THE MINERALOGY, PETROLOGY AND GEOCHEMISTRY OF THE URANIFEROUS CONGLOMERATE OF THE DOMINION REEFS MINE, KLERKSDORP AREA

by

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PART I

MINERALOGY AND PETROLOGY

(Part II, Geochemistry, in same cover)

ABSTRACT

In section A a short description is given of the minerals encountered in 129 polished sections of conglomerate from the Dominion Reefs Mine. These include:

(a) The radio-active minerals: Thorian uraninite, thorite, monazite, betafite, euxenite, leucoxene and zircon;
(b) The oxide minerals: Cassiterite, chromite, columbite and ilmenite;
(c) The sulphides: Pyrite, marcasite, galena, chalcopyrite, covellite, sphalerite, cobaltite, linnaeite and arsenopyrite;
(d) Gold and some silicate minerals.

In section B a study is made of the literature on various methods used for roundness, form and size studies, and a method is proposed which is suitable for a quantitative study of these properties in polished sections.

The results obtained when this method was applied to samples from the Dominion Reefs Mine indicate that the roundness of uraninite is in accordance with that of monazite and zircon. Uraninite in this conglomerate can therefore best be regarded as a detrital mineral concentrated together with other heavy minerals by sedimentary processes.

However, some detrital minerals, such as cassiterite and garnet, have such small mean roundness values that they could not have shared the sedimentary history of the other detrital minerals. They were probably introduced into the sediments during another cycle of sedimentation.

The same conclusions are borne out by the size distribution data. It is considered that a period of hydrothermal activity and an increase of temperature caused sulphidization and metamorphism of some minerals, the limited redistribution of gold, and corrosion and alteration of many detrital mineral grains.

CONTENTS

INTRODUCTION

The object~~s~~ of this investigation ~~were~~ was to identify the various mineral species present and to determine their association and relationship, to obtain and interpret data on roundness, shape and size, and to elucidate the geochemical relationships of the elements present in the ore. It was expected that the data obtained from these studies would throw new light on the origin of uraninite and gold in the conglomerate of the Dominion Reefs Mine.

The mineralogical examination of the conglomerate of the Dominion Reefs Mine was commenced in March, 1951, and a large portion of the work was completed in 1952. Form and size studies were made during 1961 and 1962. After having joined the Government Metallurgical Laboratory in 1962 the author was given the opportunity to complete this project.

Geochemical investigations were also made of the ore, but the results obtained will form the subject of Section II of this thesis.

The Dominion Reefs Mine is situated about 20 miles to the West of Klerksdorp in the Transvaal. This mine started its life as a gold-producer in 1888, but was subsequently closed down. In 1935 it entered its second period of activity but again proved uneconomical. When uranium production commenced in 1955 the mine entered a flourishing period, but in 1963 it had to close down as a result of overproduction of uranium. The last uranium oxide was produced on the 8th of January, 1963. The value of gold obtained at this mine is R13,000,000 and 3,000,000 pounds of uranium oxide were yielded since 1955.

PREVIOUS WORK

In his detailed work, Liebenberg (1955 p.101) described the occurrence of uraninite and other radio-active minerals in the Witwatersrand System and the Dominion Reef, and listed some of the other minerals encountered. This list was further extended in his later publications (1956, 1958, p.380, 1960, p.834).

In an unpublished MSc. thesis, Malan (1959, p.27) gave a description of the detrital minerals and sulphides which he found in the Dominion Reef.

/Uraniferous...

Uraniferous thorite was recognized by Ortlepp (1962, p.197) in the Dominion Reefs Mine. However, Taylor et al.(1963, p.329) in an article on the radio-active minerals in the Dominion Reef, stated that uranothorite was recognized by them before 1958.

Samples of monazite and conglomerate from the Dominion Reefs Mine were used by Nicolaysen et al. (1962, p.15) for age determinations and they also included a short mineragraphic description of the material utilized.

In his demonstration of hydraulic equivalence of uraninite, chromite and zircon in Witwatersrand banket, Koen also included a sample from the Dominion Reefs Mine, and he provided several photomicrographs of certain features in this sample (1961, p.35, figs. 2, 5, 6, 15).

SAMPLES USED IN THIS STUDY

The small-pebble conglomerate which is the ore of uranium and gold at the Dominion Reefs Mine occurs near the base of the Dominion Reef System. It is separated from the underlying gneissic granite by less than 60 feet of quartzite.

Two payable reefs, separated by barren conglomerate only, or also by a layer of quartzite, are distinguished, and they are known as the Upper Reef and the Lower Reef. The Upper Reef often contains a thin layer at its top, which is only one inch thick and which is virtually a consolidated black sand rich in uranium, gold and heavy minerals (See also Liebenberg, 1955, p.115).

The twenty samples used in this study consisted of large pieces of conglomerate weighing between 20 and 30 pounds. They were selected from various portions of the mine and included samples very rich in uranium and gold, as well as samples having intermediate values, and samples poor in gold and uranium. In Table I the sample numbers, the locality in the mine where they were obtained, as well as their approximate gold and uranium contents are given. The gold values were supplied as approximate values by the management of the mine. The U_3O_8 equivalent values were determined radiometrically.

/Table...

TABLE I

SAMPLES FROM THE DOMINION REEFS MINE EXAMINED

Sample No.	Section of Mine	Gold Value dwt.	U_3O_8 eq.%	Number of polished sections studied
1	Bramley 4S1, Upper Reef	130.0	5.6	12
2	Bramley 4S1, Upper Reef	100.0	3.1	5
3	Bramley 4S Dr. Lower Reef	Tr.	.021	-
4	Bramley 4N Dr. Lower Reef	Tr.	.021	-
5	Bramley 5S1 Lower Reef	1.5	.006	-
6	Bramley 7S3 Upper Reef	3.4	.194	4
7	Mackenzie 4S3 Lower Reef	Tr.	.009	-
8	Mackenzie 4S4 Lower Reef	2.2	.011	-
9	Mackenzie 6S10 Lower Reef	54.0	.148	1
10	Mackenzie 9N2 Upper Reef	Tr.	.036	1
11	Mackenzie 10S1 Lower Reef	23.4	.509	10
12	Mackenzie 11S6 Lower Reef	Tr.	.012	8
13	Mackenzie 12S5 Hanging wall	30.8	.15	5
14	Mackenzie 12S5 Hanging wall	79.5	.20	10
15	Mackenzie 15 Cross-cut West Upper Reef	1.5	.354	2
16	Mackenzie 15 Cross-cut West Upper Reef	1.5	.371	5
17	Mackenzie 15S Dr.West of Fault Lower Reef	99.0	.200	5
18	Mackenzie 15N Dr. West of Fault Lower Reef	Tr.	.018	-
20	Mackenzie 16S1 Lower Reef	Tr.	.012	-
	Concentrates of uraninite, cassiterite and sulphides			12

A further 49 polished sections of samples collected from 1945 onwards were kindly made available to the author by Dr. W.R.Liebenberg of the Government Metallurgical Laboratory. One of these, marked 451, was also used for measurements of shape and size. A concentrate of uraninite, prepared by tabling, flotation and magnetic separation, was also used.

The samples low in gold and uranium were not used for mineragraphic work and for the gathering of statistical data on size, shape and roundness, as enough grains for measurement could not be found in them. However, this very wide range of values was necessary for the

/elucidation...

elucidation of certain geochemical relationships, which will be described in Part II of this thesis.

Sample 1(a) represents a layer about 1 inch thick, capping a dark-coloured, small-pebble conglomerate (Sample 1(b)), rich in heavy minerals. From the point of view of sedimentary petrology these two samples represent layers which formed at different times and under different conditions.

The seven samples, 1(a), 1(b), 2, 6, 10, 15 and 16, which originate from the Upper Reef, are dark-coloured pebble conglomerate consisting of small pebbles of vein quartz set in a fine-grained dark matrix of quartz, sericite, chlorite, pyrite and other minerals which are usually not distinguishable with the naked eye. Sample 1(a) contains only a very few isolated small pebbles.

The samples from the Lower Reef consist of pebbles of white vein-quartz set in a matrix of yellowish-green sericite. The pebbles in any one sample are well sorted, but there is a large variation in size from sample to sample. Under the microscope it is seen that strained vein-quartz is being replaced by interstitial sericite and forms the bulk of the rocks.

The two samples from the hanging wall (Nos. 13 and 14) are similar to the samples from the Lower Reef.

In most samples evidence of shearing can be seen. Some pebbles were broken into thin slabs by parallel, closely spaced shear planes.

In some samples the pebbles are closely packed, but in others they do not even touch each other, and are separated by matrix.

TECHNIQUE

The largest portion of every sample was crushed, and a portion of the crushed material was used for the separation and identification of the minerals present in it. The heavy fractions were obtained by panning and separation by means of bromoform. These heavy fractions were further subdivided into three or more magnetic fractions.

Of the 20 samples 12 were considered to be sufficiently rich in ore minerals for study in polished sections, and 68 polished sections were prepared (Table I). An additional 12 polished sections were made from heavy mineral concentrates. A further 49 polished sections, prepared

/from...

from samples collected from 1945 onwards from the Dominion Reef were kindly made available to the author by Dr. Liebenberg. One thin section from every sample was also studied. A few polished thin sections proved useful to examine phenomena not readily visible in either polished or thin sections alone.

The usual microscopic methods were employed in studying the samples. X-rays were employed extensively for the identification of the mineral species present. Qualitative spectrographic data obtained by using the technique described by Niebuhr et al. (1959, p.93), furnished confirmative evidence in many cases.

For the study of the distribution of radioactivity use was made of autoradiographs. Ordinary x-ray film, as well as nuclear track emulsions, was employed.

The technique finally adopted for roundness, shape and size measurements will be described in section B.

SECTION A : MINERALOGY

Examination of polished sections reveals uraninite, leucoxene and sulphides as the more common constituents apart from gangue minerals. In some samples monazite is common, and smaller amounts of uranothorite, columbite, zircon, cassiterite, ilmenite, rutile, chromite, garnet, pyroxene, epidote and sphene are also present.

As sample 1(a) was used extensively for the study of the relationships of the minerals, and also for studies of roundness, size and shape, its composition was determined by means of a Swift point counter, and found to be as follows:

/Mineral...

Mineral	Percentage by volume
Quartz	30.6
Chlorite	26.2
Monazite	6.0
Uraninite	11.3
Leucoxene	12.3
Chalcopyrite	0.2
Zircon	1.1
Sericite	0.2
Cobaltite	0.2
Pyrite	10.0
Thorite	0.2
Galena	0.1
Chromite	0.5
Arsenopyrite	0.2
Cassiterite	0.3
Pyrrhotite	0.2
Covellite	0.2

A total of 3678 counts were made on three different polished sections. As the values obtained on the three different sections varied considerably, it was evident that a coarse-grained rock such as conglomerate, in which the minerals are not homogeneously distributed, cannot be adequately sampled for quantitative analysis in a few polished sections. The figures reported above must therefore be considered as a rough guide only.

I. RADIO-ACTIVE MINERALS

Most of the samples from the Dominion Reefs Mine are appreciably radio-active, and those from the Upper Reef are considerably more so than the ones from the Lower Reef.

Uraninite is the most important radio-active mineral. Appreciable amounts of monazite, thorite, columbite-tantalite, betafite, euxenite, radio-active leucoxene and radio-active zircon are also present. Only a trace of coffinite was observed.

/(a) Uraninite...

(a) Uranininite

Uraninite reaches a high concentration in the Upper Reef, and in one sample, 1(a),it forms nearly ten per cent by weight of the rock. In some samples from the Lower Reef it is present in smaller amounts, and in others it could not even be detected.

The uraninite grains are usually concentrated in fairly thin layers parallel to the bedding and locally even form clusters. Other heavy minerals, notably monazite and leucoxene, are preferably accumulated in the same layers as the uraninite (Plate I).

In the Vaal and Kimberley Reefs Koen (1961, p.28) recognized clusters of uraninite which he interpreted to be of colloidal origin. In a careful search in which 108 polished sections were systematically traversed, only one such cluster was found (Plate XI, Fig. 3). The more abundant type of cluster found in the Dominion Reefs Mine, however, is different in form. Some of the grains in this contain crystal faces, others have zircon as an original inclusion, and heavy minerals other than uraninite are also to be observed.

In samples 15 and 16 the original outlines of the uraninite grains are completely destroyed. However, in samples 1(a), 1(b) and 2 they have been excellently preserved. In some samples a fair proportion of grains displays crystal faces, as well as rounded out-lines. In Plate II six grains of uraninite are considered to represent six stages of rounding of the uraninite crystals. The uraninite grain in Fig. 6 has a Wadell roundness value of 0.76, and this value is the highest obtained for the uraninite grains in the samples studied.

In many cases cracks present in the uraninite are orientated roughly perpendicular to the outline of the grain (Plate II, Fig.3). They could be explained as sineresis cracks dating from a colloform stage in the history of the grain. The straight boundaries could then be interpreted as contact surfaces formed when spheres of unconsolidated uranium hydroxide gel came into contact with one another during growth. (Examples of colloform uraninite grains were illustrated by Ramdohr (1960, Plates 600, 605), and Maucher and Rehwald (1961, Plates 0111, 0130, 0131 and 0133)

However, an alternative explanation, favoured by the author, is that these cracks are due to an increase in volume caused by radioactive decay. In addition to such small radial cracks, tension was

/also...

also relieved by:
(i) Larger cracks traversing the grains at various angles, and in most cases filled with galena, and (ii) tiny cracks of limited extent.

The parallelism of straight portions of the grain boundaries (Figs. 1, 2 and 3, Plate II) is a further proof that these straight edges represent crystal faces and not "contact surfaces".

A few grains, evidently originally oval in shape, seem to possess overgrowths which modified their outlines. Examples of such grains are shown in Plate I (Plate I, X0.4, Y1.0 and X0.1, Y2.9), and at greater magnification in Plate XI (Figs. 1 and 2).

The impression of original oval cores ~~are~~ is suggested by roughly-concentric cracks. When viewed at high magnification these cracks are seen to be of a branching nature, and an alternative explanation, viz. that the cracks were formed as a result of differential expansion of the grain due to radio-activity then seems to be more plausible.

In Plate XI (Fig. 3) two spherical nodules are shown, the outlines of which are clearly defined. On the lower portions of these nodules an outgrowth is present, the lower boundary of which also has a nodular outline. However, in the 129 polished sections studied only one instance was observed of such spherical nodules.

The uraninite of the Dominion Reef has a fairly high content of thorium and rare earths, and in this respect it resembles the crystalline varieties of uraninite found in pegmatites (Robinson and Sabina, 1955, p.632; Berman, 1957, p. 719). If nodular uraninite was deposited in the Dominion Reef, either as overgrowths on pre-existing grains, or as nodular clusters of the type pictured in Plate XI, Fig.3, then it can be expected to be lower in thorium and rare earths, the nodular type being usual for deposits formed at lower temperatures. Therefore the study of these grains by means of an electron microprobe can be expected to yield more positive evidence as to the presence of nodular overgrowths. It is hoped to conduct such a study in the future.

A further method which may shed light on the presence of nodular overgrowths is a statistical study of roundness. Such a study was conducted in Section B, and the indications are that nodular outgrowths are actually present on some grains (p.49).

/As...

As claimed by Ramdohr (1960, p.972) samples from the Adelaide River, North-Australia, and also from Schneeberg and Shinkolobwe, contain aggregates of good crystals which grade outwards into kidney-shaped forms. Nodular overgrowths in the Dominion Reef uraninite could therefore have been original properties of detrital grains. Alternatively, they could have formed in the Dominion Reef itself after lithification.

The presence of outgrowths on original oval uraninite grains was previously recognised by Koen (1961, Plate I, Figs. 3, 4, 5 and 6). However, Koen interprets the nodular portions of the grains pictured by him as the original portions, and the parts showing crystal faces as outgrowths. This possibility is discussed later on p.59.

In heavy concentrates there are usually a few grains of a second type of uraninite characterized by numerous inclusions of galena which are larger than the inclusions to be found in the more abundant type of uraninite. This second type also has a slightly higher reflectivity than the normal type (Plate III, Fig.5).

In Table II the a_o values of unheated and heated uraninite grains obtained from concentrates, are compared. Two values, determined on two different grains are given for the ~~2nd~~ second type of uraninite. The values reported for the usual type of uraninite were obtained from about 0.5 gm. of finely ground powder on a General Electric X-ray machine and goniometer using Cu-radiation and a Ni-filter, and those for the 2nd type of uraninite with small Philips powder X-ray cameras, (diameter, 57.27 mm) using Cu-radiation, and Ni-filters on a General Electric type X-ray machine.

TABLE II

COMPARISON OF UNIT CELLS OF UNHEATED AND HEATED URANINITE GRAINS FROM DOMINION REEFS MINE

Type of uraninite	a_o (Unheated)	a_o (Heated)
Usual type	5.46 Å	5.41(1) Å 5.423(2) Å
Second type with higher reflectivity	5.45 Å	
	5.41 Å	

(1) Heated in air for 10 min. to 1000°C.

(2) Heated in nitrogen to 870°C

/Unheated...

Unheated uraninite gives a poor pattern in which back reflections are broad and faint. A sharper and more intense diffraction pattern is obtained after heating. Two different techniques were used in the heating experiments. In the first one the uraninite was heated to 1000°C in a muffle furnace, and kept at this temperature for ten minutes before cooling. After heating in air the diffraction pattern of β-UO_2 was obtained and some extra lines which are to be ascribed to ThO_2.

In the second heat treatment a thermogravimetric analysis of the Dominion Reef uraninite was carried out in an inert nitrogen atmosphere (Fig.1) This analysis was kindly done by Mr. Birkill of the Government Metallurgical Laboratory, Johannesburg. Nitrogen containing less than 10 parts per million oxygen was bubbled through pyrogallol and dried with magnesium perchlorate before entering the heating chamber.

With rising temperature a weight decrease, which amounted to 5.35% of the sample at 870°C, took place. When cooling started, the decrease in weight terminated, and a slight increase, shown by the slight upward slope of the weight curve, became apparent. It is very probable that the reaction which caused this increase started already when the temperature was still rising, but that its effect was masked by the sharp weight loss.

The weight loss may be ascribed to loss of water as well as the volatilization of some of the sulphur and lead in the galena. The slight increase may be due to a small amount of oxidation which occurred in spite of the low oxygen content of the atmosphere used. After heating, the platinum crucible was seen to be corroded by the lead present in the uraninite.

The a_0-values shown in Table II correspond to an uranium:thorium ratio of approximately 8.1 according to the graph of Robinson and Sabina (1955, p.630). The actual ratio found from a chemical analysis (Table III) is 9.34.

/Table III..

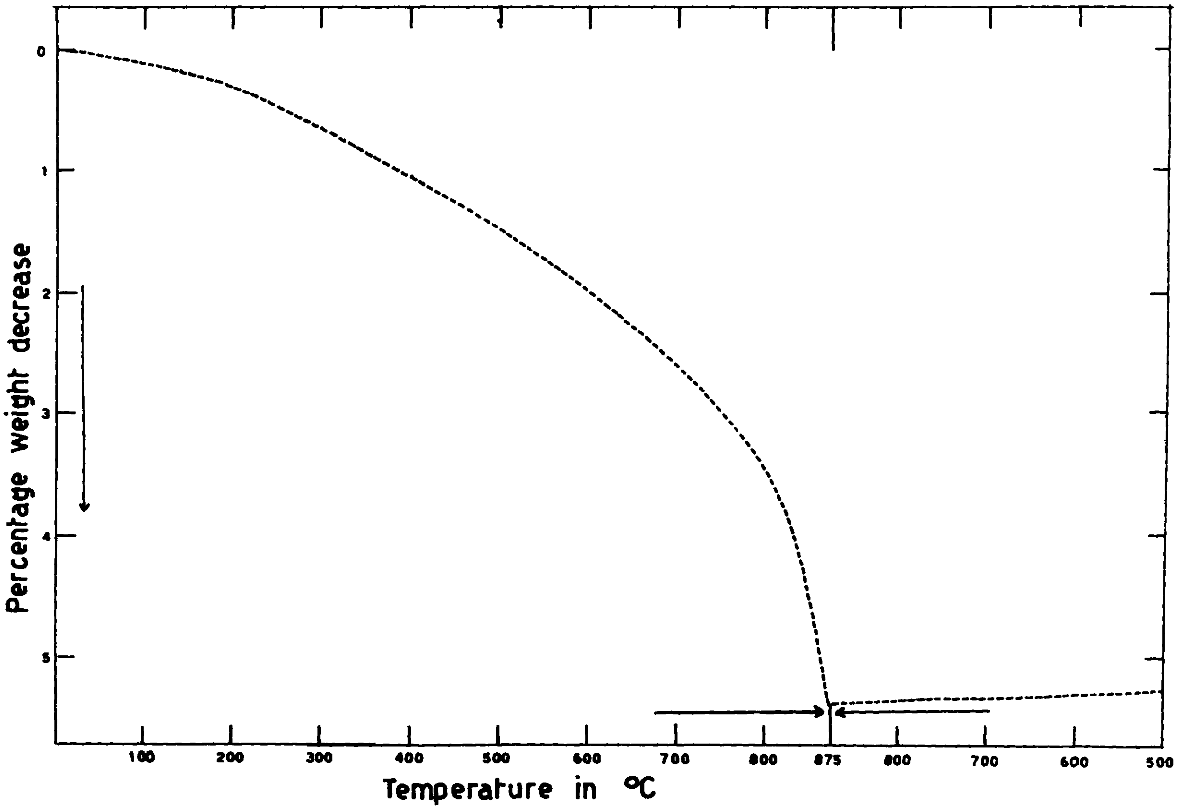

Figure 1. Termogravimetric analysis curve of uraninite from the Dominion Reefs Mine

TABLE III

CHEMICAL ANALYSIS OF URANINITE FROM THE DOMINION REEFS MINE(1)

CaO	1.30	
MnO	0.45	
PbO	14.85	
MgO	0.11	
R.E.	3.43	
UO_2	17.10	
UO_3	46.19	
ThO_2	6.52	
ZrO_2	< 0.10(2)	
SiO_2	1.43	
Al_2O_3	0.08	
Fe_2O_3	1.12	
H_2O^+	2.50	
S	1.26	
SnO_2	1.43	
TiO_2	0.40	
P_2O_5	0.09	
Total	98.26	
	- 0.63	i.e. O = 1.26 S
	97.63	

(1) Analysts : D.J.E. Venter, E.B.T. Cook, A.Watson, A. Gordon and C.Carlton, Government Metallurgical Laboratory, Johannesburg.

(2) Not included in total.

The uraninite used for this analysis was obtained from a heavy concentrate, rich in sulphides and cassiterite. The sulphides were removed by flotation, and the remaining material consisted predominantly of uraninite and cassiterite. The latter constituent could not be completely removed after repeated separation on a Franz Isodynamic Separator, and the SnO_2 reported is due to cassiterite as an impurity in the sample. The small amounts of TiO_2 and P_2O_5 may be due

/to...

to leucoxene and monazite which are major components of the heavy concentrates. These minerals could not be found in the purified uraninite by microscopic examination, but could easily have escaped detection in such small amounts.

In their study of twenty occurrences of uraninite and thorianite in the Grenville Sub-province of the Canadian Shield, Robinson and Sabina (1955, p.632) made the interesting observation that those uraninites containing 10% ThO_2 or less, and having a lattice constant less than 5.470 Å, occur in pegmatite, granite and syenite dykes which have well-defined walls but which in part may be due to replacement. The associated minerals are potassium and sodium feldspar and some quartz. Titanite, zircon, apatite and fluorite are common accessory minerals.

The thoria content and unit-cell dimension of the Dominion Reef uraninite are similar to those of this group of uraninite described by Robinson and Sabina.

In his paper on uraninite, Berman (1957, p.719) states that pitchblende differs from pegmatitic uraninite in having little or no ThO_2 and rare earths, a high CaO content and a small unit-cell size. According to these criteria the Dominion Reef uraninite again compares well with pegmatitic uraninite.

A search for inclusions other than the ubiquitous galena in the uraninite revealed a few veinlets of gold which are obviously of secondary origin, and a few tiny inclusions of zonal zircon crystals (Plate III, Fig. 6).

In sample 1(a) yellow alteration products of uraninite are present, amongst which schroeckingerite was identified with certainty. The X-ray pattern of the remaining yellow material is faint, and its constituents could not be identified. Slight effervescence with dilute hydrochloric acid, in which the material is easily soluble, indicates that small amounts of carbonate are present in addition to hydrated uranyl oxides. The vague term "gummite" can be used to designate this yellow alteration product.

The type of alteration ascribed to partial conversion of U^{+4} to U^{+6} without destruction of the crystal structure (Ramdohr, 1960, p.972) was observed in many uraninite grains; the altered material has a dull black colour and lower reflectivity than unaltered

/uraninite...

uraninite. This alteration product may replace the uraninite grains in the form of veinlets which form a trelliswork, or as irregular patches (Plate IV, Figs. 1 to 4). The uraninite grain~~s~~ pictured in Plate IV, Fig.3 ~~were~~ was used for X-ray diffraction. Before heating it yielded only the pattern of uraninite, but after heating, both the α-UO_2 and β-UO_2 patterns were present.

Another type of alteration product which is of a pitch-black colour, and in oil immersion shows brownish internal reflections, is present in many grains. In Plate IV, Fig. 4 it is shown next to the dull black material described above. In some cases it forms irregular veinlets in which galena is present as grains which are much bigger than the usual inclusions in uraninite (Plate IV, Fig. 5).

Enough material for an X-ray diffraction pattern was obtained with difficulty and the patterns yielded by this material before and after heating are faint. Although the lines could be measured, no pattern was found in the literature to match them.

In samples 15 and 16 the uraninite grains are enclosed by rims of material which contains specks of galena. These rims, when viewed by means of Ultrapak illumination are orange brown in colour and under the ore microscope they appear grey. The material yields a very faint X-ray diffraction pattern which does not conform with any one of the patterns available in the A.S.T.M. file or in bulletin 1064 of the U.S.A. Geological Survey. After heating, a stronger but completely different pattern is obtained. Some of the lines in this pattern belong to uraninite, but the remaining lines are again unidentifiable. These patterns are given in Table IV. Owing to the very small amounts of material which can be isolated and then only with difficulty, chemical information on the composition of the rims could not be obtained.

/Table IV...

TABLE IV

X-RAY DIFFRACTION PATTERNS OF REDDISH BROWN MATERIAL FORMING RIMS AROUND URANINITE GRAINS

(Small diameter Philips camera (57.27 mm. in diameter) Cu-radiation, Ni-filter. Exposed on General Electric X-ray machine)

Pattern before heating

d (Å)	I
3.39	5
3.08	10
3.01	10
2.51	1
2.29	1
2.10	10

Pattern after heating

d (Å)	I	phase
4.15	10	
3.72	1	
3.43	1	
3.34	3	
3.15	10	uraninite
3.03	1	
3.02	3	
2.72	2	uraninite
2.51	1	
2.26	10	
1.95	2	
1.93	2	uraninite
1.80	1	
1.65	2	uraninite
1.53	1	
1.39	3	
1.25	1	uraninite
1.18	4	
1.05	1	uraninite
0.925	3	"
0.912	1	"

/(b) Uranothorite...

(b) Uranothorite

Uranothorite grains are present in small amounts in most of the samples investigated. Two types of thorite could be distinguished.

The one type, formed at the expense of uraninite, was described in detail by Ortlepp (1962, p.197). It has a low reflectivity, and contains inclusions of galena similar to those in uraninite. In polished thin sections the thorite is translucent.

Unheated material yields the diffraction pattern of the included galena only. After heat treatment a thorianite pattern, having a $a_0 = 5.44$ Å, is obtained. This unit cell obviously does not reflect the true uranium : thorium ratio of the original thorite. Ortlepp reported a partial chemical analysis from which a uranium : thorium ratio of 0.26 can be calculated.

The second type of thorite is present in the form of fairly large reddish-brown grains which contain inclusions of galena, and in some grains also pyrite. The unheated material yields a diffraction pattern of galena only. After heating, the pattern of galena disappears, and the pattern of uranoan thorianite is obtained. In one case faint lines of thorite also appeared. The unit-cell of the thorianite phase so formed is 5.51 Å. This is obviously a type of ferrian thorite.

In Table V the most obvious differences between the two types of thorite are summarised.

TABLE V

COMPARISON OF THORITE AND FERRIAN THORITE ~~IN~~ FROM DOMINION REEFS MINE

	Thorite	Ferrian thorite
Colour	grey, usually translucent	reddish-brown, usually opaque
Origin	forms at expense of uraninite	present as discrete grains, of detrital origin
Inclusions	galena	galena and in some cases pyrite
Unit-cell of thorianite formed by heating	5.44 Å	5.51 Å

In plate V, Figures 1 to 4, the two types of thorite are illustrated.

/(c) Columbite...

(c) Columbite, betafite and euxenite

Columbite is present in most of the samples from the Upper Reef as grains of fairly low roundness and sphericity, many of which have been partly replaced by material having a much lower reflectivity.

Grains appearing fresh and largely unreplaced yield sharp clear columbite diffraction patterns, whereas those consisting largely of the alteration product are metamict. After heating, diffraction patterns are obtained which in some cases correspond fairly well, but not exactly, with that of betafite, and in other cases with some of the diffraction patterns reported for heated euxenite (Berman, 1955, p.187). However, this variation in the patterns obtained may be the result of the heat treatment used, and does not necessarily indicate the presence of more than one type of alteration product for the columbite. In the present study individual grains obtained from the polished sections were heated to red heat on a platinum foil in a bunsen burner.

Betafite was also reported by Ortlepp (1962, p.198), but Taylor et al. (1962, p.329) found only euxenite. The latter authors heated the metamict material to 1000°C in a neutral atmosphere.

Plate VII, Figs. 1 to 4, illustrate the typical appearance of columbite in various stages of replacement by metamict material.

(d) Monazite

Monazite is one of the most abundant heavy minerals present in the Upper Reef and is preferentially associated with uraninite. In the Lower Reef, the concentration is much lower.

In thin section the monazite is to be seen as small grains, many of them well rounded, of the order of 0.1 mm. in diameter, and always traversed by cracks.

The following optical constants were determined on grains in a concentrate of monazite from sample 1(a), using the immersion liquids described by West (1936, p.245).

$$\alpha \div \beta = 1.783 \pm 0.003$$

$$\gamma = 1.832 \pm 0.003$$

$2V\gamma = 14^{\circ}$ (determined according to Mallard's method).

The X-ray diffraction pattern shows faint lines only which could not be improved by heating. However, the pattern is unmistakenly that of monazite. The slightly low values of its refractive indices,

/and the...

and the faint X-ray pattern indicates a small degree of metamictization.

In polished section the monazite has a low reflectivity and amber coloured internal reflections. Many grains are cracked, displaced and corroded, but many well preserved grains on which roundness, size and shape can be measured, are available in some of the samples. Grains showing crystal faces are occasionally to be seen.

In Plate V, Figs. 5 and 6, and Plate VI, Figs. 1 and 2, grains of monazite are shown which attained a fairly high degree of roundness. In three of the illustrated grains crystal faces are discernible. Zoning ~~Zonation~~, such as that of the grain in Plate VI, Fig. 2, cannot be seen in polished section, but is clearly revealed in a polished thin section when a combination of transmitted and reflected light is used. Liebenberg (1956, p.382) reports 0.32% U_3O_8 and 4.74% ThO_2 in selected monazite grains from the Dominion Reef.

(e) Leucoxene

Although only a few ~~number~~ of the leucoxene grains is radio-active, the description of this mineral is included with the radio-active minerals.

In many samples leucoxene is the most abundant heavy mineral. However, in some samples from the Upper Reef both uraninite and monazite are more abundant than leucoxene.

In concentrates under the binocular microscope different colours of leucoxene from nearly white to khaki-grey, brownish, and nearly black can be seen.

According to the form of the grains three types of leucoxene can be distinguished :(i) In polished sections many grains are irregular in outline, and are interstitial to quartz and other mineral grains. (ii) A large proportion of the leucoxene grains have convex, and sometimes concave outlines, and have cores of secondary quartz. Bailey et al. (1956, p.263) described an amorphous stage in the formation of leucoxene from ilmenite. Golding (1961, p.1138) gave examples of porous silica-cemented leucoxene, some grains having colloform outlines. The type of convex and concave outlines illustrated in Plate VIII, Figs. 1 and 2, are considered therefore to represent colloform outlines which originated during an amorphous stage in the formation of leucoxene. (iii) In some samples many leucoxene grains still preserve the form of the original grains of the titaniferous mineral from which they were formed. Some of

/these...

these grains still contain inclusions of zircon which were present in the original minerals from which the leucoxene formed (Plate VIII, Fig.4).

Most of the leucoxene originated from ilmenite. The latter mineral is in fact the main host of titanium in samples 15 and 16, in which it formed as a recrystallization product of leucoxene.

A fair proportion of the leucoxene contains inclusions of galena. A number of these grains were exposed to nuclear track plates together with some grains devoid of such galena inclusions. The former type was found to be appreciably radio-active. Liebenberg (1958, p.382) reports 0.37% U_3O_8 in a leucoxene concentrate from the Dominion Reef.

Spectrographic tests show that iron and titanium are the major constituents of the leucoxene. A microchemical test using the sodium fluoride bead revealed also uranium in the leucoxene which contains inclusions of galena. Lead and boron are also indicated by the spectrograph, but as minor constituents.

The radio-active leucoxene could have formed from brannerite, sphene or davidite. Other titanium-rich radio-active minerals which may form radio-active leucoxene when they are altered by weathering are euxenite and betafite.

(f) Zircon

As many zircon grains are appreciably radio-active, this mineral is here described under the radio-active minerals.

Zircon is present in all the samples examined. It is always a minor constituent, but in sample 1(a) it reaches a concentration of approximately 1 per cent.

The most abundant type of zircon is present as euhedral purple grains which exhibit different stages of rounding, from angular to well-rounded.

A colourless type which often contains tiny inclusions of galena, and is slightly radio-active, is common in the Upper Reef. Some of these colourless grains are beautifully zoned (Plate VI, Figs. 3 and 4), and some of these zones have a lower reflectivity than the usual type of zircon. Such a decrease in reflectivity is obviously due to the lowering of the refractive index of zircon during metamictization.

Morgan and Auer (1941, p.305) found that the degree of metamictization of zircon is proportional to its radio-activity.

/They...

They recorded a refractive index as low as 1.782 for ω of highly radio-active zircon. In the zoned zircon grain shown in Plate VI, Fig. 3, the zones having the lowest reflectivity also have the largest number of inclusions of galena. If galena inclusions can be regarded as an indicator for the distribution of radio-activity, then metamictization in this crystal can be ascribed to radiation damage. Liebenberg (1958, p.382) reports 0.84% U_3O_8 in a concentrate of zircon from the Dominion Reef.

The metamict zones shown in Plate VI Fig. 4, contain only a few galena inclusions. When viewed in transmitted light these zones are completely isotropic. The refractive index is estimated to be between 1.65 and 1.70, which is considerably lower than the lowest value found by Morgan et al. (1941, p.305).

As shown by Larsen (1953, p.1118) the outer metamict zones of large zircon crystals can contain 3.9% rare earths, and then have a refractive index as low as 1.82. This changes abruptly to an inner core of fresh zircon (ω 1.918, ε 1.971) having only 0.6% rare earths.

A type of metamict zircon described under the name of ribeirite by Florencio (1954, p.159) contains 7.45% yttrium earths, 0.51% U_3O_8, and 8.43% H_2O. Its refractive index is only 1.683.

It would appear therefore that the presence of rare earths under certain conditions also causes metamictization, and the metamict zones of Plate VI, Fig. 4 may be of this nature.

These zones yield diffuse X-ray patterns in which the strongest lines for zircon can, however, still be recognized.

II. NON-RADIO-ACTIVE OXIDE MINERALS

In addition to the oxide minerals already described in the previous section, the minerals cassiterite, ilmenite, rutile and chromite were observed.

(a) Cassiterite

Cassiterite grains were encountered in some of the samples from the Upper Reef, and in sample 1(a) they reach a concentration of about one per cent by weight. Under the stereoscopic micrsscope they have an amber to dark-brown colour, and an adamantine lustre on fresh conchoidal fractures. Under the petrographic microscope the cassiterite is reddish-brown, and shows pleochroism with ε dark-brown and ω lighter-brown. Under the ore microscope it can be recognized by its

/amber...

amber-coloured internal reflections, and its anisotropism. Many grains are twinned.

The cassiterite grains usually have diameters which measure 0.10 to 0.22 mm., and most of them are elongated, and of low roundness (Plate VII, Figs. 5 and 6).

(b) Ilmenite

Ilmenite is abundant in samples 15 and 16, in which it formed by recrystallization of leucoxene. Many ilmenite grains contain remnants of leucoxene near their centres. This recrystallized ilmenite usually occurs in the form of laths and aggregates of laths, in many cases similar to rutile grains which also formed by the recrystallization of leucoxene.

Typical aggregates of ilmenite are shown in Plate VIII, Figs. 5 and 6. Rutile occurs in this ilmenite as small grains, but ilmenite aggregates, the centres of which are occupied by relatively large rutile grains, were also seen.

(c) Rutile

Apart from the rutile present as recrystallized portions of leucoxene grains, some detrital rutile was encountered. Most of the~~m~~ grains show polysynthetic twin-lamellae. They are light-yellowish in colour, and the sizes of the grains encountered are similar to those of the zircon in the same samples.

(d) Chromite

Chromite is present in small quantities in all the samples investigated in the form of small grains which often show signs of rounding. Under the stereoscopic microscope its smooth brilliant black surface and conchoidal fracture can be seen.

In polished section many chromite grains are fractured, and the fragments displaced. A greenish chlorite often accompanies this chromite, and many textures can be interpreted as replacement of chromite by chlorite.

zoning. ~~Zonation~~ of chromite grains is often evident. This ~~zonation~~ zoning in some cases post-dates the fracturing of the grains. In Plate IX, Fig. 1 a chromite grain with an outer zone of magnetite is shown. A rare type of chromite containing oriented inclusions is illustrated in Fig. 2.

Microscopic and X-ray examination reveal various types of chromite, which probably originated from different types and ages of

/host-rock...

host-rock. The most abundant type has a unit-cell size of 8.32 Å which corresponds to a chromium-rich variety.

III. SULPHIDES

The following sulphides were observed in the samples studied: Pyrite, marcasite, galena, chalcopyrite, covellite, bornite, arsenopyrite, cobaltite, linnaeite, sphalerite and molybdenite.

(a) Pyrite

Pyrite is by far the most abundant sulphide present in the Dominion Reef. Five textural types could be distinguished:

(i) In most samples the pyrite is present as isolated grains, usually between 0.1 and 1 mm. in diameter. These grains may exhibit the typical round outlines of buckshot pyrite, but in many samples their outlines are irregular. This type is associated with heavy minerals such as uraninite, monazite, leucoxene, and chromite.

(ii) Large irregular grains of pyrite not associated with heavy minerals are common in many samples. They are usually between 2 and 3 mm. in diameter, and may be closely associated with galena, chalcopyrite, covellite, bornite, linnaeite and sphalerite. In some cases these sulphides constitute inclusions in the pyrite.

(iii) Tiny veinlets or porous zones, probably formed by the partial healing of cracks, usually contain pyrite, sphalerite, galena, and chalcopyrite. These sulphides then are arranged in fairly straight lines in a zone rich in small pores and secondary quartz.

