

ECONOMIC GEOLOGY RESEARCH UNIT

University of the Witwatersrand
Johannesburg

URANIUM DISTRIBUTION IN EARLY PRECAMBRIAN
GOLD-BEARING CONGLOMERATES OF THE
KAAPVAAL CRATON, SOUTH AFRICA :
A CASE STUDY FOR THE APPLICATION OF
U-FISSION TRACK MICROMAPPING

K. THIEL, R. SAAGER and R. MUFF

• INFORMATION CIRCULAR No. 134

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ABSTRACT

In the uranium and gold-bearing pyritic quartz pebble conglomerates of the Proterozoic Witwatersrand sediments, uranium is bimodally distributed. It occurs as detrital phases, generally uraninite, and is finely disseminated in low concentrations within the matrix. The method of ^{235}U -fission track autoradiography in mica detectors was employed to investigate, specifically, the spatial distribution of the disseminated uranium in the matrix. The U-distributions in pyritic conglomerate samples from the Witwatersrand sediments were studied and compared with similar material from the Uitkyk Formation of the Archaean Pietersburg Greenstone Belt. In the samples from the Uitkyk Formation only the disseminated form of uranium was detected.

The U-micromapping revealed that uranium was repeatedly redistributed during, and/or after, regional metamorphism. A very recent U-redistribution, caused by weathering, was also observed in the Uitkyk conglomerate samples. No support was found for a synsedimentary introduction of the non-detrital uranium into the basins of deposition — an event which would require a prevailing oxidizing atmosphere during the early Precambrian.

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I. INTRODUCTION

The early Precambrian of the Kaapvaal Craton in South Africa contains several gold and uranium-bearing pyritic conglomerates, of which the gold reefs of the Witwatersrand Basin are best known. They contain the largest known gold accumulations on earth and some of the most important low-grade uranium reserves. By the end of 1975, the Witwatersrand Basin contained 49 per cent of the world's gold reserves and 17 per cent of the world's uranium reserves (van Rensburg and Pretorius, 1977). The other pyritic conglomerates of the Kaapvaal Craton have thus far been found to contain no economic mineralization.

Most scientists now agree that the Witwatersrand ores formed by sedimentary processes. Gold, uraninite, and other heavy minerals were introduced largely as detrital constituents which were, in part, redistributed during the metamorphism of the sediments. Several authors have, in the past, attempted to explain the ore formation as being due to epigenetic hydrothermal processes (Davidson, 1955; Davidson and Bowie, 1951; Graton, 1930). Furthermore, it has been suggested that most of the gold and uranium was transported in solution to the basin of deposition and was there precipitated by biochemical activity of primitive organisms (Reimer, 1975).

Of all the pyrite-bearing conglomerates on the Kaapvaal Craton, the Uitkyk conglomerates were chosen for examination because they carry minor amounts of gold. In addition, the mineralogy of the conglomerates has been studied in detail by Saager and Muff (1978a). The Uitkyk conglomerates, which are considered to be Archaean in age, form part of the Pietersburg greenstone belt, located 350 km northeast of the Witwatersrand Basin (Figure 1). The investigations by Saager and Muff (1978a) led these authors to believe that the Uitkyk Formation may have formed in a manner similar to the Witwatersrand sediments.

Recently, Simpson and Bowles (1977) re-investigated the controls of uranium mineralization in the Witwatersrand conglomerates using modern mineralogical methods. They observed a bimodal distribution of uranium, the latter occurring as allogenic grains of thorian uraninite finely disseminated in the conglomerate matrix together with phyllosilicates, porous pyrite and carbonaceous matter.

From their findings Simpson and Bowles (1977) concluded that, in the Witwatersrand Basin, uranium was deposited by transporting streams in two ways, viz. as detrital uraninite and by precipitation from solution in reducing environments. This model requires the presence of free uranyl-ions in surface waters and implies that an oxidizing atmosphere already existed during the deposition of the Witwatersrand sediments, some 2 600 m.y. ago. To explain the simultaneous presence of U^{4+} (as detrital thorium-rich uraninite) and U^{6+} (as uranyl-ion) in the oxidizing environment of the transporting waters, Simpson and Bowles (1977) argued that Th-rich uraninite is not as easily leached by meteoric waters as Th-poor uraninite. To support this explanation they quoted the occurrence of thorian uraninite as a stable detrital phase in recent unconsolidated sediments of the Indus River. However, from kinetic studies of the dissolution of uraninite, Grandstaff (1979) inferred that detrital uraninite, in recent gravels, is unstable and is not in equilibrium with its oxygen-rich environment.

The instability of detrital uraninite in an oxidizing environment is of importance since many investigators of uraniferous quartz pebble conglomerates consider a reducing earth atmosphere to be an essential pre-requisite for the fluvial transport of detrital uraninite and pyrite and thus for the formation of pyritic uraninite-bearing placers. In this context, it is interesting to note that all economically important uraninite-bearing pyritic conglomerates so far discovered (e.g. Witwatersrand, South Africa; Blind River, Canada; Jacobina, Brazil) are restricted to early Precambrian formations. Prevailing oxygen-poor atmospheric conditions during the early Precambrian are also in accordance with stable isotope studies reported by Schidlowski et al. (1975), as well as additional geological and geochemical evidence reported by Cloud (1976) and Grandstaff (1974).

A solution to this problem is of special interest to the uranium mining industry because if it can be established that the formation of uranium-bearing conglomerates does not require a reducing atmosphere, such deposits may well be expected to have formed more extensively throughout geological time than has previously been thought.

The aim of the present study was to acquire information about the mode and the time span during which the uranium deposition in the Witwatersrand and Uitkyk conglomerates took place. Emphasis was also given to the investigation of the spatial distribution of uranium in its finely disseminated form in order to establish whether this uranium was precipitated during sedimentation or whether it was re-mobilized during a later stage. An answer to this question should also permit certain conclusions to be drawn about the composition of the early Precambrian atmosphere.



Figure 1 : Geological sketch map showing portion of the South African Shield and sample locations (cover of Witwatersrand Sequence removed).

In order to reveal the micro-distribution of uranium the method of induced particle tracks was employed. This technique offers the capability of measuring element concentrations down to the ppb-range with a spatial resolution superior to common techniques including electron microprobe scanning.

II. GEOLOGY AND SAMPLE DESCRIPTION

The samples investigated originated from blanket-type horizons of the Upper Witwatersrand Sequence in the Orange Free State Goldfield (Figure 1). In addition, conglomerate samples from the Uitkyk Formation of the Archaean Pietersburg greenstone belt were also studied. Archaean granite-greenstone terranes are regarded as the provenance areas of the Witwatersrand sediments and their detrital constituents (Saager, 1973; Viljoen et al., 1970), and the Uitkyk conglomerates probably represent a primitive fore-runner of the Witwatersrand placers (Saager and Muff, 1978a).

Typical *Witwatersrand conglomerates* consist of rounded pebbles embedded in a fine-grained matrix of quartz and phyllosilicates. The polymictic conglomerates contain pebbles of vein-quartz, quartzites, chert, and banded iron-formation. Abraded grains of pyrite, chromite, zircon, uraninite, gold and other heavy minerals occur commonly in the conglomerate matrix. Generally, the conglomerates have suffered a weak regional metamorphic overprint which, according to age determinations, occurred approximately 2 000 m.y. ago, i.e. at the time of the intrusion of the Bushveld Igneous Complex (Burger et al., 1962; Wetherill, 1956).

Most of the uranium and gold in the *Witwatersrand* sediments occurs in the conglomeratic horizons (the so-called "blanket") or is associated with narrow carbon seams. According to Pretorius (1974), the *Witwatersrand* conglomerates were deposited in a braided stream environment at the interface between a fluvial and lacustrine system.

The *Uitkyk conglomerates* which form the top of the *Pietersburg Sequence* (Grobler, 1972), have a mineralogical composition similar to the *Witwatersrand* deposits (Saager and Muff, 1978a, b). They formed in a narrow, elongated, trough during a high energy pulse of sedimentation and are less mature than the *Witwatersrand* conglomerates. The *Uitkyk* conglomerates contain only traces of gold, with no uraninite yet having been reported.

In both the *Witwatersrand* and *Uitkyk* conglomerates pyrite is the most common ore mineral (Ramdohr, 1955; Saager, 1970). It occurs as three different types : (1) allogenic, compact pyrite, (2) rounded, porous or skeletal pyrite and (3) reconstituted, authigenic pyrite.

It is assumed that the skeletal pyrite variety withstood only short transport distances, or probably even formed *in situ* (Hallbauer and Utter, 1977; Ramdohr, 1955; Saager and Mihalik, 1967; Saager, 1970). The mineralogy of the uraninite and other uranium minerals has been described in detail by Liebenberg (1955) and Schidlowksi (1966, 1970).

