb-Sr isotopic systematics of these

rocks caused resetting around 2.3 and 2.1 Ga ago. Armstrong et al. (1991) also reported some relatively young zircon ages between 2.37 and 0.9 Ga, which they interpreted to represent late growth of authigenic zircon.

In a study of single zircons from altered granites in the hinterland of the basin, Robb et al. (1992; fig. 2) found that some of their data fell onto discordia lines with lower intercepts at 1089 and 962 Ma. In addition, they reported several even younger lower intercept ages between 523 and 113 Ma.

Muscovite and galena ages for authigenic growths in fractures or in shear zones (Barton et al., 1988) suggested tectonic activity with possible hydrothermal crystal growth between 2.37 and 2.05 Ga ago.

Even later magmato-tectonic activity is indicated by some Rb-Sr ages for dykes intruding strata in the Witwatersrand Basin: Schreiner and Van Niekerk (1958) reported an age of 1290 ± 180 Ma, and Van Niekerk (1962) two ages of 1330 ± 80 and 1310 ± 60 Ma for such intrusives.

^{40}Ar - ^{39}Ar stepheating and argon laser-probe dating were carried out in recent years on pseudotachylites and samples of country rocks (and minerals contained therein) from the Vredefort Dome (Reimold et al., 1990, 1992; Allsopp et al., 1991; Spray et al., 1995). Several pseudotachylite samples from a fault zone at the contact between Ventersdorp and Witwatersrand Supergroups, sampled in Central and West Rand gold mines, were also dated with the ^{40}Ar - ^{39}Ar stepheating technique (Trieloff et al., 1994). In summary, it was established that Vredefort and Witwatersrand pseudotachylites formed about 2.01 Ga ago. However, some Vredefort pseudotachylites apparently experienced later resetting 1.1 to 1.4 Ga ago (Reimold et al., 1990). According to these authors, this could either be interpreted to indicate formation of pseudotachylites at different times or post-formational argon loss due to thermal overprint. However, some evidence for the formation of pseudotachylite in post-1.6 Ga times was also recorded (Reimold et al., 1988; Spray et al., 1995). No evidence for significant post-formation argon loss was observed in the Witwatersrand pseudotachylite experiments (Trieloff et al., 1994), but minor events between 1.3 and 1.6 Ga and, possibly, even as late as 300 Ma ago are indicated by these data. Minerals and whole rock samples of country rocks from the Vredefort Dome also do not indicate significant post-2 Ga resetting. This latter observation provided a strong argument in favour of the earlier suggestion (Reimold et al., 1990, 1992) that some pseudotachylites in the Vredefort structure could have formed as late as 1.1 Ga ago.

A Rb-Sr and Pb-Pb whole rock and mineral chronological study of an alkali granitic intrusion in the area of the Vredefort Dome by Walraven and Elsenbroek (1991) provided evidence of secondary overprinting of this body at times later than 2.2 Ga ago. The Rb-Sr study, by Coetzee and Kruger (1989), of the Losberg Complex, located some 15 km northeast of the Vredefort Dome, yielded a 2042 ± 41 Ma age for this apparently Bushveld-related intrusion, but also clearly demonstrated post-2.05 Ga open system behavior, particularly of the Pb-Pb system of this body.

Robb et al. (1994) discussed U-Pb isotopic data of kerogen samples from several Witwatersrand conglomerate reefs and from peraluminous granites adjacent to the basin (original work by Allsopp et al., 1986; and Giusti, 1988; also data by Robb et al., 1994). They suggested that these data could be interpreted to indicate kerogen formation in both reefs and granites, around $2329 +69-60$ and $2380 +441-91$ Ma ago. The available data, according to Robb et al. (1994), define a "single, albeit somewhat diffuse, Pb-loss discordia suggesting an upper intercept age of circa 2300 Ma". Inspection of the data of Robb et al. (1994) further reveals that the discordia trends have lower intercepts with the concordia line at about 1.6 and 1.33 Ga.

An Rb-Sr chronological study by Pybus et al. (1994) showed that the Vredefort structure is transected by a thin, but extensive mafic sheet of 1054 ± 13 Ma age.

A number of 1250-1350 Ma ages have also been determined for alkaline and mafic rocks intrusive into the southern part of the Bushveld Complex (Brandt, 1994; Harmer, 1985).

EXPERIMENTAL PROCEDURES AND SAMPLE DESCRIPTIONS

Geochronological Method

The $\text{Xe}_s\text{-}\text{Xe}_n$ dating method, also called the Xenon Dating Technique or the Xenon-Xenon Spectrum Technique (Shukolyukov et al., 1974), is based on the spontaneous fission of ^{238}U nuclei (Shukolyukov, 1970), which results in the production of spontaneous fission Xenon (Xe_s). This method belongs to the group of so-called neutron-induced geochronological procedures that also includes the better known $^{40}\text{Ar}\text{-}^{39}\text{Ar}$ technique. In these methods the concentration of parent isotopes (^{40}K , ^{238}U) is not measured directly. Instead, a mineral is irradiated in a neutron flux to artificially produce daughter isotopes (^{39}Ar , Xe_n) from other parent isotopes (^{39}K , ^{235}U). The ratio of the concentrations of the naturally occurring isotopes to those of the neutron-induced isotopes in the sample ($^{39}\text{Ar}/^{40}\text{Ar}$, Xe_s/Xe_n) can then be used as a measure of the age of the sample.