(iv) Skeletal crystals, laths and euhedral or subhedral crystals of pyrite replace grains and pebbles of quartz in some samples.

(v) Pyritization resulting from sulphur and iron of earlier minerals was observed in a few cases. In ferrian thorite part of the iron near the circumference of some grains combined with sulphur to form tiny laths of pyrite (Plate V, Fig. 4).

Another example is a grain of leucoxene in which pyrite was formed by pyritization of the iron present in the original mineral (Plate IX, Fig. 6).

(b) Marcasite

Small inclusions of marcasite were observed in many crystals of pyrite. In most cases these marcasite grains are very small, of the

/order...

order of 1 to 2 microns across, but they may form clusters occupying areas up to 50 microns across in pyrite. Such small inclusions of marcasite can not be easily observed with low magnification. With oil immersion and at high magnifications and crossed nicols they impart a mottled appearance to those portions of the pyrite in which they are present.

Occasionally larger marcasite crystals, up to 50 microns in diameter, are present in the pyrite. In such cases the pleochroism and anisotropism typical of marcasite can easily be observed.

(c) Galena

Two textural types of galena are to be found. The most abundant type is present as tiny inclusions and veinlets in uraninite and the other radioactive minerals, which include thorite, monazite, zircon, leucoxene, columbite, betafite and euxenite.

The tiny veinlets and porous zones described under pyrite, (paragraph iii) also contain galena. This type of galena is either intergrown with pyrite and covellite, or forms discrete grains, usually in close proximity to sphalerite.

(d) Chalcopyrite, covellite and bornite

Chalcopyrite is associated with the minerals occurring in the veinlets described above. It is present as fairly large grains, which may reach dimensions of 0.1 mm in diameter. In sphalerite it occurs as tiny grains formed as a result of exsolution.

In samples 15 and 16 many chalcopyrite grains contain inclusions of linnaeite.

In samples 1(a) and 1(b) chalcopyrite has been extensively altered to covellite. In some grains of covellite remnants of chalcopyrite are to be seen.

In most cases the covellite grains form aggregates of up to 0.1 mm across. The individual grains of covellite in these aggregates are usually of the order of 0.03 mm in diameter.

Many grains of covellite contain inclusions of pyrite. A few aggregates consisting of covellite intimately intergrown with galena are present.

(e) Arsenopyrite

Arsenopyrite is a common sulphide of the Dominion Reef, and is abundant in gravity concentrates. However, its distribution is patchy and in most of the samples used in the present study it could not be detected.

/It is...

It is present as isolated grains, usually of the order of 0.1 mm. across, in samples 9, 10, 11, 17 and 18.

(f) Cobaltite

Cobaltite is present as euhedral and subhedral crystals, many of them showing round outlines, and also as anhedral grains. It is white in colour with a creamy tinge. The size of the unit-cell is 5.58 Å, and spectrographically Co, Fe and As proved to be the main constituents.

A few anhedral grains of cobaltite contained inclusions of sphalerite and gold (Plate X, Fig. 2). In many cases small grains of gold are partially enveloped by cobaltite (Plate X, Fig. 4).

(g) Linnaeite

Linnaeite occurs as irregular isolated grains or as an intergrowth with chalcopyrite. It is cream-coloured, with a reddish tinge. The largest grains observed measure only 0.10 mm. across. It is traversed by tiny cracks having the appearance of shrinkage cracks, which divide the linnaeite into tiny polygonal blocks.

In addition to its association with chalcopyrite, linnaeite was also seen with sphalerite and enveloping and replacing pyrite.

(h) Sphalerite

The sphalerite grains in the Dominion Reef are usually very irregular in outline. They occur in tiny veinlets and porous zones and are associated with pyrite, galena and chalcopyrite. Exsolution bodies of chalcopyrite are present in most grains. Sphalerite was also observed as inclusions in cobaltite where it occurs in close association with gold.

(i) Molybdenite

Only one tiny lathlike grain of molybdenite was seen in sample 1(a).

Molybdenum was detected spectrographically in most concentrates of heavy minerals rich in leucoxene, but in this mineral its presence may be due to diadochic substitution of Mo^{4+} for Ti^{4+}.

IV. GOLD

Gold occurs typically as extremely irregular hackly grains. They are usually very small. The largest ones measure only about 0.05 mm. across.

/Most...

Most of the gold is present in clusters, which may consist of only a few grains, but in some cases consist of 20 or more which vary in size from less than 5 microns to more than 50 microns in diameter. Such clusters may be distributed through two or more mineral species which are situated close to each other.

A preferential association of gold with leucoxene was noticed. The reason for this association may be that redistributed gold was precipitated in the openings of porous leucoxene together with secondary quartz.

Tiny veins of gold were observed in uraninite, monazite, quartz and phyllosilicates. In one case tiny veinlets, similar in form and size to those of galena in uraninite, are of a composite nature. The central portions consist of gold and the sides of galena. In other cases they consist only of gold.

In some cases gold is associated with sulphides, most commonly cobaltite and galena. Some tiny gold grains are partly enveloped by cobaltite or galena. Larger cobaltite crystals also contain inclusions of gold.

V. SILICATE MINERALS

Quartz and phyllosilicates make up the bulk of the conglomerate of the Dominion Reef. In addition to these, detrital heavy silicate minerals are present. Among these, garnet is the most abundant. Much smaller amounts of sphene, epidote and pyroxene were found. Zircon has already been described.

(a) Garnet

Two types of garnet were encountered, in samples 1, 10, 14, 17 and 18 a yellow garnet, and in samples 1, 2, 3, 5, 6, 7, 9, 10, 14, 16, 17 and 18 a rose-coloured one. Some grains of the rose-coloured garnet are slightly birefringent. The refractive indices, densities and unit-cell sizes of the two types of garnet are listed in Table VI.

/Table VI...

TABLE VI

OPTICAL AND PHYSICAL PROPERTIES OF TWO TYPES OF GARNET IN THE DOMINION REEF

Colour	Bright yellow	Rose
n	1.865 ± 0.003	1.824 ± 0.003
D	4.10 ± 0.05	4.20 ± 0.05
a_o	11.68 ± 0.01Å	11.52 ± 0.01Å

Using the diagrams of Winchell (1958, p.595) the following composition was derived for the garnets :

The bright yellow one : 60% almandite and 40% andradite

The rose-coloured one : 76% almandite, 20% pyrope and 4% grossularite.

The grains of garnet reach dimensions up to 1 mm. and more in diameter.

Most of them have been extensively replaced by chlorite along their peripheries and also along cracks. In some cases they have been completely replaced by chlorite. However, this replacement is not strictly pseudomorphous, but causes alterations in the original outlines. Where the garnet has been sufficiently preserved, the degree of roundness can be measured, and it may be quite high for some grains, but the mean roundness values are fairly low.

(b) Pyroxene

Pyroxene was found only in sample 7. It has a light-brown colour, and is biaxial positive, with $2V_\gamma = 40^\circ - 50^\circ$, and $n_\beta = 1.70$.

The pyroxene grains are perfectly fresh, and do not show signs of alteration. As this sample was taken in close proximity to a dyke, the pyroxene is probably authigenic.

(c) Epidote

Epidote was encountered only in samples 18 and 20. Under the binocular microscope it appears as reddish-brown grains having a short lath-like form, and a good cleavage. It is slightly altered and its refractive index could not be determined accurately, but the optical constants are as follows:

/n_β...

$n_\beta = 1.76$

Axial angle large

α : brown

β : olive green

The identification was confirmed by means of an X-ray diffraction pattern.

(d) Sphene

Sphene was found in small amounts in samples 1 and 17.

In transmitted light the grains of sphene are perfectly fresh and colourless. Under the ore microscope it is yellow-grey with many yellowish internal reflections. The grains show signs of rounding, and have a fairly low sphericity. The grain illustrated in Plate IX, Fig. 5 is slightly corroded and typical of the sphene observed in the conglomerate.

(e) Biotite and Chlorite

In the Upper Reef a greenish biotite is present as a major constituent of the matrix. In many cases it replaces garnet, chromite, and other minerals.

It shows very strong pleochroism : α light-green, and β and γ dark-green. The negative axial angle is small, and n_β is approximately 1.63.

Fairly strong lines of chromium were found in the spectrum of ~~this~~ this biotite.

In most samples the biotite has been extensively altered to a greenish chlorite.

(f) Sericite

Sericite is one of the most important constituents of the matrix of the conglomerate. It imparts a khaki-grey colour to the conglomerate of the Upper Reef. It occurs interstitially, but also replaces quartz.

/Section B...

SECTION B : STUDIES OF FORM, SIZE AND SHAPE OF MINERAL GRAINS

I. INTRODUCTION

The roundness of the grains of uraninite in the Witwatersrand banket and in the Dominion Reef has been used by "placerists" as one of the most important arguments in support of their theory. Liebenberg (1955, p.118, 169) mentions the waterworn nature of uraninite as shown by the oval and round shapes of the grains as an important argument for a detrital origin. Ramdohr (1958, p.3) believes that "Uranpecherz" came into the conglomerate as small pebbles. According to Koen (1961, p. 42) the uraninite has a well-rounded appearance and oval or roundish shape. He thinks that for the sizes in which the uraninite grains occur, too high a degree of rounding is displayed to be consistent with the detrital theory of origin, and he uses these observations as an argument for his theory that uraninite nodules were precipitated in ancient marshes and lakes and later redistributed in the conglomerates.

The three authors mentioned above studied mainly material from the Witwatersrand System, but also included samples from the Dominion Reef. Their statements therefore also apply to the latter. Davidson (1958, p.193) argues that the uraninite grains are rounded to much smaller dimensions than could be achieved by attrition in natural waters. He compares these uraninite grains to typical roundish botryoidal grains from hydrothermal deposits. Bosazza (1959, p.313) thinks that the roundness of the uraninite is of little significance. He expects small crystals of uraninite (hexoctahedrons) to appear rounded, and draws a comparison to thin sections of chromitite of the Bushveld Complex in which the chromite grains are "as well rounded as the uraninite in the banket".

It is thus evident that as far as the interpretation of the roundness of uraninite from qualitative data is concerned, an impasse has been reached.

Much is known about the processes of rounding of mineral grains by the action of water and wind from studies in the laboratory and in the field. However, before the roundness of uraninite and the accompanying minerals can be tested in order to ascertain whether it

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conforms with this knowledge, it is rather imperative that quantitative and objective data on shape and roundness should be gathered. The next chapters are devoted to the techniques used to obtain such quantitative data.

II. REVIEW OF THE METHODS APPLIED IN STUDIES OF GRAIN-FORM AND SHAPE

A study of the literature reveals that three general methods for the determination of roundness exist, namely:

(i) Classification of grains in roundness classes, usually defined as angular, subangular, subrounded, and well-rounded. This method was used for example by Guggenmoos (1934, p.429) and also by Russell and Taylor (1937, p.225). It cannot be considered to be quantitative and objective, and for the purpose of the present investigation will not be further considered.

(ii) The use of comparison standards for the evaluation of the roundness of grains. In this connection the work of Russell and Taylor (1937, p.225), Krumbein (1941, p.64), Powers (1953, p.117), and Pettijohn (1956), are of importance. Rosenfeld and Griffiths (1953, p.553) made one of these visual comparison techniques the subject of a statistical study, and came to the conclusion that values obtained from such a study may be valid, but that the results should be interpreted with caution.

(iii) Methods based on measurements are those of Wentworth (1919, p.507, 1922, p.91; 1933, p.306), Cailleux (1952, p.11), Kuenen (1956, p.354), and Wadell (1932, p.443; 1933, p.310; and 1935, p.2501). The method of Wadell is applicable to small grains, and those of Wentworth, Cailleux and Kuenen can be used to evaluate the roundness of pebbles.

The principles put forward by Wadell exerted a profound influence on the methods used for studying roundness and shape since they were first published. For this reason a resume of his method will be given below.

Wadell's method was designed for the study of loose sediments. The grains of a sediment are placed on a glass slide which is slightly shaken in order to bring every grain to rest in its most stable position, that is, with its longest and intermediate axes in a horizontal plane. The grains are illuminated by transmitted light, and their silhouettes

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are reproduced (Wadell used camera lucida drawings) to a standard size, 7 cm. in diameter. These silhouettes are used to make the necessary measurements for the calculation of roundness and sphericity.

Wadell distinguishes between the concepts of roundness and sphericity. He defines the roundness of a plane corner as r/R, where r is the radius of curvature of the corner, and R the radius of the maximum inscribed circle in the silhouette of the grain. The roundness of a grain in one plane is the arithmetic mean of the roundness values of the corners:

$$P = \frac{\sum \frac{r}{R}}{N},$$

where $\sum r/R$ is the sum of the roundness values of the corners, N is the number of corners of the silhouette, and P is the roundness of the grain.

The radii of curvature are measured by means of a transparent celluloid with circles of various diameters engraved on it.

The degree of true sphericity of a solid is defined as

$$\psi = \frac{s}{S},$$

where s is the surface area of a sphere of the same volume as the solid, and S is the actual surface area of the solid. The true sphericity is difficult to determine and in a section or silhouette, it can not be determined at all, as only two dimensions of the grain can be measured. Wadell has shown that the two-dimensional sphericity yields values close to the true sphericity of a sand grain. He defines the two-dimensional sphericity as

$$\phi = \frac{d_n}{d_c},$$

where d_n is the diameter of the circle equal in area to the silhouette and d_c is the diameter of the smallest circle circumscribing the silhouette. For grains of flat or acicular shape two-dimensional sphericity will obviously deviate much more from true sphericity than for the usually nearly ovoid sand grains.

Wadell used a polar planimeter for measuring the areas of the silhouettes, and from these values he calculated d_n for every silhouette.

/A complete...

A complete description of the principles involved and the techniques used is given in the original articles of Wadell.

The method of Wadell is very time consuming. For this reason the use of comparison standards, based on Wadell's principles, were proposed, e.g. by Krumbein, Powers and Pettijohn for the determination of roundness, and by Rittenhouse (1943 a, p.79) for the determination of sphericity. Krumbein uses sets of standard silhouettes grouped into intervals of 0.1 of roundness values. Powers and Pettijohn also use standard silhouettes; these are however, no longer grouped in equal classes, but in classes which become progressively larger as the roundness values increase.

III. THE PRECISION OF METHODS BASED ON MEASUREMENT AND ESTIMATION

The methods ~~described~~ referred to above are very attractive because of their rapidity and ease of application. However, the possibility exists that they may be less precise than previously supposed, and that the results may easily be biased. It was decided therefore to compare a method based on measurement with one based on estimation in order to evaluate their precision.

The technique of Wadell is the obvious one based on measurement and Krumbein's method was selected to compare with it. The other methods based on estimation are not suitable, as the unequal spacing of the roundness classes which they use make grouping corrections impossible.

The methods of Wadell and Krumbein were compared by regression analysis and analysis of variance techniques. The details of this study will be published elsewhere. The most important conclusions are as follows:

(a) The measuring technique of Wadell is much more precise than the visual standard method of Krumbein.

(b) The results of workers using the visual standard technique can easily be biased.

(c) One of the forms of bias is the tendency of workers to evaluate a roundness not only in terms of the visual standards, but also in terms of the grains just studied. The effect of the first grain in a set is particularly evident. This effect is known among psychologists as the "levelling effect".

/(d) A...

(d) A regression equation is necessary to convert roundness values obtained by the use of Krumbein's standards to true Wadell values. However, this equation cannot take bias into account, and does not reduce the spread of values.

It is thus evident that in a study of material, in which the interpretation is in dispute, estimating methods are best avoided.

IV. THE SIZE OF IRREGULARLY SHAPED PARTICLES

A suitable measure of size for an irregular grain remains to be considered. Wadell (1932, p.443, 1933, p.310) discussed the sizes of such particles, and came to the conclusion that the use of long intermediate and short diameters, or the arithmetic or geometric means of these is "relatively meaningless". (However, in the further discussion in this paper the geometric mean is proposed as a suitable measure). He introduced the concept of the true nominal diameter defined as the diameter of a sphere of the same volume as the particle. In sections the nominal sectional diameter defined as the diameter of the circle having the same area as the section of the grain was introduced as a measure for size. The problem of measuring grain-size was also discussed by Krumbein (1939, p.122) who came to the same conclusions as Wadell.

Referring to Wadell's definition of two-dimensional sphericity, it can be seen that the value d_n which Wadell obtained by means of a polar planimeter is in fact the nominal sectional diameter. The measurement of this value is thus a prerequisite for calculating the sphericity by means of Wadell's formula. However, if the sphericity is obtainable in any other way, the nominal diameter can be calculated from it, and the length of the grain.

Pye et al. (1943, p.29) have shown that the square root of the ratios of the short and long diameters of grain silhouettes yields values close to Wadell's two-dimensional sphericity for sand grains, particularly those of approximately ellipsoidal shape. The nominal diameter can thus be calculated from measurements of the long and short diameters of a grain by using the following formula:

/d_n=...

$$d_n = d_l \sqrt{\frac{d_s}{d_l}} = \sqrt{d_s \, d_l}$$

where d_n is the nominal diameter, d_s is the short diameter, and d_l is the long diameter of the grain. It can thus be seen that the geometrical mean of the long and short dimensions of a grain is a valid size to use, although not very precise. The long and short diameters of a grain in a section are easily measured and the above formula is the logical one to use if only these measurements are available.

An improvement in the use of long and short diameters of a grain is the use of circumscribed and inscribed circles. The formula given by Riley (1941, p.94) for sectional sphericity reads:

$$\phi = \sqrt{\frac{d_i}{d_c}}$$

where d_i is the diameter of the largest inscribed circle, and d_c the diameter of the smallest circumscribed circle of a grain. This formula yields better sphericity values than the one of Pye, and nominal diameters derived from it will therefore also be more precise. As in the case of Pye's formula, the nominal diameter is also given by a geometrical mean, but this time the geometrical mean of the smallest circumscribed circle and the largest inscribed circle of the grain:

$$d_n = \sqrt{d_i \, d_c}$$

A fourth method, slightly less elaborate than Wadell's use of the polar planimeter, and second only to it in precision, is given by Alling (1941, p.28). He projects a circle on to a grain, and adjusts its size until its area is estimated to be the same as that of the grain. The diameter of this circle equals that of the nominal diameter of the grain.

The semiautomatic instrument described by Endter et al. (1956, p.97) and now marketed by Carl Zeiss, is based on the principles advocated by Alling.

/V.Complications...

V. COMPLICATIONS RESULTING FROM THE USE OF SECTIONS IN SIZE AND SHAPE MEASUREMENT

Wadell, Pye, Riley and Alling evolved their formulae for loose grains, usually of sand size, which were sprinkled on to glass slides, and the silhouettes of which were used for the necessary measurements. Even cursory examination of the minerals in the Dominion Reef shows that the heavy mineral grains separated from the conglomerate after crushing will in many cases be broken pieces of the original grains. The methods described above can therefore not be applied to these grains. It becomes imperative to make the necessary measurements of the grains in polished section. The outlines of such grains will in general not conform to the silhouettes which would have been seen were it possible to isolate the grains,and illuminate them from below on a glass slide.

(a) Roundness and Sphericity

An empirical test was carried out in order to ascertain whether sectioning of grains have a serious effect on the roundness and sphericity values obtained from them.

Five quartz grains were selected, and their silhouettes photographed when they were orientated in their most stable position on a glass slide. In grains 2, 4 and 5 two stable positions were found, and two photographs were taken. These grains were then mounted in resin, and ground down, until sections through them were obtained. These sections were orientated randomly, and had no particular relationship to the orientation of the silhouette planes. After light polishing the grains were photographed. They were then ground down further, and the process repeated for two more sections through the grains. The roundness and sphericity were measured on each photograph using Wadell's methods. These values are shown in Table VII.

In Table VII (a) the differences in roundness values obtained by each method for each grain are evaluated statistically, and it is concluded that this experiment does not reveal any significant differences in roundness values between the two methods, There is no obvious reason why this conclusion obtained by using quartz grains, will not be true for other grains of roughly similar shape, such as the grains of heavy minerals in the Dominion Reef.

/The differences...

The differences in sphericity values obtained by each method for each grain are evaluated statistically in Table VII (b), and it is concluded that there may be a difference between sphericities measured on loose grains and in sections.

Grains, when resting in their most stable positions as required by Wadell's method, yield silhouettes of maximum area. The two-dimensional sphericities determined on these silhouettes generally will have the maximum values except in rodlike grains. In the case of flat grains the two-dimensional sphericity so determined can be much in excess of the true sphericity. It can be expected therefore that two-dimensional sphericities determined on irregular grains in their most stable positions will be higher than those determined on random sections through the grains. This is borne out by the results of Table II. The mean sphericity of all grains as determined on silhouettes, namely 0.877, is higher than that determined on sections through grains, namely 0.802. Two-dimensional sectional sphericity will in general be closer to the true sphericity than values determined on silhouettes of grains resting on their flattest sides.

The determination of roundness and sphericity values on sections through grains rather than on silhouettes of grains is thus a valid method to use.

(b) Size

The size of grains as seen in thin section is usually less than their true size. In cases where one is interested in the true size, corrections must be applied. For an individual grain no correction will be possible. For size distributions however, a number of methods for correction have been proposed. The size distributions can be represented in the form of cumulative curves drawn according to the principles discussed by Krumbein and Pettijohn (1938, p.188). Most modern workers however, prefer to use logarithmic probability curves drawn according to the method described by Otto (1939, p.64), and also by Inman (1952, p.125) for determining parameters to describe the size distribution of sediments. The cumulative curves used by these workers, and in most modern papers on mechanical analysis, show cumulative weight percentages as determined on loose sediments by sieve analysis. In cases where measurements are carried out on individual grains, number frequency

/cumulative...

TABLE VII

ROUNDNESS (P) AND SPHERICITY (Ø) VALUES DETERMINED ON SILHOUETTES AND SECTIONS OF GRAINS

	SILHOUETTES									
	Grain 1		Grain 2		Grain 3		Grain 4		Grain 5	
	P	Ø	P	Ø	P	Ø	P	Ø	P	Ø
Position 1	0.53	0.84	0.55	0.94	0.81	0.91	0.37	0.83	0.37	0.94
Position 2	-	-	0.64	0.93	-	-	0.34	0.75	0.34	0.88
Mean values	0.53	0.84	0.595	0.935	0.81	0.91	0.355	0.79	0.355	0.91
	SECTIONS THROUGH GRAINS									
	Grain 1		Grain 2		Grain 3		Grain 4		Grain 5	
	P	Ø	P	Ø	P	Ø	P	Ø	P	Ø
Section 1	0.54	0.80	0.59	0.94	0.79	0.87	0.34	0.83	0.41	0.70
Section 2	0.54	0.76	0.67	0.93	0.74	0.84	0.24	0.78	0.40	0.71
Section 3	0.48	0.70	0.68	0.85	0.71	0.84	0.19	0.78	0.26	0.70
Mean values	0.52	0.753	0.65	0.907	0.75	0.850	0.26	0.797	0.36	0.703

TABLE VII (a)

APPLICATION OF "t" TEST TO ROUNDNESS DATA OF TABLE VII*

Grain No	Roundness Values Silhouette X_1	 Section X_2	Difference $D=X_1-X_2$	Deviation $d=D-\bar{d}$	Squared Deviations d^2
1	0.530	0.520	0.010	-0.011	0.000 121
2	0.595	0.650	-0.055	-0.076	0.005 776
3	0.810	0.750	0.060	0.039	0.001 521
4	0.355	0.260	0.095	0.074	0.005 476
5	0.355	0.360	-0.005	-0.026	0.000 676
Total	2.645	2.540	0.105	0	0.013 570
Mean	0.529	0.508	$\bar{d}=0.021$		$S_D^2=0.002714$

$S_D = \sqrt{0.002714} = 0.0521$ $\quad S_{\bar{d}}^2 = \frac{0.002714}{5}$ $\quad S_{\bar{d}} = 0.0233$

$H_o: \mu_D = 0 \quad t = \frac{\bar{d} - \mu_D}{S_{\bar{d}}} = \frac{0.021}{0.0233} = \underset{\longrightarrow}{0.901}$

degrees of freedom : 4

According to tables for the distribution of t, p is slightly larger than 0.4, and the null hypothesis: $\mu_D = 0$, is accepted.
The experiment therefore does not reveal any significant difference between silhouette roundness and sectional roundness.

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* Symbols used are those of Snedecor (1961)

TABLE VII (b)

APPLICATION OF "t" TEST TO SPHERICITY DATA OF TABLE VII

Grain No	Sphericity Values		Difference	Deviation	Squared Deviations
	Silhouette	Section			
	X_1	X_2	$D=X_1-X_2$	$d = D-\bar{d}$	d^2
1	0.840	0.753	0.087	0.012	0.000 144
2	0.935	0.907 ~~0.828~~	0.028	-0.047	0.002 209
3	0.910	0.850	0.060	-0.015	0.000 225
4	0.790	0.797	-0.007	-0.082	0.006 724
5	0.910	0.703	0.207	0.132	0.017 424
Total	4.385	4.010	0.375	0	0.026 726
Mean	0.877	0.802	$\bar{d}=0.075$		$S_D{}^2 = 0.005\ 345\ 2$

$$S_D = \sqrt{0.0053452} = 0.07311 \qquad S_{\bar{d}}^2 = \frac{0.0053452}{5} = 0.001069$$

$$H_o : \mu_D = 0 \qquad t = \frac{\bar{d} - \mu_D}{S_{\bar{d}}} = \frac{0.075}{0.0327} = \underrightarrow{2.293} \qquad S_{\bar{d}} = 0.0327$$

degrees of freedom : 4

According to tables for the distribution of t, $0.1 > p > 0.05$. Indications are that the null hypothesis: $\mu_D = 0$, is not true, but a larger sample may be necessary to confirm this.

cumulative curves can be constructed. In studies of thin or polished sections neither a correct number frequency curve, nor a correct weight frequency curve is obtained. The sectional size frequency distribution will therefore be different from the number frequency and weight frequency distributions.

A study of the literature shows that factors for converting these different size frequency distributions into each other received the attention of a number of workers. The methods proposed by them are listed in Table VIII.

/Table VII ..

TABLE VIII

METHODS FOR CONVERTING SIZE FREQUENCY DATA

(a) Conversion of section size distributions into actual number frequency size distributions

Investigator	Approach
Krumbein (1935, p. 482)	Theoretical consideration of sectioned spheres.
Chayes (1950, p. 156)	Theoretical estimate of the diameter of a circle from the lengths of chords passed through the circle at random. This method duplicated Krumbein's result, but in a simpler manner.
Greenman (1951, p. 447)	Based on Krumbein's method, but extended it to obtain a total distribution.
Pelto (1952, p. 402)	Defined precision and limitations of Greenman's method.
Packham (1955, p. 51)	Applied point count method and short chord measurements to obtain the distribution, which was corrected by means of factors based on Krumbein's approach.
Vistelius (1958, p. 224)	A discussion of, and contribution to Packham's method.

(b) Conversion of section size distributions into weight or volume size distributions

Investigator	Approach
Rosenfeld et al. (1953, p.114)	Determined conversion equation empirically by studying several samples of sand.
Packham (1955, p. 53)	The approach was similar to that listed under (a) above, but the factors are different.
Roethlisberger (1955, p. 579)	Devised a simple graphical method applicable to chord measurements of concrete aggregates in the form of pebbles.
Friedman (1958, p. 394)	Determined conversion equations empirically.

(c) Number frequency to weight frequency

Investigators

Hatch (1933, p. 27)
Grassy (1943, p. 47)
Rittenhouse (1941, p. 1930)
Pye (1943, p. 85)

/In...

In the author's opinion the method of Greenman should be used for converting sectional distributions to number frequency distributions, and the regression formula of Friedman is convenient to use should it be necessary to obtain weight data from sectional size frequency distributions of particles of sand size and shape.

VI. METHOD EMPLOYED FOR THE DETERMINATION OF ROUNDNESS, SPHERICITY, AND SIZE

It has long been realised that Wadell's method, although precise, is very time-consuming. If the necessity of drawing or photographing a grain is obviated, a very large saving in time can be effected without sacrificing precision. One way of doing this, is to carry out the measurements directly under the microscope.
D.A. Robson (1958, p. 108) described a specially constructed graticule which could be fitted in an ocular of a microscope, and by means of which the necessary measurements for Wadell's roundness could be made. The diameters of Robson's graticule were spaced on a linear scale, as were the diameters of the circles engraved on Wadell's original transparent celluloid. However, the formula proposed by Wadell expressed roundness in terms of ratios of diameters. For this reason a geometric size scale will be more logical to use. In addition, a suitable geometric scale will enable one to use the same set of measurements for sphericity, roundness and size calculations.
A suitable logarithmic interval would be small enough to permit useful size measurements to be made, yet not so small that the graticule becomes unwieldy to use. A scale of which the successive members have a constant ratio of 917:1000, will enable one to measure sizes to the nearest $^1/8$th of a phi unit. *

---

* Sizes in mm. are easily converted to phi-units by using $\phi = \frac{\log \xi}{\log 2}$ where ξ is the size in mm.

/In cumulative...

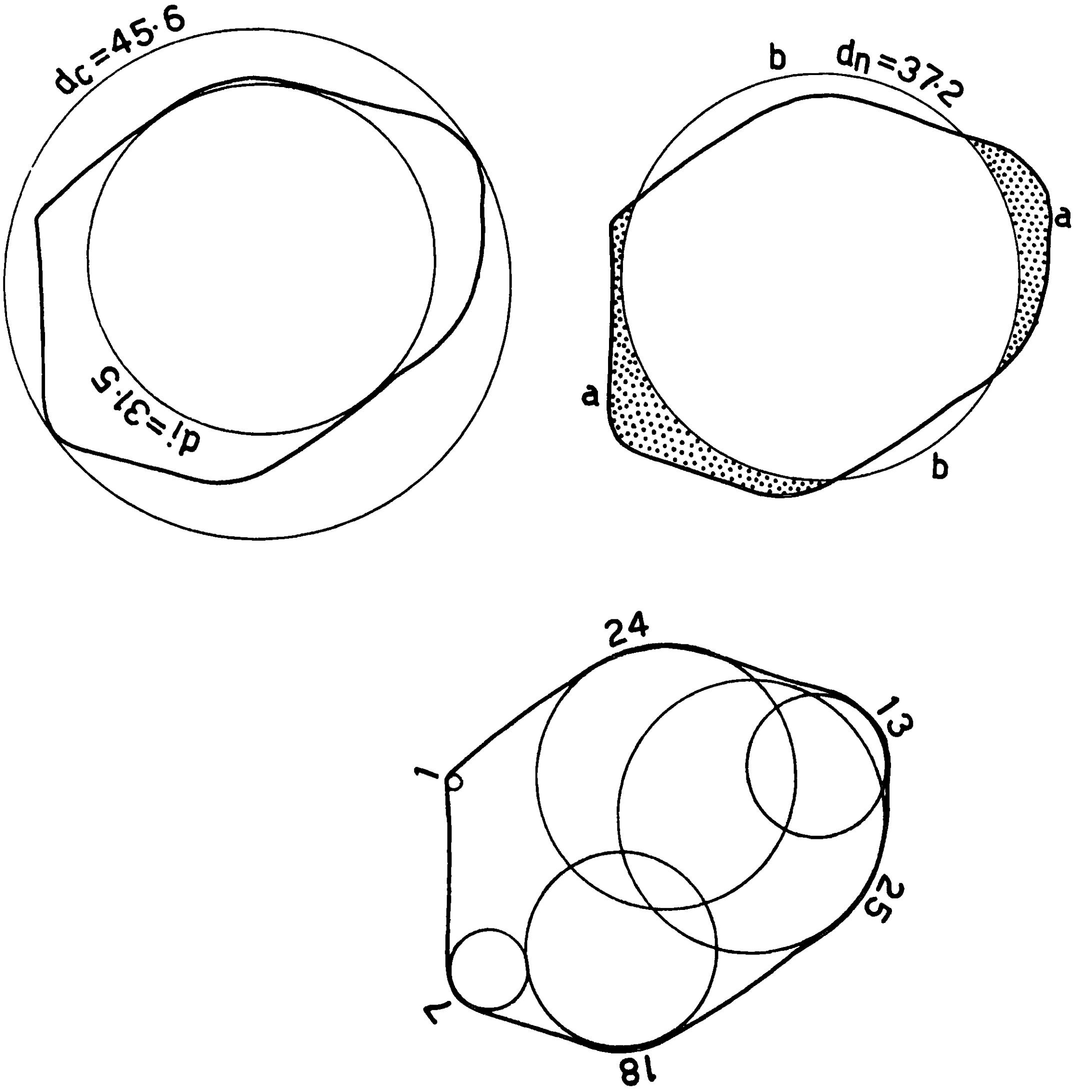


Figure 2. The method used for measuring the size, roundness, and sphericity of a grain. The upper left hand diagram shows circumscribed and inscribed circles. The circle in the upper right hand drawing has the same area as the grain section, and its diameter is the nominal diameter. The lower diagram shows radii of curvature of corners.

In cumulative curves,values are often bracketed to 1/4 phi unit. It was accordingly decided to construct a graticule with circles of the following relative sizes:

1024,	939,	861,	790,	724,	664,	609,	558,
512,	469,	431,	395	362,	332,	305,	279,
256,	235,	215,	197,	181,	166,	152,	140,
128,	117,	108,	98.7,	90.6,	83.0,	76.1,	69.8,
64.0,	58.7,	53,8,	49.3,	45.3,	41.5,	38.1,	34.9,
32.0,	29.3,	26.9,	24.7,	22.6,	20.8,	19.0,	17.4,
16.0,	14.7,	13.4,	12.3,	11.3,	10.4,	9.51	8.72
8.00,	7.34,	6.72,	6.17,	5.66,	5.19,	4.76,	4.36,
4.00							

The use of this graticule can best be explained by means of an example.

For this purpose the uraninite grain shown in Plate II, Fig. 3, will be used. In text Figure 2 (p.41) this grain is shown in outline, and the different steps of the measurement by means of circles on the graticule are illustrated.

The measurement of the circumscribed circle and the inscribed circle is easily accomplished as shown in the upper left hand drawing of Figure 2.

The method used for obtaining the nominal sectional diameter, d_n, is shown in the upper right hand drawing. The successive circles of the graticule are brought to coincide with the grain until a circle is found which has the same area as the grain. As was pointed out by Alling the most precise way of judging which circle has the correct size, is to compare the areas of the grain falling outside the circle (the stippled areas marked "a") to the areas of the circle falling outside the grain (the areas marked "b"). When the areas marked "a" equal those marked "b" , the area of the circle is equal to that of the grain, and its diameter then equals the sectional nominal diameter of the grain.

The radii of curvature of the corners of the grain are next determined. The outlines of the different circles of the graticule are brought to coincide with the periphery of the grain at the corner, until a circle is found which matches the rounded portion of the corner.

/The diameters...

The diameters of these circles for the six corners of the grain are shown in the lower drawing of Figure 2. The values obtained in the manner described above are entered in a table, an example of which is shown in Table 1X. Following Wadell, all curves having radii of curvature larger than that of the inscribed circle, are ignored. This includes re-entrant angles and concave curves.

TABLE 1X

EXAMPLE OF TABLE USED FOR ENTERING MEASURED SIZE, ROUNDNESS AND SPHERICITY DATA

d_n	d_c	d_i	dr					
			1	2	3	4	5	6
37.2	45.6	31.5	1	24	13	25	18	7

From the figures in Table 1X the following are calculated:

(a) The sectional nominal diameter in mm. is obtained from the calibration factor of the graticule for the particular objective used.

(b) The two-dimensional sphericity is obtained by the ratio of d_n/d_c, viz. 37.2/45.6, which equals 0.816.

(c) The roundness values of the individual corners are given by the ratios of d_r/d_i. The actual values found are 0.03, 0.76, 0.41, 0.79, 0.57, 0.22. The mean roundness is found by dividing the sum of these values by the number of corners measured, and the final value so obtained is 0.46.

VII. COMPLICATIONS DUE TO CORROSION AND ALTERATION

(a) Suitability of samples from the Dominion Reefs Mine for roundness, shape and size studies

Values obtained in the way as shown above can be considered to be reasonably accurate and unbiased. However, in the application of this method to ores from the Dominion Reef, difficulties are soon encountered.

A large proportion of the original grains have been altered by leaching, and deformation. Grains in which the original outlines have been partly or wholly preserved are absent or rare in some of the specimens.

/In many...

In many grains a portion of the outlines have been destroyed, but one, two, or three corners remain of which the curvature can be determined. In cases like these it is possible to determine roundness values from the remaining evidence.

It can easily be shown that in attempts to postulate the original shape of a partly destroyed corner of a grain, only a small variation of surface area will cause a big variation in roundness value. It is of course not possible to obtain objective results by such a postulating method, as the investigator is virtually free to create his own evidence. However, the sphericity and size of a grain is not seriously affected by slight alteration, and it is possible to obtain size and sphericity data from grains on which the measurement of roundness is no longer possible.

Apparently euhedral grains of zircon which may be non-metamict cores surrounded by metamict or altered material, or in some cases may even be a recrystallized zircon grain, are easily recognized, and should be ignored. Such zircon grains were rare in the samples examined.

In some cases the original shape of a grain is revealed by alteration products and lines of tiny galena crystals (Plate III, Fig. 1). In such cases it may be possible to obtain roundness and sphericity values for the original grain.

The poor degree of preservation of uraninite and other detrital heavy minerals is common to most samples not only of the Dominion Reef, but also of the Witwatersrand System which has been subjected to the same metamorphic processes.

Previous workers who used roundness in their arguments paid scant attention to the degree of preservation of the minerals. In one instance far-reaching conclusions were drawn regarding the roundness of a mineral postulated to have been amphibole, and of which nothing but alteration products remained (Koen, in press).

It may well be asked whether there are samples at all which are suitable for quantitative roundness determination. It is of course evident that in cases where quantitative measurements prove impossible or suspect, the kind of loose qualitative statements regarding roundness which were made by some previous workers should be regarded as infinitely more open to doubt.

/Of ...

Of the twenty samples listed in Table I, four are regarded as too highly altered for roundness studies. Thirteen samples are too poorly mineralized. Samples 1(a) and 1(b) contain a large percentage of excellently preserved uraninite and detrital heavy mineral grains, and are doubtlessly of the best suited samples for this kind of study that one can hope to find in the Dominion Reef and Witwatersrand Systems. Sample 2 contains well preserved grains, but they are fewer, and this sample is less suitable.

(b) Bias due to Selective Sampling

Even in the best samples many grains cannot be used for shape and roundness measurements as they have not been well enough preserved. This fact need however not be disquieting, as it is common practice to obtain statistical data from samples which form small percentages of the populations from which they are drawn. It is essential however that the data used are truly representative of the population and not biased.

It may well be argued that the object of the study is to determine as closely as possible what the roundness and forms of the original population of grains were, before metamorphism, corrosion and recrystallization of the grains took place.

A nearly perfect crystal will then be rejected by some investigators as being due to recrystallization, or even as being of secondary origin. However, it may well be a very slightly rounded crystal of original uraninite, and if this is the case, it should be included in the measured data (see Plate II, Fig. 1).

Different interpretations can also be given for grains having round corners (Plate II, Fig. 3. See also its description on page 7).

When confronted by such grains the investigator, in trying to make a decision, will be largely influenced by his previously held ideas on the origin of the minerals present, and the data selected by him will then be precisely those necessary to support his ideas, and those ignored will be the data which do not fit his theory.