In the *Uitkyk* material investigated, carbonaceous matter was found to occur predominantly as "fly-speck carbon" (Saager and Muff, 1978b). Hallbauer et al. (1977) consider the "fly-speck carbon" to be vegetative diaspores of primitive plants which existed during the deposition of the *Witwatersrand* sediments. However, this interpretation is in dispute since the granular carbon type might merely represent reworked detrital carbonaceous material (Pretorius, 1976; Zumberge et al., 1978). Other forms of carbonaceous matter in the *Witwatersrand* sediments, i.e. mats of columnar carbon (Hallbauer, 1975) and amorphous sooty textures (Hallbauer et al., 1977), were not observed in rocks of the *Uitkyk* Formation. The presence of "fly-speck carbon" in the *Uitkyk* Formation does, however, indicate that the occurrence of carbonaceous matter in the Precambrian of South Africa may be more wide-spread than commonly accepted. It has also recently been discovered in pyrite-bearing Moodies conglomerates of the Barberton Mountain Land (Saager and Muff, 1978b), which adds support to this view.

III. ELEMENT-MICROMAPPING BY INDUCED PARTICLE TRACKS

A. Principle of Particle Track Radiography

The method of heavy charged particle track radiography allows element distributions to be mapped in solids on a micron scale by means of dielectric track detectors (Fleischer et al., 1975; Price and Walker, 1963). The only pre-requisite is that the element concerned causes the emission of a heavy charged particle (e.g. fission product, ${}^4\text{He}$, ${}^3\text{H}$, etc.) during a specific nuclear reaction. Suitable nuclear reactions for this technique are, for example, $\text{D}(\text{n},\gamma)$, ${}^6\text{Li}(\text{n},\alpha)$, ${}^{10}\text{B}(\text{n},\alpha)$, ${}^{14}\text{N}(\text{n},\text{p})$, ${}^{15}\text{N}(\text{p},\alpha)$, ${}^{17}\text{O}(\text{n},\alpha)$, ${}^{235}\text{U}(\text{n},\text{f})$, ${}^{232}\text{Th}(\text{n},\text{f})$, and ${}^{239}\text{Pu}(\text{n},\text{f})$ (Fleischer et al., 1975), where particle emission occurs when the sample is still being neutron or proton irradiated.

In a few cases nuclear reactions can be used which lead to an α -emitter thus allowing element radiography *after* reactor or accelerator irradiation (Thiel and Damm, 1977; Woomer et al., 1976). Such examples are ${}^{208}\text{Pb}(\alpha,2\text{n}){}^{210}\text{Po} \xrightarrow{\alpha} {}^{206}\text{Pb}$, ${}^{209}\text{Bi}(\alpha,2\text{n}){}^{211}\text{At} \xrightarrow{\alpha} {}^{207}\text{Bi}$, and ${}^{209}\text{Bi}(\text{n},\gamma){}^{210}\text{Bi} \xrightarrow{\beta} {}^{210}\text{Po} \xrightarrow{\alpha} {}^{206}\text{Pb}$.

If an appropriate dielectric track detector is put in close contact with a polished section of the sample the emitted particles will produce a radiograph of etchable tracks in the detector foil, the track density being a measure of the element concentration.

At a given density N_{235} of ${}^{235}\text{U}$ -atoms in the sample a thermal neutron dose D will cause ρ_i induced ${}^{235}\text{U}$ -fission tracks per cm^2 on the track detector :

$$\rho_i = \sigma_f N_{235} D R_{235} \eta_{235}$$

where

σ_f = fission cross section of ^{235}U for thermal neutrons,

R_{235} = range of ^{235}U -fission fragments in the sample matrix,

η_{235} = etching efficiency, the fraction of fission fragments that cross the interface of the sample section and the mica detector and give etchable tracks in the mica.

Introducing the U-isotope ratio $I = N_{235}/N_{238}$ of ^{235}U and ^{238}U and substituting $N_{238} = \text{const } C(^{238}\text{U}) \sim \text{const } C(\text{U})$ one obtains :

$$\rho_i = \text{const } \sigma_f I C(\text{U}) D R_{235} \eta_{235}$$

where

N_{238} = volume density of ^{238}U -atoms,

$C(^{238}\text{U})$ = weight concentration of ^{238}U ,

$C(\text{U})$ = weight concentration of natural uranium, and

const = factor depending on the elemental composition of the sample.

By using a U-standard (e.g. U-containing glass) the composition of which is not too different from the sample composition, the matrix dependent factors are eliminated :

$$C_x(\text{U}) = C_s(\text{U}) \frac{I_s}{I_x} \frac{\rho_x}{\rho_s} \quad (1)$$

where the subscripts s and x refer to the U-standard and the sample of unknown U-content, respectively. For the majority of samples the natural U-isotope ratio can be used and U-determination is reduced to the measuring of two track densities.

B. "Lexan-Print" and Mica Record

There are two types of track detectors commonly used for element mapping :

- (1) Plastic foils, e.g. cellulose nitrate* or polycarbonate (Makrofol, Lexan)* and
- (2) Mica sheets, especially of high purity muscovite mica.

Plastic foils are more sensitive than mica and give etchable tracks with all charged particles at least as heavy as protons (cellulose nitrate), or alpha particles (polycarbonate) whereas mica will record only particles of masses exceeding that of ^{20}Ne .

For the micromapping of fissionable materials with thermal neutrons, both types of track detectors may be used.

In the case of plastic detectors, however, not only fission tracks are recorded, but also short alpha tracks originating from nuclear reactions like $^6\text{Li}(n,\alpha)$, $^{10}\text{B}(n,\alpha)$ and $^{17}\text{O}(n,\alpha)$. The neutron induced radiograph of a U-containing rock section recorded in "Lexan" polycarbonate exhibits (a) long fission tracks, reflecting the U-distribution and (b) high density short α -tracks outlining the mineral grain boundaries. This α -track pattern ("Lexan print") greatly facilitates U-localization on the rock section (Kleeman and Lovering, 1967; Lovering and Kleeman, 1970; Thiel and Herr, 1972). The precision of localizing uraniferous phases is limited, however, by the uncertainty of mineral outlines produced by divergently emitted α -particles and by the mechanical instability of the plastic foil in high-temperature ($\leq 80^\circ\text{C}$) reactor positions.

Thermal fading of the latent particle tracks at temperatures as low as 85°C may further complicate track evaluation (Fleischer et al., 1965). These disadvantages are overcome by the use of mica detectors which are mechanically more stable and preserve latent tracks up to temperatures of $\sim 450^\circ\text{C}$ (Fleischer et al., 1964). Since mica does not record the α -track pattern of the sample, the localization of uranium on a rock section may be achieved by a three step photographic mapping technique (Thiel et al., 1972; Thiel, 1973).

Footnote : * e.g. CA 80-15 cellulose nitrate film, Kodak Pathé, Paris, Makrofol E polycarbonate, Bayer Leverkusen, BRD.

- (1) A mica detector, containing a fine grid scratched on its rear surface, is fixed onto the polished rock section and a photomicrograph of the "sandwich" is taken showing the relative position of the grid on the mica and the mineral outlines on the sample section.
- (2) After neutron irradiation, the mica sheet is detached, etched, and again photographed to record the revealed fission track distribution.
- (3) By superimposing both pictures and matching the grid on the mica in picture (1) with the grid in picture (2) the relative position of U-fission tracks on the mica detector and uraniferous phases on the polished sample section can be determined to better than 1 μm .

IV. ANALYTICAL PROCEDURE

A. Sample Preparation

Selected samples of the two localities were prepared as polished sections ($\sim 50 \text{ mm } \phi$) or as polished thin sections ($20 \times 20 \text{ mm}$) and investigated by conventional optical methods and by electron microprobe.

Representative or particularly interesting parts of the polished sections were then separated for uranium distribution studies. For our investigation a total of six polished sections and two polished thin sections were used (cf. Table 1). The polished sections were embedded in cylindrical pellets of Araldit D resin* (13 mm ϕ , 4,5 mm thick) exhibiting the polished surface of the embedded samples.

TABLE 1

DESCRIPTION AND LOCALITIES OF THE SAMPLES INVESTIGATED (c.f. Figure 1)

NO.	LOCALITY	REMARKS
MR ⁺ , 11 ⁺ 12 ⁺ , 13 ⁺ D1 ⁺⁺	Uitkyk Formation, Pietersburg Schist Belt Mt. Robert Gold Mine, N. Transvaal	Quartz pebble conglomerate, weathered surface samples
WW ⁺ D2 ⁺⁺	Basal Reef, Orange Free State Goldfield F. S. Geduld Mine, S.A.	Quartz pebble conglomerate, fresh underground samples
10 ⁺	"B"-Reef, Orange Free State Goldfield F. S. Geduld Mine, S.A.	Quartz pebble conglomerate, fresh underground samples

+ polished section

++ polished thin section

B. Reactor Irradiation

Polished sections and thin sections were carefully cleaned in acetone and covered with high purity muscovite mica foils of 50 μm thickness which served as track detectors during n-irradiation. The U-content of the mica foils was determined, by several test runs, to lie in the sub-ppb-range. To make sure that the relative position of the sample section and the detector foil did not change during sample processing and n-irradiation, the mica was fixed to the edge of the Araldit pellet or thin section glass carrier using a small amount of fast hardening Technovit** resin.

The sample/mica "sandwiches" were then clamped in stacks by means of high purity aluminium brackets and neutron irradiated in the thermal column of the FRJ-2 reactor at KFA, Jülich.