The $\text{Xe}_s\text{-}\text{Xe}_n$ dating method has been described in detail by a number of authors (e.g., Shukolyukov et al., 1974; Teitsma et al., 1975; Teitsma and Clarke, 1978; Shukolyukov and Meshik, 1987). This method possesses the same advantages as the $^{40}\text{Ar}\text{-}^{39}\text{Ar}$ technique. Firstly, it has the capacity to provide at least some chronological information per experiment, even in a case of partial loss of the daughter isotope, because of its suitability to stepwise degassing of the sample. Second, only isotopic ratios and no absolute concentrations have to be measured in order to determine an age (Shukolyukov et al., 1974). In addition, the $\text{Xe}_s\text{-}\text{Xe}_n$ method has a number of advantages in comparison with the $^{40}\text{Ar}\text{-}^{39}\text{Ar}$ technique: (1) it is much easier to irradiate samples for $\text{Xe}_s\text{-}\text{Xe}_n$ dating, because only a relatively small thermal neutron flux and no irradiation with fast neutrons are required; (2) because of an extremely small background (Clarke concentration) of Xe in the Earth, excess or trapped radiogenic Xe_s does not present a problem, in contrast to the $^{40}\text{Ar}\text{-}^{39}\text{Ar}$ method, where significant disturbances due to excess ^{40}Ar frequently occur; (3) $\text{Xe}_s\text{-}\text{Xe}_n$ ages can be calculated in three different ways, making use of different isotopic ratios, whereas in the case of Ar dating only one radiogenic isotope (^{40}Ar) can be used.

The Xe_s - Xe_n dating method has previously been successfully applied to dating of zircon (Shukolyukov et al., 1979; Krylov et al., 1993; Shukolyukov et al., 1993), pitchblende (Shukolyukov and Meshik, 1987, 1988; Leveque and Meshik, 1991), and other uranium or uranium-bearing minerals.

In this study, uraninite concentrates, as supplied from the various gold mines, were further enriched by diligent handpicking in order to: (1) reduce the mass of the samples to be irradiated, and, hence, to delimit the required activity and resulting post-irradiation cooling duration; and (2) to remove as many contaminating grains as possible. Samples were irradiated in the thermal column of the MIFI research reactor of the Moscow Engineering Physics Institute in two batches: samples BV (from Blyvooruitzicht Gold Mine) and SN (Sub Nigel Gold Mine) in the first and samples FSG (Free State Geduld Gold Mine) and VCR (Kloof Gold Mine) in the second. In each irradiation the Witwatersrand samples were accompanied in the same ampoule by three specimens of "Berdiaush" zircon of well-known age (Krylov et al., 1993) serving as monitors of the thermal neutron flux. The ratio of thermal to fast neutrons exceeded 5 000, so that contributions from ^{238}U and ^{232}Th fission products were negligible. Variation of the neutron flux along the ampoule was determined to be < 1.5%, which represents the uncertainty of the absolute flux determination.

A month after irradiation the samples were introduced into the gas extraction system and baked out overnight at 200°C. The step-wise heating procedure was then carried out in a double-vacuum, high-temperature oven with Ta crucibles. The temperature was increased from 450 to 1800 °C in 7 to 11 temperature steps, each time degassing for 30 minutes. After a sample had been fully degassed, the temperature in the furnace was increased to 2000 °C, under vacuum, to reduce the "memory" effect. The "hot" blank after baking was less than $5 \cdot 10^{-14}$ cc STP of ^{132}Xe .

A classical gas separation system with two Ti-Zr getters and two activated charcoal traps was used for the separation of Xe from active gases and from He, Ne, Ar, and, partially, Kr.

Xe isotopic analyses were carried out on a MI-1201 directional-focusing high-resolution mass spectrometer operated in static vacuum mode. In order to increase the precision of isotopic analyses, the triple dilution technique was applied. The isotopic composition of the tracer is shown in Table 1. Application of the triple isotope dilution technique also allows avoidance of numerous measurements of standards, which would normally be carried out for mass discrimination correction.

Data processing involved correction of the measured Xe isotopic ratios for mass discrimination and air contribution, which normally amounted to less than a few per cent of radiogenic Xe. Thereafter, on the basis of the difference in isotopic composition of Xe_s and Xe_n (Table 1), Xe_s/Xe_n ratios and apparent ages were calculated for each degassing step. The uncertainty of measured isotopic ratios (1σ) was 0.2 to 0.6%, depending on the relative amounts of xenon available in the samples.