It is thus clear that any attempt to select only those grains thought to reveal the original condition will introduce bias, and data obtained from such grains only are worthless.

/Certainly...

Certainly, the decision as to which grains are suitable for measuring and which ones should be rejected cannot be left to the whims of the investigator.

The criterion used in the present study was based on the state of preservation of the grains examined. If more than fifty percent of the outlines of a grain was not corroded, the data which could be measured on it were used. If more than fifty percent of the outline of the grain was destroyed, no roundness, shape or size data were measured on it.

By using this criterion, bias due to selective sampling was avoided.

However, if processes such as recrystallization, the presence of colloform grains, etc. played a role in the history of the grains, the roundness data will include their effects. It will be shown later on how the data can be tested for such effects, and if present, freed from them.

This approach, viz. the interpretation of data after having obtained them, and not while obtaining them, is of course the only tenable one.

(c) Bias due to selective corrosion

If the process of corrosion prefers rounded grains above angular ones, or vice versa, the method above will include data which are not representative of the population which existed before corrosion took place.

In order to test for this possibility, data on the state of preservation of the outlines of every grain studied were obtained.

Three types of outlines are distinguished, viz. rounded outlines, crystal edges, and corroded outlines. It is possible to represent the relative proportions of these types of outline on a ternary diagram by means of a single point for every grain studied. It was decided to divide the ternary diagram into fifteen classes (Fig 3). After a short study a grain could usually be allocated to the correct class without actually measuring the different types of outline.

After studying from 100 to 300 grains in this way, and recalculating to percentages, a diagram such as those of figures 4 and 5 can be constructed. Only the uraninite and monazite of samples 1(a) and 1(b) were studied in this way, and 3 polished sections of sample 1(a) and 2 of sample 1(b) were used.

/Referring...

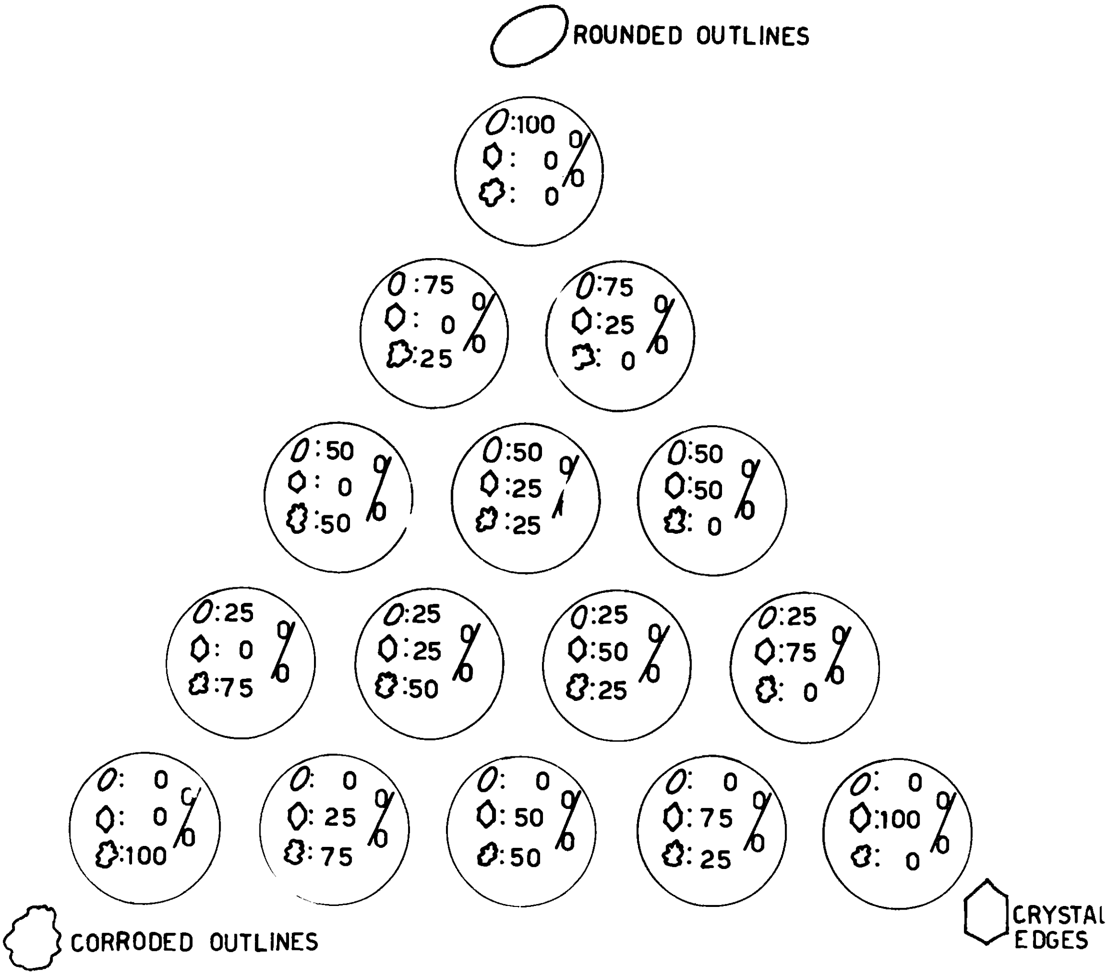


Figure 3. Fifteen classes into which a ternary diagram can be divided, and in which a grain can be classified according to its outlines

Fig. 4(a)

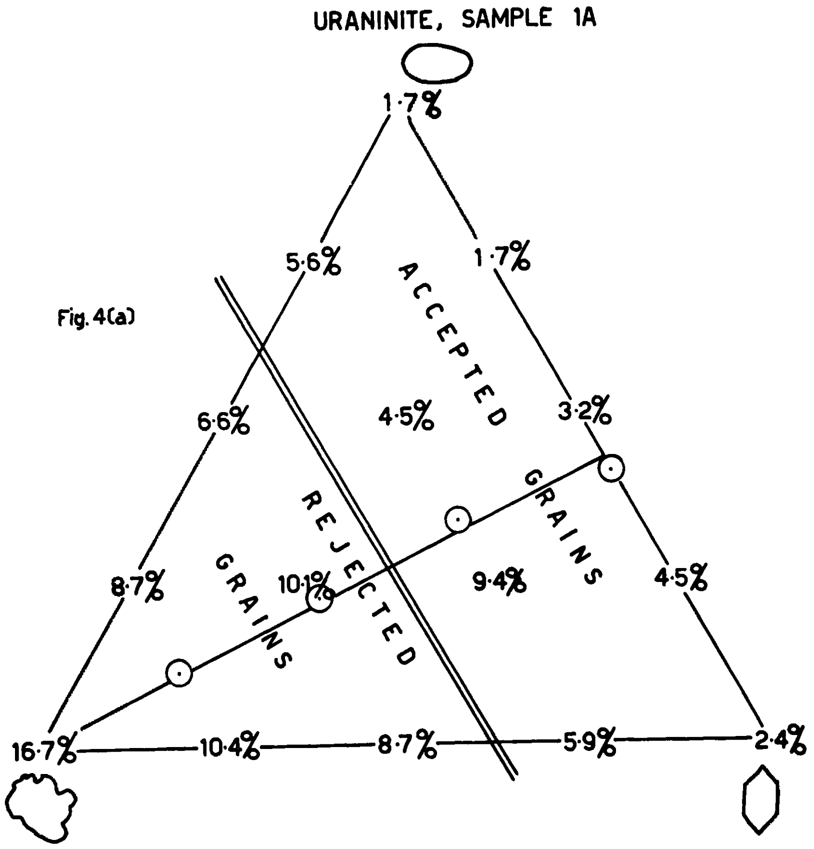


Fig. 4(b)

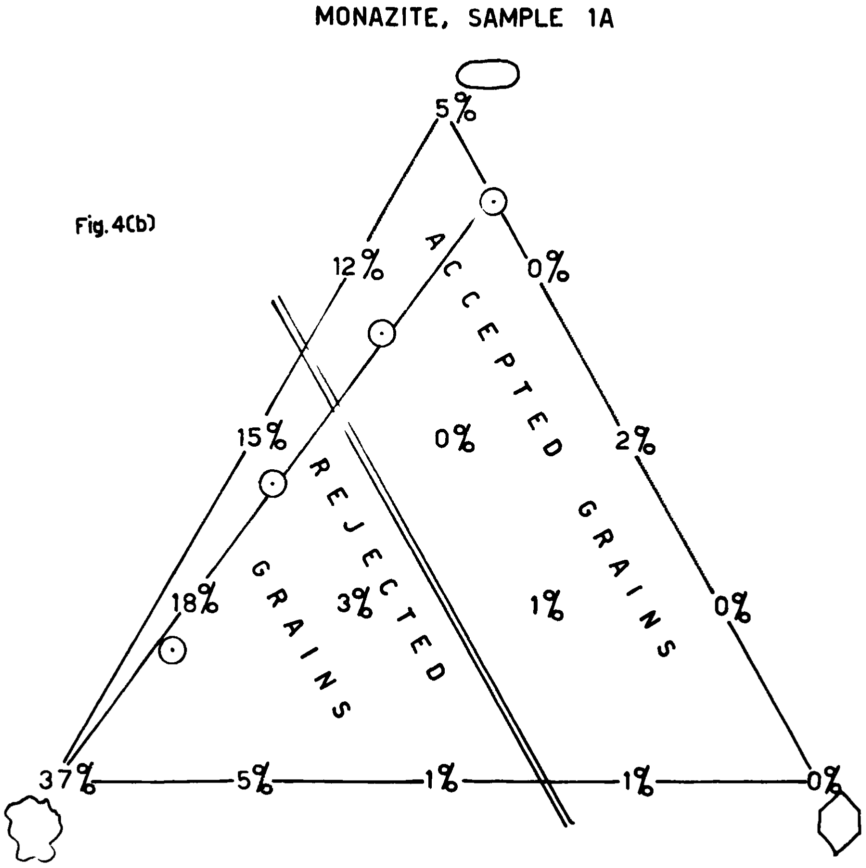


Figure 4. The distribution of uraninite and monazite grains in sample 1(a) according to their outlines. The number of grains used in each case are: For fig. 4(a) 287 uraninite grains. For fig. 4(b) 100 monazite grains.

URANINITE, SAMPLE 1B

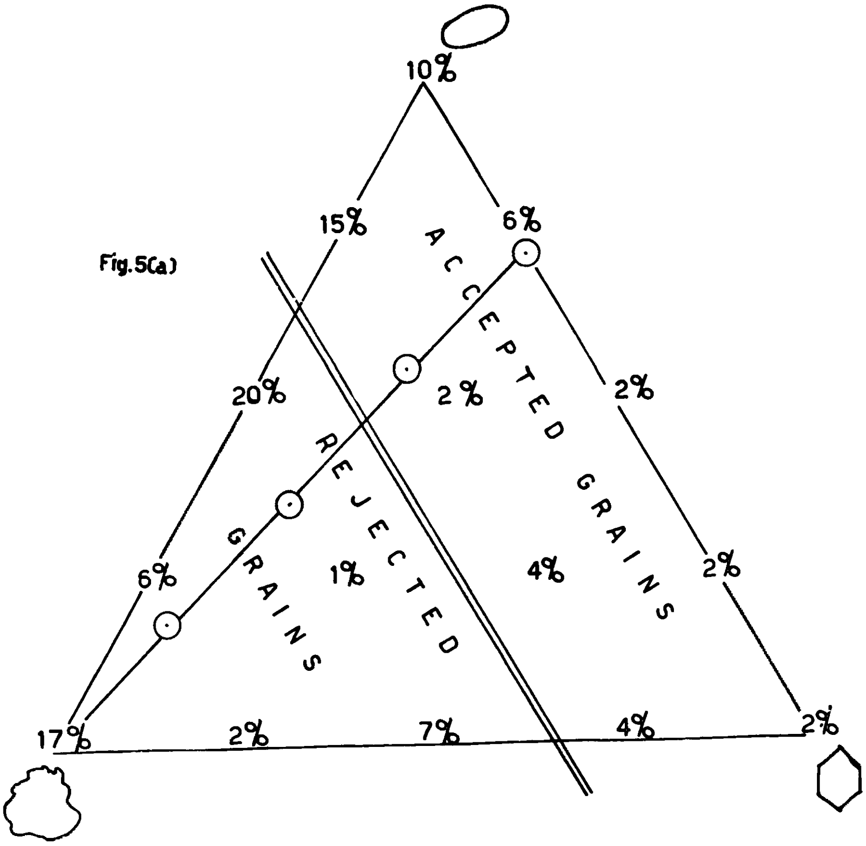


Fig.5(a)

MONAZITE, SAMPLE 1B

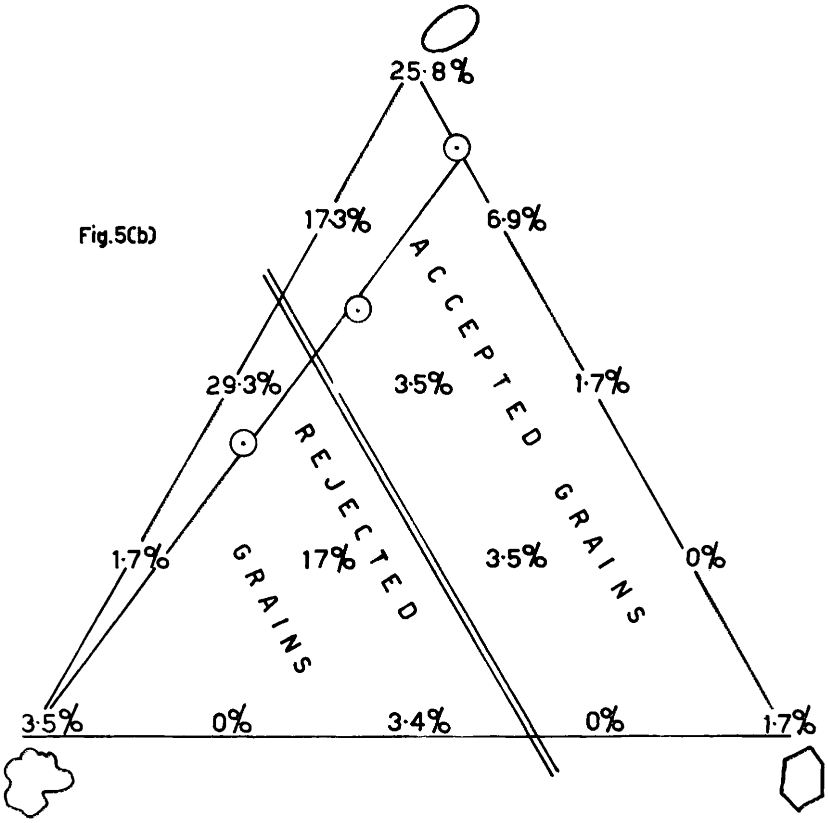


Fig.5(b)

Figure 5. The distribution of uraninite and monazite grains in sample 1(b) according to their outlines. The number of grains used in each case are: For fig. 5(a) 100 uraninite grains. For fig. 5(b) 116 monazite grains.

Referring to Fig. 4(a), which contains the data for the state of preservation of the outlines of uraninite in sample 1(a), it can be seen that in the five classes on the right hand side diagonal (i.e. from 100% rounded outlines to 100% crystal edges) 13.5% of the uraninite grains are classified. For these five classes a mean percentage of rounded outlines can be calculated. This percentage is indicated by the circled point.

Similarly, in the second diagonal line from the right, in the four classes having 25% corroded outlines in common, 25.4% of the uraninite grains are classified, and the percentage of rounded outlines can be calculated in a similar way. This process can be repeated for the three classes having 50% corroded outlines in common, and the two classes having 75% corroded outlines in common. The results of these calculations are given in Table X.

TABLE X

PERCENTAGES OF ROUNDED OUTLINES AND CRYSTAL EDGES IN CLASSES HAVING COMMON PERCENTAGES OF CORRODED OUTLINES, URANINITE, SAMPLE 1(a)

Classes	% rounded outlines	% crystal edges
0% corroded outlines	42.2	57.8
25% corroded outlines	34.65	40.35
50% corroded outlines	22.95	27.05
75% corroded outlines	11.375	13.625

Originally, when no corroded grains were present, all the uraninite would have been allocated to the five classes on the right hand diagonal. Due to corrosion some of these grains were removed from these classes, and moved in the direction of the corner representing corroded outlines.

/Now...

Now, if the process of corrosion favoured, say, rounded grains, more rounded grains would have been removed from the classes on the right hand side diagonal towards the corner representing corroded outlines. The percentage of rounded outlines on the right hand side diagonal would decrease, and amongst the grains removed, the percentage of rounded grains would be higher than in the original population.

On the other hand, if the process of corrosion did not favour rounded grains, or vice versa, the probability of attack of a rounded outline will be proportionate to its percentage in the original population. The proportions of rounded outlines to crystal edges in the preserved and corroded classes can then be expected to be substantially the same.

In Fig. 4(a) this can be tested for by ascertaining whether the points representing the outlines,as listed in Table X,lie close to a straight line through the corner representing 100% corroded outlines. Inspection of Fig. 4(a) shows that this is indeed the case.

Similar plots for uraninite in sample 1(b) and monazite in samples 1(a) and 1(b) (Figs. 4(b), 5(a) and 5(b))show that in all cases the points lie close to straight lines. This is taken as proof that in these four cases no bias due to selective corrosion is present. This result is not at all surprising, as differences in physical or chemical properties which could make rounded grains more susceptible to chemical attack than angular ones, or vice versa, were not observed.

Another interesting phenomenon revealed by Figs. 4(a) and 5(a) is that in sample 1(a) crystal faces are more abundant on the uraninite grains than in sample 1(b). As shown later on (Figs. 8 and 9; Table XIII) the size distributions of the uraninite fractions in these two samples are also quite different, in that sample 1(b) contains a larger percentage of coarse grains. If for arguments sake the placer hypothesis is accepted, this would indicate that in the uraninite as released from the source rocks, more of the finergrains were bounded by crystal faces, and the sorting according to size and shape had the effect of concentrating more grains bounded by crystal faces in sample 1(a) than in 1(b).

On the other hand, if the crystal faces are due to a process of recrystallization which took place before the grains were corroded, then the different percentages of grains with crystal faces may be due to different degrees of recrystallization.

/If...

If recrystallization is invoked, it must have preceeded the period of corrosion as crystal faces and rounded outlines were corroded to a similar degree.

However, as shown later (p. 60) a process of recrystallization <u>in situ</u> is regarded as very unlikely.

(d) Bias due to additional roundness populations

Roundness of grains can be ascribed to, or modified by a variety of processes. In Table X1 some of these are listed.

TABLE X1

PROCESSES WHICH MAY CAUSE, OR MODIFY, ROUNDNESS OF GRAINS

(a) Processes concurrent with the formation of the grains

(i) In crystals, round corners may be due to oscillatory development of crystal forms.

(ii) Grains formed from gels may show colloform outlines.

(b) Processes post-dating the formation of the grains

(i) Mechanical abrasion, i.e. the grinding action of wind and water.

(ii) Chemical attack.

(iii) Modification of roundness by recrystallization.

(iv) Modification of roundness by colloform overgrowths.

Cases doubtlessly exist where more than one type of rounded outline are present on grains, or where in a population of grains, different processes causing or modifying roundness were at work. Quantitative data determined on such populations will not reflect the effect of the major process only, but will be modified by the additional processes. This will be called bias due to additional roundness populations.

Pye (1943, footnote on p. 101) lists roundness as one of the properties of sand grains which are normally distributed. Pye's statement refers only to sediments, but it is not impossible that roundness data of particles which are not of sedimentary origin, also follow the normal distribution.

/However...

However, as the roundness scale of Wadell extends only from 0 to 1, Pye's statement should be slightly modified. It will be more correct to state that roundness data fit a normal curve, excepting at the lower and higher limits, where it is truncated. When plotted on probability paper, roundness data lie on a straight line, with only the lowest and highest values deviating.

In Figs. 6(a) and 7(a) the roundness data for monazite in samples 1(a) and 1(b) were plotted in this way. Roundness values of single corners were used, as the average value of a grain may mask extreme values of e.g. very angular or very round corners. It is evident that the roundness data of monazite fits the normal distribution well, except for the highest and lowest values.

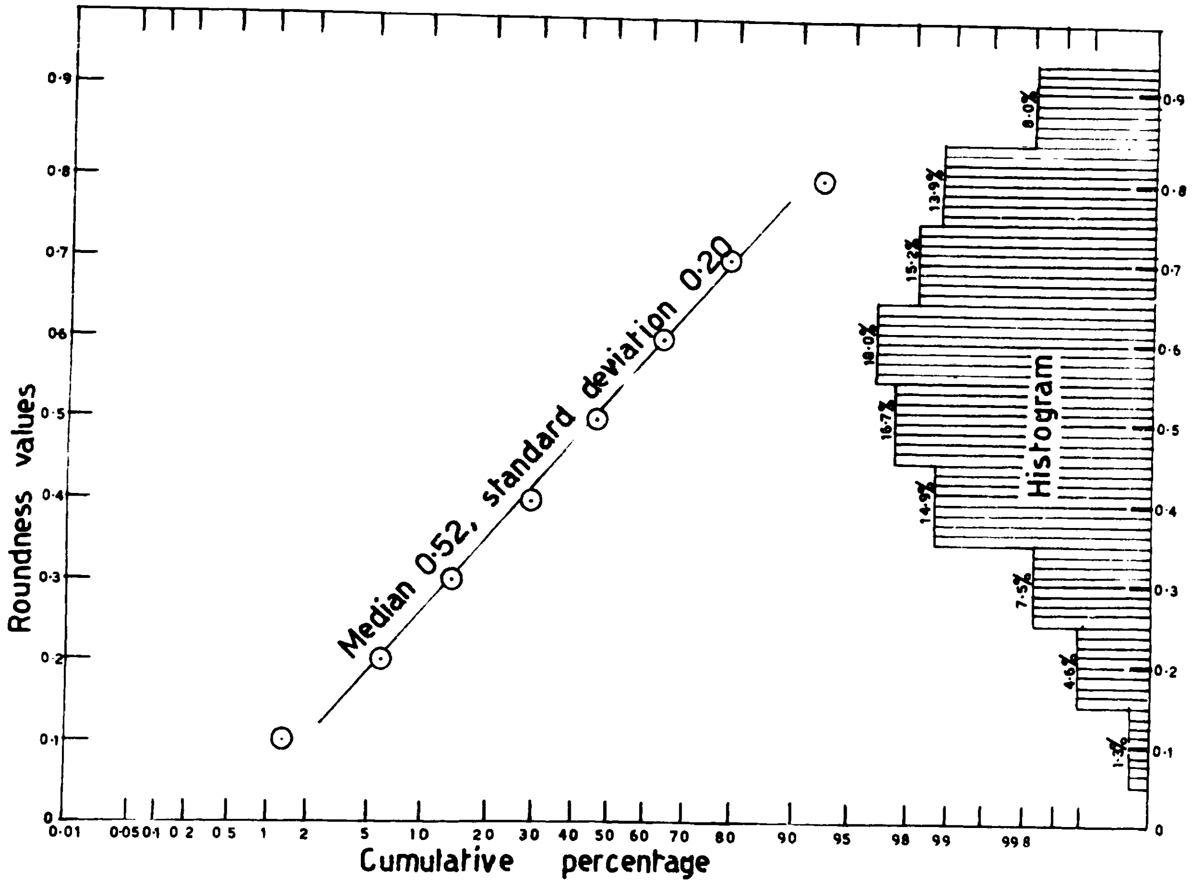
If a second population of different mean and/or standard deviation were present, this would cause a deviation in the straight line relationship. Therefore the monazite data show no bias due to additional roundness populations.

In Figs. 6(b) and 7(b) the roundness data of uraninite in samples 1(a) and 1(b) are shown as plotted on probability paper. The measured values (circles) do not lie on straight lines. The histograms shown on the right hand side of each diagram suggest the presence of two modes in each case.

The method of Harding (1949, p. 141) was accordingly applied, and it was found possible to resolve the data into two normal distributions in each case. They are shown as black lines on the diagrams, and are marked "population A" and "population B" respectively. The resultants of these populations (stippled lines) fit the measured data well. It is also possible to deduce the median roundness values and standard deviations of the two populations in each case.

In these diagrams roundness values measured on single corners were plotted. If roundness values of complete grains (i.e. the mean values of the corners of the grains) are plotted in a similar way, then population B becomes imperceptible. Population B is characteristic of certain corners of grains, therefore, and when mean values for grains are taken, their effect is masked.

/Amongst...



URANINITE, SAMPLE 1A.

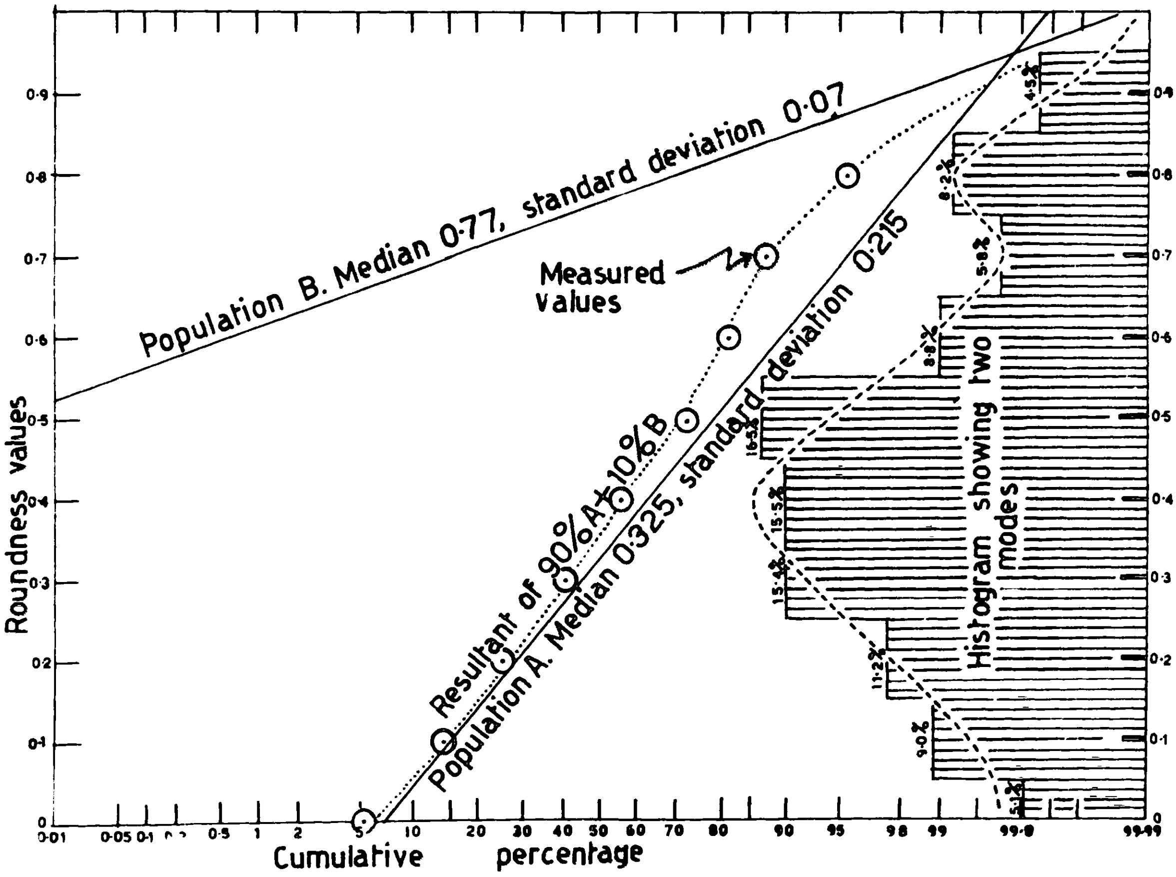
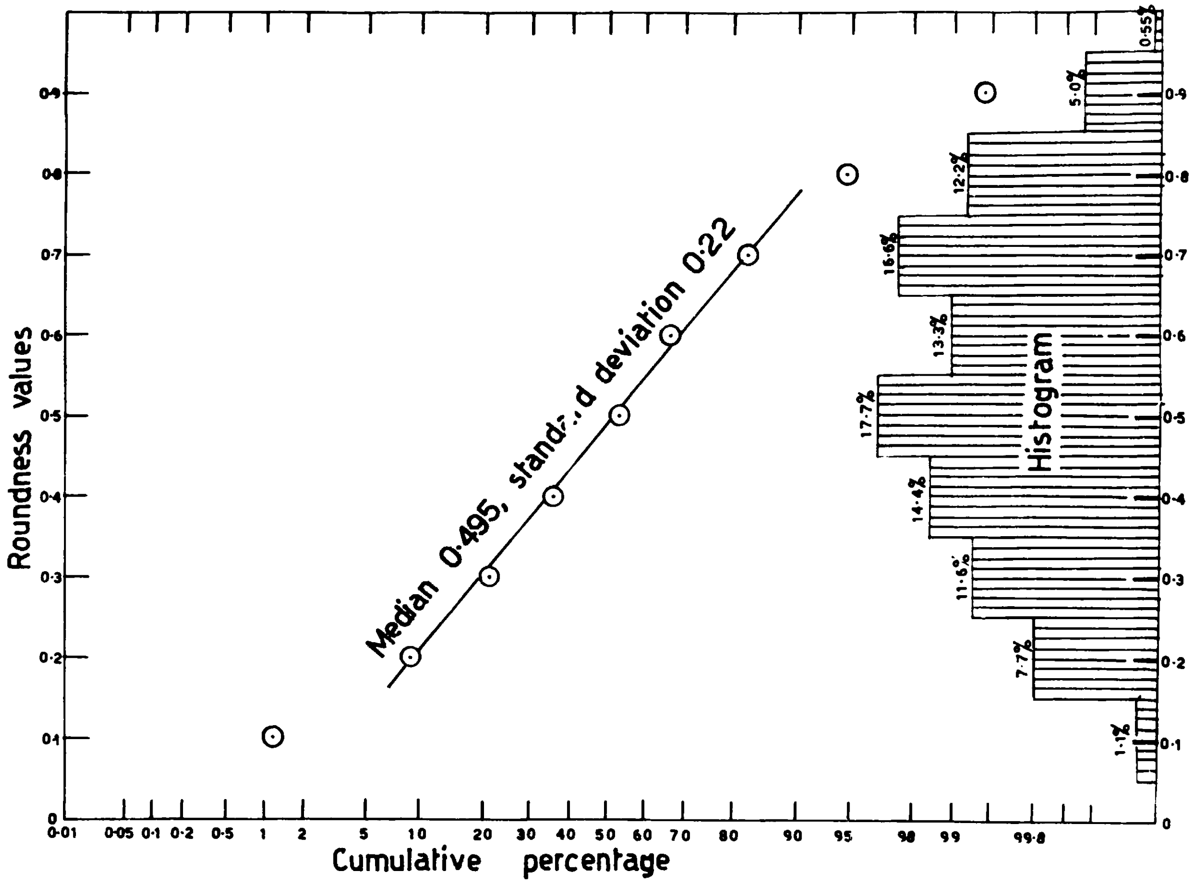


Figure 6. Roundness data of monazite and uraninite in sample 1(a) as plotted on probability paper

MONAZITE, SAMPLE 1B



URANINITE, SAMPLE 1B.

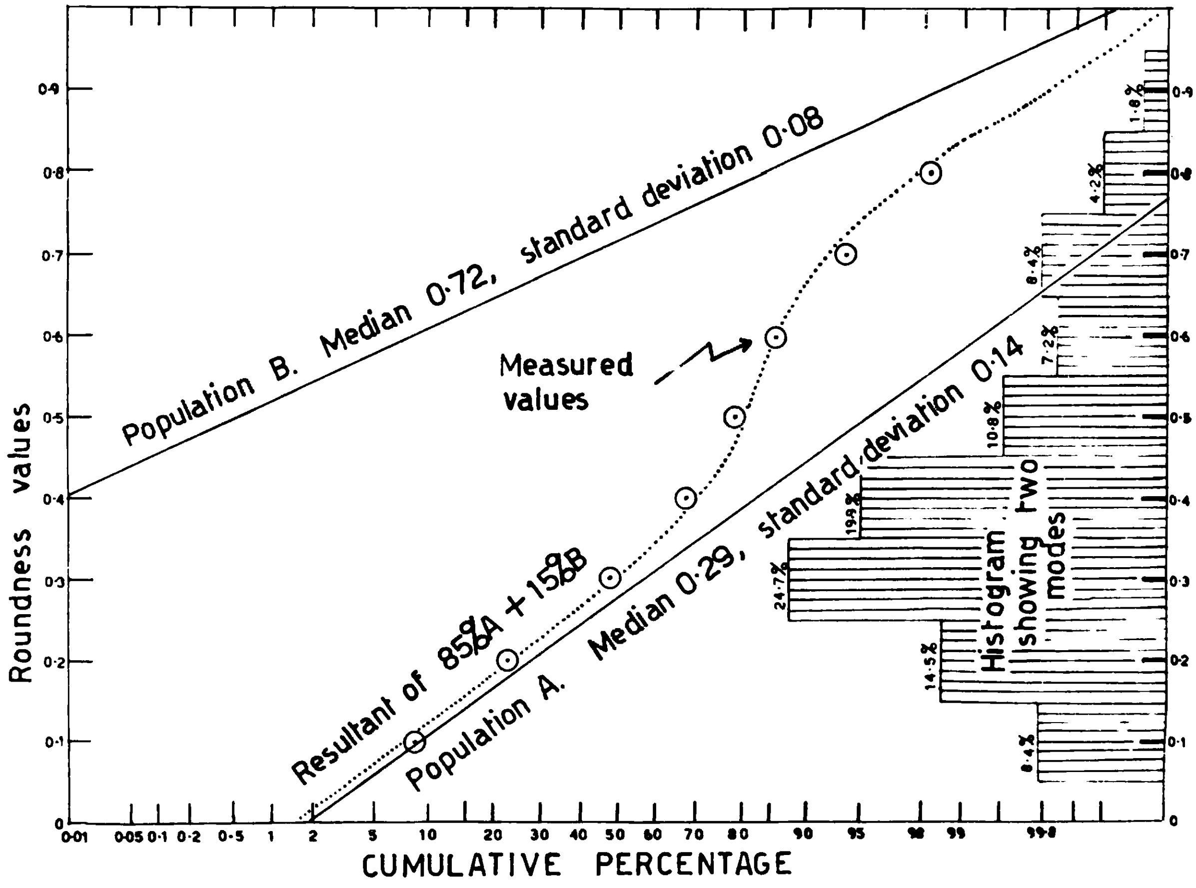


Figure 7. Roundness data of monazite and uraninite in sample 1(b) as plotted on probability paper

Amongst the possible causes of the rounded outlines listed in Table X, colloform overgrowth is regarded as the most plausible explanation. As shown previously (p. 8) certain microscopic features also suggest the presence of overgrowths.

The effect of the population B corners on the mean roundness values of uraninite listed in Table X11 was an increase of 0.05 to 0.07. In order to apply a correction, 0.05 to 0.07 should be deducted from the mean roundness values for uraninite in Table X11.

Grains of low roundness usually have more corners and protuberances than ones of high roundness. Therefore a median, or even a mean value, based on corners will be weighted towards the lower value. This weighting effect is eliminated if the mean roundness value for every grain is used.

The seeming discrepancy of lower median roundness values and larger standard deviations in Figures 6 and 7 when compared with the values listed in Table X11 is due to the fact that the former was based on individual angles, and the latter on grains.

V111. RESULTS AND INTERPRETATION

The results of roundness, form and size studies of heavy minerals are listed in Tables X11, X111 and X1V. These figures must be clearly understood to reflect upon the grains as they exist today. As shown in section V11 no bias due to selective corrosion is present, but the uraninite data may have a slight bias, of the order of 0.05 to 0.07 too high, which is due to an additional roundness population.

The standard deviation of the mean roundness and sphericity values listed can easily be evaluated by dividing the standard deviations of the populations by the square root of the number of grains measured. The standard deviations of the mean roundness value for uraninite in sample 1(a), e.g., can be given as ± 0.011, and for the mean sphericity value as ± 0.0072.

Examples of cumulative size distribution curves are given in text figures 8, 9 and 10.

(a) The degree of roundness of grains in the Dominion Reef

In order to interpret the roundness values given in Table X11 the following facts concerning rounding processes must be taken into account.

/Table X11...

TABLE X11

ROUNDNESS AND SPHERICITY VALUES FOR VARIOUS MINERAL SPECIES IN SAMPLES FROM THE UPPER REEF, DOMINION REEFS MINE

Sample No.	Uraninite					Monazite					Chromite					Leucoxene				
	Roundness		Sphericity			Roundness		Sphericity			Roundness		Sphericity			Roundness		Sphericity		
	M*	S*	M	S	N*	M	S	M	S	N	M	S	M	S	N	M	S	M	S	N
1(a)	0.45	0.17	0.76	0.088	150	0.60	0.15	0.78	0.082	160	Shattered		(0.78)	0.059	33	Forms destroyed				
1(b)	0.42	0.16	0.76	0.074	66	0.56	0.14	0.75	0.083	66	and									
2	0.38	0.16	0.77	0.045	33	0.50	0.14	0.78	0.070	33	replaced		grains	shattered		(0.4)		(0.72)	0.086	33
6	corroded		(0.75)	0.096	33	(0.4)	0.15	(0.75)	0.086	40	No roundness		(0.81)	0.075	33	Most grains not suitable				
16	Corroded		(0.79)	0.060	33						measurements		(0.80)	0.086	33					
451	0.36	0.16	0.79	0.062	54						possible		(0.77)	0.065	33					

Sample No.	Zircon					Garnet					Cassiterite					Columbite				
	Roundness		Sphericity			Roundness		Sphericity			Roundness		Sphericity			Roundness		Sphericity		
	M	S	M	S	N	M	S	M	S	N	M	S	M	S	N	M	S	M	S	N
1a	0.32	0.15	0.82	0.091	50	(0.3)		(0.79)	0.066	33	(0.26)	0.08	0.76	0.11	33					
1b			0.75	0.075	33	(0.3)		(0.77)	0.067	33	(0.2)	0.10	0.79	0.12	33					
2	0.31	0.21	0.80	0.064	33	(0.4)		(0.83)	0.045	33								0.74	0.060	33
6	(0.31)	0.12	0.77	0.075	33			(0.81)	0.055	33								(0.76)	0.065	33
16	(0.32)	0.12	0.78	0.071	33			(0.78)	0.12	33								0.77	0.070	33
451	0.24	0.12	0.76	0.086	50	(0.7)		(0.82)	0.18	33								0.76	0.076	33

* M: arithmetic mean S: standard deviation N: number of grains measured
Values appearing in brackets are considered to be less accurate than the unbracketed ones due to a high degree of corrosion or cataclasis.