Since the highest U-contents to be expected (lying in the per cent-range) had been estimated based on the electron microprobe examination, thermal neutron doses between $5,4 \times 10^{12}$

Footnote : * CIBA Geigy, Wehr/Baden, BRD.

** Kulzer & Co. GmbH, Bad Homburg, BRD.

and $1,9 \times 10^{15} \text{n/cm}^2$ were chosen in order to cover U-contents from several per cent down to a few hundred ppb by obtaining appropriate track densities. For medium and low U-contents NBS-glasses SRM 612 containing $37,88 \pm 0,08 \text{ ppm U}$ were included in each stack as U-standards. For extremely high U-concentrations, special phosphate glasses* having UO_2 -contents between 1 to 10 per cent, were taken as standards. Since the epithermal and fast neutron flux was low, compared to the thermal flux

$$(\phi_{\text{epi}}/\phi_{\text{therm}} \leq 2,8 \times 10^{-3}, \phi_{\text{fast}}/\phi_{\text{therm}} \leq 5,6 \times 10^{-4}),$$

the interference of n-induced fission of ^{232}Th could be neglected. The thermal flux within a single stack varied by a factor of 1,6 to 2,5, depending on the size of the stack (30 to 50 mm).

This flux variation, as well as n-flux depression and n-self-absorption, could be accounted for by the inserted standard glasses.

C. Track Evaluation

After n-irradiation the mica foils were carefully detached from the samples and etched for 5 minutes in 48 per cent HF at 26°C . During sample processing, a series of more than 1 200 mosaic micrographs of the sample surface, the mica/sample "sandwiches", and the fission track record of the etched mica foils, were taken employing the localizing technique already mentioned (Thiel et al., 1972; Thiel, 1973). Track counting was done either visually or automatically by means of an Optomax image analyzing system** using optical microscopy (Leitz Ortholux microscopes). High track densities, between $\sim 10^7$ and $\sim 10^9 \text{ cm}^{-2}$, were measured by means of a scanning electron microscope (Jeol JSM P-15***). Absolute U-concentrations C(U) were calculated according to relation (1) accounting for the uranium isotope ratio $\text{U}_{235}/\text{U}_{238} = 1$ of the NBS glasses which is different from the natural isotope ratio of the samples ($I_{\text{standard}} = 2,392 \times 10^{-3}$, $I_{\text{sample}} = 7,106 \times 10^{-3}$) (Cowan and Adler, 1976). Error calculations include counting statistics of the track densities, ρ_{sample} and ρ_{standard} , recorded over sample and glass, and the given uncertainty of the U-concentration of the standard glass.

V. RESULTS

The samples investigated from the Witwatersrand conglomerates were found to contain at least two different types of *uraninite*, a feature also reported by Feather (1976). These include :

- (1) An oval-shaped phase of detrital origin. This variety of *uraninite* is fractured in most cases and shows intimate association with galena formed from radiogenic lead (Plate 1A). The margins of the rounded *uraninite* grains often show alteration, suggesting partial chemical dissolution during metamorphism of the sediments. According to Feather (1976), this feature indicates that attrition during sedimentary transport was not the only abrasive mechanism to create this variety of oval-shaped *uraninite*.
- (2) *Uraninite* was also observed as anhedral inclusions in carbonaceous matter. These inclusions seem to be fragments of larger grains which were partially replaced by the carbon.

The other important uranium mineral found in the Witwatersrand samples is *brannerite*, an alteration product of detrital *uraninite*. *Brannerite* is frequently associated with phyllosilicates, porous pyrite, quartz and leucoxene. Age determinations (Pb-Pb method) carried out by means of an ion microprobe on *brannerite*, and reported by Featuer (1976), yielded an age of 1 810 m.y. which is somewhat lower than previously reported ages of $2\,040 \pm 60$ m.y. (Burger et al., 1962) and $1\,945 \pm 25$ m.y. (Köppel and Saager, 1976) for the period of lead loss and reconstitution of the uranium minerals in the Witwatersrand sediments.

In the Uitkyk samples no *uraninite* grains were found and *brannerite* was found as discrete particles (Plate 1B) in only one polished section (Saager and Muff, 1978a). Detailed mineralogical and geochemical observations on *uraninite*, *brannerite*, zircon, monazite and other uranium-bearing minerals in the Witwatersrand sediments have been published by a number of authors (Feather, 1976; Feather and Koen, 1975; Grandstaff, 1979; Hiemstra, 1968; Liebenberg, 1955; Ramdohr, 1955; Schidlowksi, 1970).

Employing the method of uranium fission track mapping, the present work was focused on the low U-concentrations associated with carbonaceous matter, leucoxene, matrix minerals and pyrite.

Footnote : * Schott and Gen., Mainz, BRD.

** Micro Measurements Ltd., Cambridge, UK.

*** Jeol Ltd., Tokyo, Japan.

A. Carbonaceous Matter

Since only the "fly-speck" type of carbon was found in the Uitkyk Formation (Plate 1C and Plate 2B) the other two types of carbonaceous matter which occur in the Witwatersrand sediment (viz. the columnar and the amorphous carbon) were excluded in the present investigation. The fission track distributions of discrete grains of "fly-speck carbon" indicate overall U-contents of approximately 4 per cent (cf. Table 2). Remarkably high U-values in this type of carbonaceous matter were also reported by Feather and Koen (1975) from the Witwatersrand rocks and by Saager and Muff (1978b) from the Uitkyk conglomerates. Carbonaceous matter may readily fix uranium from solution by reduction and/or absorption (Doi et al., 1975; Hostetler and Garrels, 1962; Langmuir, 1978; Szalay, 1964; Van der Weijden and Langmuir, 1976) and in sandstone-type uranium deposits it constitutes an important factor in the enrichment of uranium (Adler, 1970; Harshman, 1970). In addition to these mechanisms of uranium concentration it was suggested by Hallbauer et al. (1977), that the primitive organisms of the Witwatersrand, now represented by the carbonaceous matter, were able to assimilate uranium and other heavy metals by biogenic processes (Hallbauer, 1975).

TABLE 2

U-CONTENT OF THE MOST ABUNDANT MINERALS IN THE SAMPLES INVESTIGATED

MINERAL	UITKYK-FORMATION	BASAL/"B"-REEF	ANALYSES
QUARTZ	1 ppb - 6,3 ppm	1 ppb - 525 ppm	7
POROUS PYRITE ⁺)	10 ² ppm range	5 ppm - 320 ppm	5
COMPACT PYRITE	1 ppb - 2 ppm	1 ppb - 1 ppm	4
LEUCOXENE ⁺)	108 ppm - 557 ppm	256 ppm - 2,78 %	23
CLAY-MINERALS	6,5 ppm - 0,90 %	0,55 % - 0,69 %	8
ZIRCON ⁺⁺)	10 ² ppm range	0,11 % - 0,84 %	5
CHROMITE ⁺⁺)	10 ¹ ppm range	8,8 ppm - 250 ppm	5
CARBONACEOUS MATTER	1,22 % - 1,90 %	1,56 % - 4,9 %	11 68

+) Without U-rich inclusions

++) U inhomogeneous within individual grains

The "fly-speck carbon" studied displays an internal texture of oval-shaped structures (Plate 2A). Microprobe analyses revealed that the boundary material between the structures contained up to 40 per cent uranium, 4 per cent thorium, some titanium, and traces of lead (Saager and Muff, 1978a, b). These uranium distribution patterns and contents are supported by the fission track record (Plate 2B). However, some grains of "fly-speck carbon" exhibit much lower U-contents, the uranium being distributed more homogeneously in these cases.

In some instances the U-contents associated with fine matrix material were found to decrease with increasing distance from the "fly-speck carbon" (cf. Plate 3,I). Such uranium gradients point to the presence of reducing micro-environments around carbonaceous material. Within these reducing halos uranium was either precipitated from percolating hydrous solutions or it could not be oxidized to the mobile hexavalant state because it was already fixed.

B. Leucoxene

An unusual feature, observed in the samples, is the invariable association of uranium with leucoxene. The latter generally forms porous, earthy aggregates (Plate 3,II), or fine disseminations (Plate 4,I). Some of the leucoxene was mobilized during, or after, the consolidation of the sediments. This is indicated by numerous narrow fractures in larger quartz grains now filled with leucoxene. Although it is difficult to distinguish leucoxene from brannerite by optical and X-ray methods, Feather and Snegg (1978) were able to show by electron microprobe analysis that the "uraniferous leucoxene" in the Witwatersrand ores, first described by Liebenberg (1955), is an admixture of leucoxene, anatase and tiny irregularly

distributed brannerite needles. These admixtures probably formed in the basins of deposition by hydrothermal-metamorphic alteration of detrital uraninite and Ti-oxides such as rutile, ilmenite and titaniferous magnetite. Another explanation for the close association of uranium with leucoxene was given by Yermolayev (1971) who pointed out that adsorption processes during diagenesis and blastesis may cause U-enrichment of leucoxene. Furthermore, this is supported by the work of Van der Weijden and Langmuir (1976) and Kochenov et al. (1965) who showed experimentally that uranium can be adsorbed by iron hydroxides and titanium-oxyhydroxides.