Sample Descriptions

The four samples studied in this investigation were obtained in the form of heavy-mineral concentrates from the respective gold mines. The sample from the Carbon Leader Reef on Blyvooruitzicht Gold Mine consisted of uraninite and minor pyrite, platinoids, cobaltite, some free gold (occasionally associated with tetrahedrite), galena, chalcopyrite, and gersdorffite, as well as some Al-, Fe-, and Cu-filings, contaminants derived from the milling-grinding stage. Uraninite was partially spotted with galena, which also filled some cracks or occurred as larger inclusions in uraninite. Some uraninite grains contained gold specks which were clearly introduced during secondary events. Few grains indicated partial alteration to coffinite along cracks or enclosed specks of kerogen. Figures 4a-c provide an indication of the mineralogical composition of the raw material of this sample.

The uraninite concentrate of the Main Reef on Sub Nigel Gold Mine consisted mainly of uraninite, spotted with galena, with minor amounts of cobaltite, arsenopyrite, some free gold (less commonly as inclusions in uraninite, cobaltite, and arsenopyrite), platinoids, and rare Cu- and brass-filings. Figures 4d and e represent this sample prior to further purification of uraninite.

The uraninite concentrate of the Basal Reef (Fig. 4f and g) from Free State Geduld Gold Mine was mainly made up of uraninite grains, some of which were contaminated by minute gold specks, and minor amounts of the following minerals (listed in order of abundance): galena, arsenopyrite, cobaltite, polycrystalline gersdorffite aggregates, platinoids, few grains of free gold, and rare pyrite. This sample was not contaminated by metal filings.

The VCR sample from Kloof Gold Mine was obtained by treatment of a reef sample with HF. This sample did not contain any free uraninite. All uraninite was situated in < 1 mm-diameter kerogen nodules. Pyrite, zircon, and gold were present in this sample as trace amounts. Extensive petrographic work on Ventersdorp Contact Reef samples from Central and West Rand gold mines (Reimold and Boer, unpubl. data) resulted in only very little uraninite being observed in reef specimens. All the uraninite that was seen represented secondary mineralization in the form of fracture or interstitial fillings in, or between, other phases. Uraninite was occasionally observed together with galena, secondary pyrite or arsenopyrite, and secondary Ti-bearing minerals, all of which constituted part of the pervasively altered matrix in the reef conglomerate or in the footwall quartzite.

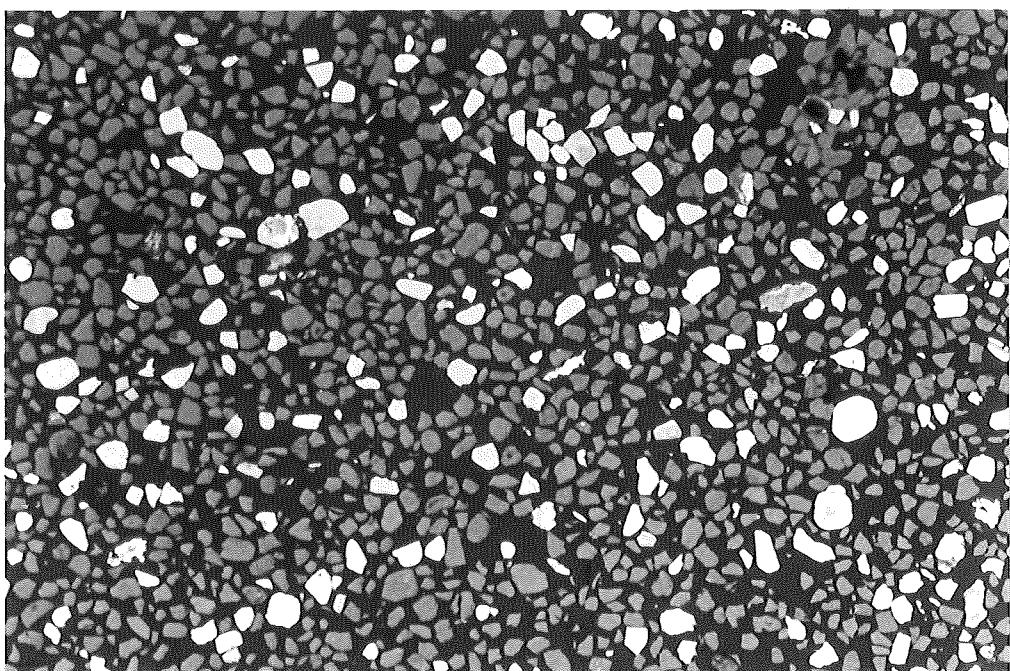
RESULTS AND DISCUSSION

The results of the isotopic analyses are shown in Table 2. Xe_s/Xe_n ratios and apparent ages for each temperature step were calculated by two independent methods using the $^{134}Xe/^{136}Xe$ and $^{131}Xe/^{136}Xe$ ratios, respectively. The $^{132}Xe/^{136}Xe$ ratio was not used, as the differences between Xe_s and Xe_n concentrations for this ratio are too small (Table 1), and the precision of ages calculated via this ratio would not be sufficient.