TABLE X111

SIZE DISTRIBUTION DATA (IN phi UNITS) FOR VARIOUS MINERAL SPECIES IN SAMPLES FROM THE DOMINION REEF

Sample No.	Uraninite			Monazite			Chromite			Leucoxene			Zircon			Garnet			Cassiterite			Columbite		
	M	(25)	(75)*	M	(25)	(75)	M	(25)	(75)	M	(25)	(75)	M	(25)	(75)	M	(25)	(75)	M	(25)	(75)	M	(25)	(75)
1(a)	3.23	2.86	3.75	3.20	2.82	3.61	3.65	3.14	4.26	3.48	3.00	4.02	3.30	2.96	3.69	(0.60	0.20	1.40)	2.86	2.17	3.30		-	
1(b)	3.04	2.63	3.41	2.76	2.37	3.10		-		2.10	1.79	2.60	(3.38	2.90	3.84)	(0.98	0.70	1.50)	2.78	2.22	3.34		-	
2	3.15	2.48	3.66	2.75	2.30	3.07		-		2.37	1.93	2.80	3.28	2.93	3.74	(0.64	0.00	1.24)		-		2.50	2.12	2.85
6	3.96	3.65	4.25	3.32	2.96	3.68	3.48	3.26	3.70		-		3.40	3.06	3.75		-			-		(3.14	2.74	3.54)
16	3.55	3.06	3.82		-		3.36	2.88	3.86		-		3.70	3.56	4.26	(3.10	2.46	4.20)		-		(3.37	3.06	3.70)
451	3.36	3.00	3.78		-		3.76	3.28	4.26		-		3.98	3.56	4.40	(0.57	0.47	1.30)		-		(2.58	2.08	3.04)

* M is the median size,(25) is the first quartile,(75) is the last quartile

Values in brackets are not as precise as the unbracketed ones.

The data is based on the measurement of between 100 and 200 grains in each case.

TABLE XlV

MEDIAN GRAIN SIZES* IN MM. FOR VARIOUS MINERAL SPECIES IN SAMPLES FROM THE UPPER REEF, DOMINION REEFS MINE

Sample No.	Uraninite	Monazite	Chromite	Leucoxene	Zircon	Garnet	Cassiterite	Columbite
1(a)	0.107	0.109	0.080	0.090	0.102	(0.660)	0.138	-
1(b)	0.121	0.148	-	0.233	(0.096)	(0.507)	(0.146)	-
2	0.113	0.149	-	0.194	0.103	0.642	-	(0.177)
6	0.064	0.100	0.090	-	0.095	-	-	(0.113)
16	0.085	-	0.097	-	0.077	(0.117)	-	(0.097)
451	0.097	-	0.074	-	0.063	(0.674)	-	(0.167)

* Values in brackets are not as precise as the unbracketed ones.

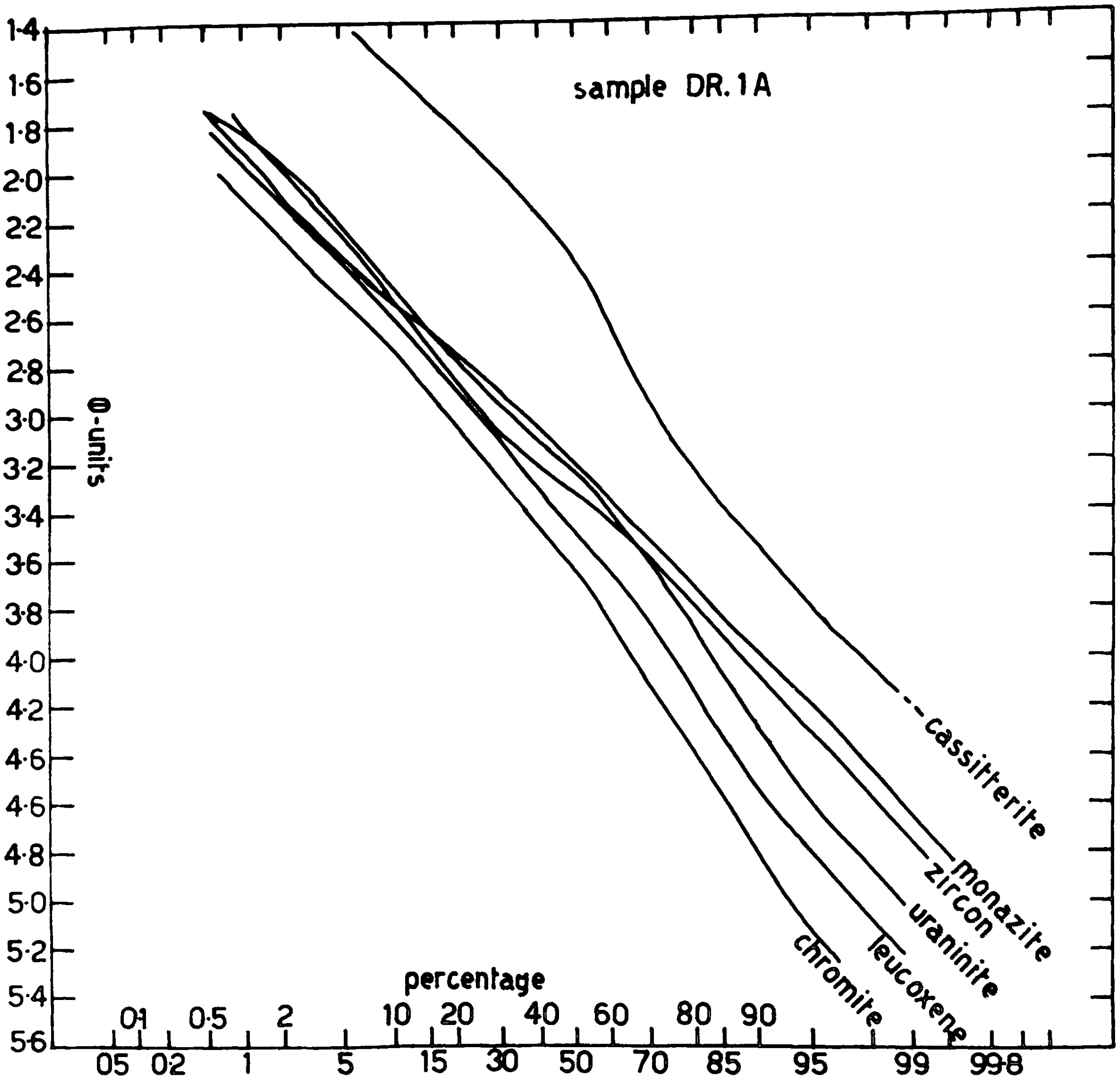


Figure 8. Grain size distributions of various minerals in sample 1(a)

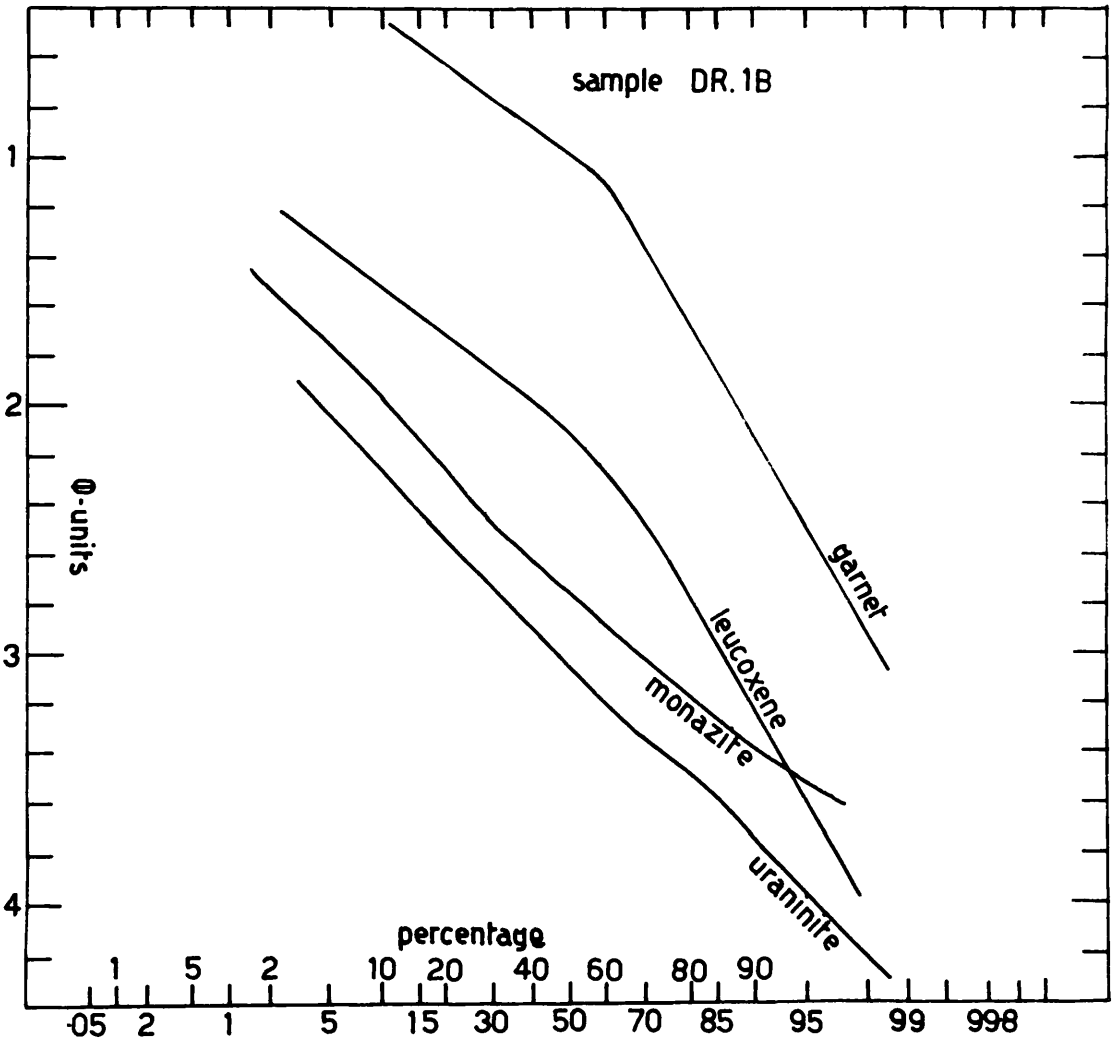


Figure 9. Grain size distributions of various minerals in sample 1(b)

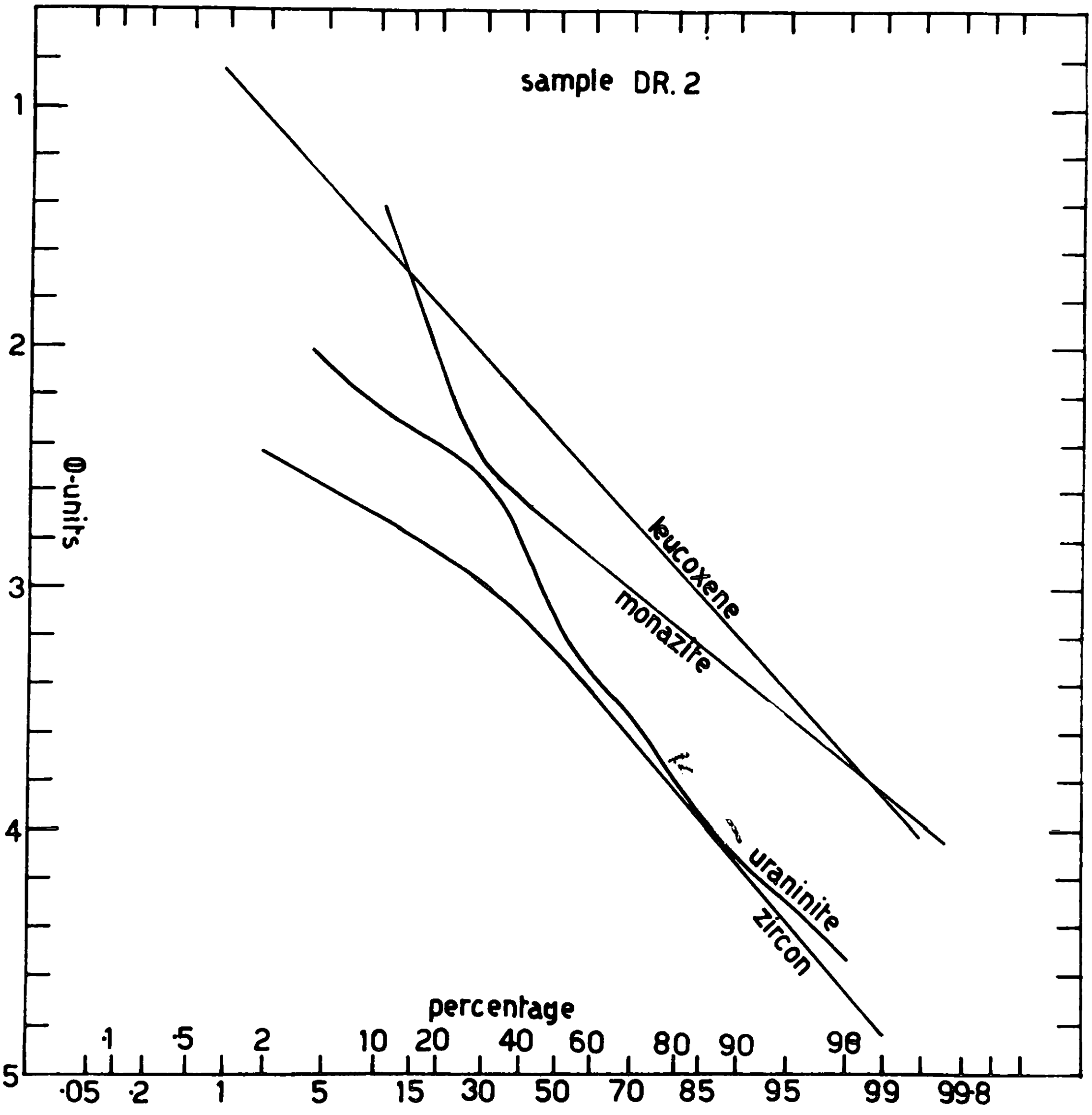


Figure 10. Grain size distributions of various minerals in sample 2

1. If rounding were caused by mechanical action (for instance the abrasive action of wind or water) the larger grains of a particular mineral would have been rounded at a faster rate than the smaller ones. However, if rounding is caused by chemical action, the smaller grains will be attacked at a faster rate than the bigger ones.
(Kuenen, 1955, p. 142; 1956, p. 336; 1958, p. 47; 1959(a),p. 172 and 1959(b) p. 14;Twenhofel, 1945,p. 59; Galloway, 1919, p. 270; Knight, 1924, p. 107; McCarthey, 1933, p. 205).

It was accordingly decided to ascertain whether the coarser uraninite grains are rounder than the finer ones.
In Table XV the degrees of roundness are shown for the two halves of the uraninite size distribution in samples 1(a) and 1(b).

TABLE XV

COMPARISON OF ROUNDNESS VALUES OF COARSE HALVES AND OF FINE HALVES OF GRAIN-SIZE DISTRIBUTIONS

Sample No.	Median Grain Size	Roundness of Coarse Half	Roundness of Fine Half
1(a)	0.107 mm.	0.36	0.32
1(b)	0.121 mm.	0.39	0.32

The differences in roundness values between the coarse and fine halves of the grain size distributions listed in Table XV are significant at the 95% confidence level. The results of this test indicate that the differences in roundness between coarse and fine uraninite grains in samples 1(a) and 1(b), although small, are consistent with the theory that rounding was caused mechanically. However, similar tests carried out on the monazite fractions failed to reveal significant differences between the coarse and fine fractions.

/2. Certain...

2. Certain mineral species are more resistant to wear than others. This may be ascribed to differences in hardness, brittleness, elasticity, cleavages and flaws. It was shown for instance that monazite wears away at a faster rate than uraninite in an attrition cell containing water (Koen, 1958, p. 183). This was ascribed to the greater hardness of uraninite as compared with that of monazite.

However, since he published this paper, Koen (personal communication) has come to the conclusion that the results of his experiment cannot be quantitatively true for abrading conditions in natural waters. The main reason is that the relative rates of diminution are highly dependant on the pulp density in the attrition cell, which, in Koen's tests was arbitrarily fixed at 50%, and which differs from conditions in natural waters.

In their work on comminution kinetics Fuerstenau et al. (1963, p. 5) showed that the initial rate of formation of material finer than a given grain size is a constant, which is characteristic of the material, the size being considered, and the comminution method. If the process of mechanical rounding is considered as a method of comminution, different relative rounding rates can be expected for the same minerals under different conditions.

The abrasion of quartz sand formed the subject of a detailed study by Kuenen (1959, p. 172). He found it difficult to ascribe the roundness of most natural sands to the action of water alone. During its journey from the land to the sea the chances are that detrital particles are exposed to the action of the wind, and since abrasion by wind proceeds at a rate many times larger than that in water, the major amount of abrasion could be ascribed to wind action. Even on present-day shores wind is an important distributing and rounding agent for sand grains. During Precambrian times, before the advent of land plants, the landscape most probably had a desert-like aspect, and wind abrasion could have played a more important role than today.

In an attempt to test the relative roundnesses shown today by different heavy mineral species against the theory of mechanical abrasion, due consideration must therefore be given to the possibility of action by both wind and water.

/The relative...

The relative rates of rounding under such extremely different conditions may well be very different. Experimental data are completely lacking.

It was decided therefore to test the relative rates of rounding of monazite and uraninite.

Aeolian abrasion was simulated by introducing similarly sized monazite and uraninite grains, obtained by crushing and sieving large pieces of the mineral, into a funnel-shaped glass vessel, and adding an excess of sand grains. The vessel was closed at the top by means of fine wire mesh. A stream of air was blown upwards through the stem. This carried the mineral particles upwards to the top of the vessel, after which they started to fall back. During the period of fall many particles collided with rising ones, and this was the main cause of attrition. The very fine dust formed by this process escaped through the wire mesh. At intervals samples were taken, and the roundness measured.

Abrasion was also affected in a water cell, using a stirrer which rotated at 3,500 revolutions per minute. The cell was charged with a mixture consisting of 0.6 cc. uraninite grains, 0.6 cc. monazite grains, 9 cc. quartz sand and 150 cc. water.

Results of these experiments are listed in Table XVl. The large difference in relative rounding rates in the first two experiments listed, in which uraninite from Shinkolobwe was used, may be ascribed to the brittleness of the uraninite. This uraninite, although it still gave a weak x-ray pattern corresponding to UO_2, has been completely oxidised to U_3O_8, as proved by chemical analysis. The wind-attrition was caused mainly by impact, and the rounded grains attained a frosted appearance. Under such conditions a brittle material can be expected to wear away faster than material of the same hardness which is not brittle. In the water cell grinding and rubbing played the major role in the abrasion of the grains which attained a smooth appearance. During these conditions brittleness could not play such an important role.

The Vickers hardness numbers measured in sample 1(a) are as follows:

Zircon	1380
Uraninite	660
Monazite	770

/The hardness...

TABLE XV1

RESULTS OF ARTIFICIAL ATTRITION ON URANINITE AND MONAZITE

Rounding Agent	Mineral	Vickers * Hardness	Roundness attained
Water abrasion for 32.45 hours	Uraninite from Shinkolobwe	610	0.46
	Monazite from Houtenbek	800	0.45
Wind abrasion for 6 hours	Uraninite from Shinkolobwe	610	0.56
	Monazite from Houtenbek	800	0.43
Wind abrasion for 12 hours	Uraninite from Great Bear Lake	1000	0.38
	Monazite from Houtenbek	800	0.46
Wind abrasion for 10 hours	Uranothorianite from Phalaborwa	1110	0.42
	Monazite from Houtenbek	800	0.45
Wind abrasion for 14 hours	Uranothorianite from Phalaborwa	1110	0.45
	Monazite from Houtenbek	800	0.54

* Vickers hardness numbers determined by means of a Leitz Miniload hardness tester, using a load of 15 gm. for the indentations on monazite, and 50 gm. for uraninite, uranothorianite and zircon. The results were rounded off to significant figures only.

The hardness value obtained for uraninite is surprisingly low, and certainly does not reflect the original hardness of this mineral. The low value may be due to the abundant inclusions of galena, and the presence of small cracks in the uraninite grains.

Although the results listed in Table XV1 do not permit the prediction of the exact rounding rates to be expected in nature, one can state with confidence that monazite can be expected to attain a higher roundness than unweathered uraninite, and zircon according to its hardness, a lower roundness, provided that they are of roughly similar sizes, that they were abraded in the same physical environment, and that, when they were supplied to the sediments, they had roughly similar low roundnesses. After weathering and oxidation the hardness of uraninite can decrease appreciably to values lower than that of monazite. Such weathered uraninite may be rounded at a higher rate than monazite.

The relative roundnesses of uraninite, monazite and zircon as shown in Table X11 are in accord with the results obtained from artificial attrition experiments and hardness measurements.

The prima facie interpretation of the relative roundnesses of monazite, uraninite and zircon as they stand in Table X11 therefore, is that the roundnesses are the result of mechanical attrition which affected the three mineral species at the same time.

A further possibility to be considered is that uraninite was deposited as poorly crystalline nodules in ancient marshes, and that they were later reworked and deposited as detrital grains by wave action (Koen, 1961, p. 28). An extensive search was conducted in the 129 polished sections available, and in only one instance was a cluster of nodules observed (Plate X1, Fig. 3). The rarity of undoubted nodules makes it necessary to postulate processes of metamorphism whereby
(i) the original clusters of nodules were practically eliminated, and
(ii) replaced by recrystallized grains, in many cases having the appearance of partly rounded crystals, and exhibiting a degree of roundness fortuitously in accordance with a theory of mechanical abrasion.

But what is the evidence that can be advanced in support of such a theory?

(i) The elimination of nodular grains. It would be extremely difficult to prove that all the grains originally present in the conglomerate were nodules, which were subsequently eliminated. The rare nodules actually present may represent original detrital nodules as previously pointed out on page 8.

/They...

They could have been formed also in ancient marshes as claimed by Koen. They could have been deposited in the sediments before lithification by groundwater, and finally, they could have been formed in the rocks after lithification during a process of metamorphism, during which portions of uraninite grains could have been taken into solution and redeposited closeby. The mere presence of rare clusters of nodules can therefore not be cited as evidence that all the grains originally present were nodules.

Even if all the original grains were nodules,.then their elimination by processes of corrosion should have left abundant evidence as to their former existence. The grain shown in Plate III, Fig. 1, for instance, has been partially corroded, yet the traces of the original outlines are still discernible. If a large number of nodules were eliminated in this way without leaving traces of their former existence, then severe processes of metamorphism and recrystallization have to be invoked.

(ii) Recrystallization. Four photomicrographs were published by Koen (1961, Plate 1, Figs. 3,4,5 and 6) in which he interprets the presence of crystal faces as being due to recrystallization. As pointed out by Davidson (1961, p. 47) "...the statement that euhedral or subhedral cubic crystals of uraninite are due to recrystallization of detrital spheroidal particles in situ is no more than an opinion ..." A study of Koen's photographs reveals that more than one interpretation is possible for the grains pictured. The morphology of the grains shown by him in Figures 3 and 6 may simply be due to corrosion. In Figures 4 and 5 the nodular portions may be the later overgrowths, and not vice versa, as pointed out previously (p. 8). Moreover, the shape of the grains may be that of original detrital grains. Ramdohr (1960, p. 972) mentions the presence of both crystals and colloform uraninite in the same veins.

The nodular forms of uraninite usually deposited in hydrothermal veins and by groundwater actually are cryptocrystalline, the crystallite size being dependant on the temperature of formation (Miller, 1958,p.530). If such nodules are recrystallized, many nuclei will be available, and the crystallite size will increase until an aggregate of small grains are formed. An example of a similar process is afforded by amygdales of chalcedony which recrystallize to quartz aggregates.

/Outgrowths...

Outgrowths on such crystallographic discontinuous grains forming an aggregate would yield, if anything, small crystal faces and not straight edges to the entire aggregate. It seems theoretically impossible for a nodule having a number of nuclei to form on recrystallization a single crystal especially under conditions of low grade metamorphism.

A further possibility is the formation of idioblasts, independantly from the nodules. In this case textures can be expected somewhat similar to those of the iron ores of the Pretoria Series. In these ores perfect idioblasts of magnetite are present in an environment of poorly crystallized nodules (ooliths) of iron oxides (Schweigart, 1965, Figs. 2 and 5).

The tendency of uraninite to form idioblasts will be stronger or weaker than that of other minerals also present within the Dominion Reef depending on its position in the idioblastic series of minerals (Harker, 1950, p. 39 and 199; Ramberg, 1952, p. 131: Turner and Verhoogen, 1960, p. 594).

In arenaceous rocks the oxide minerals rutile, haematite, ilmenite and magnetite are listed high up in the idioblastic series. Unfortunately no empirical data could be found on the position of uraninite in the idioblastic series, and one must resort to indirect evidence, such as that afforded by the ionic packing index as defined by Fairbairn (1943, p. 1310). He studied the correlation of packing index with morphological development of porphyroblasts, and came to the following conclusion: "It is seen that a general decrease in packing and bonding indices coincides with decrease in ability to develop crystal faces. The agreement is imperfect in detail, but little more could be expected in view of the unavoidable lack of precision in arranging a series of this kind." (Fairbairn, 1943, p. 1369).

The packing indices of the oxide minerals quoted above are as follows: Rutile, 6.6; haematite, 6.6; ilmenite, 6.4; and magnetite, 5.9. These values were obtained from Fairbairn's list (1943, p. 1371). Assuming ionic radii of 1.32Å for oxygen and 1.05 Å for uranium (Bragg, p. 114) and a unit cell size of $a_o = 5.46$Å (Palache et al. p. 610) the packing index of uraninite was calculated and found to be 5.9. The indications of the packing index therefore are that uraninite cannot be expected to show a greater ability to develop idioblasts than rutile or ilmenite for instance.

/If anything...

If anything, its ability should be lower. In the Dominion Reef both rutile and ilmenite were formed by the recrystallization of leucoxene. The recrystallised rutile has little tendency to form crystal faces in the same samples where uraninite crystals were observed (Plate VIII, Fig. 2, and Plate II, Figs. 1 to 6). In samples 15 and 16 recrystallized ilmenite has a tendency to develop crystal faces (Plate VIII, Fig. 6). However, in these same samples uraninite does not show crystal faces, but corroded reaction rims (Plate III, Figs. 3 and 4).

It would appear therefore that the assumption that uraninite can recrystallize in the same environment in which rutile remains xenoblastic must be regarded as unfounded.

(b) The Sphericity of Grains in the Dominion Reef

A study of Table X11 shows that the sphericity values of all the grains suitable for measurement are between 0.72 and 0.83.

Sphericity values for uraninite and monazite correspond closely.

Zircon has a surprisingly high sphericity.

(c) The Grain Size of Heavy Minerals in the Dominion Reef

In Table X111 the grain size of the heavy minerals which were suitable for measurement are listed. In some cases the mineral was altered and corroded to such an extent that the grain size measured on it must be regarded as of low accuracy. In other cases only some twenty or thirty grains suitable for measurement were encountered, and the precision of the mean value listed is fairly low. Such values which are somewhat suspect are nevertheless included in the table, but the figures are given in brackets.

Various workers have given attention to the size relationships of associated heavy minerals in detrital sediments. Rubey (1933, p. 3) assumed that settling velocity was the important factor which determined the relative sizes of associated heavy minerals. He deduced hypothetical size distributions for minerals of different specific gravities, and described the principle involved as "hydraulic equivalence".

Rittenhouse (1943 (b), p. 1725) attributed variations in size distributions to varying hydraulic conditions, variations in the hydraulic equivalence, relative availability of the different size grades and other factors.

/He...

He ascribed the formation of heavy mineral concentrations to the selective removal of light minerals from material newly arriving in a river.

McIntyre (1959, p. 278) made use of the principles proposed by Rittenhouse to explain deviations of empirically determined mean sizes in three sedimentary layers on a beach from expected values.

Koen (1961, p. 35, 36, 37) gave eleven examples from the Dominion Reef, the Carbon Leader, the Basal Reef, the Vaal Reef and the Kimberley Reef where the sizes actually measured on uraninite, zircon, chromite, and in some samples quartz, correspond closely to values expected from the theory of hydraulic equivalence. Koen preferred samples in which particles of uraninite were more or less homogeneously distributed throughout the ground mass. He did not use sections showing clusters of numerous uraninite grains.

In order to test the relative sizes of the associated heavy minerals listed in Table Xlll for hydraulic equivalence, use was made of the formula developed by McIntyre. This formula states that

$$\phi_A - \phi_B = \frac{1}{X} (\log_2 D_A - \log_2 D_B)$$

where ϕ_A and ϕ_B are the grain sizes in phi units for mineral A and mineral B respectively, D_A and D_B are the respective densities from which 1.0, the density of water, was subtracted, and X is a factor depending on the grain size of the particles. For grains below 0.1 mm. (i.e. above 3.3 phi units) Stokes' law will be valid, and X equals 2. Rubey's Impact law applies to grains larger than 1 mm. and for these X is equal to 1. For grains between 0.1 mm.and 1 mm. in size (between 3.3 and 0 phi units) McIntyre used a formula which he calls "the linear approximation hydraulic equivalence formula". Grains in this size range have settling velocities somewhere in between values as calculated by Stokes' law and the Impact law. For these grains McIntyre used a value of 1.5 for X.

In this paper a graphical method, based on McIntyres formula, is used as an aid to visualize grain size relationships, and to compare them with with theoretical values calculated from hydraulic equivalence principles.

/For...

For this purpose the logarithms to the base 2 of D is calculated for each mineral, where D is the density from which 1 was subtracted. These values as well as the values calculated for $X = 1.5$ and $X = 2$, are given in Table XV11.

From these values a diagram is constructed with $1/X$ on the horizontal scale, and $\frac{1}{X}\log_2 D$ for the various minerals on the vertical scale. Points for each mineral species will lie on a straight line, as shown in Figures 11 and 12. The distances between these lines represent differences between ϕ values in MacIntyre's formula for all values of X between 1 and 2.

TABLE XV11

DENSITIES AND DATA FOR CONSTRUCTING HYDRAULIC EQUIVALENCE DIAGRAMS

Mineral	Density	D	$\log_2 D$	$\frac{1}{1.5}\log_2 D$	$\frac{1}{2}\log_2 D$
Uraninite	9.0	8.0	3.000	2.000	1.500
Cassiterite	7.0	6.0	2.585	1.723	1.293
Columbite	6.0	5.0	2.322	1.548	1.161
Monazite	5.1	4.1	2.036	1.357	1.018
Zircon Ilmenite Chromite	4.7	3.7	1.888	1.259	0.944
Garnet	4.2	3.2	1.678	1.119	0.839

To test for hydraulic equivalence, mean or median sizes are plotted on a strip of paper, and this paper laid vertically on the diagram at the appropriate value of $1/X$, when it will be immediately apparent whether the empirical values obtained correspond with the theoretical ones on the graph. For grains below 0.1 mm. the right hand side, where X is equal to 2 is used. For grains larger than 1 mm. the left hand side, where X is equal to 1 is used. For grains between these sizes the values of X between 1 and 2 should be used.

/In...

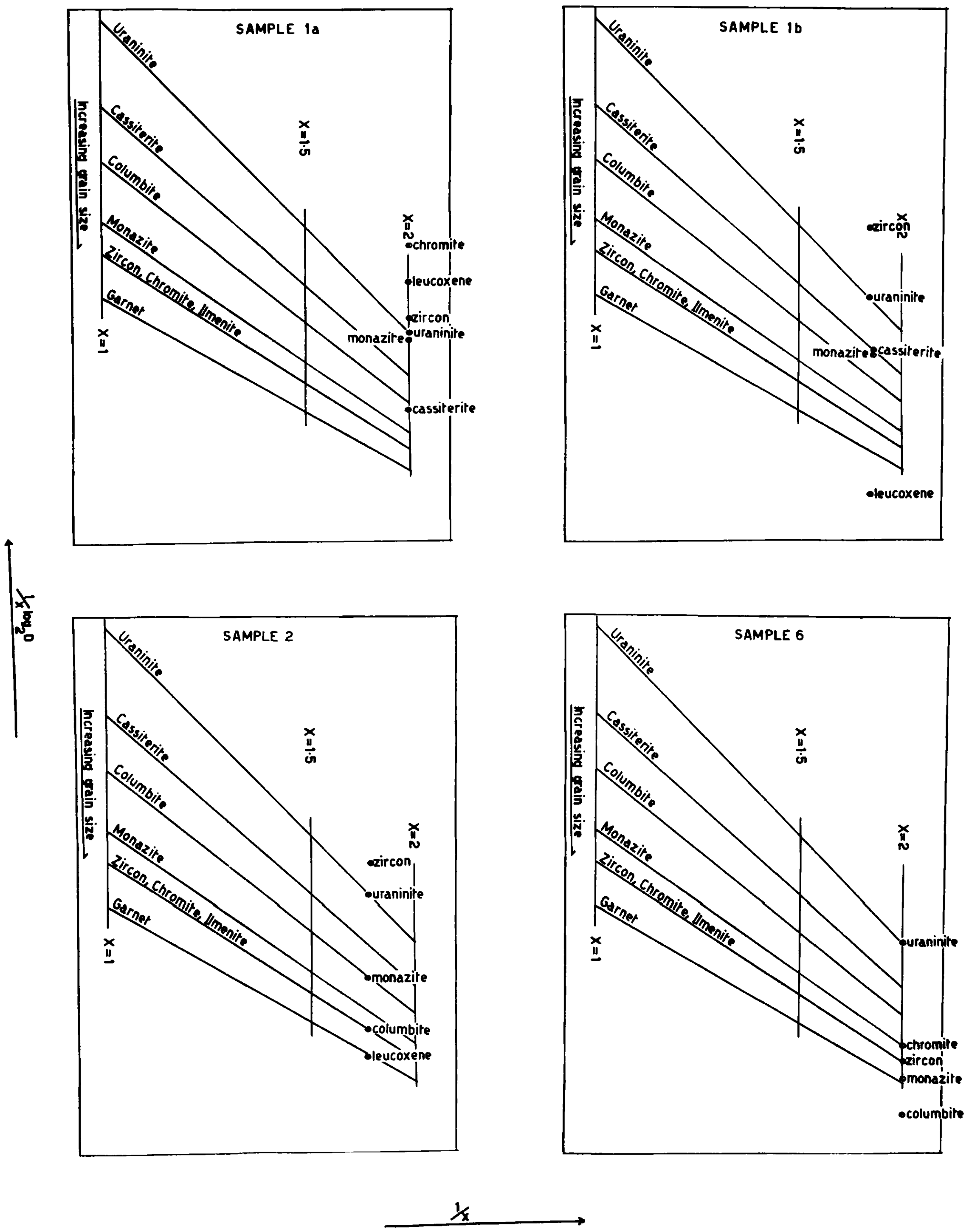


Figure 11. Tests for hydraulic equivalence in samples from the Dominion Reefs Mine

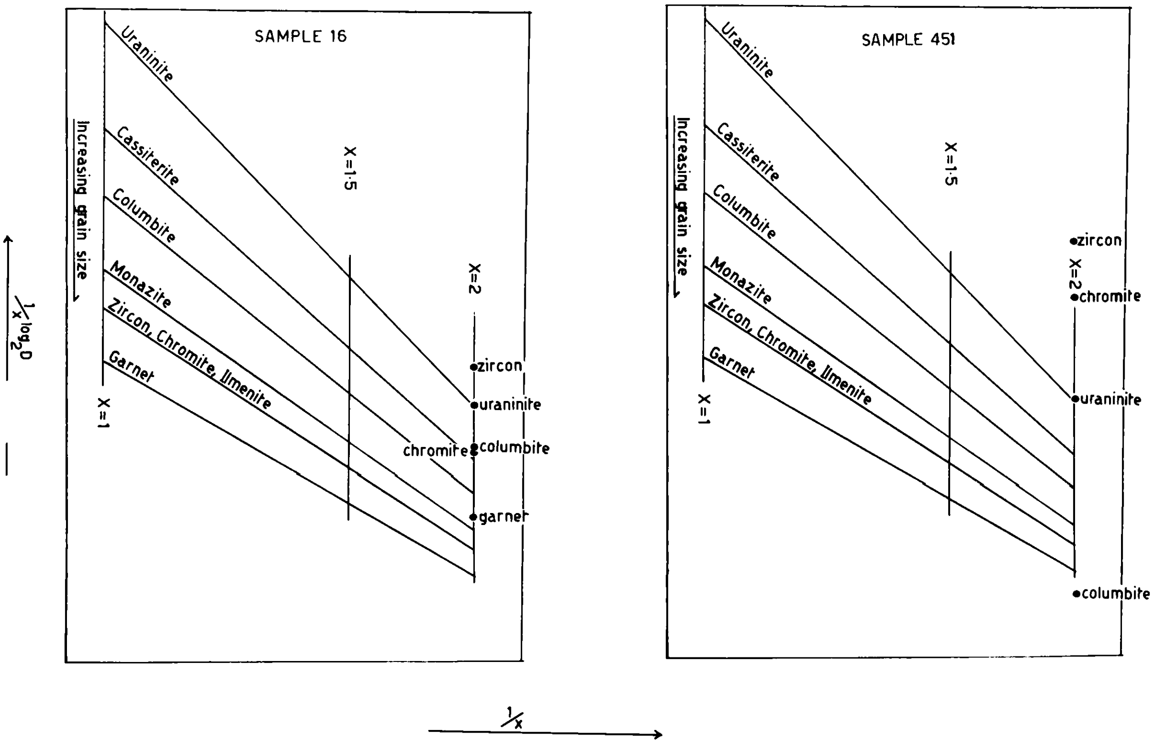


Figure 12. Tests for hydraulic equivalence in samples from the Dominion Reefs Mine

TABLE XV111

GRAIN SIZE RELATIONSHIPS OF MINERALS IN THE DOMINION REEF AS COMPARED WITH HYDRAULIC EQUIVALENT VALUES BASED ON URANINITE

Sample No.	Zircon	Chromite	Leucoxene	Monazite	Cassiterite	Garnet	Columbite
1a	Too fine	Much too fine	Much too fine	Too fine	Too coarse	Much too coarse	-
1b	Much too fine	-	Much too coarse	Too fine	As expected	Much too coarse	-
2	Too fine	-	Much too coarse	Too fine	-	Much too coarse	Too coarse
6	As expected	As expected	-	As expected, but slightly too coarse	-	-	Too coarse
16	Too fine	Too fine	-	-	-	Too fine	Too fine
451	Too fine	Much too fine	-	-	-	Much too coarse	Much too coarse

In Figs. 11 and 12 points are shown as obtained for samples 1(a), 1(b), 2, 6, 16 and 451. The point for uraninite is matched with the uraninite line in each case, as the object is to test whether hydraulic equivalence between uraninite and other mineral species was attained. From these figures the following relationships were deduced:-

Sample 1(a). The median grain sizes are all quite close to, or larger than, a value of 3.3 phi units, with the exception of cassiterite and garnet, which are coarser. Comparison is made on the right hand side of the hydraulic equivalence diagram where X is 2. Compared with uraninite, garnet and cassiterite are much coarser than expected. The point for garnet at 0.60 phi units is not shown, as it falls outside the diagram at the bottom.

Monazite, zircon, leucoxene and chromite are all much finer grained than expected. The median values for monazite and zircon are very close to that for uraninite.

Sample 1(b). Comparison of sizes is made near the right hand side of the hydraulic equivalence diagram. In this diagram the median size of cassiterite corresponds closely with its expected value. Zircon and monazite are finer than expected, and garnet and leucoxene coarser than expected. (The point for garnet falls outside the limits of the diagram).

Sample 2. Much the same relationships as 1(a) and 1(b) ~~is~~ are shown, with monazite and zircon too fine, and garnet, leucoxene and columbite too coarse.