The uranium distribution in a typical "uraniferous leucoxene" aggregate is shown in Plate 4, I. The fission track pattern reveals local uranium clusters due to the above-mentioned admixture of brannerite needles and leucoxene. Areas of a more uniform U-concentration, within the leucoxene in this case, suggest U-enrichment by adsorption from hydrous solution rather than by admixture of uraniferous minerals.

C. Matrix Minerals

Finely disseminated uranium was found to be associated with the phyllosilicates of the conglomerate matrix (Plate 4, I), i.e. with chlorite, sericite, muscovite, pyrophyllite, and occasionally biotite, kaolinite, and an unidentified 7 Å mineral (Fuller, 1958; Liebenberg, 1955; Simpson and Bowles, 1977). In the fine-grained quartz, forming the bulk of the matrix minerals, no uranium was detected.

Several samples of the Uitkyk Formation contain allogenic pyrite grains surrounded by low pressure areas which abound with metamorphically grown chlorite needles (Saager and Muff, 1978a). As revealed by the fission track radiographs, these chlorite seams in the pressure shadows exhibit considerable amounts of uranium at their peripheries (Plate 4, II).

The association of uranium with the phyllosilicates montmorillonite and kaolinite is well known (Rogers and Adams, 1974), and has been investigated experimentally by Doi et al. (1975), and by Goldsztaub and Wey (1955), who found that these minerals readily adsorb uranium. Doi et al. (1975) pointed out that chlorite, in particular, is a highly effective uranium concentrator, but did not provide quantitative data for its U-concentrating ability. Based on these experimental results it is suggested by the present authors, that the uranium concentrations associated with the phyllosilicates of the conglomerate matrix and the metamorphic chlorite minerals in low pressure shadows are due to the adsorption of uranium from circulating hydrous solutions.

The samples from the Uitkyk Formation, all being collected from the weathered zone, generally exhibit thin coatings of Fe-hydroxides around the larger conglomerate components caused by descending alteration. The Witwatersrand samples were not subjected to surface weathering, and thus do not display these coatings of secondary alteration material. Fe-hydroxides are known to be potential uranium adsorbers (Doi et al., 1975; Langmuir, 1978), and therefore, in the Uitkyk Formation, some of the uranium enrichments found around larger conglomerate constituents are probably due to adsorption by rims of supergenic Fe-hydroxides rather than to adsorption by phyllosilicate minerals.

D. Pyrite

Uranium concentrations of less than 1 ppm have been found in most massive pyrites, either of detrital or authigenic origin. In cases where isolated fission track stars over pore-free pyrite domains can be correlated with small visible ($\geq 1 \mu\text{m}$) inclusions, the track densities indicate U-concentrations of several thousand ppm (Plate 5, I) pointing to the presence of uraniferous phases.

In concretionary, porous pyrite the pores are filled with silicate minerals such as chlorite, sericite, quartz, pyrophyllite and chloritoid. These pores commonly contain diffusely disseminated uranium of a concentration reaching a few hundred ppm. Occasionally, over some pores, high-density fission track stars are recorded, which indicate the presence of fine-grained U-minerals such as brannerite or probably coffinite.

The very thin U-coatings found around both detrital and metamorphically grown Witwatersrand pyrites are distinctly discontinuous (Plate 5, II). They differ from the above-mentioned diffusely adsorbed uranium associated with the secondary iron-hydroxide coating on detrital grains of weathered samples from the Uitkyk conglomerates.

Track analyses of the mica detectors, using scanning electron microscopy, clearly reveal that the discontinuous uraniferous rims on Witwatersrand pyrite are made up of discrete grains (Plate 6, I) occasionally exhibiting crystal shaped outlines. Because of their high U-contents in the per cent range these grains are assumed to represent U-minerals. They are, however, too small ($> 1 \mu\text{m}$) to be identified.

Simpson and Bowles (1977) also observed uranium-rich rims on alloogenetic Witwatersrand pyrites which they ascribed to an association of uranium with secondary clay minerals forming a marginal overgrowth to the pyrites. They considered this relationship to be an important control in the deposition of uranium in the Witwatersrand sediments.

Typical U-concentrations measured in a sample from the Witwatersrand deposit (Plate 5, II) are : 0,5 ppm U for the large quartz pebbles, $1,48 \pm 0,09$ per cent U for the uraniferous skin around pyrites, and $5,0 \pm 0,3$ ppm U for areas of pore free pyrite. The mean ratio for the corresponding uranium contents obtained from different parts of five concretionary pyrites is $1 : 2,5 \times 10^4 : 5$ (quartz : skin : pyrite).

The ranges of U-contents of different mineral phases associated with the finely disseminated uranium are listed in Table 2.

VI. DISCUSSION

This work, in association with the fission track studies of Simpson and Bowles (1977), supports a bimodal occurrence of uranium in the Witwatersrand sediments, viz., as uraninite grains and in a finely dispersed form together with matrix minerals. Most authors agree that the uraninite is a detrital component of the Witwatersrand conglomerates. The formation of the finely dispersed uranium in the matrix is, however, still controversial. The period of formation of the uranium dispersion is particularly problematical, although Simpson and Bowles (1977) suggested that the U-disseminations are precipitates which formed during sedimentation, i.e. approximately 2 600 m.y. ago.

A much younger post-sedimentary redistribution of the uranium, during a metamorphic event which also resulted in the formation of authigenic sulphide minerals, is envisaged by several authors (Ramdohr, 1955; Schidlowksi, 1966, 1970). Lead isotope measurements carried out on sulphide minerals of the Witwatersrand sediments indicate that these redistributions coincide with a period of regional metamorphism caused by the intrusion of the Bushveld Igneous Complex some 2 000 m.y. ago, resulting in a total lead loss and a reconstitution of the uraninite (Burger et al., 1962; Köppel and Saager, 1976; Wetherill, 1956).

The U-distribution patterns of Witwatersrand and Uitkyk conglomerates, derived from the present study, yield no unequivocal evidence for a synsedimentary introduction of uranium in solution and its subsequent precipitation in the depository basin as proposed by Simpson and Bowles (1977).

The present study also does not allow the period of U-distribution to be confined to a single event. Rather it indicates a redistribution of uranium during, or after, the period of metamorphism. This is supported by the following findings :

- The presence of uraniferous phases forming rims on metamorphically grown pyrites.
- Uranium enrichments on the boundaries of chlorite seams which developed during the metamorphic overprint in low pressure areas around alloogenetic pyrites.
- The presence of an admixture of fine-grained leucoxene and brannerite, the latter being a metamorphic mineral.

A redistribution of uranium, particularly during the metamorphic event, is furthermore manifested by the abundant occurrence of so-called uraninite "ghosts" in the polished sections from Witwatersrand ores. The uraninite "ghosts" consist of fine-grained brannerite seams, the latter still displaying the outlines of the original shape of the water-worn abraded uraninite parent grains (Plate 6, II and III).

According to Schidlowksi (1966) these uraninite "ghosts" are due to the formation of refractory brannerite seams on the periphery of the uraninites and the subsequent leaching of the central portion of the grains. The grain interiors were then replaced by silicates and authigenic ore minerals (Plate 6, III).

Simpson and Bowles (1977) maintained a synsedimentary introduction of finely disseminated uranium into the Witwatersrand Basin but, contradictory to this statement, these authors provided indications suggesting a late U-redistribution in the Witwatersrand basin and report in particular that :

- (1) the uranium was incorporated into clay-mineral overgrowths which formed during diagenesis and metamorphism on alloogenetic pyrite, and that

- (2) regional metamorphism "tended to reduce the thickness of the uraniferous rim, which is best developed where weathered pebbles are relatively unaltered".

These features underline the fact that uranium was redistributed during regional metamorphism which would have obliterated primary syngenetic relationships. Such alterations of primary U-distributions cannot be regarded as isolated cases since this study, and previous investigations, indicate that metamorphic U-remobilization was common in the Witwatersrand Basin.

This study has also yielded clear evidence for a later post-metamorphic redistribution of uranium. This is shown by the occurrence of hairline cracks (< 1 mm to < 1 cm in length and ~ 5 to ~ 50 μm in width) filled with U-bearing phases and intersecting several silicate and ore mineral grains (cf. Plate 6, IV). The crack fillings exhibit inhomogeneous U-concentrations and some are even completely devoid of uranium which possibly reflects their different ages (cf. Plate 3, I). Based only on U-distribution patterns the time period of the post-metamorphic U-mobilization (possibly a multi-stage process) can, however, not be exactly defined.

U-contamination of the cavities and fractures in the sample sections by uraniferous dust abraded during the polishing procedure can also be positively ruled out because of the presence of completely uranium-free fractures and pores appearing in close vicinity to uranium-rich pores and cracks (Plate 5, I). Further evidence for a non-artificial origin of the U-content of fractures is given by the micron scale variation of the U-concentration within a single hairline crack, the U-content often changing abruptly at grain boundaries intersected by the crack (cf. Plate 6, IV). This feature can possibly be attributed to different chemical influences of the intersected mineral phases on the precipitation of uranium.