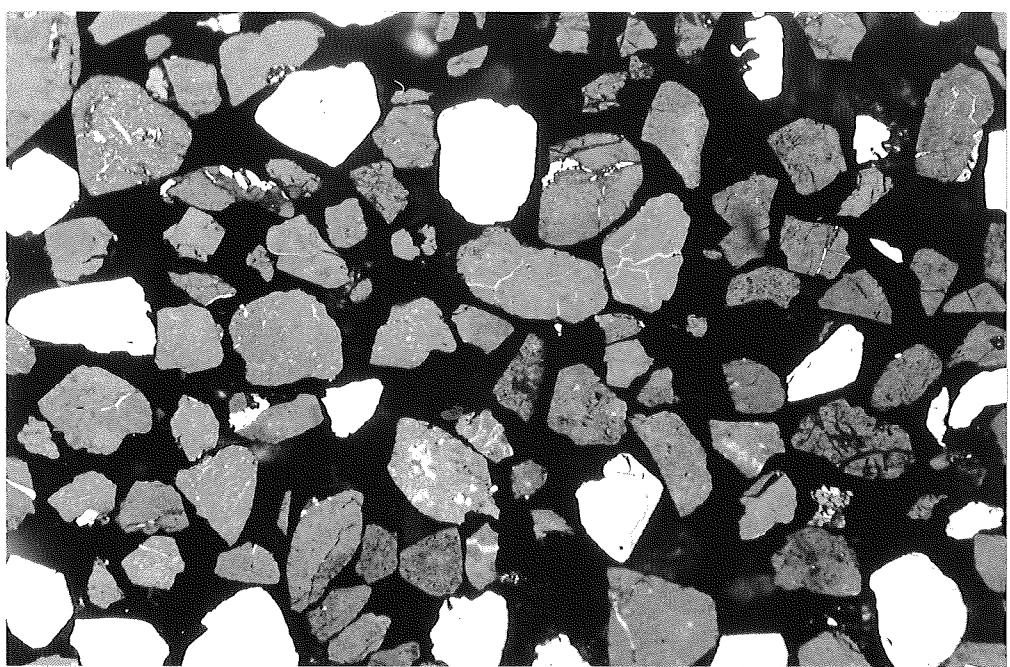
The age spectra obtained are shown in Figure 5. All these spectra have a typical "step-up" pattern, but in each case a high-temperature plateau can be defined. Integrated plateau ages

Figure 4: Example of a uraninite concentrate obtained from the Sub Nigel Gold Mine. (a) Low magnification image of highly concentrated uraninite, with contaminants of platinoids, cobaltite, arsenopyrite, gold and copper filings (all pale grey). Magnification: 5x. (b) Concentrate of uraninite (grey) and arsenopyrite (white) grains. Small specks and fracture fillings of galena (white) and black spots of coffinite are visible in uraninite grains (magnification: 20x). (c) Intragranular alteration of uraninite to coffinite (black) - magnification (50x).

(a)



(b)



(c)