Sample 6. Zircon and chromite have the expected grain sizes within the limits of experimental error. Monazite is only slightly coarser than expected. Only columbite, which is much too coarse, deviates considerably from its expected value.

Sample 16. Zircon, chromite, columbite and garnet are all finer than expected compared with uraninite. However, the relationship between columbite and garnet closely corresponds to the requirements of the hydraulic equivalence principles.

Sample 451. Zircon and chromite are once more finer, and columbite and garnet coarser than expected.

The relationships for the samples examined are summarised in Table XV111.

/Size...

(d) Size Distributions

Examples of size distributions determined in polished sections for various mineral species are given in Figs. 8, 9 and 10. These figures were plotted in such a way that low phi values appear at the top and high phi values at the bottom. The relationships revealed in these figures are as follows:-

Fig. 8 (Sample 1(a)). A family of closely similar cumulative curves are shown for uraninite, leucoxene, chromite, zircon and monazite. The curve for cassiterite is of a different form, and displaced in the direction of the coarser grain size. Garnet is not shown, but most of its cumulative curve falls outside the diagram at the top.

Fig. 9 (Sample 1(b)). The cumulative curves for uraninite and monazite are fairly similar and lie close to each other. Those for leucoxene and garnet are of a different form, and are displaced in the direction of coarser grain sizes.

Fig. 10 (Sample (2)). The curves are dissimilar, and no particular relationship is revealed.

A few types of cumulative curves can be recognized:

1. The distribution is lognormal or nearly lognormal. The best example of a lognormal distribution in the figures above is the one for leucoxene in sample 2. This size distribution plots as a perfectly straight line on probability paper. The distribution for uraninite in sample 1(b) approaches lognormality quite closely.

2. The majority of the size distribution curves trend gently upwards or downwards, and show slight secondary undulations. If the method of Harding (1949, p. 141) is applied to such curves they can usually be resolved into two normal distributions, the sums of which correspond closely with the observed cumulative frequencies. However, they can also be regarded as the differences of two normal grain-size distributions.

3. Examples are also shown of curves of grain-size distribution which can be regarded as consisting of two straight lines of different slopes joined together in a curve. This type of distribution is shown by garnet and leucoxene in sample 1(b), and by monazite in sample 2.

/When...

When analysed according to the method of Harding they do not yield satisfactory solutions. They can therefore not be regarded as sums of, or differences between, lognormal distributions. The garnet and leucoxene curves of sample 1(b) both show deficiencies of coarse material, and the monazite of sample 2 has an excess of coarse material.

IX. DISCUSSION

The results of roundness, size and shape determinations show that the minerals studied can be divided into two distinct groups.

The first group, consisting of monazite, zircon and chromite is usually finer-grained than would be expected from hydraulic equivalence considerations based on the size of associated uraninite grains. Their mean roundness values, as far as they could be determined, are in accord with one another and with that of uraninite, assuming that they were mechanically abraded under identical conditions.

Their cumulative size distributions are similar in most of the samples examined.

The second group, consisting of garnet, cassiterite, and columbite, is usually coarser-grained than anticipated when compared with uraninite on a basis of hydraulic equivalence principles. Taking their sizes into account their mean roundness values are usually much lower than anticipated. Cumulative size distributions of the members of this group of minerals are usually not similar to each other nor to those of the first group.

The mean sphericity values of all the minerals studied fall between narrow limits. The sphericity values of uraninite and monazite correspond closely.

In only one sample was hydraulic equivalence between mineral species evidently attained.

The theory of hydraulic equivalence cannot explain the relationships as described above. The formation of clusters or thin layers consisting of single species of heavy minerals or mixture of different species, as well as the close sorting of mineral grains according to size and shape are additional phenomena not explained by the theory of hydraulic equivalence.

/In...

In a deposit of sand in which the various mineral species have grain sizes as expected from the theory of hydraulic equivalence, concentration of the heavy minerals can be effected by the selective removal of coarse material, which consists predominantly of the light minerals, but may include the coarse portions of the heavy fractions. The removal of coarse material will cause the original lognormal grain size distributions to deviate from lognormality, and the median grain size will be decreased. These are exactly the phenomena observed for the first group of minerals, as well as leucoxene in some samples.

McIntyre (1959, p. 278) gave a similar explanation for the fact that quartz in the samples studied by him was finer than expected from the theory of hydraulic equivalence.

He states that: "Quartz also is much finer than expected. This may be due to the differential removal of coarse quartz. This may come about in the following way: the heavy minerals, being smaller than the associated quartz, would tend, under the action of swash and backwash, to hide behind the larger grains of quartz, and so there would be a greater proportion of quartz grains available at the surface of the deposit than hydraulically equivalent heavy minerals. Therefore, after reworking, the resulting size distribution of quartz would tend to have a smaller mean than that predicted from the linear approximation model."

Evans (1939, p. 28) determined empirically that the swash and backwash on a beach cause extensive sorting of beach material. In the beach slope the finer sediments are found near the swash line, and the coarser at the plunge line where the moving water has its greatest velocity. He states that the segregation of heavy minerals by waves and current takes place mostly in the upper part of the swash. The controlling factors are their fineness and high specific gravity. Grains between 0.25mm. and 0.60 mm. occur near the median line between the swash line and plunge line, and are transported most rapidly by beach drifting.

The processes described above can account for the following phenomena: (a) The deviation from lognormality of grain size distributions. It can be expected that, in cases where the above processes were operative, the cumulative size distributions will show deficiencies of coarse material.

/A decrease...

(b) A decrease in the median sizes of the lighter minerals in the deposit. These minerals will be finer than expected from hydraulic equivalence calculations based on a heavier mineral in the deposit.

(c) Concentration of heavy minerals relative to light ones.

However, such a close sorting according to size and shape as shown by sample 1(a) for uraninite and monazite, and to a lesser extent zircon, chromite, and leucoxene, are not satisfactorily explained. The formation of thin layers of closely sorted heavy minerals, as well as clusters, as are shown in Plate I, is also left unexplained.

Recently there appeared two articles by Moss (1962 and 1963), which must be regarded as one of the most important, if not the most important contribution to the understanding of the formation of sandy and pebbly deposits, which appeared during the last two decades, and in which a mechanism is described which can explain the features mentioned above.

On the basis of shape studies, he recognized three types of grain populations in sediments, which he calls A, B and C.

The grains forming a population A show good sorting, and a lognormal size distribution. They show an even gradation, over a small size range, from relatively small equant particles to somewhat larger, more elongated ones. The nature of the material of which the grains are composed have but little effect on their sizes and shapes, as they appear in this type of population. This type of population appears typically on beaches in the intertidal zone.

The grains forming population B occur associated with populations A in sediments laid down by unidirectional currents. They are of finer grain size than the associated population A grains, and occur interstitially between these grains, which shielded them from the current.

Population C consists of the relatively coarse grains occurring in coarser sediments.

The original articles by Moss should be studied for a clear understanding of the processes involved. In the next paragraph a portion of Moss' description of the formation of concentrations of dense particles by moving water is quoted verbatim: (Moss, 1963, p. 323).

/A relatively...

"A relatively small particle will project upwards less than its neighbours on the bed, and, consequently, the intergranular cavities of which it forms a side will provide less shielding to newly arriving potential Population A particles than will other intergranular cavities on the bed. Thus the addition of particles of the dense mineral to the bed will cause relatively smooth, particle-repellent spots to form on the bed surface. Newly arriving quartz particles that land on such spots are very likely to be lifted off or even rolled away for a diameter or two, but, for reasons already given, newly arriving particles of the dense mineral are much more likely to rest stably on these spots. The addition of relatively small particles of the dense mineral to Population A therefore further their already favored deposition, and the process has attributes analogous to those of a chain reaction. Provided that the quartz-repellent spots on the bed are sparsely distributed, quartz-dominated Population A will continue to be deposited between them and to eliminate them by lateral encroachment. But if the concentration, in the traction carpet, of suitable particles of the dense mineral exceeds a certain critical value, quartz-repellent spots will become more and more common until they unite. The dense mineral will gain control of the bed surface, and a new lamina, the growing Population A of which rejects nearly all quartz grains and dense particles that are too large or too small to rest stably on the bed, is initiated. This dense mineral Population A continues to grow until the supply of suitable particles from the traction carpet becomes too small to keep pace with the unloading requirements of the current; then a new quartz-dominated lamina is seeded on the bed surface in the manner previously described. The degree of segregation of dense minerals that this process can achieve is virtually astounding in some cases and will vary with the shape and specific gravity relationships of the minerals concerned. Two or more minerals, individually too uncommon to become segregated, can separate out together, and very rare minerals can become relatively concentrated in the company of others with similar combinations of specific gravity and shape characteristics".

/The process...

The process, as described by Moss, adequately and elegantly explains the puzzling features observed in many samples from the Upper Reef, viz. the close association of particles of similar size and shape of uraninite and monazite, with no regard to the requirements of hydraulic equivalence, as well as the occurence of clusters and layers of heavy minerals (Plate 1). These clusters therefore are "traction clogs", and have no connection with a colloform state.

Small amounts of clayey material, which could have been present originally in the Dominion Reef Sediments, can be regarded as representative of Moss Population B.

The presence of the second group of heavy minerals, viz. garnet, cassiterite, and columbite remains to be considered. As pointed out previously, these minerals (a) are coarser than expected when compared to uraninite, monazite, zircon and chromite, (b) have lower mean roundness values than expected, and (c) have cumulative size distributions showing no relationship to those of the other heavy minerals.

Two possible explanations can be given for the features mentioned above:

(a) That the roundness exhibited by monazite, zircon, and particularly uraninite was not caused by mechanical attrition, and no relationship need exist between roundness values in the two groups of minerals.

(b) That the minerals cassiterite, garnet and columbite did not share the complete sedimentary history of uraninite, monazite and zircon, but were added to the sediments at a later stage.

In considering possibility (a): The roundness of uraninite, monazite and zircon could have been caused by some of the processes mentioned in Table X. The roundness shown by garnet, cassiterite, and columbite, on the other hand, can be due to mechanical attrition. Therefore no relationship need exist between the roundness values of the two groups of minerals.

Recently Koen (in press) has focussed attention on an instance where the roundness exhibited by uraninite in the Witwatersrand banket is higher than that of much larger chlorite pseudomorphs after amphibole associated with it.

/Koen...

Koen uses this observation as an argument for his theory that uraninite does not owe its roundness to mechanical abrasion, but was precipitated as nodules in ancient marshes.

However, as shown in the previous pages, the roundness of uraninite, monazite and zircon in the present samples agree with the theory of mechanical abrasion.

In considering possibility (b): The relatively coarse and unrounded minerals cassiterite, garnet and columbite did not necessarily share the complete sedimentary history of uraninite, monazite and zircon, but could have been added at a later stage.

(i) In this case the lower roundness values can be considered to reflect the shorter period of sedimentary transport and reworking to which these minerals were subjected.

(ii) The larger grain sizes could be due to the availability of only relatively coarse material and the non-availibility of much material of a grain size similar to that of uraninite, monazite and zircon, as well as the short period of attrition and the sorting action of currents and waves to which they were subjected.

(iii) The dissimilar cumulative curves could also be ascribed to the availability of only relatively coarse material.

It is of interest to note that previous workers also suggested a dual source for Witwatersrand sediments (Simpson, 1951, p. 129; Fuller, 1960, p. 842).

The debris from a nearby landmass, furnishing the garnet, cassiterite and columbite, could have been added during the major cycle of sedimentation, or also during a period of erosion, in which case they shared only the relatively short period of reworking and concentration of heavy minerals following this event. This second possibility appears attractive if the following points are considered.

1. As shown by Rao (1957) erosion due to storms, regression of beaches and annual changes in sea-level is often followed by the formation of well-sorted heavy mineral sands. The processes involved are sorting by powerful waves, and transportation by along-shore currents.

2. Sharpe (1949) has shown that in the Witwatersrand, concentrations of economic minerals appear in the sedimentary cycle where disconformities are present.

/Reworking...

3. Reworking of sediments which may already contain concentrations of heavy minerals, will cause the availability of more heavy minerals of suitable size and shape in the traction carpet, so that more extensive heavy mineral laminae can be built according to the process described by Moss.

CONCLUSIONS

1. Uraninite is considered to be a detrital mineral, originating most probably from pegmatite. This is borne out by the following observations:

(a) Its composition (high in thorium and rare earths) and unit cell size correspond to pegmatitic uraninite, and are different from that of pitchblende.

(b) Many uraninite grains are present as partially rounded crystals.

(c) Zircon is present as a primary inclusion in some uraninite grains.

(d) The degree of rounding of uraninite is in accord with that of undoubted detrital grains such as monazite and zircon.

(e) In samples in which heavy mineral concentration can be ascribed to sorting according to size and shape, uraninite has a closely similar size and shape to that of monazite and some other heavy minerals.

(f) In one sample in which sorting according to size and shape did not play a dominant role, the sizes of uraninite, zircon, chromite and monazite show the relationships as expected from the theory of hydraulic equivalence.

2. Debris containing garnet, cassiterite, and columbite was probably added to the beach deposits in which uraninite, monazite, zircon and chromite were already present. This occurred most probably during a period of erosion of the sediments followed by further concentration of heavy minerals.

3. Before, during and after consolidation, weathering processes caused corrosion and alteration of some mineral species. The alteration of ilmenite and other titanium-rich minerals to leucoxene took place during this stage. Uraninite and zircon may have been corroded at the same time.

/4. At...

4. At a later date a period of metamorphism and hydrothermal activity caused alteration of many mineral species:

(a) An unidentified mineral originated from uraninite at this stage; it envelopes many uraninite grains.
(b) Uranothorite was formed at the expense of uraninite.
(c) Some leucoxene recrystallized into a mixture of ilmenite and rutile.
(d) Chromite, and to a lesser extent cassiterite, were cataclastically shattered. Chromite and garnet were partially replaced by phyllosilicates.
(e) Sulphidization of some iron-bearing minerals to form pyrite, and of some radiogenic lead to form galena, took place.
(f) Gold was redistributed.
(g) Extensive corrosion by solutions, during which uraninite, chromite, monazite, zircon and garnet were all attacked (simultaneously or successively with changing conditions) took place.

5. The formation of alteration products of uraninite occurred fairly recently under the influence of meteoric water. Schroeckingerite and "gummite" were formed. Covellite formed from chalcopyrite at this stage.

XI. ACKNOWLEDGMENTS

The author has great pleasure in thanking Professor J. Willemse and Dr. W.R. Liebenberg for encouragement, guidance and stimulating discussions. He is also indebted to Dr. G.M. Koen for criticism and suggestions which improved the manuscript, but hastens to point out that Dr. Koen can in no way be held responsible for the various opinions and interpretations in this thesis which conflict with his own views.
Mr. R.S. Birkill kindly carried out a thermogravimetric analysis of uraninite, and Mr. H. Stoch supervised the chemical analysis of uraninite.

In 1962 the author joined the staff of the Government Metallurgical Laboratory, and the Director Dr. R.E.Robinson, kindly allowed him to continue this study, and to make use of the facilities of the laboratory. The author wishes to express his gratitude also to them.

EXPLANATION OF PLATES

PLATE I

Mosaic of an area 2.5 mm. by 3 mm. of a polished section of a layer about 1 mm. thick, in which uraninite and other heavy minerals are concentrated. The 75 grains of uraninite pictured range in size from slightly less than 0.1 mm. to 0.3 mm. across. Although many grains possess frayed and irregular outlines, a fair percentage is well preserved.

The 14 monazite grains (marked M) range in size from slightly less than 0.1 mm. in diameter to slightly more than 0.2 mm. in diameter. Most monazite grains are well preserved.

It is apparent that in this sample uraninite and monazite are not hydraulically equivalent.

The 8 grains of leucoxene (marked L) are irregular in outline but of a similar size as ~~to~~ monazite. Grains having typical porous centres occupied by secondary quartz are shown at X1.4, Y2.8; and X2.2, Y0.4.

One grain of cassiterite (Ca) at X1.3, Y2.3 is not typical in having a very high sphericity.

Remnants of garnet (Ga) partly replaced by chlorite are present at X1.2, Y2.7; and X0.7, Y1.3.

Pyrite (P) and chalcopyrite (C) are visible at X1.8, Y2.3.

A grain of gold (G) partly surrounded by galena at X1.0, Y2.6, is shown in more detail in Plate X, Fig. 3.

The cluster of uraninite grains at X1.1 and Y 1.0 is reminiscent of the type of cluster described by Koen (1962). However, a monazite grain (X1.2, Y0.7) is present in this cluster. This cluster of uraninite cuts with a concave boundary into the quartz grain just north of it, illustrating the often observed phenomenon that in many samples uraninite retains its shape better than quartz.

The high concentration of heavy minerals of similar size and shape shown here ~~shown~~ is regarded as a "traction clog".

Photographic data: Photo's taken with water immersion. The scales at the left and bottom are in mm.

Plate I

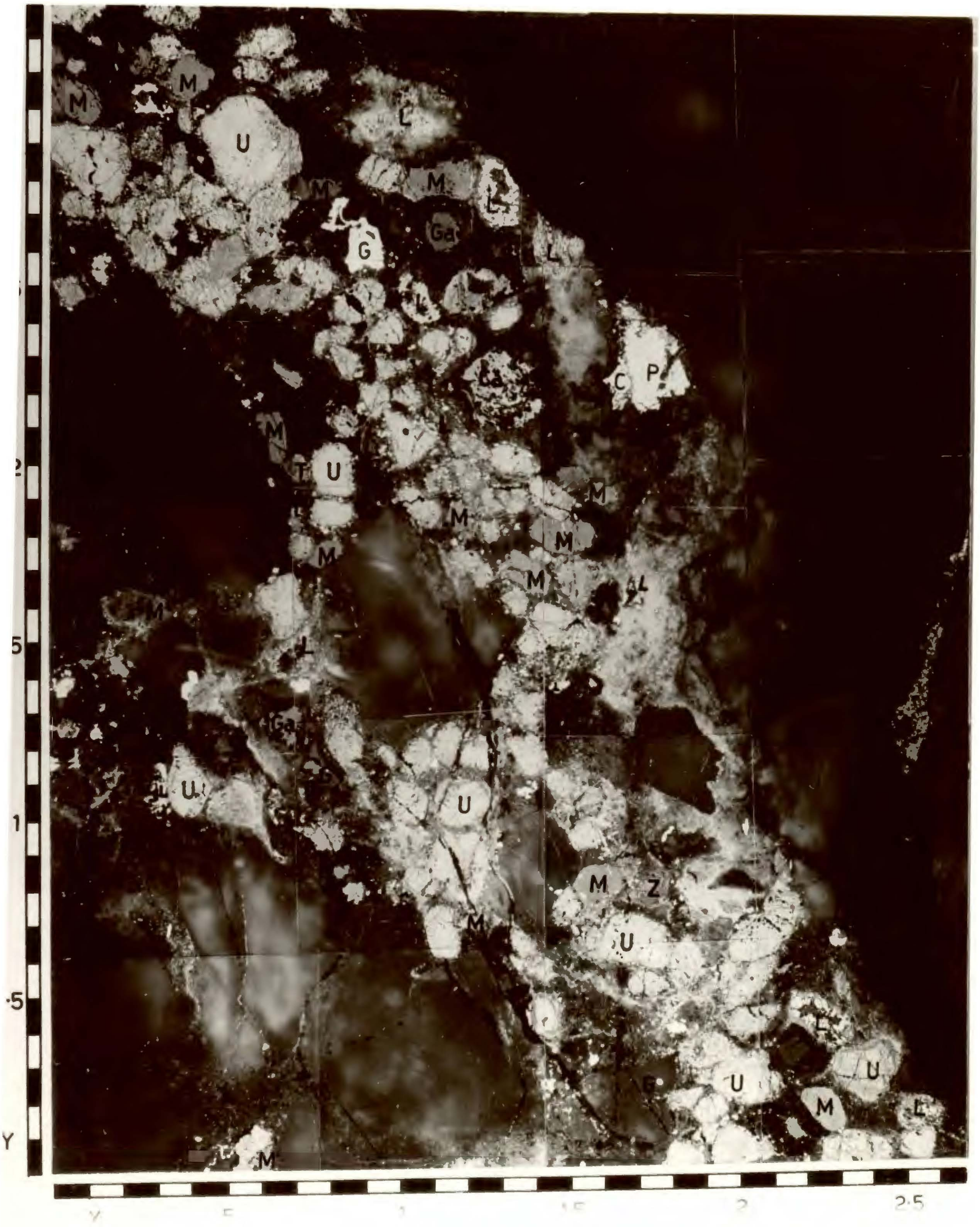


PLATE II

Six crystals of uraninite on this plate exhibit six stages of rounding.

In Fig.1 the grain is bounded by crystal planes. It is a moot point whether signs of rounding are present in this crystal or not.

In Fig. 2 four crystal faces form most of the boundary. The two upper corners have Wadell roundness values of 0.24 and 0.56, but the degree of roundness of the two lower corners is very small.

In Fig. 3 six straight portions of the boundary are here interpreted as crystal faces. This grain was used as an example to explain the method of roundness and shape measurement used in this paper (See text Fig.2, p.41).

In Fig. 4 four crystal faces are present, and the corners have roundness values as follows: The large curved boundary at 2 o'clock, 0.76; the sharp corner at the bottom, 0.08; the obtuse corner at 8 o'clock, 0.64; the obtuse corner at 10 o'clock, 0.64; the remaining round protuberance at 11 o'clock, 0.32.

In Fig. 5 four crystal faces are still discernible, but a large portion of this grain is already bounded by curved surfaces. The roundness values of its corners are: At 2 o'clock, 0.96; clockwise from this corner, 0.54, 0.54 and 0.31 respectively. The lower left hand portion of this grain was slightly corroded, obscuring the very fine detail of its boundary.

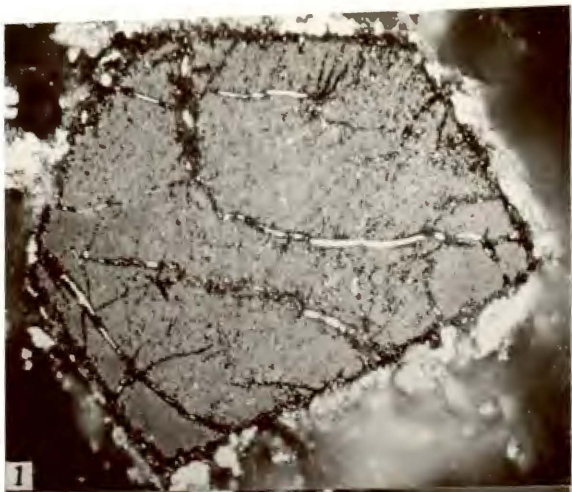
The grain in Fig. 6 is almost completely bounded by curves. The small straight portion of the lower boundary is a remaining portion of a crystal face. The curved boundary at 12 o'clock has a roundness value of 0.96; the one at 4 o'clock, 0.60, and the one at 9 o'clock 0.64. This grain has the highest degree of rounding encountered amongst the several thousands studied in the Dominion Reef.

Small radial cracks are displayced by the grain of Fig.3, and also in Figs. 4 and 6. Large cracks traverse the grains at various angles, and are filled by galena in most instances. In Fig. 5 many tiny cracks of limited extent are present.

Photographic data: All photo's taken with oil immersion. The magnifications are :

Fig. 1 : 1070 x; Fig. 2 : 420 x; Fig.3 : 490 x; Fig. 4 : 350 x;
Fig. 5 : 360 x; Fig. 6 : 350 x.

Plate II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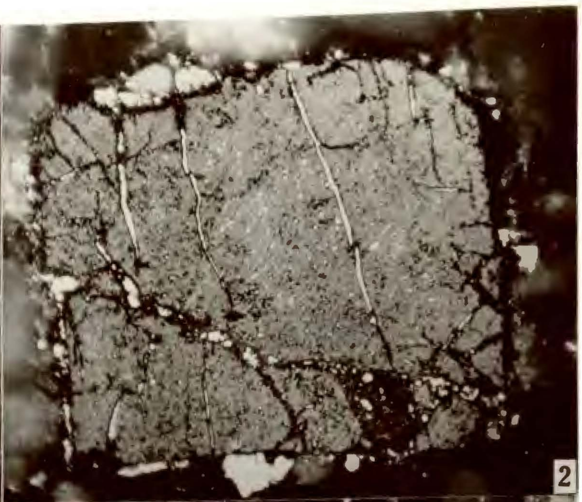
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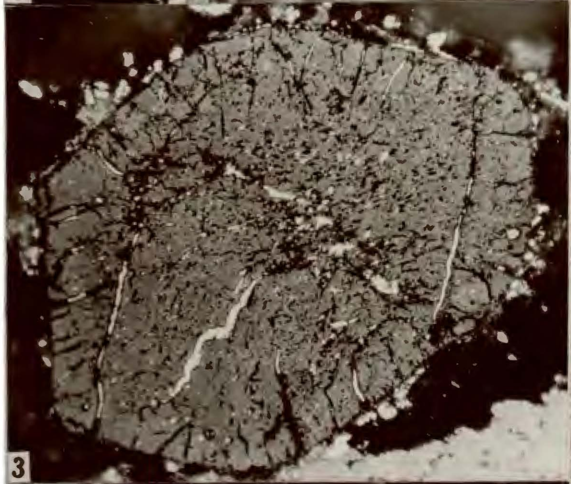
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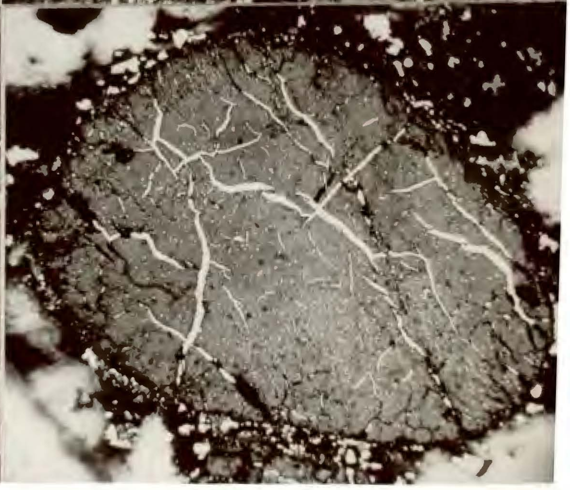


PLATE III

Figs. 1 and 2 : Two corroded uraninite grains. In Fig. 1 the area occupied by the original grain is still visible due to yellowish alteration products. In this case the size and roundness of the original grain can still be measured. In Fig. 2 the lower portion of the grain has been destroyed, and no evidence remains of the degree of roundness of this portion. If an attempt is made to draw the original outline by "extrapolation" bias will be introduced. However, as the inscribed circle can still be measured fairly accurately, roundness values can still be obtained for the three remaining corners.

Figs. 3 and 4 : Reaction rims of unidentified material as described on p.13. In Fig. 3 a large portion of the uraninite core of the grain was altered to pitch-black material.

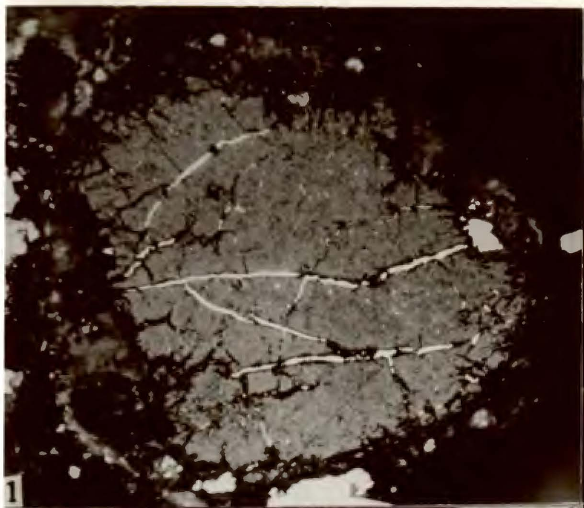
Fig. 5 : A second type of uraninite compared to the usual type. It has a distinctly higher reflectance and is characterized by numerous inclusions of galena which occur in the form of small equant grains rather than veinlets.

Fig. 6 : Inclusion of a tiny zircon crystal in uraninite. Zircon is the most abundant type of primary inclusion encountered in the uraninite grains.

Photographic data: All photo's taken with oil immersion. The magnifications are :

Fig. 1 : 220 x; Fig. 2 : 470 x; Fig. 3 : 490 x; Fig 4 : 510 x;
Fig. 5 : 585 x; Fig. 6 : 1070 x.

Plate III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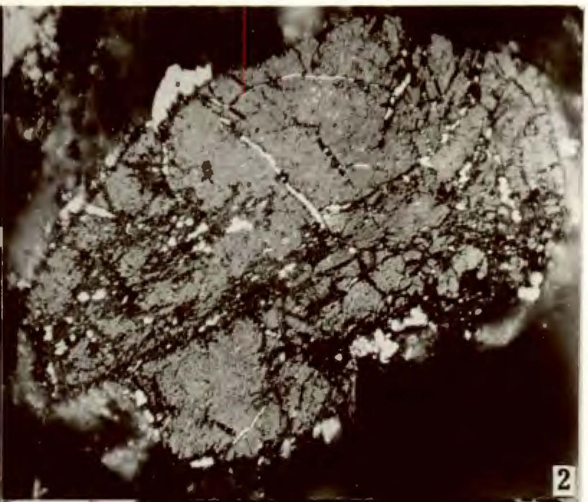
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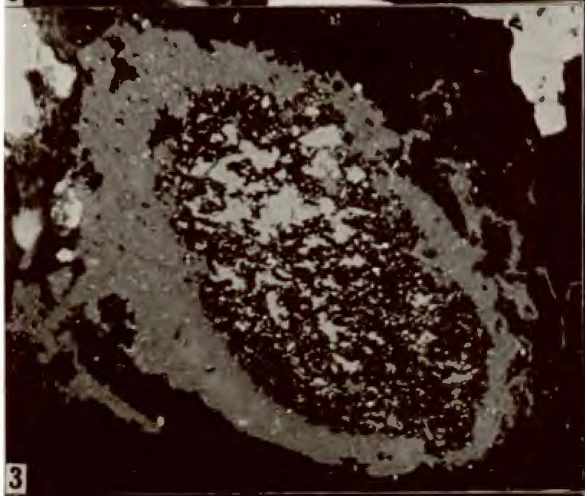
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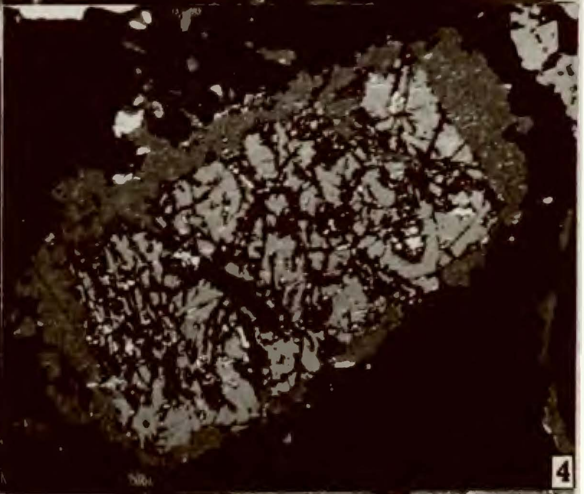
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5


6

PLATE IV

Three types of alteration products. Figs. 1 to 3 : A dull black material with low reflectivity replaces uraninite in the form of oriented veinlets forming a trelliswork (Fig. 1), or in veinlets of random orientation (Fig. 2). In the grain pictured in Fig. 3 replacement proceeded from the outside towards the centre, and only the core contains unaltered uraninite. This type of alteration can be ascribed to a partial conversion of U^{+4} to U^{+6} without destruction of the crystal structure. It still yields the diffraction pattern of uraninite.

Figs. 4 and 5 : Another type of alteration product, which is black, but under oil shows brownish internal reflections. It occurs as irregular elongated bodies, in many places partly lined by galena, and also contains abundant fairly big inclusions of galena.

Fig. 4 : Both types of alteration products in the same grain.

Fig. 6 : A portion of a veinlet consisting of orange "gummite". Remnants of uraninite visible in it.

Photographic data: All photo's taken with oil immersion. The magnifications are :

Fig. 1 : 240 x; Fig. 2 : 480 x; Fig. 3 : 240 x; Fig. 4 : 400 x;
Fig. 5 : 820 x; Fig. 6 : 270 x.

Plate IV

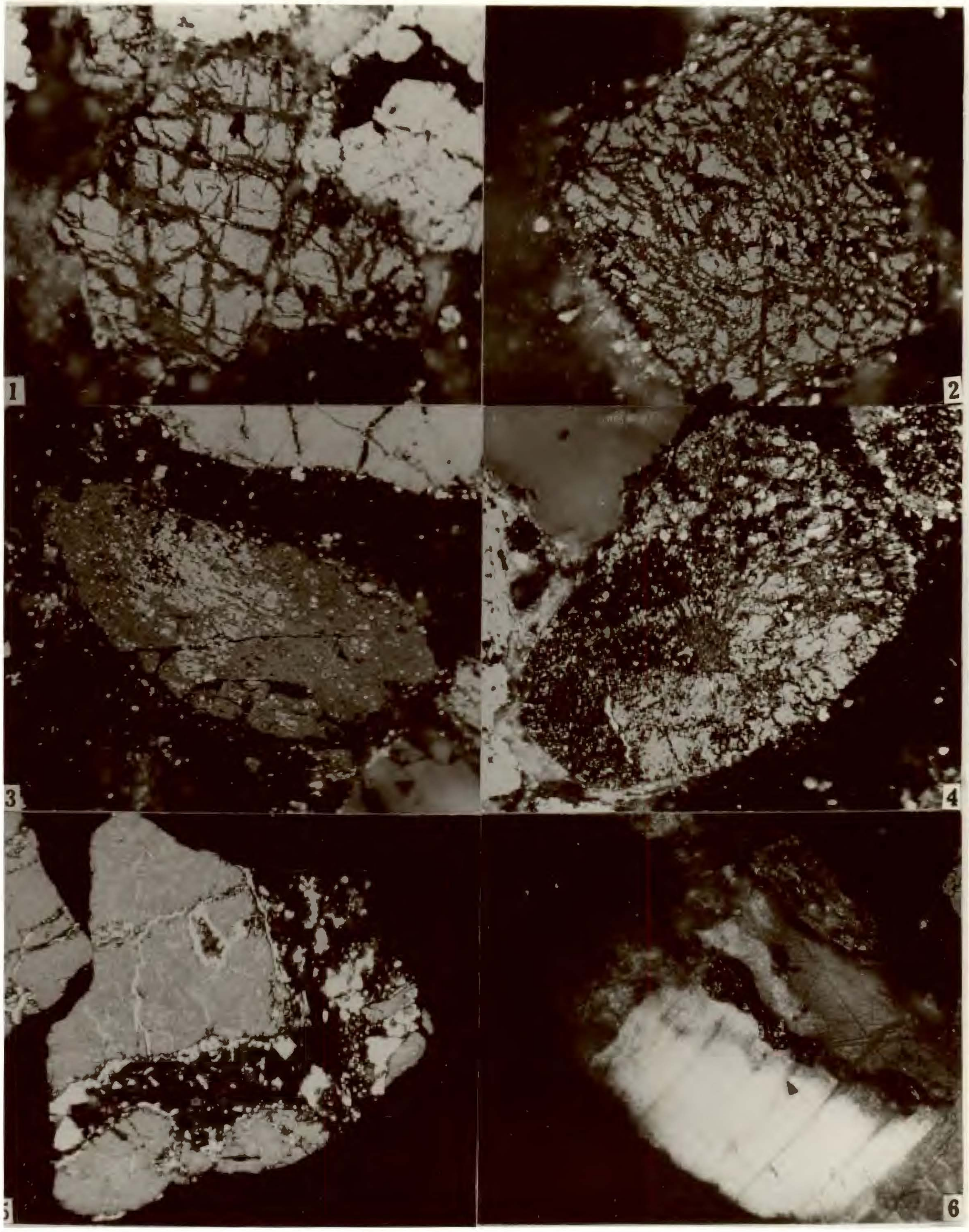


PLATE V

Figs. 1 to 4. Two types of thorite: The most common type (Figs. 1 and 2) is colourless, usually translucent, and in some places forms at the expense of uraninite. The grain of roughly square outline (Fig. 1) was photographed with a combination of transmitted and reflected light. Fig. 2 : The same type of thorite by reflected light only. The two grains of uraninite in the upper left and right corners have been partly replaced by thorite.

Figs. 3 and 4 : Ferrian thorite. In Fig. 3 a reddish-brown opaque grain with many inclusions of galena is pictured. In Fig. 4 a reddish-brown grain is shown in which some portions near the middle have not been so extensively altered. These portions yielded the X-ray pattern of thorite in addition to that of thorianite after heating. The small white lathlike inclusions which are abundant at the upper left hand and the lower right hand corners consist of pyrite, thought to have formed by the reaction of a portion of the iron of the ferrian thorite near its boundary with sulphur. Tiny white inclusions of galena are also present.

Photographic data:

All photo's taken with oil immersion. The magnifications are: Fig. 1 : 645 x; Fig. 2 : 470 x; Fig. 3 : 120 x; Fig. 4 : 179 x.

PLATE V, FIGS. 5 AND 6, AND PLATE VI, FIGS. 1 and 2.

These four figures illustrate grains of monazite. Fig. 5 shows a fairly typical grain which attained a high degree of rounding, and is traversed by parallel cracks. Plate V, Fig. 6, and Plate VI, Figs. 1 and 2 show remains of crystal faces. A monazite crystal sectioned roughly parallel to (010) so as to cut the (100) and (101) faces will have the type of outline shown in Plate VI, Figs. 1 and 2. The zonation in Fig. 2 could only be shown in polished thin section by combining transmitted light with reflected light.

Photographic data: All photo's taken with oil immersion. The magnifications are:

Plate V: Fig. 5 : 196 x; Fig. 6 : 450 x; Plate VI: Fig. 1 : 500 x; Fig. 2 : 380 x.