Indications of a very recent type of U-mobilization were observed in the samples from the Uitkyk Formation which were collected from surface exposures. Although the overall mineralogical composition of the Uitkyk conglomerates is almost identical to that of the Witwatersrand reefs, these samples did not contain uraninite. The higher U-concentrations, recorded generally, occur in association with rounded grains of carbonaceous matter. The absence of uraninite in the Uitkyk samples may be due to recent dissolution of detrital uraninite by highly oxidizing meteoric waters. Only in the reducing micro-environment of the "fly-speck carbon", was uranium either stable against its removal by oxidizing waters, or alternatively the carbonaceous matter precipitated uranium from the solutions that had leached uraninite in the weathered portion of the Uitkyk sediments. In the unweathered samples from the Witwatersrand Basin such pronounced leaching of uraninite was not observed. However, it is known that U-bearing Witwatersrand conglomerates are free of uranium minerals due to weathering at their surface outcrops (J. Grootenboer, personal communication, 1978).

The presence of U-containing carbonaceous matter in deeply weathered Archaean conglomerates may thus serve as a tool to detect potential prospecting targets (Saager et al., 1979).

VII. SUMMARY AND CONCLUSION

The fission track patterns of all the samples investigated provide strong evidence for a bimodal U-distribution in the Witwatersrand sediments. The uranium occurs as detrital U-minerals, particularly uraninite, or as fine dispersions in close association with carbonaceous matter, pyrite, leucoxene and fine-grained phyllosilicates in the conglomerate matrix. In the Uitkyk samples, which originate from the weathering zone, only the finely dispersed uranium is present, whereas detrital uranium minerals are presumed to have been recently leached out by descending meteoric waters.

From the fission track patterns obtained, and from mineralogical and textural considerations, it is concluded that the finely disseminated uranium suffered repeated remobilization within the consolidated conglomerates. The following stages of U-transport can be distinguished :

- (1) Syn-metamorphic redistribution of uranium

This is indicated by mineral phases grown during the period of regional metamorphism and having incorporated uranium during their growth.

- (2) Post-metamorphic uranium mobilization

This is evidenced by hairline cracks which intersect metamorphic minerals and are filled with uraniferous material.

- (3) Recent uranium transport by oxidizing meteoric waters

In the surface samples of the Uitkyk conglomerates this recent U-transport is manifested by the occurrence of supergenic iron-hydroxides carrying adsorbed uranium and by the absence of uraninite in the weathering zone. Similar patterns may also possibly be found in Witwatersrand samples collected from surface outcrops.

This multi-stage redistribution of uranium obliterates all syngenetic textures and therefore no unequivocal evidence for a synsedimentary introduction of uranium into the basins of deposition as mobile U^{6+} can be deduced. Thus the observed bimodal U-distribution in the Witwatersrand conglomerates is not necessarily due to synsedimentary uranium transport and does not permit the inference to be made that oxidizing atmospheric conditions prevailed during the time of sedimentation, i.e. the early Precambrian. In particular, one may not conclude that economic uraniferous quartz pebble conglomerates should occur in sediments younger than 2 000 m.y.

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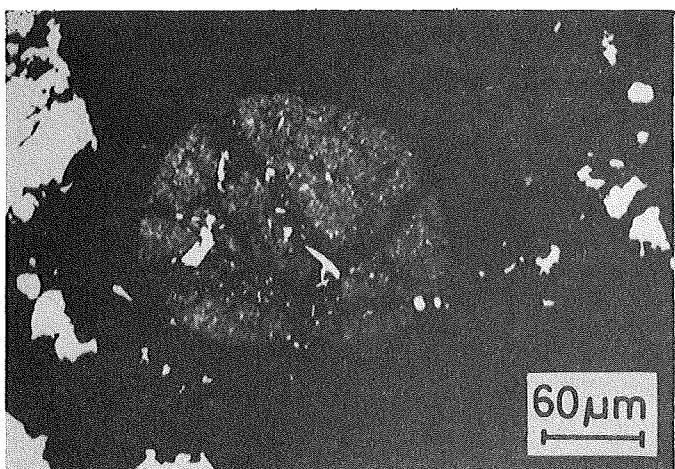
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PLATE 1

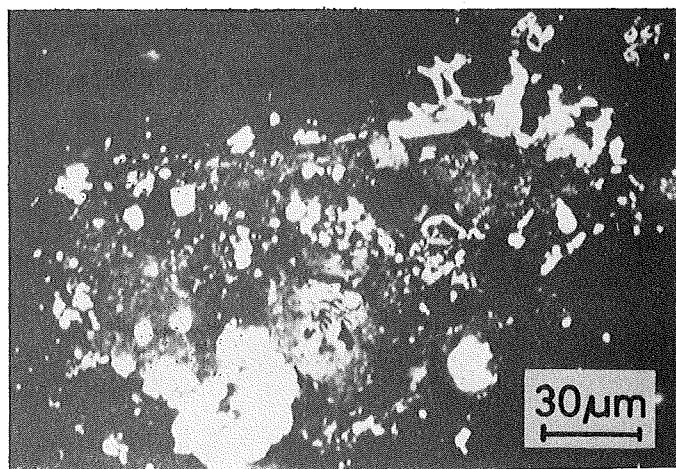
- A. Rounded grains of uraninite showing dustings and crack fillings of radiogenic galena (white). Larger ore minerals outside the uraninite grain are authigenic pyrite (white). Basal Reef, Free State Geduld Mine, Witwatersrand; oil immersion.
- B. Porous aggregate of brannerite (grey) with authigenic pyrite (white). Uitkyk Formation, Mt. Robert Gold Mine, Pietersburg Schist Belt.
- C. Scanning electron micrograph of a granular type carbon ("fly-speck carbon") from the Uitkyk Formation, Mt. Robert Gold Mine, Pietersburg Schist Belt.

PLATE 1

A



B



C

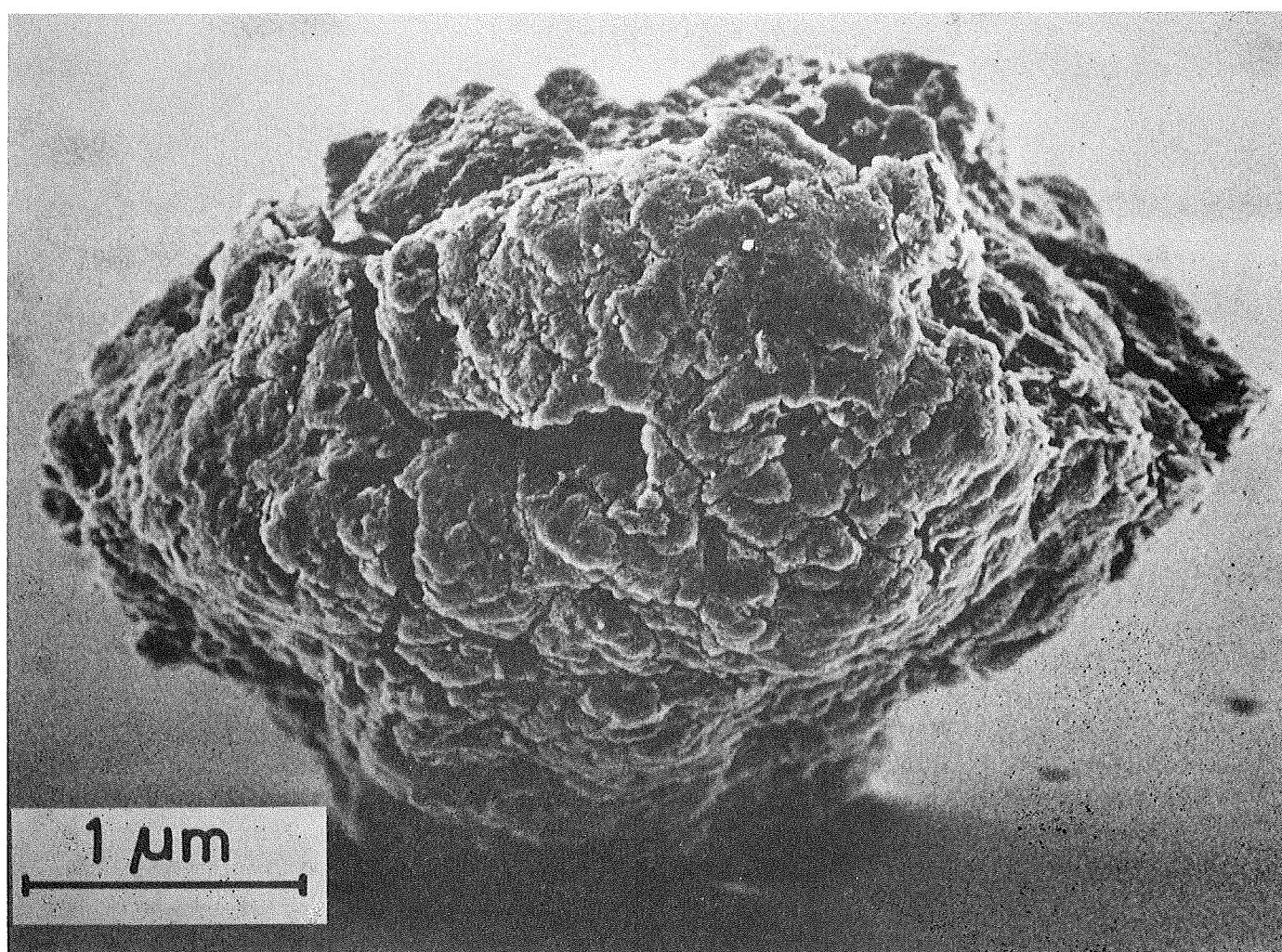
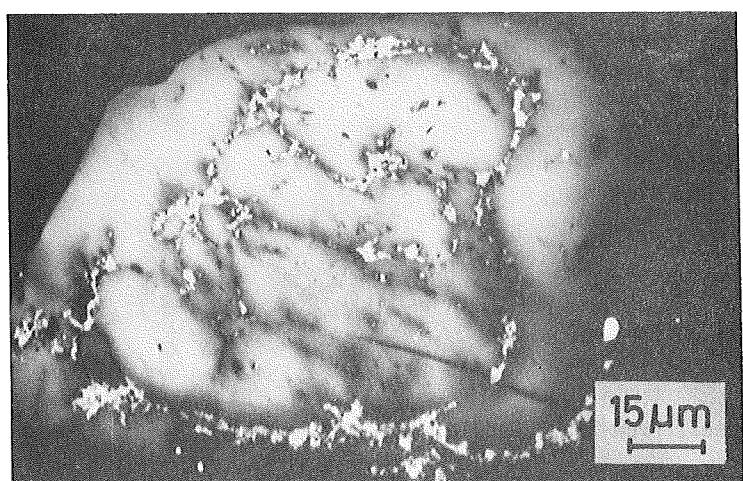