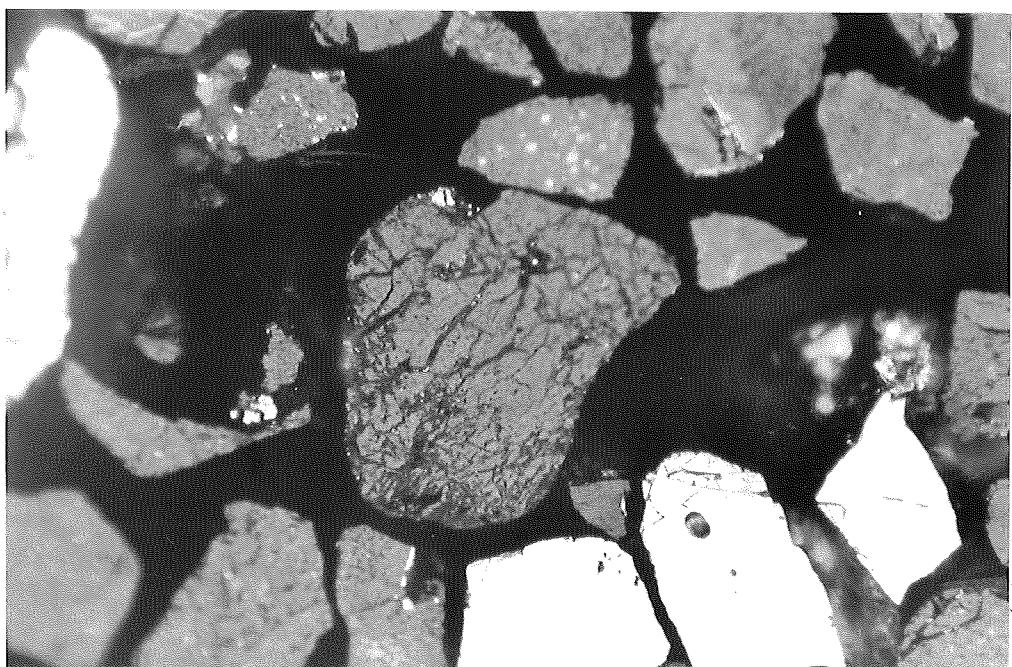


Table 1. Isotopic composition of xenon produced from spontaneous fission of $^{238}\text{U}(\text{Xe}_8)$, neutron induced fission of $^{235}\text{U}(\text{Xe}_n)$ and atmospheric xenon (Xe_{atm}), and of the tracer used for isotopic dilution (Xe_{tr}).

	136_{Xe}	134_{Xe}	132_{Xe}	131_{Xe}	130_{Xe}	129_{Xe}	128_{Xe}	126_{Xe}	124_{Xe}
Xe_s	= 1000	825	578	82	-	-	-	-	-
Xe_n	= 1000	1238	680	456	-	105*	-	-	-
Xe_{atm}	= 1000	1176	3031	2394	458	2981	216	9.97	10.7
Xe_{tr}	-	-	-	-	-	6	976.5	1004.7	= 1000

* this isotope has the 17.2 Ma precursor ^{129}I , so that after artificial irradiation the ratio $^{129}\text{Xe}/^{136}\text{Zr}$ will be 0.

Table 2. Isotopic composition of Xe released from samples during stepwise heating, concentrations of Xe components, and apparent ages

T, °C	$^{136}\text{Xe} = 1000$					Sample FSG, 2.25mg					apparent ages, Ma $t_{134/136}$ $t_{131/136}$
	^{134}Xe	^{132}Xe	^{131}Xe	^{130}Xe	^{129}Xe	$^{136}\text{Xe}_s$	$^{136}\text{Xe}_{n'}$	$^{136}\text{Xe}_a$	$^{136}\text{Xe}_{n'}$	$^{136}\text{Xe}_a$	
1	2	3	4	5	6	7	8	9	10	11	
450	1234	1182	869	97.3	625.						
610	1249	683	457	-10.4	-	274	1.8	-47	-4		
800	1219	824	582	29.9	202.3						
900	1221	670	456	8.6	14	698	4.5	73	0		
1020	1216	713	477	6.8	45.1						
1200	1216	674	448	.5	136	3486	22.7	97	40		
1500	1139	686	401	5.3	32.3						
1800	1138	659	378	-2.0	4.5	154	1.0	545	454		
	1112	677	379	5.1	32.7						
	1081	651	321	2.0	13.1						
	1081	640	312	.0	3125						
	1071	654	354	8.6	46.7						
	1069	609	315	-9.1	1118	1716	11.2	1138	999		
	1072	674	350	6.3	37.7						
	1070	641	322	-3.1	1069	1714	9.3	1122	928		
											Total
						6552	15643	100	707		

Table 2. Isotopic composition of Xe released from samples during stepwise heating, concentrations of Xe components, and apparent ages
 (continued)

T, °C	$^{136}\text{Xe} = 1000$					Sample VCR, 4.2mg	apparent ages, Ma		
	^{134}Xe		^{132}Xe	^{131}Xe	^{130}Xe		$^{136}\text{Xe}_n$	$^{136}\text{Xe}_n$	$t_{134/136}$
	1	2	3	4	5		7	8	9
500	1156	715	398	9.1	58.9				
800	1156	668	357	-.5	2009	1540	20.2	427	615
1000	1039	663	321	7.6	47.9				
1200	1036	623	286	-1.5	893	996	13.1	1519	1337
1500	1109	731	447	19.3	126.4				
1600	1106	630	361	.6	173	427	5.6	789	578
1800	1054	716	381	14.9	96.9				
	1050	638	313	-.2	1036	1427	18.7	1342	1014
	1036	654	314	5.5	35.4				
	1034	625	289	-.6	1077	1206	15.8	1546	1300
	1041	668	335	7.4	48.8				
	1039	629	302	.6	840	1027	13.5	1480	1145
	1050	673	352	8.3	53.9				
	1047	629	314	.0	740	1007	13.1	1376	1005
	Total				5227	7630	100		1120