Plate V

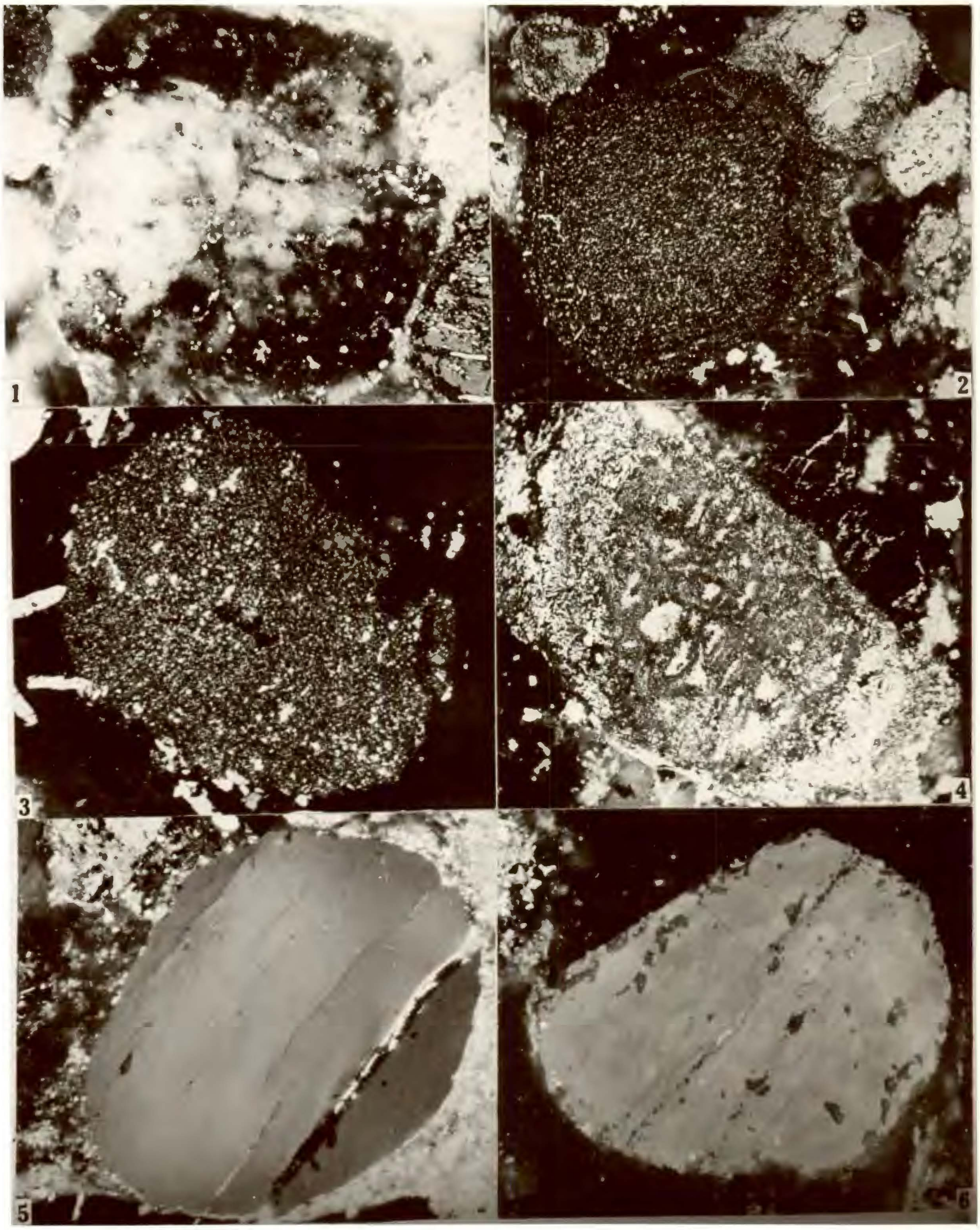


PLATE VI

Figs. 3, 4, 5 and 6 : Various types of zircon. Figs.3 and 4 show grains of a colourless well zoned type of zircon. Some of the zones in Fig. 3 contain many inclusions of galena. Studies by means of autoradiographs proved that such grains are radio-active.

In Fig. 4 zones of malacon are seen which do not contain inclusions of galena.

Fig. 5 illustrates a rounded grain of a purple type of zircon which is also fairly abundant in the Dominion Reef.

Many grains of zircon have been well preserved, and they may exhibit various stages of rounding as shown in Figs. 3, 4, and 5. Many grains have been extensively altered and corroded as shown in Fig. 6.

Photographic data: All photo's were taken with oil immersion. The magnifications are:

Fig. 3 : 430 x; Fig. 4 : 360 x; Fig. 5 : 400 x; Fig.6 : 440 x.

Plate VI

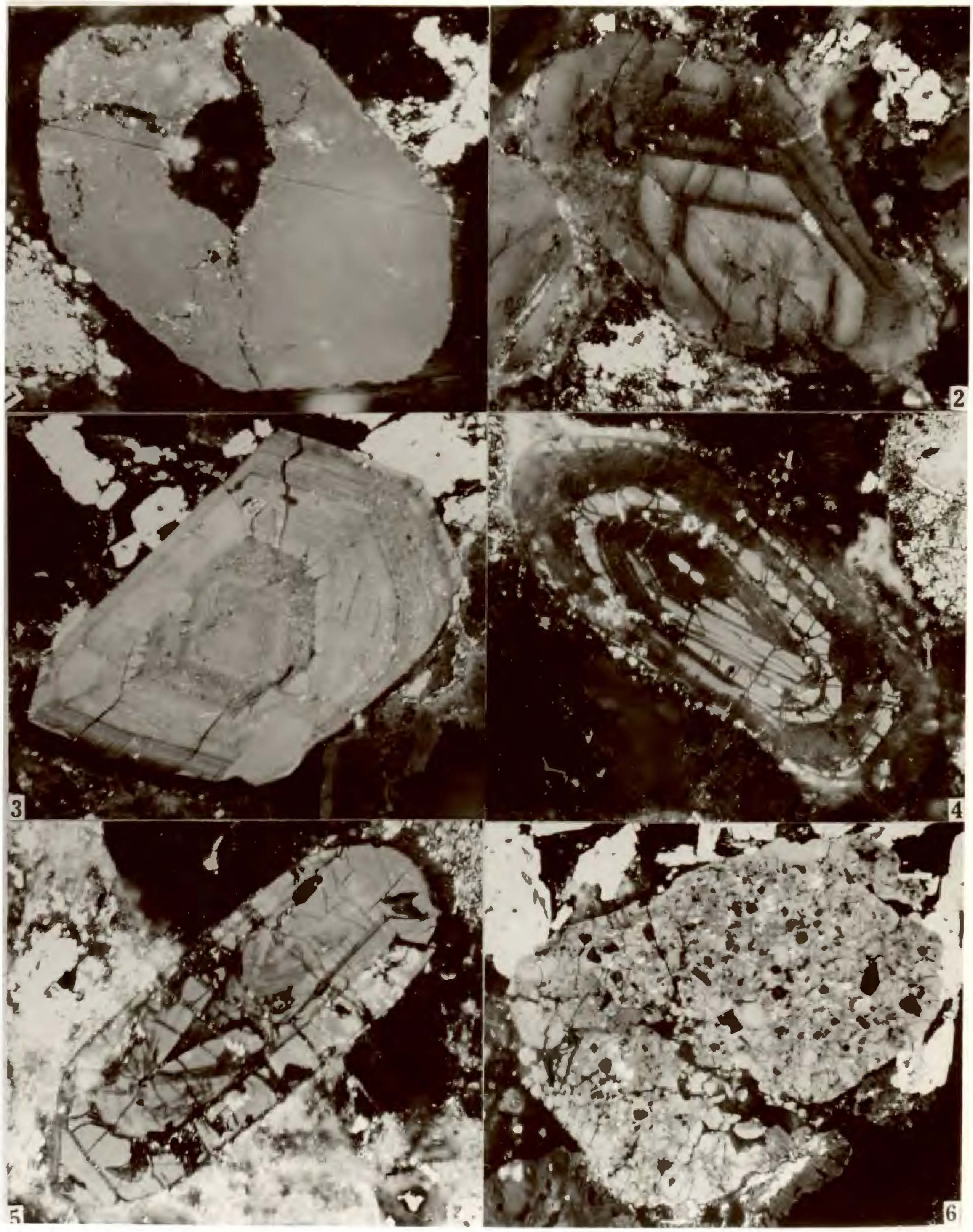


PLATE VII

Figs. 1 to 4: Grains of columbite partly replaced by metamict material. In Fig. 1 only a thin zone at the border of the grain has been replaced. In Figs. 2, 3 and 4 progressively more advanced stages of replacement are shown. The grain in Fig. 4 consists completely of metamict betafite. It is surrounded by light-yellowish material (white in the photograph). The fairly low sphericity and roundness shown by these grains are typical of the larger grains of the columbite in the specimens examined.

In Figs. 5 and 6 typical grains of cassiterite are shown. The grain illustrated in Fig. 5 has been fairly well rounded, but the degree of rounding exhibited in Fig. 6 is low. Twinning such as that shown in Fig. 5 is common. The elongated grains of fairly low sphericity as is shown here are typical of the larger cassiterite grains.

Photographic data: All photo's taken with oil immersion. The magnifications are :

Fig. 1 : 385 x; Fig. 2 : 464 x; Fig. 3 : 83 x; Fig. 4 : 200 x; Fig. 5 : 470 x; Fig. 6 : 236 x.

Plate VII

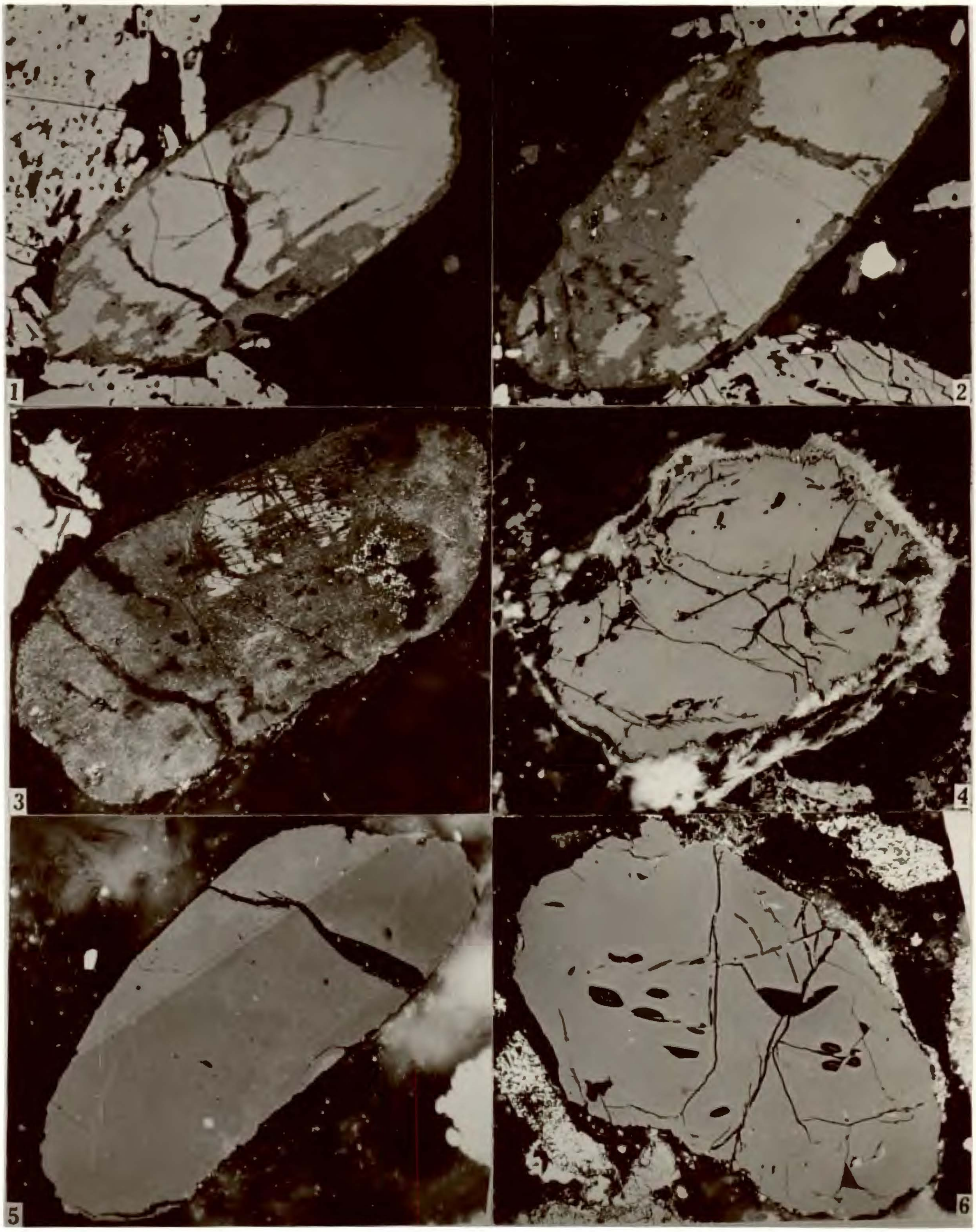


PLATE VIII

Figs. 1 to 4: Different types of leucoxene. In Fig. 1 an aggregate of leucoxene grains is bounded by convex and concave surfaces thought to be of colloidal origin. The central portion of this aggregate is occupied by secondary quartz (black).

In Fig. 2 many of the same features of Fig. 1 are shown. However, the leucoxene in this aggregate has been recrystallized to rutile. This photograph was taken with partially crossed nicols, and the anisotropism of the individual grains of rutile in the aggregate is evident from the differences in brightness.

Figs.3 and 4 : A type of leucoxene which contains tiny inclusions of galena. As shown by autoradiographs, this type of leucoxene is appreciably radio-active. In Fig. 4 the original form of the mineral from which the leucoxene was formed was partially retained. This grain contains inclusions of zircon altered to malacon in the upper left hand corner and near the right edge. If the radio-active leucoxene is heated strongly, brannerite can be synthesized.

Figs. 5 and 6 : Ilmenite formed at the expense of leucoxene. Many remnants of leucoxene (black) are still present in these aggregates. Rutile which has a slightly higher reflectivity than the ilmenite is distributed through the aggregate as tiny grains.

The aggregate shown in Fig. 5, which is composed of lathlike grains of ilmenite and a little rutile, still shows the outlines of the original grain.

Photographic data: All photo's taken with oil immersion. The magnifications are:

Fig. 1 : 360 x; Fig. 2 : 280 x; Fig. 3 : 92 x; Fig. 4 : 224 x;
Fig. 5 : 325 x; Fig. 6 : 252 x.

Plate VIII

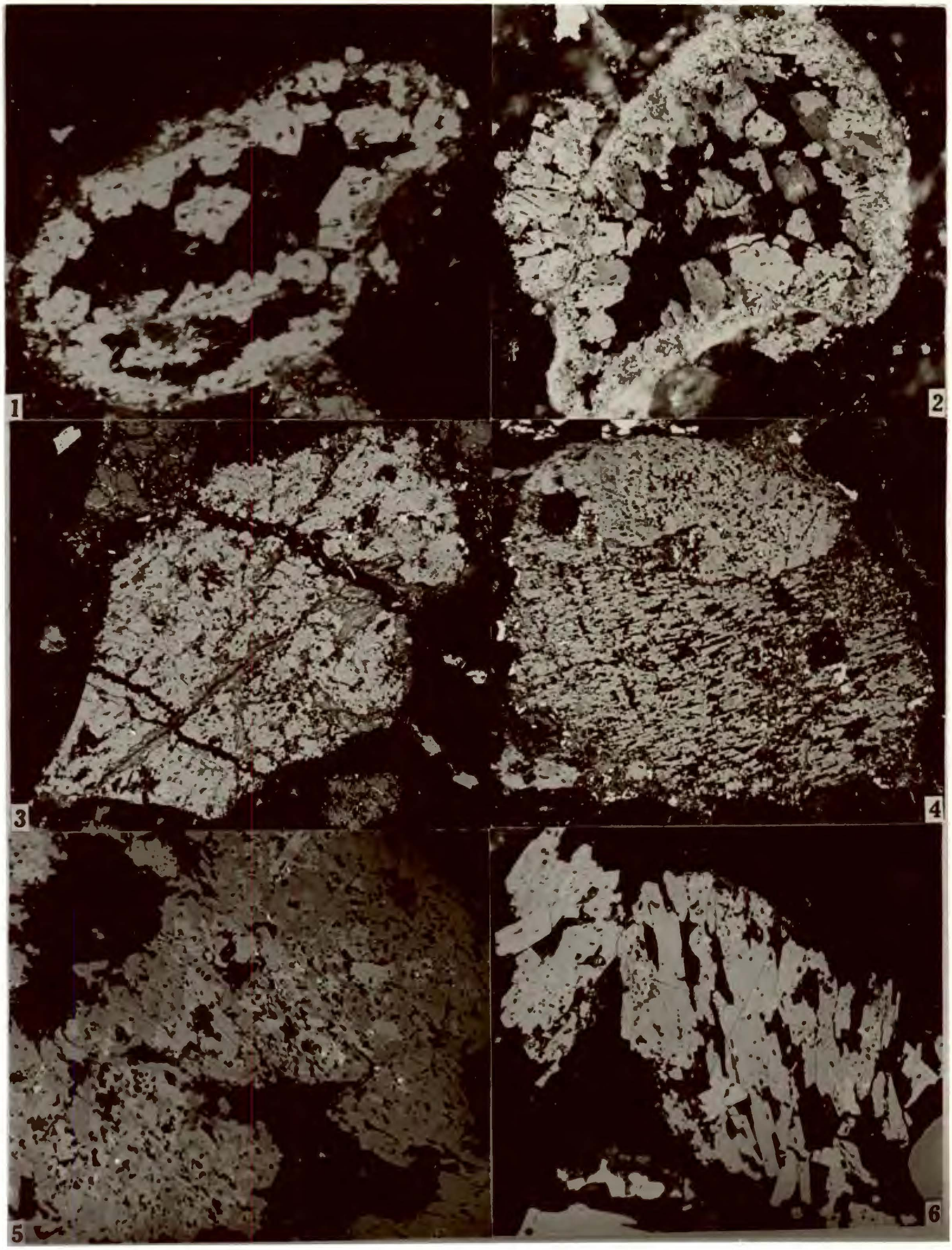


PLATE IX

Figs 1 and 2: Two types of chromite. The grain in Fig. 1 is surrounded by a thin zone of material which has a higher reflectivity, and is considered to be magnetite. The shattering as shown by the irregular cracks which traverse the grain is a common feature of the chromite in the Dominion Reef. In some cases these cracks are lined by the pale material forming the outer zone in Fig. 1. The tiny orientated white and dark inclusions in Fig. 2 are thought to be haematite and quartz.

Photographic data:

Both photographs were taken with oil immersion. The magnifications are:

Fig. 1 : 432 x; Fig. 2 : 825 x.

Figs. 3 and 4: Typical grains of cobaltite. Straight crystal faces, round corners, and concentric cracks as well as cubic cleavage planes are exhibited by most grains. The round corners can be ascribed to oscillatory development of cube and pyritohedron faces. (See also Plate X for examples of anhedral cobaltite).

Photographic data:

Both photographs were taken with oil immersion. The magnifications are:

Fig. 3 : 342 x; Fig. 4 : 721 x.

~~In~~ Fig. 5: A detrital grain of sphene is shown. The dark embayments were caused by chipping during polishing.

Photographic data:

Oil immersion, magnification : 355 x.

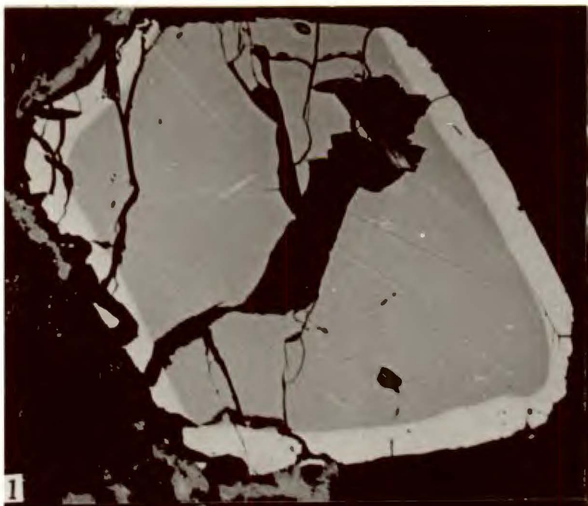
Fig. 6: Pyrite (white) intimately intergrown with tiny grains of rutile, and also enveloping some rutile grains (light grey). The dark material with lighter grey specks at the middle of the grain consists of leucoxene.

This assemblage is thought to have originated as follows: (a) Alteration of an original iron-titanium mineral (probably ilmenite) to iron-bearing leucoxene. (b) Pyritization of some of the iron at a later date, and recrystallization of a portion of the titanium dioxide in the leucoxene to rutile.

Photographic data:

Oil immersion, magnification : 131 x.

Plate IX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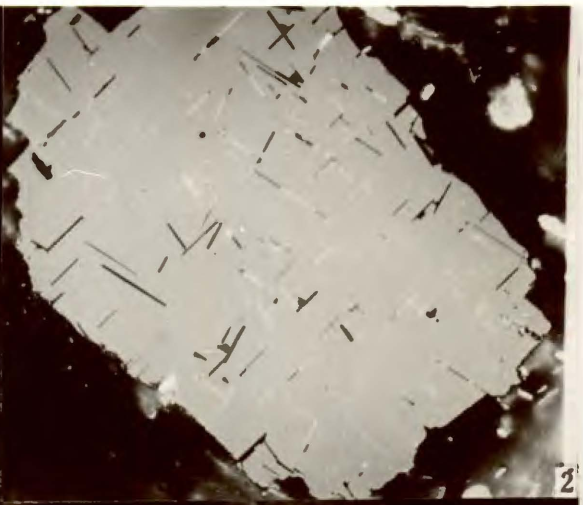
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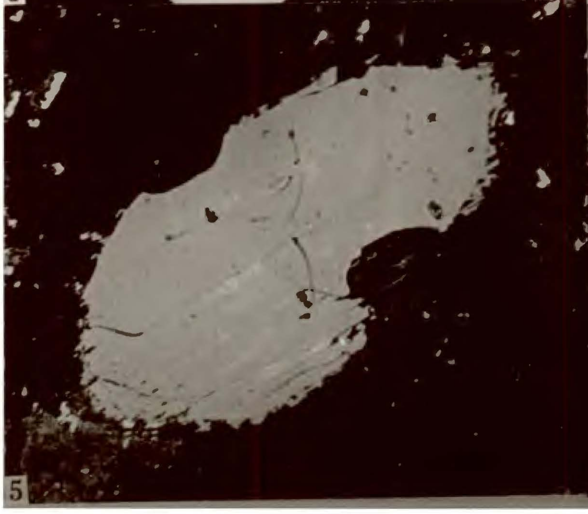
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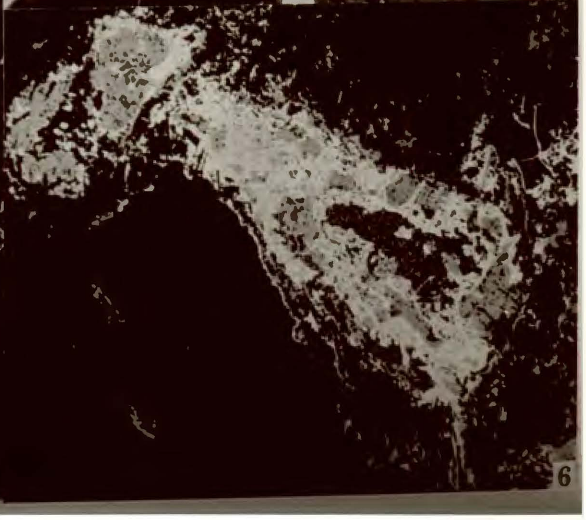

6

PLATE X

Fig. 1: ~~shows~~ A pyrite grain which contains many slightly tarnished inclusions of galena. A few inclusions of chalcopyrite are also present in some of the grains. In some samples inclusions of galena in pyrite are bigger, and bounded by straight planes.

Photographic data:

Oil immersion, magnification: 257 x.

Fig. 2 : An anhedral grain of cobaltite containing large inclusions of sphalerite (dark grey) and also a tiny grain of gold (not visible in the photograph).

Photographic data:

Oil immersion, magnification: 486 x.

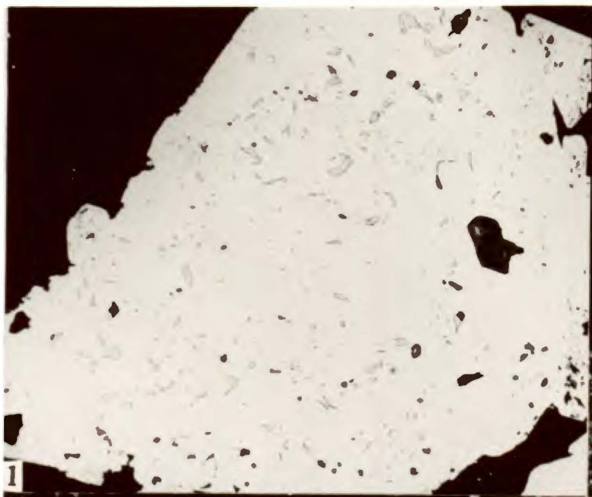
Figs. 3 to 6: Various textural types of gold. In Figs. 3 and 4 gold is associated with sulphides forming overgrowths on the gold grains. In Fig. 3 the overgrowth consists of galena, and in Fig. 4 of cobaltite. Fig. 5 shows some members of a cluster of gold grains in leucoxene. Fig. 6 illustrates a typical hackly grain of gold which is here intimately associated with leucoxene.

Photographic data:

All photo's were taken with oil immersion. The magnifications are:

Fig. 3 : 350 x; Fig. 4 : 1271 x; Fig. 5 : 1714 x;
Fig. 6 : 917 x.

Plate X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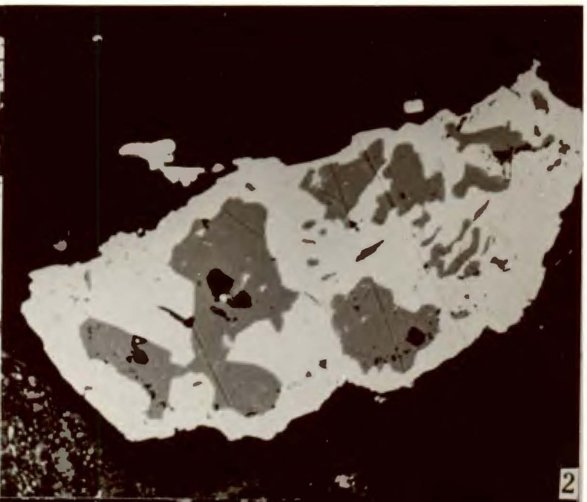
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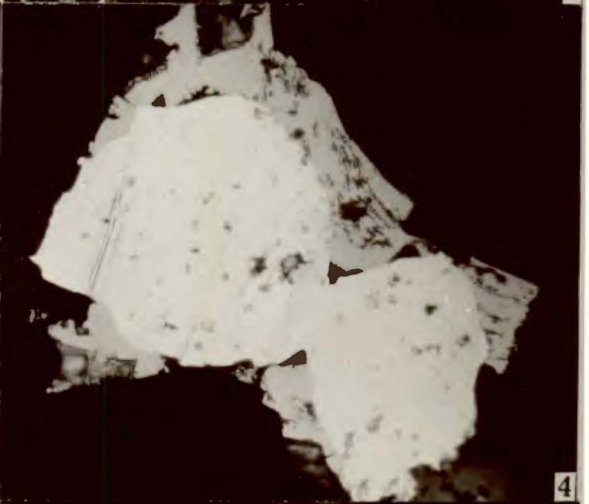
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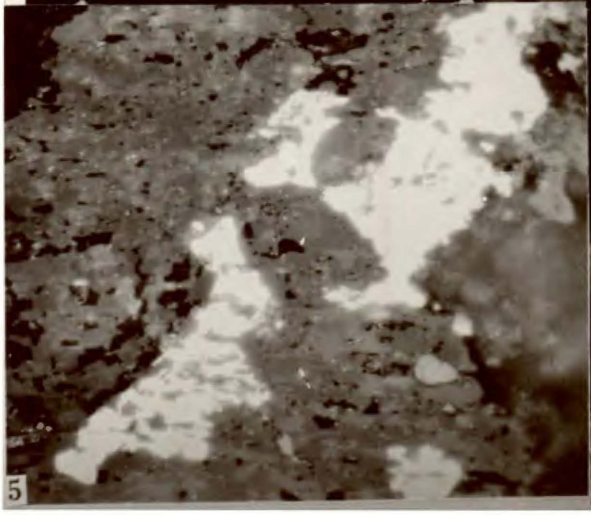
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6

PLATE XI

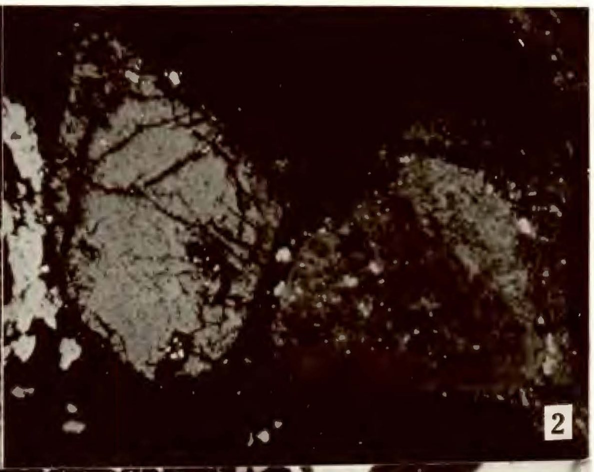
~~In~~ Figs. 1 and 2: Two grains of uraninite are shown which contain cracks bounding roughly ellipsoidal cores. The same grains are shown in Plate I (X0.4, Y1.0; X0.1, Y2.9). One possible interpretation is that the cores represent original grains, and the borders overgrowths. However, the cracks may also be the result of differential increase in volume due to radio-activity.

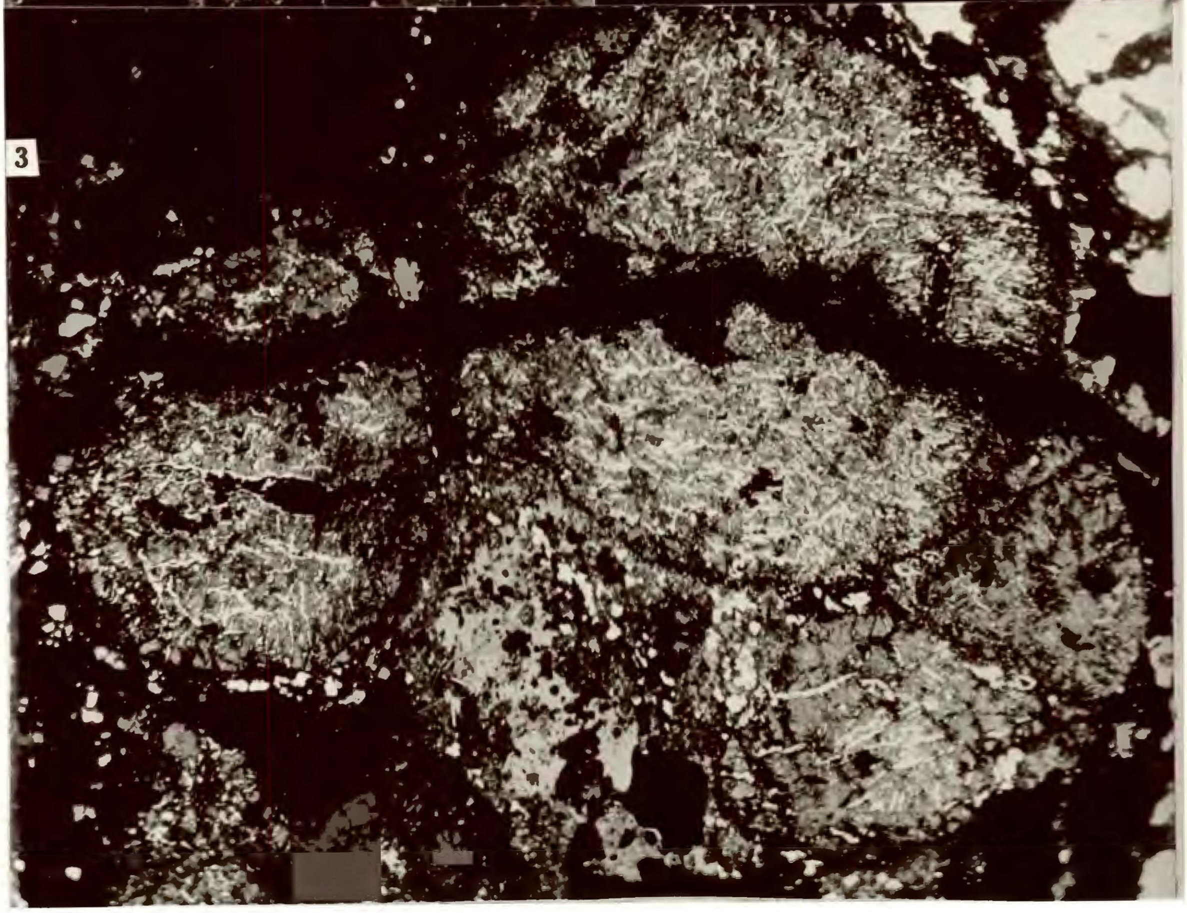
~~In~~ Fig. 3: A cluster consist~~ing~~s of two spherical nodules joined together by a nodular overgrowth at the bottom ~~is pictured.~~ Such clusters are very rare, the example being the only one found in 129 polished sections of the Dominion Reef ores.

Photographic data: All photo's were taken with oil immersion. The magnifications are :

Fig. 1 : 144 x; Fig. 2 : 226 x; Fig. 3 : 256 x.

Plate XI





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PART 2

GEOCHEMISTRY

ABSTRACT

Spectrographic techniques are described for the determination of the following elements present in the uraniferous conglomerate from the Dominion Reefs Mine: Ti, Zr, Cr, Fe, Pb, As, Sn, Cu, Ag, Zn, Nd, Pr, U, Ce, La, Y, Th, Nb, Mo, Co and Ni.

From the quantitative data obtained correlation coefficients were calculated for 101 pairs of elements, and high degrees of correlation were found between elements present in detrital minerals and uranium. A relatively low degree of correlation exists between gold and uranium.

It is considered that sedimentary concentration of heavy minerals and the subsequent limited redistribution of gold provides the most satisfactory explanation for the geochemical pattern of element distribution that was found.

CONTENTS

I. INTRODUCTION

In Part 1 of this thesis the mineralogy of the conglomerate of the Dominion Reefs Mine was described in detail. It was concluded that one suite of minerals present in the conglomerate is of detrital origin, and another showed signs of a hydrothermal redistribution. These conclusions were based on mineral associations, measurements of size and shape, and chemical evidence. Some of these conclusions could be put to the test by obtaining quantitative data for mineral concentrations in the various samples. The usual mineralogical methods, viz. separation, weighing and counting of heavy minerals, and modal analysis by means of a point counter were also tested but found to be of too low a precision for the purpose envisaged.

The method based on heavy mineral separation gave unsatisfactory results, as some of the mineral grains have been shattered, and others are of too friable a nature. The conglomerate is well cemented, and severe grinding techniques have to be applied to pulverize it. A large proportion of some of the mineral species is accordingly lost as slime. The method of point-counting is unsatisfactory because of its very low precision for minor constituents of a rock.

It was accordingly decided to apply a spectrographic method of analysis to these samples in order to obtain quantitative data on the ore. As the mineralogy of the samples studied is well known, the hosts of the various elements present could be determined readily

For the purpose of spectrographic analysis 20 samples of conglomerate were used. Of these, 7 originated from the Upper Reef, 11 from the Lower Reef, and 2 from the hanging-wall. Details of these samples are given in Table XII, and also in part I of this thesis, (Table I).

II. TECHNIQUES

A. QUALITATIVE ANALYSIS

As a preliminary survey qualitative spectrographic analyses were carried out on the samples, as well as on concentrates of minerals obtained by superpanning, separation by heavy liquids, flotation, and magnetic separation by means of a Frantz Isodynamic Separator.

/For the...

For the spectrographic analyses, a large Hilger quartz/glass spectrograph was available. The samples were loaded as powders into hollow copper and graphite anodes. Pointed cathodes of the same composition as the anodes were used. The samples were usually burnt to completion.

In cases where cyanogen bands masked some of the analysis lines, arcing was done in an atmosphere of carbon dioxide, by which process the cyanogen bands are eliminated.

The results of the qualitative analyses are given in Table I.

TABLE I

RESULTS OF QUALITATIVE SPECTROGRAPHIC ANALYSES OF SAMPLES FROM THE DOMINION REEFS MINE

Sample No.	As*	Bi	Co*	Cr*	Ge	Li	Mn	Mo*	Na	Nb*	Ni*	Pb*	Sn*	Th*	U*	V	Zn*
1(a)	+	tr	+	+	–	–	+	tr	tr	–	+	+	+	+	+	–	–
1(b)	–	–	+	tr	–	–	+	–	–	–	tr	+	+	tr	+	–	–
2	–	–	+	+	–	–	+	+	tr	tr	+	+	+	+	+	–	+
3	–	–	–	+	–	tr	–	tr	tr	tr	–	+	tr	tr	tr	–	tr
4	–	–	–	tr	–	tr	tr	–	tr	tr	tr	tr	tr	tr	tr	–	–
5	–	–	–	tr	–	tr	–	tr	tr	–	–	tr	tr	–	tr	–	tr
6	–	–	tr	+	–	–	+	–	tr	tr	tr	+	+	+	tr	–	tr
7	–	–	tr	tr	–	–	tr	tr	tr	tr	tr	tr	tr	–	tr	tr	tr
8	–	–	tr	tr	–	–	tr	tr	–	+	tr	tr	tr	–	tr	tr	tr
9	tr	–	tr	tr	–	–	tr	tr	–	+	tr	+	+	–	tr	tr	tr
10	+	–	tr	tr	–	–	+	–	–	+	tr	+	+	–	tr	–	–
11	+	–	+	tr	–	–	tr	–	–	+	tr	+	+	tr	+	–	–
12	+	–	+	tr	–	–	tr	tr	–	+	tr	+	+	–	tr	–	+
13	tr	–	+	tr	–	–	tr	–	–	+	tr	+	+	tr	tr	–	+
14	tr	–	tr	tr	–	–	tr	–	–	+	tr	+	+	tr	+	–	+
15	tr	–	+	+	–	–	+	tr	+	+	tr	+	+	+	+	–	–
16	tr	–	+	+	–	–	+	tr	+	+	tr	+	+	+	+	–	–
17	+	–	+	tr	tr	–	+	tr	–		tr	+	+	tr	+	–	–
18	tr	–	–	tr	–	–	tr	tr	–	+	tr	tr	tr	–	tr	–	–
20	–	–	–	tr	–	–	–	+	–	–	tr	tr	tr	–	tr	–	–

Elements absent or below the limits of detection of the method used:
Ba, Be, Cs, Ga, Hg, In, Os, Pt, Sb, Sr, W.

Strong spectral lines of the following elements were found in all samples:
Al, Ca, Fe*, K, Mg, Si, Ti*, Zr*

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*Quantitative results are also given for those elements marked with asterisks (Table V).

B. QUANTITATIVE ANALYSIS

(1) Sample preparation and spectrographic procedure

For quantitive work a sample of rock weighing 10 pounds, was crushed, and coned and quartered in order to obtain about one pound of representative powder. This was ground down in a clean mechanical mullite mortar under alcohol for one hour. A portion of this powder was heated in a porcelain crucible in a muffle furnace to 800°C, and kept at this temperature for one hour. After cooling, and the determination of weight loss, it was again ground for a further hour. The powder obtained was kept for the analysis of the non-volatile elements. For the volatile elements, portions of the unheated powder was used.

The object of the heat treatment was to eliminate most of the sulphur of the sulphides which have an adverse matrix effect. Conversion of most of the sulphides to oxides rendered the composition of the samples closer to that of the standards, which consisted of oxides of the relevant elements in a quartz base.

The dried powders were kept in a dessicator until required.

The small amounts of sample and internal standard were weighed on a Mettler micro-balance in plastic vials which fitted an apparatus known as a Wig-L-Bug and used for mixing. Electrodes of national spectrographic graphite were cut to shape on a lathe and purified by heating to a bright red heat. The powders were loaded by firm tamping.

A direct current arc was used to volatilize the samples in the anode. Current strength and gap width were controlled manually. Processing of the spectrographic plates was done in a Jaco processing unit at a constant time and temperature.