PLATE 2

- A. Polished section of "fly-speck carbon" with oval-shaped internal structures. Tarnished phases within the carbon consist mainly of a mixture of galena, brannerite, leucoxene and uraninite. A small grain of authigenic pyrite (white) occurs on the right hand edge of the carbon grain. The same carbon grain is also shown in Plate 2 B (b).
"B"-Reef, Free State Geduld Mine, Witwatersrand; oil immersion.
- B. Polished sections of "fly-speck carbon" from the Uitkyk Formation (a) and the "B"-Reef, Witwatersrand (b-c). White grains are pyrite. The corresponding U-distribution recorded on the mica detectors is given by the scanning electron micrographs (d-f). The characteristic oval-shaped internal structure of the "fly-speck carbon" in all grains is reflected by the U-distribution pattern. The white rims especially apparent in (f) are caused by preferential electron emission due to the etch relief of the mica detector. These rims therefore do not indicate higher U-contents. Photographs (b) and (e) are also described in Plates 2A and 3A.

PLATE 2

A



B

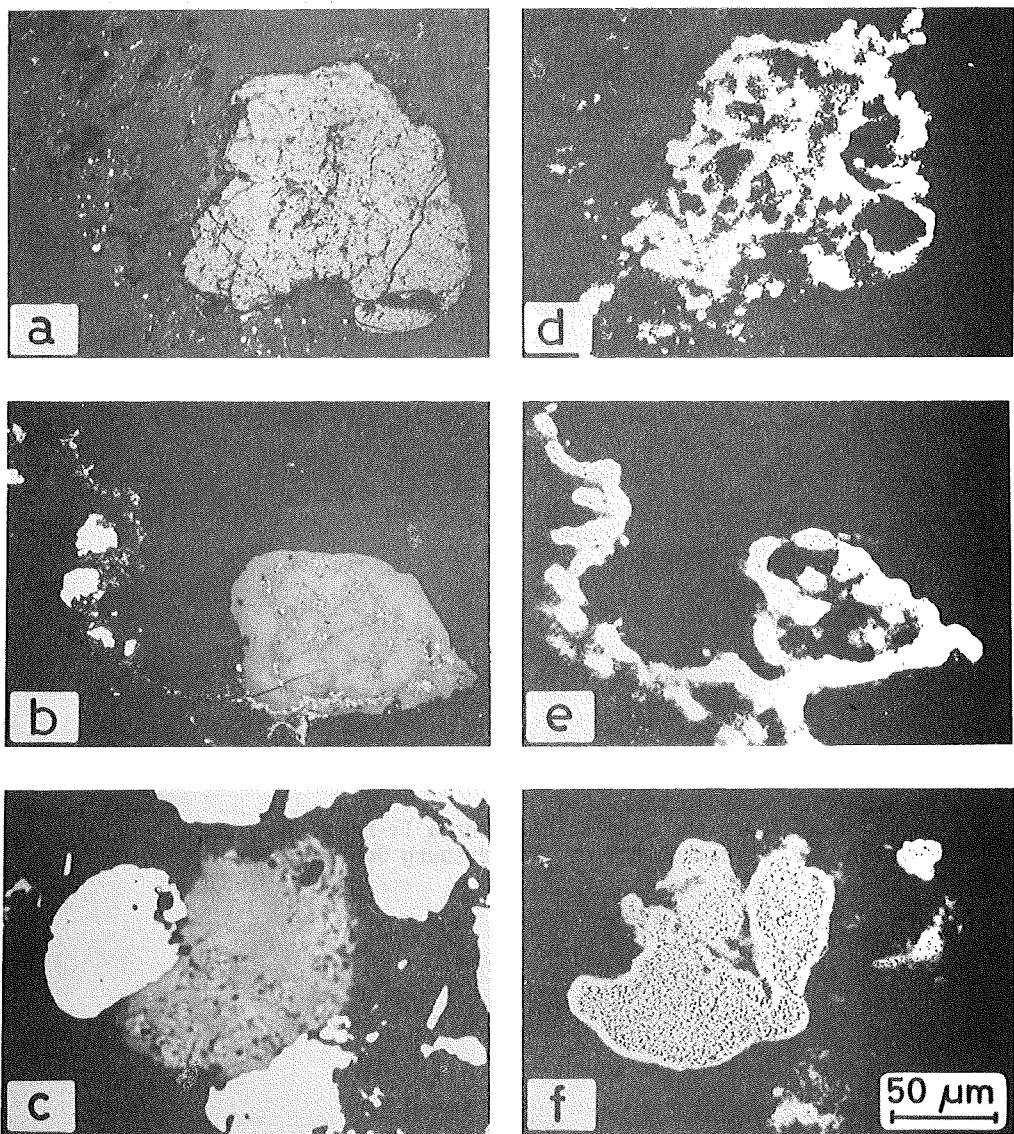


PLATE 3

I. Sample 10, "B"-Reef, Free State Geduld Mine, Witwatersrand.

- A. Polished section showing carbonaceous matter (CM), leucoxene (LX) and portion of large porous pyrite (white, right hand side) with inclusions of chloritoid laths (grey). Note cross-cutting hair-line crack.
- B. Mica detector showing corresponding U-distribution. The highest U-concentrations are associated with carbonaceous matter and with matrix minerals occurring between large quartz grains. The outline of the porous pyrite is marked by fission track clusters indicating somewhat lower U-contents. In the leucoxene aggregate the uranium is diffusely distributed except for the four fission track stars which may represent brannerite admixtures.
The inset diagram gives U-contents measured along the leucoxene-bearing matrix. Note the overall decrease of U-concentrations with increasing radial distance from the central carbon grain.

II. Broken aggregates of leucoxene alteration after a Ti-Fe-oxide mineral.
Uitkyk Formation, Mt. Robert Gold Mine, Pietersburg Schist Belt;
oil immersion.