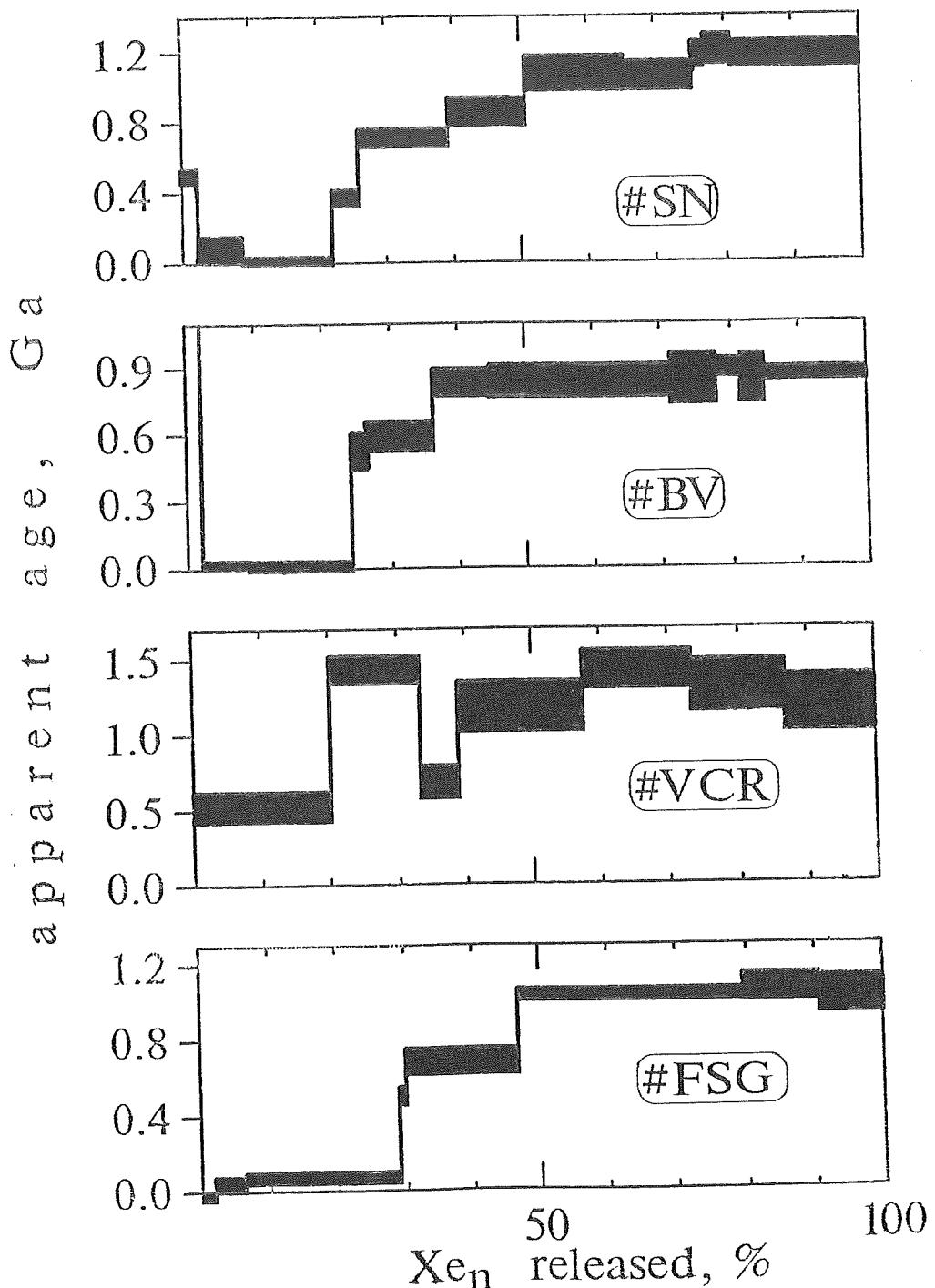


Figure 5: Xenon release patterns for the four uraninite samples from Witwatersrand gold mines in terms of percentages of $^{136}\text{Xe}_n$ released at each temperature step versus apparent ages calculated for each fraction of Xe released (compare text for detail). BV = Blyvooruitzicht; SN = Sub Nigel; FSG = Free State Geduld; VCR = Ventersdorp Contact Reef from Kloof Gold Mine.

are 0.84 ± 0.04 Ga for 64% of Xe_n for sample BV, 1.035 ± 0.05 Ga for 53% of Xe_n for sample FSG, 1.12 ± 0.05 Ga for 49% of Xe_n for sample SN, and 1.26 ± 0.02 Ga for 61% of Xe_n for sample VCR. These ages were calculated as weighted mean values of apparent ages of the individual temperature steps assigned to a plateau. The weights are proportional to the Xe_n amounts in corresponding steps. Not a single apparent age was in excess of about 1550 Ma. The spectra also do not indicate significant xenon loss at times later than the ages corresponding to the partial plateaux.