Identification of the spectrum lines and densitometry was carried out on a Steinheil photometer, and the necessary calculations, based on the methods of Kaiser (1941, p.1 and 1948, p. 297) were done on a Respectra calculating board.

Each sample was analysed in triplicate, and the final values recorded are the geometric means of the three results.

/The ...

The following three different spectrographic procedures were applied to each sample:

(a) Copper oxide was used as an internal standard, and graphite as buffer for the determination of Ti, Cr, Fe, Zr, Co, Ni, and Nb. This procedure differed from that of Jaycox (1955, p.347) in that no GeO_2 was used to dilute the samples. It is considered that the large amounts of SiO_2 present in the samples and standards had a dilution effect similar to that of GeO_2.

(b) As the internal standard, indium was employed with K_2SO_4 as buffer for the determination of Pb, Sn, Cu and Ag, following Ahrens et al. (1961, p.188).

(c) Zirconium served as internal standard and graphite as buffer for the determination of Nd, Pr, Ce, La and Y.

Details of these methods are given in Tables II, III and IV.

TABLE II

OPERATING CONDITIONS FOR THE DETERMINATION OF Co, Cr, Fe, Nb, Ni, Ti AND Zr

Standards:	Quartz mixed with specpure oxides of the relevant elements.
Internal Standard and buffer:	9 parts of black copper oxide (CuO) mixed with 20 parts graphite. This was mixed in a ratio of 1:1 with the sample or the standard.
Samples:	Ground, heated and reground portions of Dominion Reefs Mine conglomerate. This consisted predominantly of quartz. Many of the elements in the heated sample were present as oxides and the composition of the sample was thus similar to that of the standards. (An exception is Zr, present as zircon).
Anode:	Cut from "National" spectrographic graphite. Crater 4 mm. deep by 2.5 mm. in diameter, and walls of 0.5 mm. Purified by heating to redness.
Cathode:	Pointed "National" spectrographic graphite, purified by heating to redness.
Optics:	Quartz.
Exposure:	For 2 minutes at 8 amp. in a CO_2 atmosphere, using D.C. excitation. After the exposure the whole sample was consumed, and all the refractory ZrO_2 was distilled off.
Gap:	Manually maintained at 10 mm. The middle 7 mm. was photographed.
Emulsion:	Ilford N 50.
Development:	In ID2 diluted with 4 parts water, for 3 minutes at 24°C.

/Table 2...

TABLE II(Continued)

Line pair	Regression equation	Standard deviation S_x	Relative error E	Approximate detection limit
$\frac{\text{Co } 3453}{\text{Cu } 3307}$	y=.6528+.8974x	.0599	8.0%	30 ppm. CoO
$\frac{\text{Cr } 4254}{\text{Cu } 3307}$	y=.3494+.7565x	.0789	10.5%	30 ppm. Cr_2O_3
$\frac{\text{Fe } 3407}{\text{Cu } 3307}$	y=-.8819+.9755x	.0528	7.0%	1000 ppm. Fe_2O_3
$\frac{\text{Ni } 3492}{\text{Cu } 3307}$	y=.5691+.8597x	.0511	6.8%	30 ppm. NiO
$\frac{\text{Ti } 3371}{\text{Cu } 3307}$	y=-.1323+.8601x	.0703	9.4%	1000 ppm. TiO_2
$\frac{\text{Zr } 3438}{\text{Cu } 3307}$	y=.1230+.8386x	.1395	18.9%	100 ppm. ZrO_2

/Table 3.....

TABLE III

OPERATING CONDITIONS FOR THE DETERMINATION OF Ag, Cu, Pb, and Sn.

Standards:	Quartz mixture containing 10% muscovite. The muscovite was added to allow for the small amounts of alkali metals present in the sericite and chlorite in the samples of ore. To this was added known quantities of Ag, Cu, Pb and Sn.
Internal standard and buffer:	K_2SO_4 containing 2% In_2O_3. One part of this mixture was added to 100 parts of sample.
Sample:	Ground portions of Dominion Reefs Mine conglomerate. This consisted predominantly of quartz. The elements Cu and Pb were present mainly as sulphides, Sn as an oxide, and Ag both in gold and in sulphides.
Anode:	Cut from 'National" spectrographic graphite. Crater 4mm. deep and 2.5 mm. in diameter, and walls of 0.5 mm., purified by heating to redness.
Cathode:	Pointed "National" spectrographic graphite, purified by heating to redness.
Optics:	Quartz.
Exposure:	For 1 minute at 7 Amp, using D.C. excitation. The exposure was discontinued after complete distillation of the alkali elements.
Gap:	Manually maintained at 10 mm. The middle 7mm. was photographed.
Emulsion:	Ilford N 50.
Development:	In ID2 diluted with 4 parts water, for 3 minutes at 24°C.

/Table 3...

TABLE III (Continued)

Line pair	Regression equation	Standard deviation S_x	Relative error E	Approximate detection limit
$\frac{\text{Ag } 3280}{\text{In } 3256}$	Due to self reversal no regression equation calculated			1 ppm. Ag
$\frac{\text{Cu } 3274}{\text{In } 3256}$	$y=2.9190+.7589x$	.0411	5.5%	3 ppm. CuO
$\frac{\text{Pb } 2833}{\text{In } 3256}$	$y=1.0674+.5981x$	.0703	9.0%	10 ppm. PbO
$\frac{\text{Sn } 3175}{\text{In } 3256}$	$y=1.6939+.7256x$	.0416	5.6%	3 ppm. SnO_2

TABLE IV

OPERATING CONDITIONS FOR THE DETERMINATION OF Ce, La, Nd, Pr and Y

Standards:	Mixture of 90% quartz and 10% muscovite. To this was added known quantities of the oxides of the rare earths analysed for.
Internal standard and buffer:	Specpure graphite powder containing 0.1% ZrO_2. One part of the sample was mixed with ten parts of the graphite-zirconia mixture. In a few samples Zr was already present in significant amounts, and to these less ZrO_2 was added in order to maintain the total Zr-content at a constant level.*
Sample:	Ground, heated, and reground portions of Dominion Reefs Mine conglomerate. Most of the rare earths in the samples were present in the phosphate mineral, monazite.
Anode:	Cut from "National" spectrographic graphite. The crater was 6 mm. deep with a concave bottom, and 5 mm. in diameter. The crater walls were 0.5 mm. thick. This crater took 0.1 gm. of the sample-graphite mixture.
Cathode:	Graphite rod, 6 mm. thick, and having an ellipsoidal point.
Optics:	Glass.
Exposure:	For 90 secs. at 20 Amp. in an atmosphere of CO_2 Due to the high current intensity used, selective volatilisation was counteracted as is also proved by the fact that no glass beads were formed. During the exposure about 90% of the powder loaded into the anode was consumed.
Gap:	Manually maintained at 5 mm. The middle 3.5 mm. was photographed.
Emulsion:	Ilford N 50.
Development:	In ID2 diluted with 10 parts of water, for 10 minutes at 24°C.

*** Sample 1(a) was first diluted with pure quartz powder. In samples 1(a), 2, 15 and 16 dilution was used to minimize the effect of the low precision of the spectrographically determined Zr values.**

TABLE IV (Continued)

Line pair	Regression equation	Standard deviation S_x	Relative error E	Approximate detection limit
$\frac{\mathrm{Ce}\ 4138}{\mathrm{Zr}\ 3999}$	$y = .0527 + .9093x$	.0405	5.4%	.1% CeO_2
$\frac{\mathrm{Ce}\ 4134}{\mathrm{Zr}\ 3999}$	$y = .0962 + .9173x$	.0387	5.1%	.1% CeO_2
$\frac{\mathrm{La}\ 4334}{\mathrm{Zr}\ 3999}$	$y = .6963 + .9207x$	.0469	6.3%	.05% La_2O_3
$\frac{\mathrm{Nd}\ 4061}{\mathrm{Zr}\ 3999}$	$y = .1307 + .9325x$	.0516	6.9%	.03% Nd_2O_3
$\frac{\mathrm{Nd}\ 4109}{\mathrm{Zr}\ 3999}$	$y = .1153 + .8977x$	.0475	6.3%	.03% Nd_2O_3
$\frac{\mathrm{Pr}\ 4101}{\mathrm{Zr}\ 3999}$	$y = .2007 + .8987x$	.0577	7.7%	.1% PrO_4
$\frac{\mathrm{Y}\ 4375}{\mathrm{Zr}\ 3999}$	$y = 1.2313 + 1.1078x$	.0531	7.1%	30 ppm. Y_2O_3

In these tables (II,III,IV) the regression equation is expressed as:

$$y = \alpha + \beta x,$$

where y is $\log \dfrac{\text{Intensity of analysis line}}{\text{Intensity of internal standard line}}$

x is the log of the concentration of the element analysed for, and α and β are constants.

The constants α and β for every line pair used were calculated according to the formulas:

$$\beta = \frac{\sum_{i=1}^{n}(y_i - \bar{y})(x_i - \bar{x})}{\sum_{i=1}^{n}(x_i - \bar{x})^2}$$

$$\alpha = \bar{y} - \beta\bar{x}$$

where $\bar{y}$ and $\bar{x}$ are the average values for y and x (Shaw et al. 1954, p. 111).

The standard deviations and relative errors were calculated as follows:

For the three y-values obtained for sample 1, a mean value $\bar{y}$, and three deviations, $(y_1 - \bar{y}_I)$, $(y_2 - \bar{y}_I)$, and $(y_3 - \bar{y}_I)$ were calculated. The same procedure was adopted with samples 2, 3, and so on to sample m, and deviations $(y_4 - \bar{y}_{II})$, $(y_5 - \bar{y}_{II})$ .... to $(y_n - \bar{y}_m)$ were obtained.

The standard deviation of y was then obtained from:

$$S_y = \sqrt{\frac{\Sigma_1^n (y - \bar{y})^2}{n - m}}$$

where n is the total number of analyses, and m the total number of sample means used.

The standard deviation of x was derived from:

$$S_x = \frac{S_y}{\beta}$$

The relative error based on the geometric mean of three determinations, was calculated as follows:

$$E = \frac{100\ \text{antilog}\{(\bar{x} + Sx) - \text{antilog}\ (\bar{x} - Sx)\}}{2\sqrt{3}\ \ \text{antilog}\ \bar{x}}$$

(See Shaw, 1960, p. 467)

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This simplifies to:

$$E = 28.8684 \quad \frac{z^2 - 1}{z}$$

where z is the antilog of S_x.

This "relative error" is thus closely related to the relative deviation, $\frac{100\ S}{\sqrt{n}\ \bar{x}}$, usually reported as a measure of precision in spectrographic analysis. The relative deviation is considered to be an unrealistic measure of precision for spectrographic data, as it takes no cognisance of the lognormal distribution of the spectrographic error (Ahrens, 1961, p.118).

From exposures on moving plates it was established that the elements Co, Cr, Fe, Ni and Ti, when arced according to the conditions listed in Table II had volatilization trends similar to that of Cu. The intensity of the analysis lines reached a maximum soon after arcing was commenced, and decreased to small values just before the entire sample was consumed. The use of copper as an internal standard for these elements was therefore justified.

The distillation trends of Zr (Fig. 1) and Nb differed appreciably from that of Cu. The intensity of their analysis lines increased slowly as arcing proceeded, and when nearly all of the more volatile elements were consumed, their intensities increased sharply towards a maximum. The analysis lines persisted for a short time even after the entire sample was consumed. Accordingly arcing was prolonged for a few seconds after completion of the samples.

Even when using this total distillation method the use of Cu as an internal standard for Zr was not as successful as with Ti, Cr, Fe, Co and Ni, as proved by the relative errors listed in Table II. The development of a better spectrographic method for Zr was not considered to be justified for the present investigation, as the variation in Zr-values from a low value of 0.21% to a high value of 1.65% in the different samples is of such a high magnitude that compared to it the spectrographic error can be regarded as insignificant.

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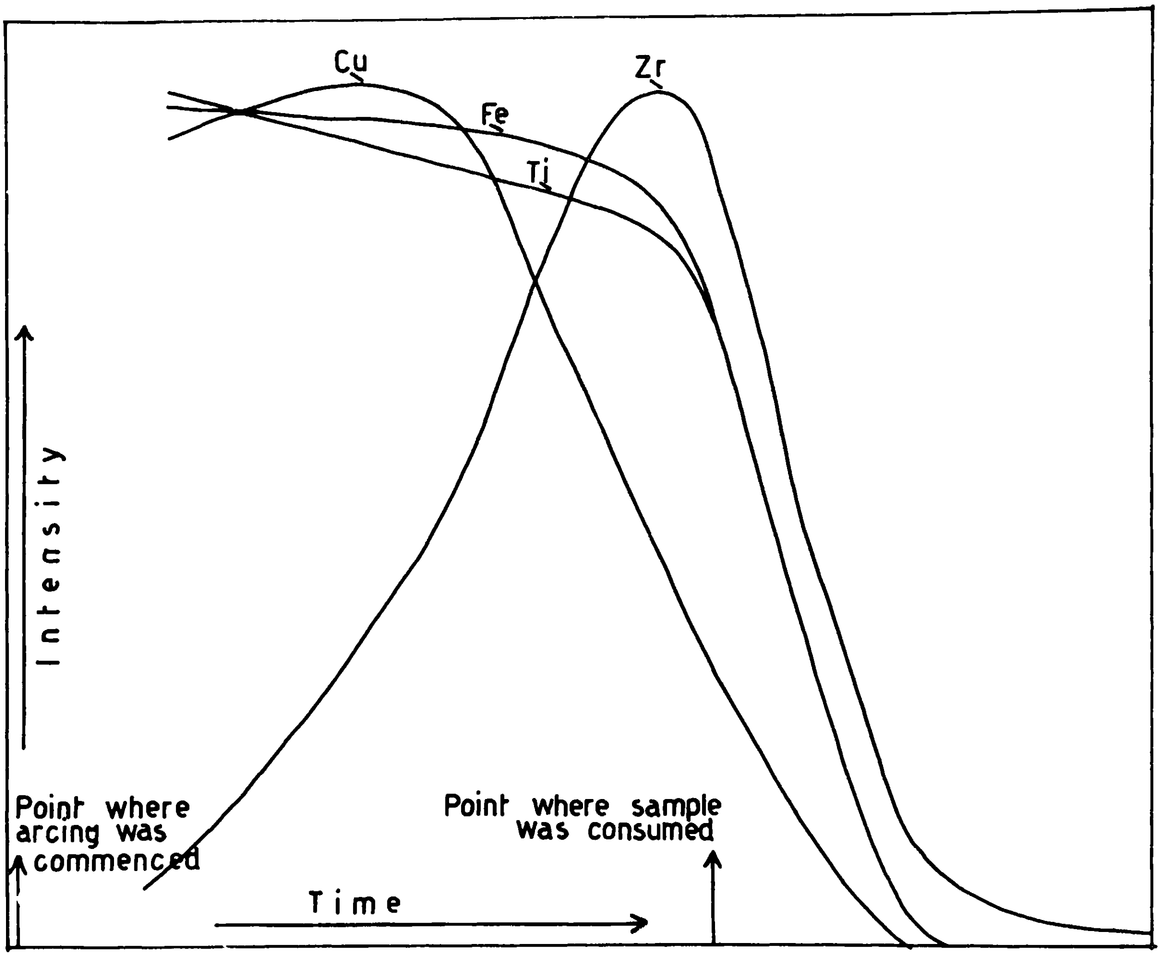


Figure 1. Volatilization trends of Cu, Fe, Ti, and Zr when arced according to the conditions specified in Table 2

In the method applied to the volatile elements listed in Table III several difficulties were encountered. Some of these elements (Zn and As) had unsatisfactory detection limits, and could be determined in only a few samples. The excitation of the volatile elements was also affected appreciably by variations in the matrix. The large amounts of sulphur present as sulphides in some samples had an adverse effect on the precision. Heating of the samples to drive off the sulphur is not advisable, as some of the volatile elements could be distilled off simultaneously.

Various methods had been proposed in the literature to minimize the matrix effect. It is claimed for instance that the Stallwood jet (Stallwood, 1954; Hawley et al., 1956) improves precision and accuracy. However, this involves a considerable loss of sensitivity.

The addition of large amounts of buffers is a technique often resorted to. Several buffers consisting of alkali metal salts were tried, but the concomitant loss of sensitivity due to dilution made the determination of many elements in the present suite of samples impossible. The results obtained by using the method described by Ahrens (1954) are listed in Table V. The precision obtained for the volatile elements given in Table III is satisfactory, but the concentrations of As and Zn in many of the samples were below the limits of detection.

The rare earth elements listed in Table IV are present in appreciable amounts in many of the samples. Large amounts of C were added as buffer, and as shown by exposures on moving plates selective volatilization was absent when the samples were burnt at 20 amps. in an atmosphere of CO_2. This method resulted in detection limits of only 0.03% to 0.3% for most of the rare earths except for Y. The detection limits of 30 ppm. for this element enabled its determination in all the samples.

(2) Contamination

Contamination is a possible cause of inaccuracy and to avoid this hazard it was necessary to take special precautions. To test for contamination pure quartz crystals were ground at intervals in the same mortars and under the same conditions as the samples, and portions of the quartz powder were analysed spectrographically. The following elements were usually detected: Al, Fe, Cu, B and Mg.

/The aluminium......

The aluminium was evidently derived from the mullite mechanical mortar used for the fine grinding. As aluminium was not analysed for quantitatively, the trace amounts of this element added to the samples did not affect the results.

The small amounts of iron were evidently derived from the tools used in crushing the samples. In every case the amount of iron by which the quartz was contaminated was estimated to be of the order of XO ppm. As the samples of conglomerate were easier to crush, and contained less jagged tough splinters of quartz than the pure quartz crystals, the level of contamination of the sample is considered to be less. The smallest amounts of iron found in the conglomerate samples, viz. 4290 ppm. is already of such a magnitude that the contribution of iron by contamination can be regarded to be negligible.

Copper lines were just visible in some instances. The source of the copper contamination is unknown. It is considered that the contribution of copper by contamination may be significant in those samples in which less than 5 ppm. of CuO were found. In Table V all copper values thought to be suspect due to contamination appear in brackets.

Boron and magnesium were not analysed for, and the presence of their lines in the spectra did not cause any complications. They could have been present in the electrodes employed.

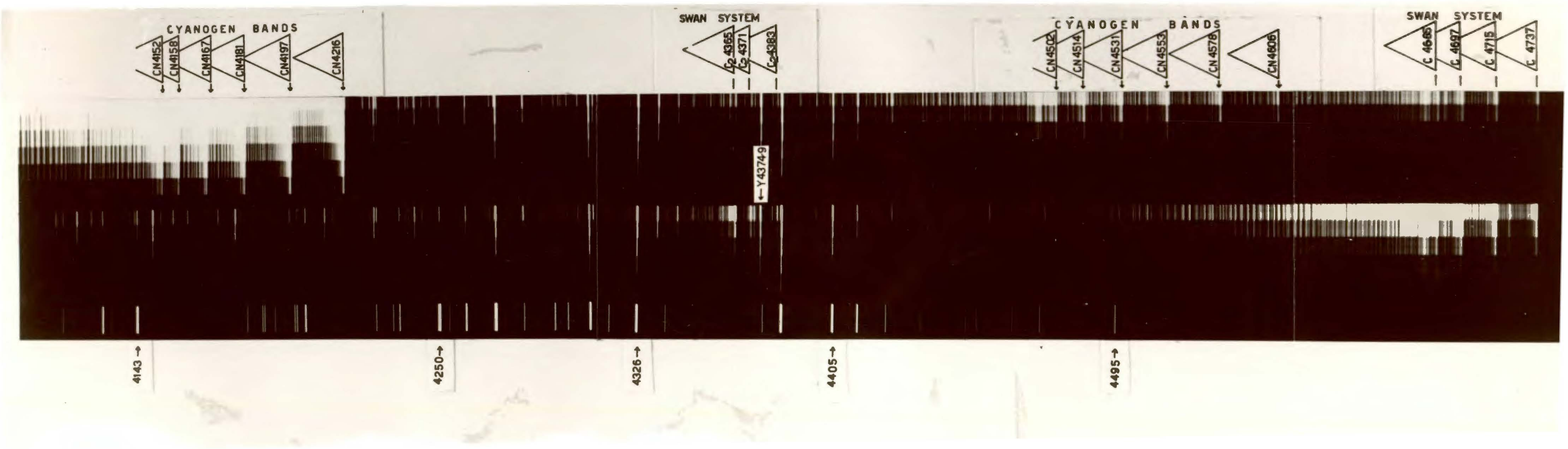
(3) Coincidenses of lines

The lines used in the analysis and listed in Tables II, III, and IV were chosen only after careful consideration of possible coincidences of lines of other elements also present in the samples. As pointed out by Ahrens (1961, p. 73) some publications contain errors because line coincidences were overlooked. One cause of this type of mistake is that too much weight is attached to the relative intensities listed for lines in the M.I.T. Tables.

In a few examples the use of analysis lines which contain interfering lines could not be avoided and in those examples a correction was applied according to the method described by Ahrens (1961, p. 167). Examples are Zr 3999.0 which had to be corrected for Ti 3999.0, Cu 3273.96 which coincides with Fe 3274.4, and Y 4374.9 which coincides with a C_2 line.* The latter example will be described in more detail below.

* (The iron line (Fe 3174.96) coinciding with Sn 3175.0 had a negligible effect in the samples employed

Plate I



Three spectra: (a) The cyanogen bands (upper spectrum) developed when graphite electrodes are arced in air.

(b) The elimination of the cyanogen bands when arcing is done in an atmosphere of CO_2 (middle spectrum). Under these conditions the Swan System of C_2 bands is intensely developed, and one of its rotational compounds interferes with Y4374·9.

(c) The lower spectrum is of iron, for purposes of comparison.

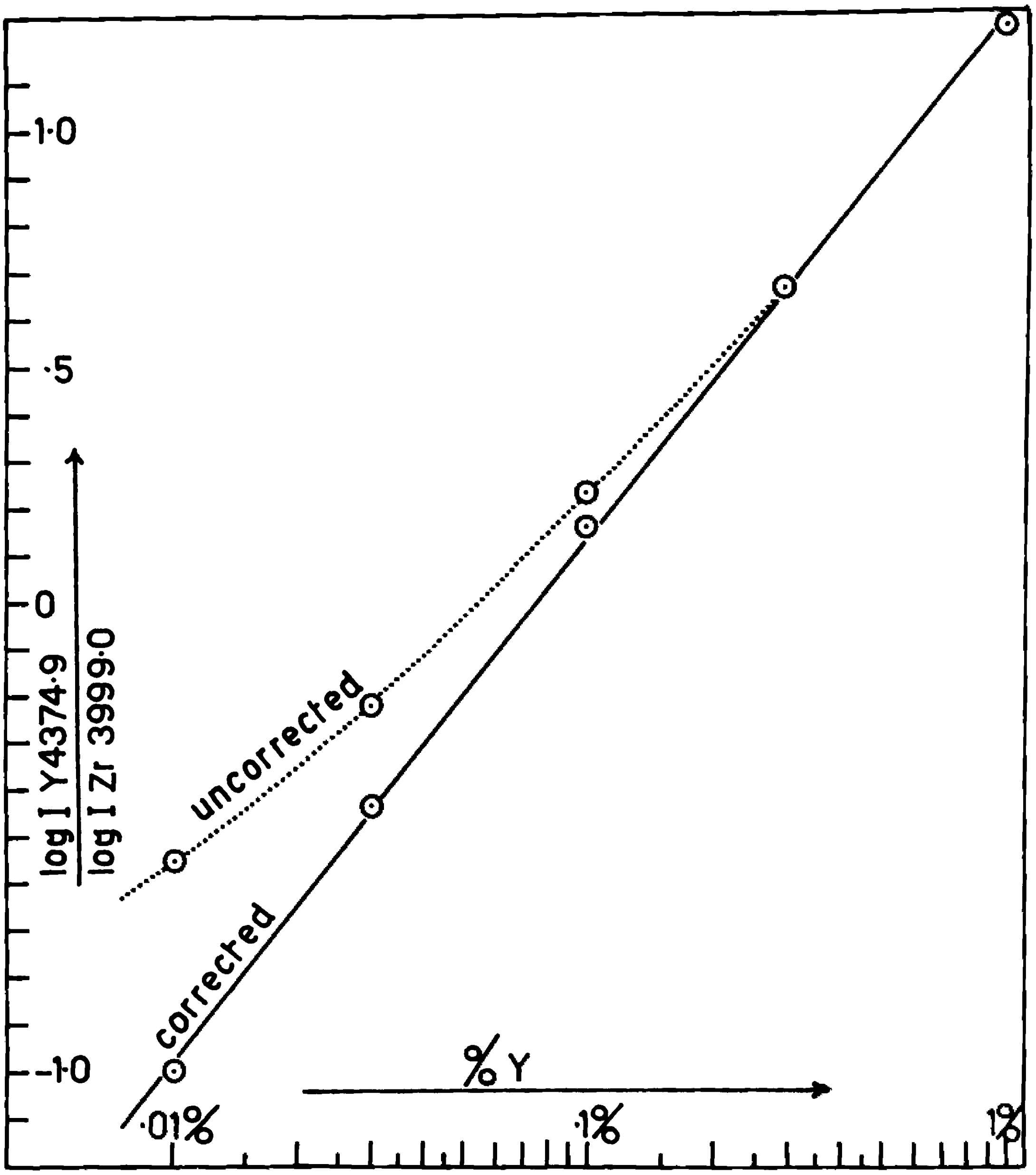


Figure 2. Working curve for Y4375· showing its form after correction was made for a coincidence of a rotational compound of the Swan System of C_2 bands. Each point plotted is the mean of three values.

Some lines of the rare earths fall within the cyanogen bands. Arcing was therefore carried out in an atmosphere of CO_2, which resulted in the elimination of the cyanogen bands. Under the conditions of arcing however, the Swan System of C_2 bands were developed intensely (Plate 1). One of the rotational compounds of the system having its head at 4382.5Å and degraded towards the violet end of the spectrum coincides with Y 4374.9. Correction could be applied by measuring a nearby compound at about 4374.5Å, which is of nearly the same intensity as the interfering compound. The intensity ratio of these compounds was first determined on spectra containing no Y-lines and this ratio was then subsequently employed for the correction (Fig. 2).

III. RESULTS OBTAINED

The results of all the spectrographic analyses, as well as chemically determined P_2O_5, ThO_2, and U_3O_8 are listed in Table V. The gold values were supplied by the management of the Dominion Reefs Mine.

Values thought to be somewhat suspect, due to matrix effects, contamination effects, or values close to the threshold of detectibility are also included in Table V but are given in brackets.

IV. DISCUSSION OF RESULTS

A. INTRODUCTION

Previous workers on the Dominion Reef based their theories mainly on stratigraphical and mineralogical evidence and also on the available geochemical evidence which was mainly limited to the distribution of gold, uranium, and to a certain extent, pyrite and platinoids. Although a vast amount of literature is available on the genesis of the Witwatersrand ores in general, the work dealing specifically with the Dominion Reef is of a very limited nature and geochemical work is almost lacking, except for the elements mentioned above.

The most salient features of previous theories can be summarized as follows:

/The detrital...

TABLE VI

LOGARITHMS OF CONCENTRATIONS OF ELEMENTS IN VARIOUS SAMPLES FROM THE DOMINION REEFS MINE (BASED ON THE RESULTS GIVEN IN TABLE 5)

Sample No.	Au log dwts.	CuO log %	CoO log %	Cr_2O_3 log %	Fe_2O_3 log %	NiO log %	PbO log %	P_2O_5 log %	SnO_2 log %	ThO_2 log %	TiO_2 log %	U_3O_8 log %	ZrO_2 log %	Y_2O_3 log %.	CeO_2 log %	La_2O_3 log %	Nd_2O_3 log %	PrO_4 log %
1(a)	2.11	-1.09	-0.55	-0.35	0.98	-0.61	-0.89	0.08	-0.49	0.28	0.83	1.00	-0.22	0.03	0.51	0.27	0.18	0.08
1(b)	-	-1.25	-1.89	-1.77	0.43	-1.69	-0.76	-1.05	-1.89	-1.30	-0.40	-0.47	-1.39	-1.29	-0.92	-1.12	-	-
2	1.00	-1.03	-0.95	-0.42	1.21	-0.87	0.05	0.04	-1.25	-0.19	0.50	0.44	-0.22	-0.19	0.32	0.05	-0.13	-0.51
3	-	-3.32	-	-1.40	-.22	-	-1.67	-0.92	-3.34	-1.57	-0.31	-1.85	-1.16	-1.64	-0.89	-1.09	-	-
4	-	-3.29	-	-1.97	0.21	-	-2.51	-1.40	-3.03	-2.10	-0.63	-1.82	-1.55	-1.24	-	-	-1.12	-
5	0.18	-3.25	-	-1.91	0.10	-	-2.91	-1.30	-3.37	-2.10	-0.67	-2.70	-1.51	-2.34	-	-	1.03	-
6	0.53	-4.35	-1.61	-1.09	0.86	-1.54	-1.08	-0.57	-2.56	-1.09	0	-0.92	-0.89	-1.04	-0.43	-0.66	-0.77	-0.89
7	-	-2.56	-2.47	-1.95	-.37	-2.18	-3.16	-1.15	-3.36	-2.52	-0.65	-2.22	-1.67	-2.21	-	-	-1.04	-
8	0.34	-2.64	-2.17	-1.89	1.00	-2.07	-2.71	-	-3.15	-1.92	-0.19	-2.22	-1.53	-2.00	-	-	-	-
9	1.73	-2.03	-1.96	-2.18	0.04	-2.14	-1.58	-	-2.78	-2.84	-0.66	-1.92	-1.78	-1.77	-	-	-	-
10	-	-2.37	-1.55	-1.82	0.86	-1.62	-1.87	-	-2.71	-1.92	-0.31	-1.59	-1.46	-1.55	-	-	-	-
11	1.37	-2.79	-1.34	-2.02	-.31	-1.49	-0.75	-1.05	-2.44	-1.12	-0.44	-0.38	-1.68	-1.28	-	-	-	-
12	-	-1.80	-1.78	-1.98	0.43	-1.93	-1.27	-	-2.92	-1.92	-0.55	-2.05	-1.55	-1.80	-	-	-	-
13	1.49	-3.05	-1.51	-1.70	0.43	-1.64	-0.92	-	-2.32	-1.77	-0.51	-1.11	-1.32	-1.60	-	-	-	-
14	1.90	-2.04	-1.58	-1.92	0.32	-1.69	-0.96	-1.22	-1.22	-1.52	-0.51	-0.85	-1.56	-1.47	-	-	-1.07	-
15	0.18	-2.39	-1.31	-0.62	0.96	-1.32	-1.22	-0.12	-2.64	-0.68	0.48	-0.77	-0.28	-0.60	0.07	-0.17	-0.35	-0.68
16	0.18	-2.62	-1.38	-0.77	0.88	-1.35	-1.35	-0.12	-2.63	-0.70	0.45	-0.72	-0.27	-0.55	0.12	-0.14	-0.30	-0.64
17	2.00	-3.47	-	-	-	-	-1.38	-	-2.32	-1.40	-	-0.77	-	-1.26	-	-	-	-
18	-	-3.14	-1.95	-1.94	0.04	-2.11	-2.52	-	-3.16	-2.10	-0.58	-0.82	-1.68	-2.12	-	-	-	-
20	-	-3.56	-2.11	-1.83	-.28	-2.04	-2.92	-	-3.49	-2.10	-0.51	-2.05	-1.40	-2.28	-	-	-	-

1. The Detrital Theory

The following different variants of the detrital theory are propogated by its exponents.

(a) As conceived by Liebenberg (1955, p. 167), and Ramdohr (1958, p. 42) the minerals uraninite, gold and most of the sulphides are of a detrital origin, but the gold and sulphides show the results of later metamorphic influences which induced them to change their original detrital forms by plastic flow and limited hydrothermal redistribution, whereas uraninite, chromite, cassiterite and monazite released strain by cataclasis.

The uraninite grains are considered as having been derived from granitic rocks, pegmatites, and high temperature veins.

The interpretation advanced in Part I of this thesis is in accordance with the views expressed by Liebenberg, Ramdohr and also in general, by Malan (1959, p. 58).

It must be emphasized, however, that none of the detritalists could escape from the conclusion that certain elements, as they now appear in the Witwatersrand and Dominion Reef beds, were brought in hydrothermally. In the samples described in Part I of this thesis the present author also came to the conclusion that the sphalerite, a small portion of the galena, and also some pyrite were introduced by hydrothermal solutions. These minerals were deposited in tiny veinlets and porous zones and did not permeate the conglomerate. The galena and pyrite occurring outside these veinlets are of another textural variety and are not considered to be genetically connected with the veinlets. As this hydrothermal action only affected small portions of the conglomerate, and were of a limited extent, lateral secretion may be called upon to explain their origin, as opposed to hydrothermal solutions which originated from a deep-seated magma.

(b) Another variant of the detrital theory was proposed by Koen (1961, p.23). He interpreted the uraninite as representing detrital nodules precipitated in ancient marshes, and redistributed by wave action. Koen expressed no views on the origin of the sulphides and the gold. However, he is of the opinion that chromite and zircon in the conglomerate are detrital minerals.

Koen's views can thus be regarded as adapting the detrital theory to an origin from terrestrial waters by precipitation of the uraninite.

(2) The Groundwater and Hydrothermal Theory

In addition to the theory advanced by Koen as outlined above, Davidson, (1964 (a), p.175, 1964 (b), p.177), also subscribes to the idea that uraninite was precipitated from groundwaters. However, according to him, precipitation occurred in the conglomerate after its deposition. Similar views were proposed by Derry (1960, p.926) and Joubin (1960, p.1751) for the uraninite in the Blind River ores, Canada.

Two further modes of origin were suggested by Davidson for the other constituents of the Dominion Reef, viz. a hydrothermal origin for gold and sulphides (Davidson, 1962, p.159) and a detrital origin for chromite and zircon. Davidson's theory can, therefore, strictly be regarded as polygenetic.

B. SYMPATHETIC VARIATIONS

The geochemical data presented in Table V includes the distribution of a number of elements in addition to uranium, gold and pyrite. As will be shown below a clear conception of the processes which gave rise to this pattern of element distribution may shed new light on the origin of the host minerals.

The authors referred to above are unanimous on one point only viz. that the minerals zircon, chromite, monazite, cassiterite, and the ilmenite from which leucoxene was derived are of detrital origin. Furthermore they subscribe to the premise that elements which display a sympathetic variation have a common origin (Liebenberg, 1955, p. 215 Davidson, 1957, p. 678, Koen, 1961, p. 42). These two premises were accordingly accepted as guiding principles in trying to interpret the distribution pattern of the elements listed in Table V.

/Previous.......

(1) Previous Work

The words "sympathetic variation" or "sympathetic relationship" have been used by various workers on the Witwatersrand ores in the past, most commonly in connection with gold and uranium values in the banket (Liebenberg, 1955, p. 215; Simpson, 1951, p. 131; Malan, 1959, p. 83). These authors evidently accept the term to mean that in the auriferous conglomerate an increase in gold is associated with an increase in uraninite and vice versa.

According to Ramdohr (1958, p. 42)".. the rare components like gold, "Uranpecherz", and platinoids, are often concentrated together, which of course would be expected according to every placer theory." However, Koen (1961, p.42) claims"... wide fluctuations in the relative abundance of uraninite and other heavy detrital minerals (as opposed to the virtually constant ratio between minerals such as zircon and chromite in the same reef)".

The statements of the authors favouring a sympathetic relationship of gold and uranium are usually based on the critical examination of polished sections. In some instances attempts are made to illustrate thi sympathetic relationship by means of histograms (Malan, 1959, Plate 2). In these instances the tendency of an increase in, say, uranium values in the ores to be associated with an increase in, say, gold values, is disturbed by other variations, and a critical reader may be reluctant to agree that a sympathetic relationship does exist, or that it is significant. The conclusion in these examples that such a relationship is significant must be regarded as purely subjective.

As shown by Papenfus (undated thesis, p. 53) the ratio of the monthly production of gold and osmiridium at Government G.M. Areas and New State Areas tends to remain constant as long as milling is confined to one reef only. Papenfus interpreted this nearly constant ratio as proof that gold and osmiridium grades varied sympathetically.

Another term "sympathetic ratio", used by Cousins (1956, p.95) is defined by him as the similarity in distribution curves of two sets of values when plotted as cumulative logarithmic values on a probability scale. Such a similarity simply means that the dispersion of the logarithms of the two sets of values, which could for instance be

/expressed.....

expressed as standard deviations, is the same. The "sympathetic ratio" used by Cousins is thus completely different from what should be understood by the words "sympathetic variation" as used by Liebenberg and others. In the final issue, the judgement as to whether the curves as plotted by Cousins are sufficiently parallel, or show sufficiently similar patterns or not, is also subjective. The relationship which may exist between pairs of values obtained from the same samples is lost by Cousins' treatment. In the opinion of the author it is a moot point whether similar dispersions in two sets of values necessarily indicate a common or a similar causality, or even a sympathetic variation. It is too probable for unrelated sets of values to have similar dispersions, as also shown, for instance, by Koen (Discussion of paper by Cousins, 1956).

Sympathetic variations in the sense as used by Liebenberg can most clearly be illustrated by means of scattergrams as illustrated in Figs. 3 and 4. In these diagrams logarithms of the values are used for the following reasons:

(a) On a linear scale variations of the same size, say, x%, of small values and large values do not appear to be of the same size. This distortion is eliminated on a logarithmic scale.

(b) In Cousins' paper (1956, p. 107) the examples of cumulative frequency curves of ore values for deposits of hydrothermal, pneumatolytic, magmatic and placer origin, as well as gold, pyrite, and uranium values from the Witwatersrand, all conform closely with the lognormal distribution. It is highly probable that the value distributions of the minor components of the Dominion Reef are also lognormal, and they should therefore be handled on a logarithmic scale.

In Table VI the logarithms of the values of Table V, expressing all the values as percentages, are listed. For gold, the logarithms of the dwt.-values are given. The elements Ag, Mo, Nb and Zn are omitted, as all the values reported for these elements are regarded as approximate only. The logarithmic values can now be used to prepare scattergrams, such as the ones shown in Figs. 3 and 4.

/(2) Correlation....