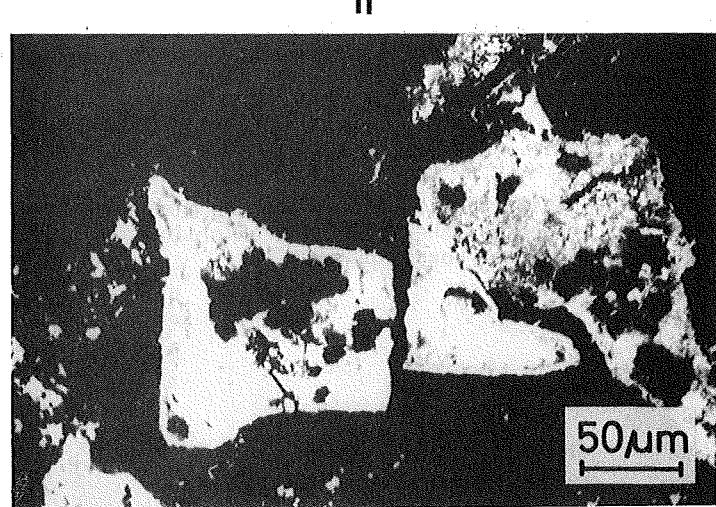
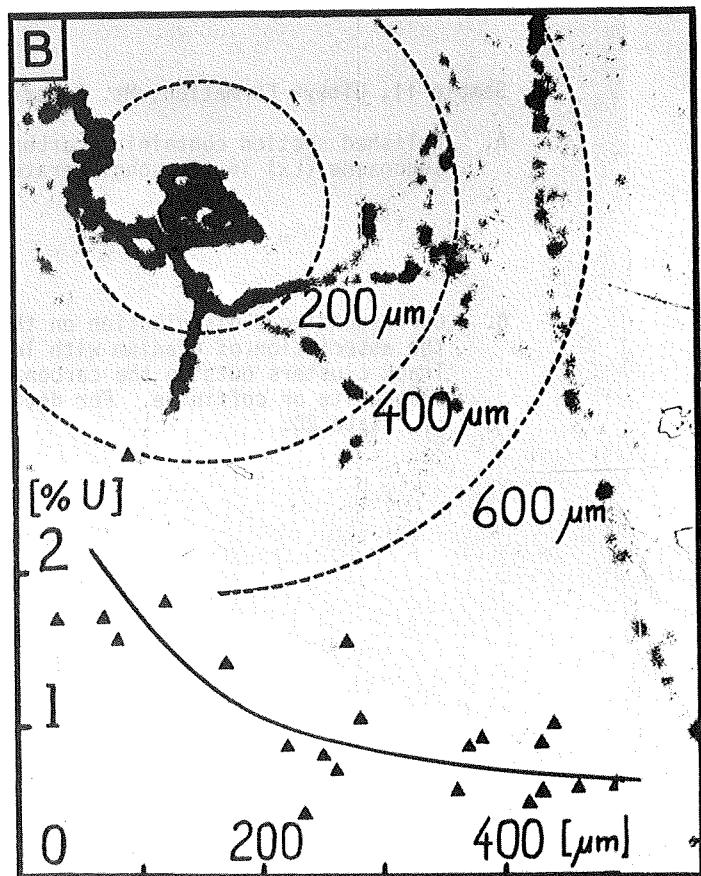
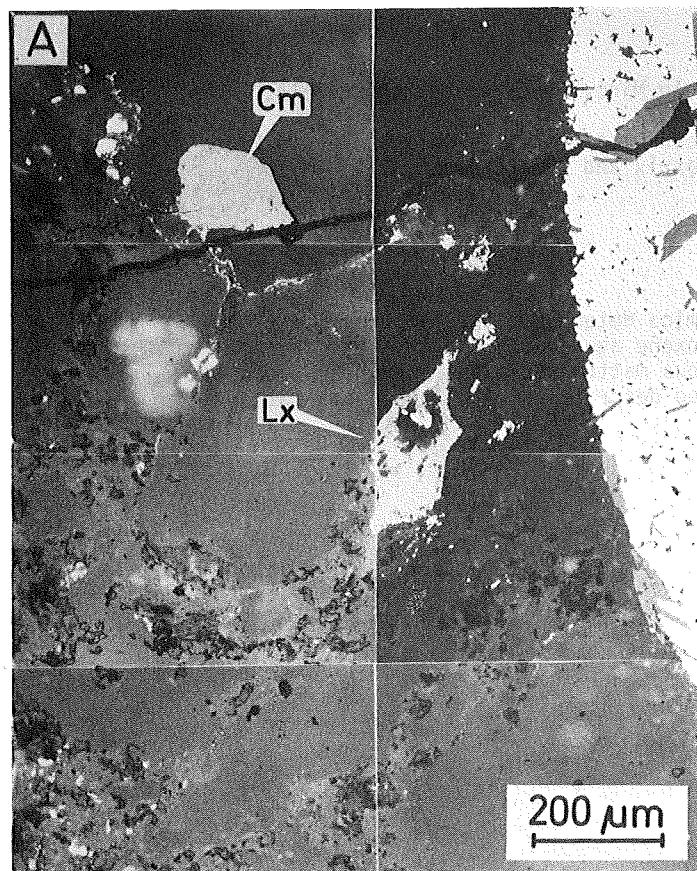


PLATE 4

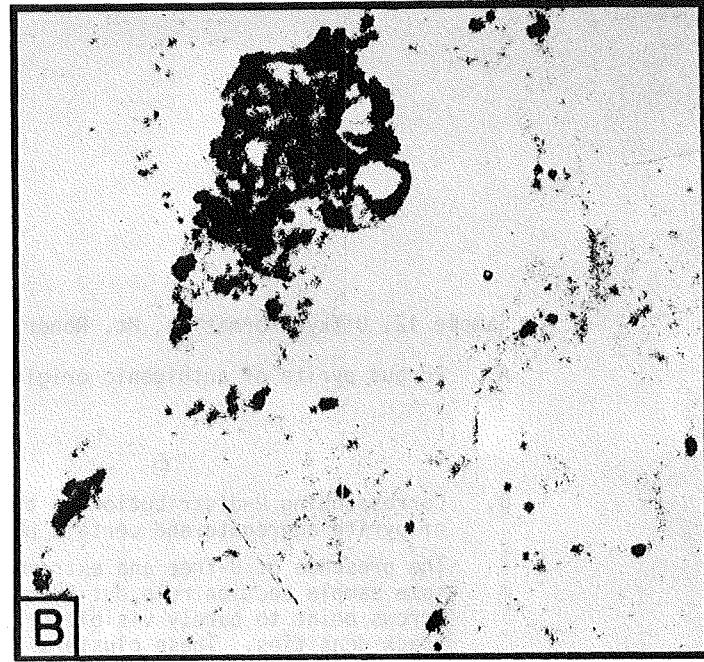
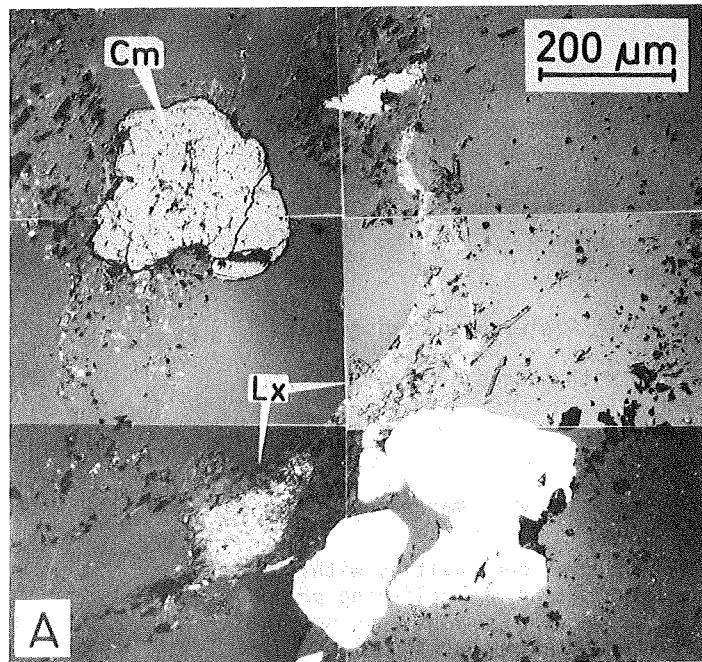
I. Sample 11, Uitkyk Formation, Mt. Robert Gold Mine, Pietersburg Schist Belt.

- A. Polished section containing carbonaceous matter (Cm, same grain as in Plate 2B), leucoxene (Lx) in the conglomerate matrix and pyrite (white).
- B. Corresponding U-distribution on the mica detector. This figure illustrates the association of uranium with leucoxene aggregates and phyllosilicates. Track clusters outside the carbonaceous matter represent U-phases, probably brannerite or coffinite. For detailed description of carbonaceous matter cf. Plate 2B.

II. Sample 10, "B"-Reef, Free State Geduld Mine, Witwatersrand.

- A. Porous pyrite grains (white) associated with metamorphically grown chloritoid laths (grey). The pyrite grain near the centre shows a low-pressure seam (arrows) filled with chlorite.
- B. The corresponding U-distribution pattern on the mica detector indicates U-concentrations at the peripheries of the chlorite seam. High U-concentrations also occur marginal to the porous pyrites.