A first-order observation is that the ages obtained are much younger than the *a priori* expected values of about 2 or 2.3 Ga. This assumption was based on: (1) the earlier chronological data, especially the Ar geochronological results (Allsopp et al., 1991; Trieloff et al., 1994; and Spray et al., 1995); (2) the conclusion that major metamorphic and hydrothermal overprinting in the realm of the Witwatersrand Basin took place at about 1950–2050 Ma ago; and (3) the U-Pb data for kerogen by Robb et al. (1994). A possible reason for the results obtained could be partial losses of Xe_s from the uraninites. However, all previously developed criteria for the reliability of $\text{Xe}_s\text{-}\text{Xe}_n$ ages (Shukolyukov and Meshik, 1987, 1988) indicate that this is not the case. In reality, all Xe release patterns (Fig. 6) have demonstrated the presence of high temperature peaks with maxima at temperatures above

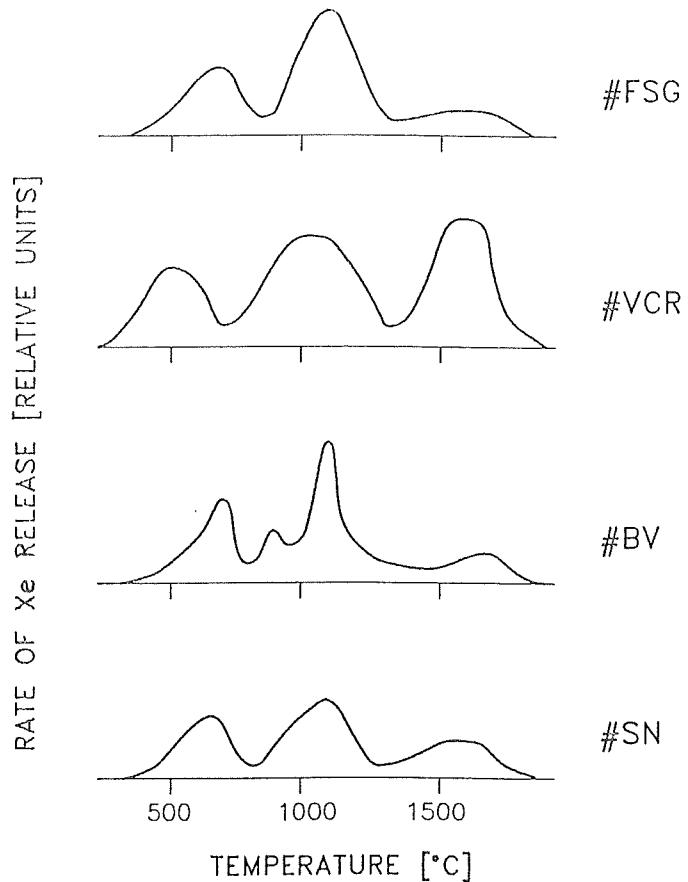


Figure 6: Diagrammatic presentation of the relative amounts of radiogenic xenon released at the different temperature steps during step-wise heating of the uraninite samples.

1500 °C. In addition, it should be noted that the release of xenon was monitored at temperatures above 450 to 500 °C, whereas all knowledge regarding post-Ventersdorp (post-2.7 Ga; Armstrong et al., 1991) metamorphism in the Witwatersrand Basin indicated conditions of low-temperature metamorphism - generally related to Bushveld or Vredefort activity just prior to 2 Ga - under < 400 °C conditions. All plateaux in these age spectra are much wider (49 to 64%) than the 30% which are normally regarded as an indicator of high retention of radiogenic Xe.

Another possible reason for the disturbed age spectra could be the presence of naturally occurring neutron-induced xenon, which is often observed in uranium veins (Shukolyukov, 1970). However, the absence of ^{129}Xe (compare Table 2), which would be a sensitive indicator of such a component, unequivocally proves that no significant amount of Xe_n was accumulated naturally in these samples since their formation.

In principle, recoil effects could also disturb $\text{Xe}_s\text{-Xe}_n$ age spectra, but such effects should increase, and not decrease, with the increase of apparent ages. In these cases, small recoil losses of reactor-induced Xe could explain the somewhat higher ages in the 500 °C fractions for the VCR and SN samples (Fig. 5). It is, however, impossible to explain the relatively young plateau ages formed during this process by the high-temperature fractions.

Another possible explanation of these results could be the effects caused by the admixture of other minerals, such as galena, pyrite, or gold. In the case of U-Pb dating, galena and other minerals may dramatically influence the isotopic age of a sample. In complete contrast, the $\text{Xe}_s\text{-Xe}_n$ dating method, when applied to U-bearing minerals, is absolutely insensitive to any admixtures that do not contain uranium because of the extremely low xenon background in these minerals. Furthermore, should any of the other phases that might have been co-irradiated with the uraninites used in this study contain significant amounts of uranium it must be assumed that the uraninite and other mineral inclusions were formed or reprecipitated at the same time. This assumption is supported by extensive petrographic evidence from the various reefs sampled.

Consequently, it is concluded that the $\text{Xe}_s\text{-Xe}_n$ ages obtained for the Witwatersrand uraninite samples correspond to some real geological event. It was recently shown by Shukolyukov and Meshik (1987, 1988) that $\text{Xe}_s\text{-Xe}_n$ ages of pitchblendes and uraninites from hydrothermal deposits do not correspond to the primary formation age of these minerals, but to the age of the final stage of hydrothermal activity that affected such ores. This is probably due to the fact that these minerals easily dissolve and can then be redeposited, more or less *in situ*, sometimes without any visible petrographic indication.

Regarding the sampling localities in the arc around the Vredefort Dome, no obvious relationships exist between age data and relative distances between sampling sites and the Vredefort Dome, or the relative distances to the Bushveld Complex to the northeast of the study area. A comparison between relative plateau ages and stratigraphic settings of the four samples also does not reveal a correlation between xenon ages and relative depth of reefs.

Recent chronological work on samples from the region of the Witwatersrand Basin and its environs has shown that the latest *regional* resetting event recorded in the isotopic

data base appears to be the 2.024 Ga Vredefort event. However, later disturbance or strong resetting to younger ages around 1.1-1.4 Ga were also repeatedly demonstrated. It could be speculated that these late disturbances were caused by magmatic (Pybus et al., 1994) or tectonic events, but it appears certain that hydrothermal activity played a role (Spray et al., 1995). Allsopp et al. (1991) and Trieloff et al. (1994) indicated that even later geological activity, perhaps as late as the 180-190 Ma (Fitch and Miller, 1984; Reimold et al., 1995) Karoo volcanism, could have affected the argon isotopic record of their samples and this possibility can not be completely discarded. However, in the light of the chronological record presented above, it appears more likely to assume localized, but regionally detectable, magmato-tectonic activity throughout the central part of the Kaapvaal Craton between 1000 and 1300 Ma ago. This period coincides with the major accretionary activity along the southern and eastern margins of the Kaapvaal Craton, as for example reviewed by Jacobs et al. (1993). It is suggested that this event induced tectonism and resulted in magmatic activity in the region of the Witwatersrand Basin and the southern parts of the Bushveld Complex (Brandt, 1994).

Petrographic observations on the Ventersdorp Contact Reef from Central and West Rand gold mines have indicated that gold and uraninite were largely remobilized. The direct relationship between Witwatersrand pseudotachylite formation, dated at about 2.17 Ga (Trieloff et al., 1994), and remobilization of gold, uraninite, and other minerals, strongly suggest that autometasomatism at that time was responsible for extensive, perhaps basin-wide, alteration and re-crystallization of these important minerals. Coincidence of these pseudotachylite ages with that of the Vredefort event led Reimold (1994) to suggest that this catastrophic impact event triggered the hydrothermal activity throughout the Witwatersrand Basin. In conclusion, the current chronological data base suggests at least two extensive hydrothermal events in the region of the Witwatersrand Basin, at about 2 and 1 - 1.2 Ga. Whether they were preceded by an earlier, basin-wide thermal or hydrothermal event at about 2.2 - 2.3 Ga ago, as suggested by some of the earlier data and some recent ^{40}Ar - ^{39}Ar dating of minerals from mafic intrusives from the Vredefort structure (Reimold et al., 1995), needs to be further pursued.

CONCLUSIONS

This U-Xe dating study of four uraninite separates from gold mines in the Witwatersrand Basin revealed that the ages of all four separates were completely reset at times between 0.85 and 1.3 Ga ago. Apparently, geological activity during this time, coinciding with the Namaqua-Natal Thrust Front activity at the southern and eastern margins of the Kaapvaal Craton, also occurred in the interior of the craton.

It is surprising that these Xe dating results only pinpoint an about 1-1.3 Ga geological event, whereas the results of earlier argon dating and, also, the various Rb-Sr and U-Pb studies on Witwatersrand rocks and minerals, did not supply any firm evidence for this late activity. There are only three possibilities that could account for these findings:

1. only the uraninite, but no other, K-bearing phase, was affected by this 1.1-1.3 Ga event. In the light of the regional resetting of the Xe systematics in samples from four sites covering an area of more than 200 km in diameter, this possibility appears

- remote;
2. the argon isotopic record could be more insensitive to the processes that led to the apparent isotopic re-equilibration of uraninite than the xenon isotopic system. A comparative experimental investigation of this possibility appears warranted. It is noteworthy that the U-Pb data from Witwatersrand kerogen (Robb et al., 1994) also indicate possible resetting at times later than 2 Ga ago, namely during the period from 1.6 to about 1.4 Ga; and
 3. it appears possible that, while this late activity affected a large region, the effects might have been comparatively localized - such as in the vicinity of intrusive bodies and in faults and fault zones. In this context it should be recalled that it was suggested previously (Reimold and Boer, 1993; Reimold, 1994) that the laterally extensive bedding-faults of the Witwatersrand Basin could, in particular, have been effective channels for fluidization. The high porosity reef horizons could have been effective fluid conduits. The study of Spray et al. (1995) has also demonstrated that relatively young 1.4 Ga ages for Vredefort pseudotachylites could be the result of *local* hydrothermal alteration, only along the vein margins. Obviously the narrow, but deformed contact zone between the pseudotachylite and the unaffected country rock acted as a channel for fluids.

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