PLATE 4



II

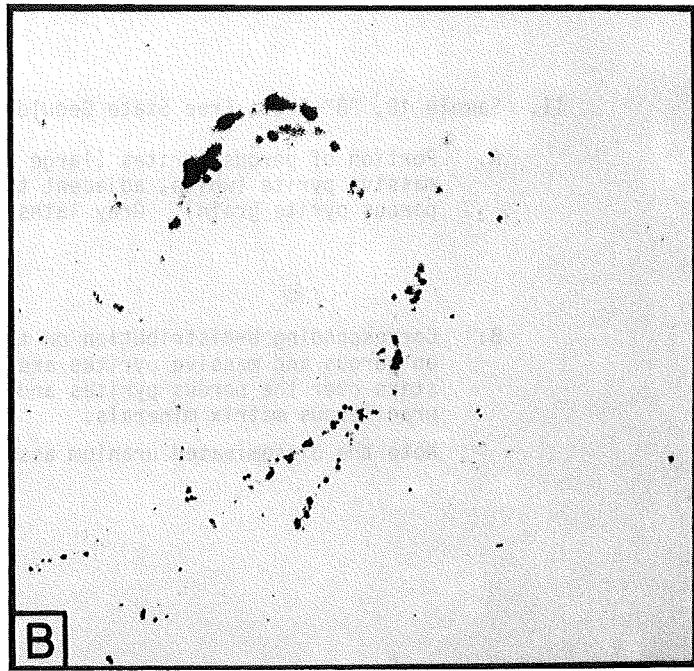
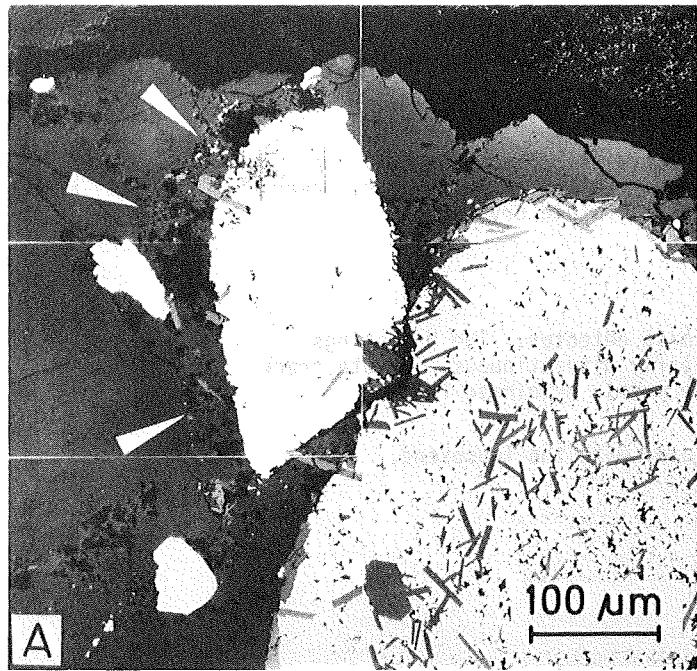


PLATE 5

I. Sample 12, Uitkyk Formation, Mt. Robert Gold Mine, Pietersburg Schist Belt.

A. Porous pyrite of authigenic origin (white) with gangue minerals (grey).

B. Corresponding U-distribution on the mica detector with marked outlines of pyrite aggregate and certain pores.

The presence of U-free and extremely U-rich pores and cavities within the same sample section rule out U-contamination during the polishing procedure. Arrows point to barely visible or submicroscopic inclusions causing high track densities. These clusters indicate U-contents reaching almost 1 per cent.

II. Sample 10, "B"-Reef, Free State Geduld Mine, Witwatersrand.

A. Portion of porous pyrites (large white grains), leucoxene (Lx) and massive pyrite (white, adjacent to leucoxene and to left rim of lower porous pyrite grain). Grey laths are chloritoid crystals.

B. Corresponding U-distribution on the mica detector. The U-coatings on porous and massive pyrites are clearly discontinuous. Minute track stars over the porous pyrites and the matrix originate from uraniferous matrix minerals.

Note the disseminated uranium associated with the leucoxene.

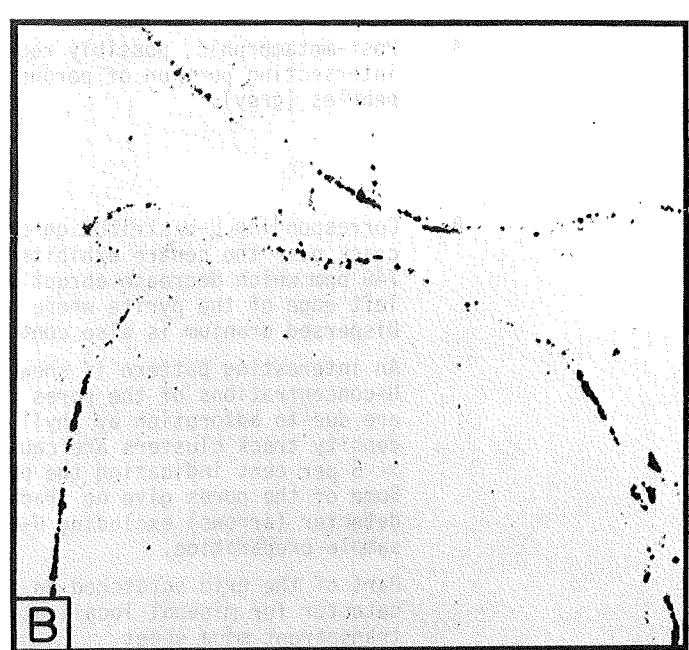
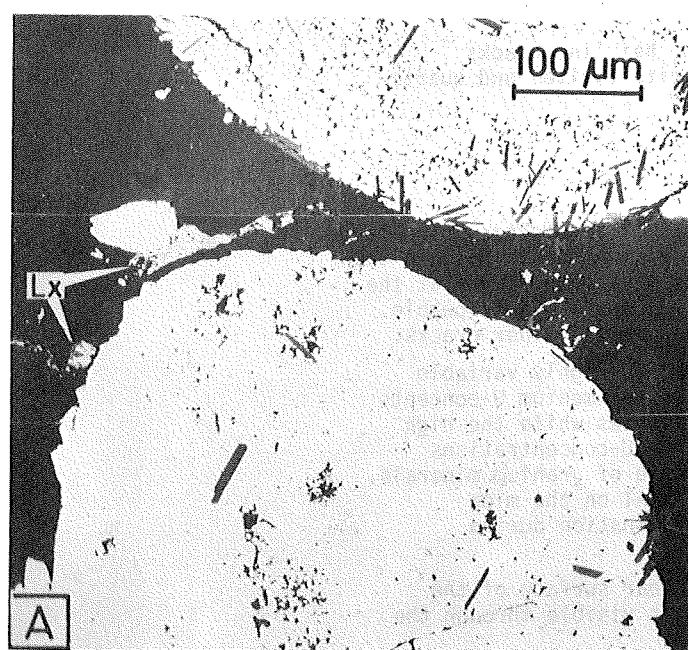
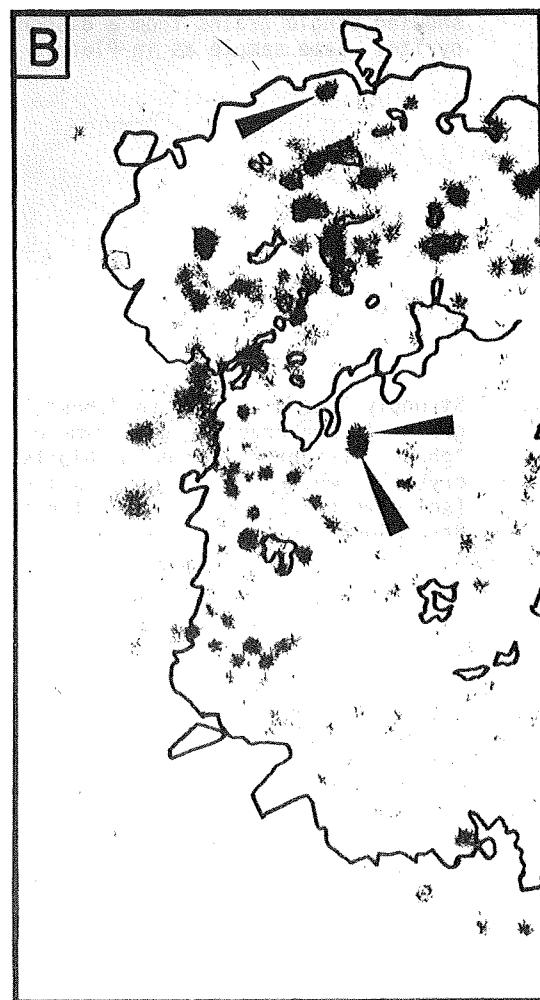


PLATE 6

- I. Scanning electron micrograph of part of the mica fission track detector showing single grains from a discontinuous uraniferous rim around porous pyrite. Same sample as in Plate 5 II.
- II. Strongly leached uraninite "ghosts". The shape of the original uraninite grains is preserved by thin rims of brannerite (grey). The large uraninite "ghost" (right hand side) exhibits a conspicuous aureole of authigenic gold crystals (white). Rounded cataclastic chromite (dark grey) and pyrite (solid white) are other constituents.
Basal Reef, Free State Geduld Mine, Witwatersrand; oil immersion.
- III. Brannerite forming a uraninite "ghost".
The leached central part of the "ghost" is now filled with authigenic gold (central grain in photograph) possessing a round inclusion of gersdorffite. Other constituents are pyrite (white) and chromite (grey).
- IV. Sample WW. Basal Reef, Free State Geduld Mine, Witwatersrand.
 - A. Post-metamorphic, possibly recent, hairline cracks intersecting portion of porous pyrite (white) and quartz pebbles (grey).
 - B. Corresponding U-distribution on the mica detector. The large crack near the centre exhibits uranium contents of more than 740 ppm which decrease abruptly to less than 90 ppm at the left edge of the pyrite where it adjoins a quartz pebble. Dispersed uranium is also contained in the minor cracks.
An interesting pattern is shown by the highly variable U-concentrations of the pores : low and medium U-contents are due to adsorption by phyllosilicates while the high density track clusters are caused by U-concentrations \gg 5 per cent indicating the presence of uranium minerals. Some of the pores give no track signal on the mica detector (arrows) excluding U-contamination during sample preparation.
Part of the grid scratched on the rear surface of the detector for mineral localization is visible through the transparent mica sheet.