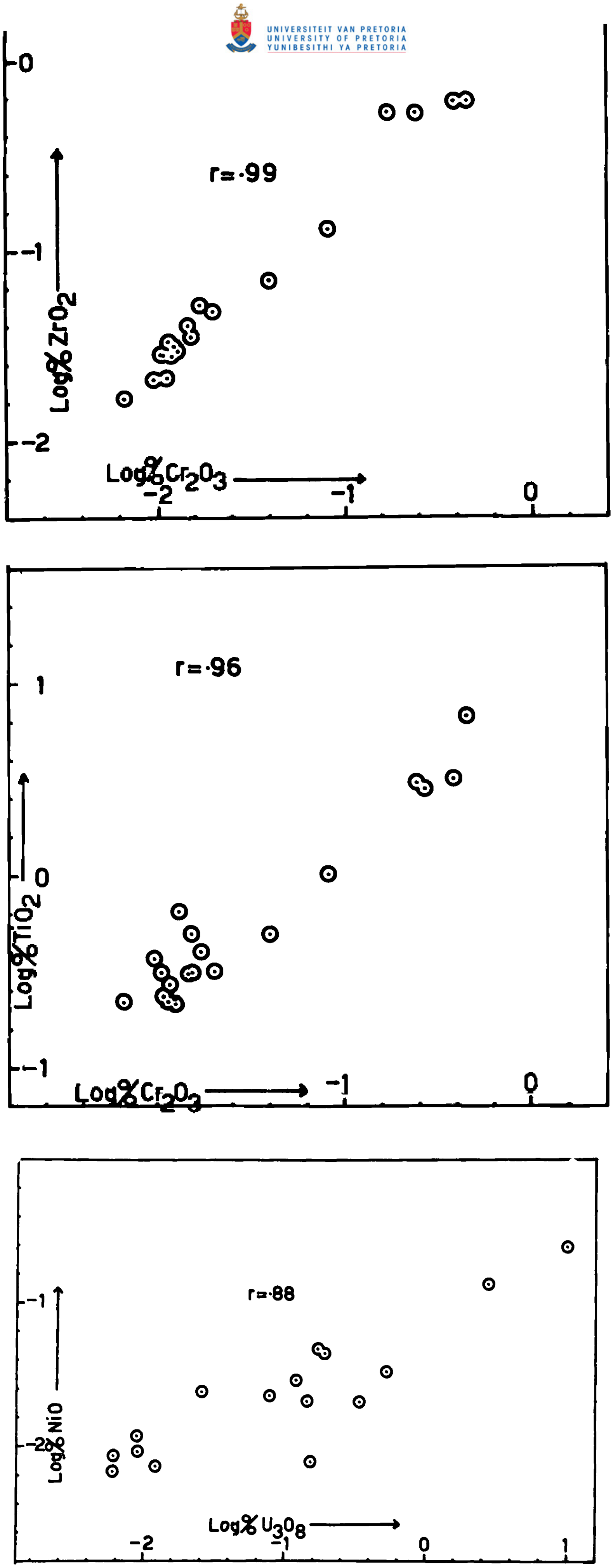


Figure 3. Scattergrams illustrating high correlations of Zr and Cr, Ti and Cr, and Ni and U.

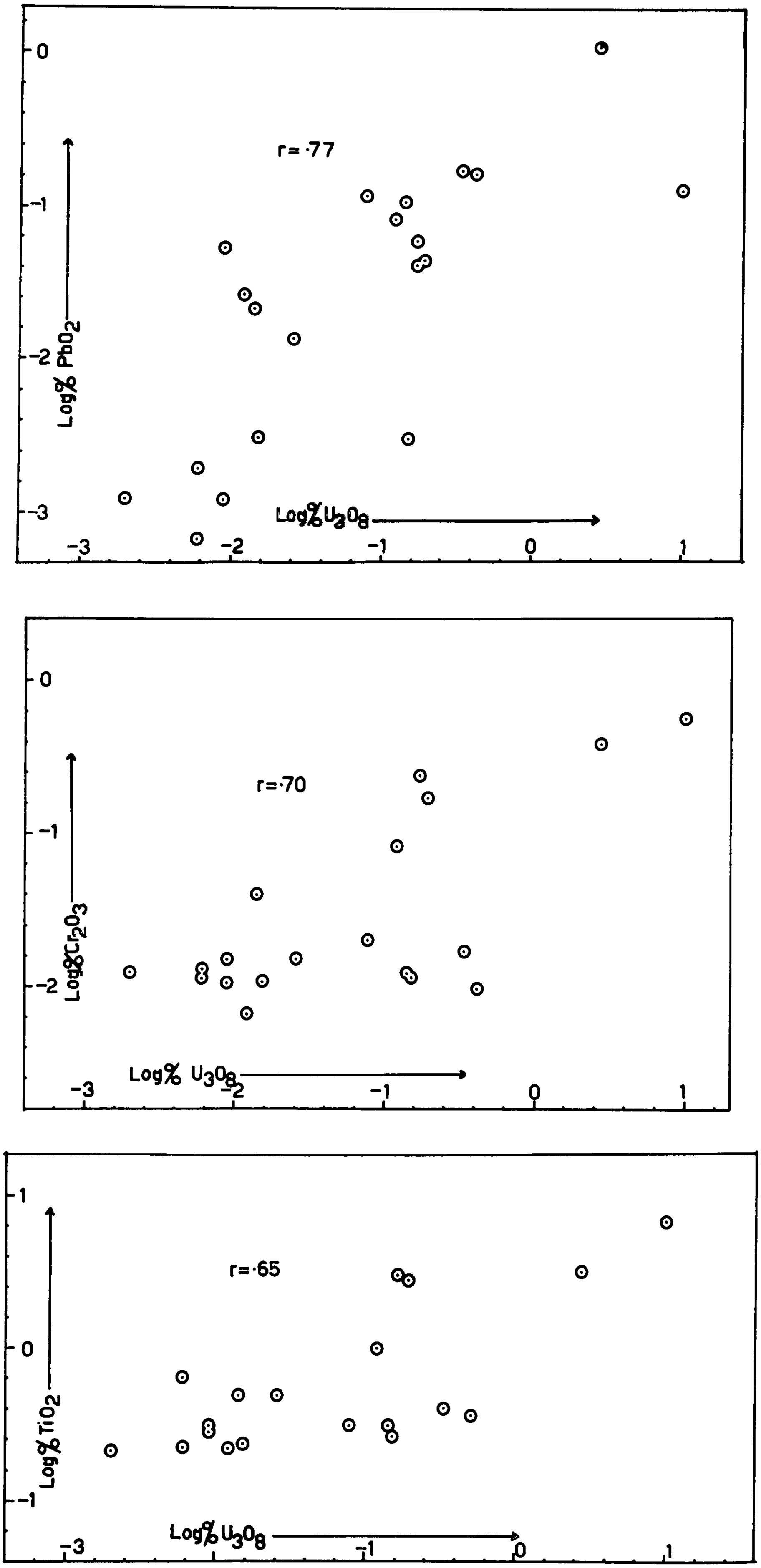


Figure 4. Scattergrams showing correlations of Pb and U, Cr and U, and Ti and U. The degrees of correlation are lower than in figure 3.

TABLE V

RESULTS OF SPECTROGRAPHIC AND CHEMICAL ANALYSES OF SAMPLES FROM THE DOMINION REEFS MINE

Sample No.	Ag ppm.	Au * dwt. /ton	CuO ppm.	CoO ppm.	Cr_2O_3 ppm.	Fe_2O_3 %	NiO ppm.	PbO_2 ppm.	P_2O_5 (1) %	SnO_2 ppm.	ThO_2 (1) ppm.	TiO_2 %	U_3O_8 (1) %	ZnO ppm.	ZrO_2 ppm.	MoO ppm.	Nb_2O_3 ppm.	Y_2O_3 %	CeO_2 %	La_2O_3 %	Nd_2O_3 %	PrO_4 %
1(a)	(10)	130	815	2830	4500	9.58	2500	1300	1.2	3200	19000	6.61	9.98	-	16500	-	-	1.07	3.25	1.86	1.53	1.19
1(b)	-	-	560	130	170	2.67	210	1700	0.09	130	500	0.40	0.34	-	410	-	-	0.051	0.12	0.076	-	-
2	(30)	100	928	1110	3800	16.1	1400	11100	1.1	570	6500	3.15	2.78	(290)	6100	(500)	-	0.65	2.07	1.13	0.74	0.31
3	-	tr.	(4.8)	tr.	400	0.61	-	210	0.12	(4.6)	270	0.49	0.014	-	690	-	-	0.023	0.13	0.081	-	-
4	-	tr.	(5.1)	-	110	1.64	-	31	0.04	(9.4)	80	0.24	0.015	-	280	-	-	0.058	-	-	0.076	-
5	-	1.5	(5.6)	-	120	1.26	-	12	0.05	(4.3)	80	0.21	0.002	-	310	-	-	0.0046	-	-	0.094	-
6	(2)	3.4	(.44)	250	820	7.24	290	830	0.27	27	810	1.00	0.12	-	1300	-	-	0.091	0.37	0.22	0.17	0.13
7	(3)	tr.	28	(34)	110	0.43	(66)	69	0.07	(4.3)	(30)	0.22	0.006	-	2100	-	-	0.0062	-	-	0.091	-
8	-	2.2	23	(68)	130	9.92	(85)	20	n.d.	(7.1)	120	0.65	0.006	-	300	-	(300)	0.010	-	-	-	-
9	-	54	93	110	65	1.10	(73)	260	n.d.	17	(20)	0.22	0.051	(70)	160	-	(200)	0.017	-	-	-	-
10	(1)	tr.	43	280	150	7.18	240	130	n.d.	20	120	0.49	0.026	-	350	-	(600)	0.028	-	-	-	-
11	(2)	23.4	16	460	95	0.49	320	1800	0.09	36	750	0.36	0.42	-	210	-	(1100)	0.053	-	-	-	0.090
12	(3)	tr.	16	170	100	2.69	120	540	n.d.	12	120	0.28	0.009	(700)	280	-	(400)	0.016	-	-	-	-
13	(4)	30.8	9.0	310	200	2.71	230	1200	n.d.	48	170	0.31	0.078	(170)	480	-	(600)	0.025	-	-	-	-
14	(6)	79.5	90	260	120	2.07	200	1100	0.06	610	300	0.31	0.14	(250)	280	-	(1000)	0.034	-	-	0.086	-
15	(2)	1.5	41	490	2400	9.17	480	600	0.75	23	2110	2.99	0.17	(30)	5200	(200)	(4000)	0.25	1.18	0.67	0.45	0.21
16	(2)	1.5	24	420	1700	7.53	450	450	0.75	23	2000	2.81	0.19	(30)	5400	(200)	(3000)	0.28	1.31	0.72	0.50	0.23
17	(3)	99	(3.3)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	48	400	n.d.	0.17	n.d.	n.d.	-	n.d.	0.055	-	-	-	-
18	-	tr.	(7.2)	110	110	1.10	(78)	30	n.d.	(6.9)	80	0.26	0.15	-	210	-	(200)	0.0076	-	-	-	-
20	-	tr.	(2.8)	(77)	150	0.53	(91)	12	n.d.	(3.3)	80	0.31	0.009	-	400	(100)	-	0.0052	-	-	-	-

* : Values obtained by fire assay

(1): Values obtained chemically

The remainder of the values was obtained spectrographically

n.d: Not determined

- : Below the limits of detection

(2) Correlation Coefficients

In order to express the intensity of the relationship as revealed in the scattergrams quantitatively, and to judge objectively whether this relationship is significant, use was made of the product-moment correlation coefficient. Estimates of the coefficient were calculated according to the formula:

$$r = \frac{N\sum_{0}^{N} X_1X_2 - \sum_{0}^{N} X_1 \sum_{0}^{N} X_2}{\left\{N\sum_{0}^{N} X_1^2 - \left(\sum_{0}^{N} X_1\right)^2\right\}^{\frac{1}{2}} \left\{N\sum_{0}^{N} X_2^2 - \left(\sum_{0}^{N} X_2\right)^2\right\}^{\frac{1}{2}}}$$

where r is the estimate of the correlation coefficient, X_1 and X_2 are the logarithms of the concentrations of element 1 and 2 determined on the same sample, and N is the number of value pairs used (Young, 1962, p.130) The correlation coefficients always have values lying between +1 and -1. A value of +1 indicates that a perfect quantitative relationship exists between the two sets of data used, and a value of -1 indicates that a perfect inverse relationship exists. Values of the correlation coefficient above 0.7 (or below -0.7) can be regarded as indicating a high degree of correlation, whereas for values below about 0.5 (or above -0.5) the degree of correlation would be weak. A value of 0 indicates the absence of correlation.

A clear distinction must be made between the actual correlation coefficient ρ of a population, and an estimate of the correlation coefficient, r, which is calculated by using a limited sample of the population.

The precision of the estimate, r, of the correlation coefficient is dependant on the number of samples used, and is easily evaluated from the data given in Table VII, using standard methods, for instance as illustrated by Snedecor (1961, p.175). Choosing conditions comparable to those of many of the values of Tablo VII viz. $r = 0.80$ and $N = 20$, the 95% confidence limits of the correlation coefficient can be shown to be: $0.755 \leq \rho \leq 0.838$, where ρ is the correlation coefficient, as opposed to its estimated value, r. For the values calculated from a smaller number of value pairs, i.e. where N is much smaller than 20, the 95% confidence limits will be farther apart than in the example above.

/In order...

In order to judge whether an estimate of the correlation coefficient is significant or not, use was made of tables published by Fisher and Yates (1957, p. 59). The levels of significance obtained were also entered in Table VII. A level of significance of 0.1% indicates that the chances of obtaining an estimate r, of the correlation coefficient as high as the one actually listed in Table VII from the limited set of data used, assuming that the variations are actually random and unrelated, is less than 1 in 1000. Similarly, levels of significance of 1%, 5% and 10% indicate chances of 1 in a 100, 1 in 20 and 1 in 10 of obtaining such a high r value where ρ is actually 0. Where the chances are less than 1 in 10, the letters n.s. were entered, meaning that the estimated correlation coefficient is not significant.

However, in cases where the estimated correlation coefficient found is not significant, the assumption cannot be made that no correlation exists. It simply means that the data available are not sufficient to establish whether such a correlation does or does not exist. Similarly, a high r value calculated from a few pairs of values may not be significant, or may have a low level of significance, whereas a low r value calculated from many sets of values may be very highly significant.

By using the above method, the judgement as to whether a correlation is significant or not is thus no longer subjective. In this sense also, a sympathetic variation of two elements can be defined as a positive correlation which can be shown to be significant.

Reference to Table VII shows that many of the 101 pairs of elements listed possess significant correlations, 45 of them on the 0.1% level.

(3) Interpretation of Correlations

In the interpretation of the significant correlations due attention should be given to the following possible causes:

(i) Elements present in the same host minerals can be expected to exhibit a high degree of correlation.

(ii) Elements introduced simultaneously into the ore by the same mechanism can be expected to have significant correlations.

Elements having closely similar geochemical properties usually will occur in the same mineral species and partake in the same

/processess....

TABLE VII

ESTIMATED CORRELATION COEFFICIENTS FOR 101 PAIRS OF ELEMENTS IN 20 SAMPLES OF CONGLOMERATE FROM THE DOMINION REEFS MINE

	Au	Cu	Co	Cr	Fe	Ni	Pb	P	Sn	Th	Ti	U	Zr	Y
Au		.33 12 n.s.	.29 10 n.s.	-.15 11 n.s.	-.31 11 n.s.	.12 10 n.s.	.43 12 n.s.	-.05 8 n.s	.64 12 5%	.01 12 n.s.	-.13 11 n.s.	.42 12 n.s.	-.30 11 n.s.	.10 12 n.s.
Cu			.41 16 n.s.	.29 19 n.s.	.38 19 n.s.	.44 16 10%	.48 20 5%	.41 12 n.s.	.65 20 1%	.38 20 10%	.38 19 n.s.	.47 20 5%	.29 19 n.s.	.44 20 10%
Co				.77 16 .1%	.59 16 2%	.96 16 .1%	.78 16 .1%	.76 9 2%	.80 16 .1%	.88 16 .1%	.76 16 .1%	.87 16 .1%	.75 16 .1%	.54 16 5%
Cr					.68 19 1%	.87 16 .1%	.52 19 5%	.98 12 .1%	.55 19 2%	.89 19 .1%	.96 19 .1%	.70 19 .1%	.99 19 .1%	.86 19 .1%
Fe						.64 16 1%	.47 19 5%	.82 12 1%	.52 19 5%	.56 19 2%	.75 19 .1%	.51 19 5%	.70 19 .1%	.68 19 1%
Ni							.72 16 1%	.84 9 1%	.80 16 .1%	.95 16 .1%	.87 16 .1%	.88 16 .1%	.85 16 1%	.59 16 5%
Pb								.59 12 5%	.77 20 .1%	.70 20 .1%	.52 19 5%	.77 20 .1%	.51 19 5%	.75 20 .1%
P									.51 12 10%	.90 12 .1%	.99 12 .1%	.73 12 1%	.97 12 .1%	.87 12 .1%
Sn										.74 20 .1%	.59 19 1%	.85 20 .1%	.51 19 5%	.75 20 .1%
Th											.91 19 .1%	.86 20 .1%	.87 19 .1%	.90 20 .1%
Ti												.65 19 1%	.96 19 .1%	.88 20 .1%
U													.66 19 1%	.84 20 .1%
Zr														.86 19 .1%

TABLE VII(continued)

	Y	Ce	La	Nd	Pr
Y		.98 7 .1%	.98 7 .1%	.93 9 .1%	.90 5 5%
Ce			1.00 7 .1%	.99 5 1%	.87 5 5%
La				.99 5 1%	.88 5 5%
Nd					.93 5 5%

KEY

U ↑ U and Co: pair of elements tested for correlation.

Co ←

.87 : Estimated correlation coefficient, r

16 : Number of pairs used for calculation

1% : Level of significance, i.e. the chance that r will be greater than the values given.

n.s. : Not significant

processes of concentration. Thus two or more causes may contribute to a high correlation in some cases.

(a) Elements which have common host minerals

The rare earth elements Y, Ce, La, Nd and Pr are present in the minerals monazite, uraninite, uranothorite, columbite, betafite, euxenite, leucoxene, and zircon. Of these monazite is responsible for by far the largest portion of the rare earths. The high estimated correlation coefficients for the rare earth pairs in Table VII can thus be ascribed to their similar geochemical properties, and their presence in the same host minerals.

Due to their poor detection limits obtained by using the equipment and methods described, the elements Ce, La, Nd and Pr could be determined in only a few samples. Y on the other hand was measurable in every sample used. As Ce, La, Nd and Pr all have high correlations with Y, the distribution of Y was taken as an indication of the distribution of the other rare earths.

The pair Y and P show a high correlation, which is due to the fact that most of the Y is present together with P in monazite.

(b) Elements which have detrital minerals as hosts

As stated previously the samples from the Dominion Reefs Mine contain a suite of typically and undoubtedly detrital minerals. The distribution of these minerals can be ascertained by determining the distribution of an element present mainly in one mineral species, and largely absent from the other minerals. Such minerals and elements are the following:

Host mineral	Element
Cassiterite	Sn
Chromite	Cr
Leucoxene	Ti
Monazite	P
Zircon	Zr

Although leucoxene must strictly be regarded as authigenic, there can be little doubt that the Ti from which it was formed was originally introduced as part of an allogenic mineral or minerals, among which ilmenite was probably the most important.

Ten possible correlations for the five elements mentioned above can be calculated; these are listed in Table VIII.

/Table VIII..

TABLE VIII

ESTIMATED CORRELATION COEFFICIENTS FOR ELEMENTS PRESENT IN DETRITAL MINERALS

	P	Sn	Zr	Ti
Cr	.98 .1%	.55 2%	.99 .1%	.96 .1%
P		.51 10%	.97 .1%	.99 .1%
Sn			.51 5%	.59 1%
Zr				.88 .1%

Six of these pairs have high estimated correlation coefficients which are significant at the 0.1% level.

The highly significant correlations can be attributed to the following: Similarly sized detrital minerals are concentrated in sediments by the same processes. In traction clogs (Moss, 1963, p. 324) the concentration of a rare heavy mineral may actually be dependant upon the presence of much more of another heavy mineral species of similar size. Closely sympathetic relationships of these minerals will result if their relative availabilities remain constant. Such a very close relationship of the minerals ilmenite, rutile and zircon was found for instance by Nel et al. (1960, p. 153) in black sands from Umgababa.

The four pairs which have Sn as one component possess fairly low estimated correlation coefficients, between 0.51 and 0.59, which, depending on the number of pairs used in the estimation, is significant only at the 1% to 10% levels. Since all the relevant minerals are obviously detrital, possible explanations of these lower, though significant correlations are the following:

(i) The availability of cassiterite relative to that of chromite, monazite, zircon and ilmenite (which furnished leucoxene) could have fluctuated. A possible reason for such a fluctuation could be that the cassiterite was supplied to the sediments from a different

/Source....

source, relatively close to the basin of deposition, whereas chromite, monazite, zircon and ilmenite were contributed from a more distant region. During transportation these minerals had ample opportunity to become well mixed, and to become sorted to similar grain sizes. The cassiterite, having been added at a later stage, had not been well mixed with the other heavy minerals before its final deposition.

Such a dual source was advocated also from size and form studies (Part I of this thesis).

(ii) The grain-size distribution of cassiterite could have been so different from that of the associated heavy minerals, that the concentration processes, which are size-sensitive, acted somewhat differently on the former. Such different size distributions were actually demonstrated in some of the samples from the Dominion Reef. (Part 1 of this thesis).

(c) Elements which have a chalcophile tendency

Elements which appear in the ores in the form of sulphides are Cu, Co, Ni, Pb and the main portion of the Fe. The host minerals of these elements are the following.

Element	Host minerals
Cu	Chalcopyrite, covellite, bornite.
Co	Cobaltite, arsenopyrite, linnaeite, pyrite.
Fe	Pyrite, marcasite, small amounts of other sulphides, leucoxene, ilmenite and chromite.
Ni	Arsenopyrite, linnaeite, cobaltite, pyrite.
Pb	Galena.

The estimated correlation coefficients of these elements are listed in Table IX.

TABLE IX

ESTIMATED CORRELATION COEFFICIENTS FOR ELEMENTS WHICH HAVE A CHALCOPHILE TENDENCY

	Co	Fe	Ni	Pb
Cu	.41 n.s.	.38 n.s.	.44 10%	.48 5%
Co		.59 2%	.96 .1%	.78 .1%
Fe			.64 1%	.47 5%
Ni				.72 1%

/Of these....

Of these elements the estimated correlation coefficient of Co with Ni is by far the highest. In nature, Co and Ni are usually closely associated. According to Rankama and Sahama (1952, p. 585) this close geochemical coherence is ascribed to their position in the periodic system.

In polished section the linnaeite is closely associated with chalcopyrite, and both the linnaeite and some of the cobaltite grains are of such an irregular form that the impression is gained that they are of hydrothermal origin. However, arsenopyrite and most of the cobaltite occur as discrete grains, which may conceivably indicate detrital grains. Ramdohr (1958, p. 23) claims that in the Witwatersrand both arsenopyrite and cobaltite occur as definite "Gerölle", and therefore are of a detrital origin.

If the host minerals of Co and Ni were introduced as detrital minerals, then it can be expected that their concentrations will vary sympathetically with that of Cr, P, Sn and Zr. In Table X correlation coefficients for these elements are listed.

TABLE X

ESTIMATED CORRELATION COEFFICIENTS OF CHALCOPHILE ELEMENTS WITH THOSE PRESENT IN DETRITAL MINERALS

	Cr	P	Sn	Ti	Zr
Co	.77 .1%	.76 2%	.80 .1%	.76 .1%	.75 .1%
Ni	.87 .1%	.84 1%	.80 .1%	.87 .1%	.85 .1%
Cu	.29 n.s.	.41 n.s.	.65 1%	.38 n.s.	.29 n.s.
Fe	.68 10%	.82 1%	.52 5%	.75 .1%	.70 .1%
Pb	.52 5%	.59 5%	.77 .1%	.52 5%	.51 5%

The estimated correlation coefficients of Ni and Co with the elements present in the detrital minerals are highly significant, and indicate that they could also have been introduced in detrital minerals. The small amount of cobaltite and linnaeite which appear to be of hydrothermal origin are either present in such small concentrations that

/they.......

they have a very slight effect on the correlation coefficient, or they show the effects of a limited hydrothermal redistribution.

It has been claimed by Davidson (1962, p. 159) that there is good evidence in the Dominion Reef that hydrothermal metallization has been super-imposed on a suite of heavy detrital minerals. If Ni and Co is regarded as belonging to this hydrothermal mineralization, then a process which caused the preferential deposition of Ni- and Co-minerals in sediments rich in heavy detrital minerals must have been in operation. Such a process could have been controlled by the texture or the chemistry of the rocks. This possibility will be discussed later on.

The mode of occurrence of the copper minerals are in accordance with a hydrothermal origin, or hydrothermal redistribution. The estimated correlation coefficients with Cr, P, Ti and Zr (Table X) are rather low, and for the suite of samples studied, they are not significant. An unexpectedly high estimated correlation coefficient of Cu with Sn was, however, found, and this is significant at the 1% level.

The association of cassiterite with chalcopyrite in high-temperature hydrothermal veins is well known. This association may be maintained after a cycle of weathering in different ways:

(i) Both minerals could have been deposited as detrital minerals although it is hardly conceivable that copper minerals as we know them would survive a detrital cycle. As the typical form of the copper minerals as they appear today is not that of a detrital mineral, a further assumption has to be made namely that they were reconstituted by hydrothermal action which caused only a limited redistribution, or by plastic flow (Liebenberg, 1955, p. 203).

(ii) During the normal cycle of weathering Cu goes into ionic solution, whereas cassiterite remains in the detritus. Both elements may be transported by water; the Cu in solution, and the cassiterite mechanically. Finally the cassiterite may be deposited as a detrital heavy mineral, and Cu may be precipitated in sapropelic mud, or trapped in associated hydrolyzate sediments, from which it is later redistributed by hydrothermal action or lateral secretion. That this process can cause a high degree of correlation of Cu with Sn will be difficult to

/prove.....

prove, particularly as the correlation of copper with the other detrital minerals is of such a low magnitude.

That the concentration of Cu in the Dominion Reef is connected in one way or another with the concentration of Sn and U seems highly probable, but the exact nature of the mechanism must remain conjectural, until more evidence is obtained. If a portion of the Cu as well as the Sn and U are of detrital origin this relationship would be explained.

The correlation of Fe with the elements present in the detrital minerals is partly due to the presence of a portion of the Fe in leucoxene, ilmenite and chromite. Using the theoretical Fe: Ti and Fe: Cr ratios of ilmenite and chromite, a correction was made in the Fe values, and a new correlation coefficient was calculated for Ti and that portion of the Fe not present in leucoxene, ilmenite and chromite. The value so found is 0.58. Although this value is considerably lower than the value obtained with uncorrected Fe values, it is still significant at the 1% level. This residual correlation is ascribed to the type of pyrite known as buckshot pyrite, which was found to be associated with the heavy minerals. The fact that the estimated correlation coefficients with the detrital minerals is fairly low is thought to be due to the presence of a considerable amount of large irregular pyrite grains which could be of hydrothermal origin in many of the samples studied.

The major portion of the Pb is probably of radiogenic origin. The distribution of the Pb through the samples clearly is in conformity with the distribution of U, and elements which have a high correlation with Pb also have a similar or higher correlation with U. The problem of interpreting the significant correlations of certain elements with Pb and U can therefore be regarded as identical, and will be considered in the next section.

(d) Uranium and thorium

As shown previously (p. 26), the elements present in different detrital minerals may show high correlations. In addition, elements which are of hydrothermal origin, and belong to the same mineralization, may also show significant correlations. Thus Bieler et al. (1960, p.363) in their study of 50 samples of uranium-bearing veins of "siliceous f" in the Boulder Batholith, Montana, found that the elements

/Fe, As,....

Fe, As, Co, Mo, and Pb had correlations with U which were significant at the 1% level, and Ba, Cu and Ni had correlations with U at the 5% level of significance.

A test may therefore be conducted to indicate the origin of U and Th by ascertaining whether they show higher correlations with the elements present in detrital minerals or with the elements present in minerals appearing to be of hydrothermal origin.

In Table XI the estimated correlation coefficients of U and Th with the elements present in detrital minerals and the chalcophile elements are listed.

TABLE XI

ESTIMATED CORRELATION COEFFICIENTS OF U AND Th WITH ELEMENTS OF DETRITAL MINERALS AND ALSO WITH CHALCOPHILE ELEMENTS

	Cr	P	Sn	Ti	Zr	Co	Ni	Cu	Fe
U	.70 .1%	.73 1%	.85 .1%	.65 1%	.66 1%	.87 .1%	.88 .1%	.47 5%	.51 5%
Th	.89 .1%	.90 .1%	.74 .1%	.91 .1%	.87 .1%	.88 .1%	.95 .1%	.38 10%	.56 2%

The highest estimated correlation coefficients are shown with those elements present in the detrital minerals, and also with Co and Ni, which have previously been shown to follow the detrital minerals closely. With Cu and Fe, which are present in minerals partly showing the effects of a hydrothermal origin or redistribution, the correlation is much lower, and significant only at the 2 to 10% levels. Even this low degree of correlation must be ascribed to the low degree of correlation of Fe and Cu with the detrital minerals, rather than to a genetic relationship with U and Th.

The geochemical evidence is therefore overwhelmingly in favour of a genetic relationship of uranium and thorium with the detrital minerals.

(e) Gold

As shown in Table VII the estimated correlation coefficients of Au with the other elements are not significant, with the exception of Sn. For the pair Au and Sn an estimated correlation coefficient of 0.64 was found, which is significant at the 5% level.

/Many.....

Many previous workers commented on the sympathetic relationship of Au and U in the Witwatersrand and Dominion Reef ore. Thus Simpson (1951, p.131) points out that: "From the assay of some half a million samples covering the whole Witwatersrand System, it has been established that a relationship exists between the occurence of Au and U in these sediments".

"This relationship is quite close in some areas, but in others becomes more erratic. In all cases the more persistent element is U, and this is subjected to fluctuations in content which tend to take place sympathetically with variations in Au content".

The statement by Liebenberg (1955, p. 215) which is based on data which was already published in 1948, confirms the contention of Simpson quoted above. However, Liebenberg is careful to point out that "exceptions to these statements are relatively common".

It may well be that the Dominion Reef, which is well known for its erratic Au mineralization, is one of the areas where this sympathetic relationship of Au and U is not well developed. If the correlation of Au with U is weak, then it may not be detected as a significant correlation by only 12 samples (Table VII). For this reason correlation coefficients were also calculated for the data given by Malan (1959, Plate 2), and found to be 0.49, with a significance at the 0.1% level. These data therefore confirm the relatively low correlation of Au and U in the Dominion Reef, and enough values were available to show that this relationship is highly significant.

(f) The bearing of rock texture on mineralization

Since the original contribution by Reineke in which he convincingly demonstrated an increase in the average Au content as the average diameter of the larger pebbles increases in ores obtained from Government Areas (Reineke, 1928, p. 103), many writers have made statements on the bearing of ore texture on mineralization in the Witwatersrand. Malan (1960 p. 59) has also claimed that higher Au values are found in the coarser-grained conglomerates of the Lower Dominion Reef. In many cases the evidence for these statements are not presented in such a way that one can easily evaluate the intensity of the relationship.

/The.....

The bearing of rock texture on gold values has been used by placerists as evidence of a sedimentary control, and consequently as an argument in favour of a detrital origin. The hydrothermalists on the other hand maintain that the texture is obviously related to the original permeability of the rocks, which would then form favourable loci for the deposition of hydrothermal minerals, and, incidentally also explains the association of ore minerals with detrital heavy mineral grains. Thus, Davidson (1957, p. 682) argues: "That detrital chromite and zircon should be found in the same parts of a reef as gold and uraninite, is not at all contrary to hydrothermal ideas for it is precisely these heavy minerals in more than ordinary concentration which would mark zones of scour possessing more than ordinary permeability".

If the high degree of correlation of U with heavy minerals should be due to a textural control, a close correlation of the heavy minerals with texture, and also U tenor with texture is to be expected. The same applies to Co and Ni.

Correlations were accordingly calculated for maximum pebble size and pebble content on the one hand with tenors of U, Au, Cu, Co and Cr on the other hand. The pebble data are listed in Table XII, and the estimated coefficients of correlation in Table XIII. The values listed in the same horizontal line as percentage pebbles are estimates of the product-moment correlation coefficient, and were calculated according to the formula on page 22. The values listed in the same horizontal line as maximum pebble size are estimates of Spearman's rank correlation coefficient, calculated according to the formula:

$$R = 1 - \frac{6\Sigma\, d^2}{n(n^2-1)}$$

where d is the difference in ranks assigned to the pebble data corresponding to increasing sizes, and to element contents corresponding to increasing amounts (Snedecor, 1961, p. 191).

It is evident that all the estimated correlation coefficients of element content with maximum pebble size are negative. However, due to the small number of samples used the estimated correlation coefficients are significant only in the cases of Au at the 1% level, and Cr at the 5% level, both in the Upper Reef. The indications are therefore that

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TABLE XII

MAXIMUM SIZE AND PERCENTAGE OF PEBBLES IN CONGLOMERATES FROM THE UPPER AND LOWER DOMINION REEF

Upper Reef		
Sample No.	Percentage pebbles, to nearest 5%	Maximum size pebble observed
1(a)	5	2 mm,
(b)	50	30
2	15	20
6	50	18
10	60	30
15	15	28
16	50	25
Lower Reef		
3	35	25 mm.
4	50	45
5	50	30
7	80	40
8	50	35
9	70	45
11	30	12
12	70	37
17	45	40
18	70	45
20	65	65

TABLE XIII

ESTIMATED CORRELATION COEFFICIENTS OF U, Au, Cu, Co, AND Cr CONTENTS WITH MAXIMUM PEBBLE SIZE AND PERCENTAGE PEBBLES IN DIFFERENT SAMPLES

Upper Reef					
	U	Au	Cu	Co	Cr
Maximum Pebble size	-.54 n:7; n.s.	-.89 n:7; 1%	-.07 n:7; n.s.	-.54 n:7;n.s	-.75 n:7; 5%
% Pebbles	-.81 n:7; 5%	-.64 n:5; n.s.	-.51 n:7; n.s.	-.84 n:7; 2%	-.84 n:7; 2%
Lower Reef					
	U	Au	Cu	Co	Cr
Maximum Pebble size	-.06 n:11; n.s.	-.25 n:11; n.s.	-.15 n:11;n.s	-.32 n:7;n.s.	-.08 n:10;n.s.
% Pebbles	-.37 n:11; n.s.	.06 n:5; n.s.	0.44 n:11;n.s.	-.72 n:7; 10%	-.44 n:10;n.s.

in the Upper Reef, the relationship of gold tenor with maximum size of pebble is the opposite to that found by Reineke (1928, p. 103) for ores from Government Areas.

Most of the estimated correlation coefficients of pebble content with element content are negative. Exceptions are Au and Cu in the Lower Reef, for which the values obtained are not significant. In the Upper Reef values were found which are significant for U at the 5% level, and for Co and Cr on the 2% level. In the Lower Reef only one significant value was obtained, viz. that for Co on the 10% level. The indications are therefore that there is a weak negative correlation of element tenor with pebble content.

One possible reason for this negative correlation is that it may be due to a dilution effect by the pebbles. The ore minerals are present only in the matrix and if the amount of ore minerals in the matrix vary only between fairly close limits, the tenor of ore minerals will have a negative correlation with pebble content. It is possible to test for this possibilityby recalculation of the percentages of the elements in terms of the matrix, and by using these figures to calculate new correlations. The results obtained in this way were close to those of Table XII, and indicate that the dilution effect of the pebbles is relatively unimportant.

The proponents of the hydrothermal theory generally assume increasing mineralization with increasing permeability. As argued by Graton (1930, p. 50) the permeability can be expected to increase with pebble size, and also with the amount of pebbles. On the basis of this theory the correlation coefficients of hydrothermally introduced elements with pebble size and pebble content can be expected to be positive. In the Dominion Reef this is not the case, and the relationship of rock texture with mineral content does not vindicate the hydrothermal theory.

V. GENERAL DISCUSSION OF ELEMENT RELATIONSHIPS

The overall picture emerging from this study of the relationship between elements in the Dominion Reef is one indicating a control by sedimentary processes of concentration. High degrees of correlation exist between elements present in detrital minerals and U, Th, Ni, and Co. Precisely those elements which appear to be of hydrothermal

/origin.....

origin, or are present in minerals which could have been hydrothermally redistributed, viz. Au and Cu, have estimated correlation coefficients with each other as well as with the elements of detrital origin which are not significant, or have a low significance on the 5% or 10% levels only. The only exception, viz. the pair Cu and Sn which have an estimated coefficient of correlation which is significant on the 1% level, has already been discussed.

The theory that Co and Ni are of hydrothermal origin and that their high degree of correlation with the elements present in detrital minerals is the result of a textural control could not be vindicated. The same is true of Davidson's theory (1964, p.175) that the Dominion Reef uraninite was derived from andesitic and rhyolitic lavas and tuffs, which rapidly released uranium to groundwater on devitrification, and that the groundwater deposited uraninite in the Upper and Lower Reefs in close proximity to the detrital minerals.

The detrital theory as advocated by Liebenberg (1955, p. 167) and Ramdohr (1958, p. 46) is strongly supported by the geochemical evidence. The claims made by Liebenberg (1955, p. 215) viz. that a sympathetic relationship exists between uraninite, gold and heavy detrital minerals, were confirmed, and it was established that a sympathetic variation obtains also for many other element pairs in the Dominion Reefs Mine.

One of the more important conclusions to be drawn from the pattern of element distribution is that, at the time of deposition of the Dominion Reef, the exogenic geochemical cycles of the elements Co, Ni, U, and to a lesser extent Cu and Fe differed from the cycles of today in that larger proportions went into the resistates.

Several students of ore genesis have found geochemical patterns of element distribution for Precambrian rocks which differ from those of today. The most recent are Lepp et al.(1964, p. 1025), who studied Precambrian iron formations. They postulated an oxygen deficient atmosphere in order to explain the differences between the Precambrian and younger iron formations. However, Davidson (1964, p. 1025) is of the opinion that to postulate different conditions in Precambrian times from those of today is to violate the doctrine of uniformitarianism.

Such a departure from the doctrine of uniformitarianism must be viewed with caution. However, a too rigid adherence to a doctrine

/may.....

may easily lead its followers astray. It cannot be denied for instance that the advent of the antroposphere caused changes in the exogenic cycles of many elements which are of such a magnitude that the elucidation of pre-antroposphere conditions may prove extremely difficult in some cases. Examples that can be cited are the changes in the composition of the water of the main rivers since intensified agricultural and industrial activities commenced all over the world. Another event that must have had far reaching effects on the geochemical cycles of many elements was the advent of land flora, which must have produced marked changes in

(i) the zone of weathering,
and
(ii) in the terrestrial waters.

(i) The effect of land flora on the zone of weathering

The major mechanical influence of the land flora would be the protection of the soil cover from rapid erosion over large portions of the globe, with the consequence that the underlying rocks would be exposed to air and moisture for much longer times. Prolonged reaction times would increase the importance of hydration, hydrolysis, oxidation and reduction, carbonation and solution.

The introduction of organic material initiated biological activity of plants and micro-organisms in the soil. The pH values in the soil would be changed by the oxidation of organic material to organic acids, by the production of CO_2 and by oxidizing sulphur.

(ii) The effect of land flora on terrestrial waters

The two major changes brough about by land-flora on the composition of water in rivers and streams would be the increase in the amount of dissolved salts, and the lowering of the pH.

From the above it is clear that the overall effect of a land flora would be to increase the amount of chemical weathering and to decrease the amount of mechanical weathering. In the exogenic cycles of the elements, the resistates would have played a much more dominant role before the advent of a land flora than under present conditions; a conclusion which agrees with the pattern of element distribution in the Dominion Reef.

/The......

The absence of a land flora during Dominion Reef times may not have been the only cause for the relative importance of mechanical weathering. One or more other causes may have contributed. The survival of uraninite as a detrital mineral was ascribed for instance to a non-oxidizing atmosphere by Liebenberg (1955, p. 173) and also by Louw (1954, p. 226), to a process of glacial erosion by Wiebols (1955, p. 368), to the absence of land flora by Bain (1955, p.239), and by Ramdohr (1958, p. 43) to refractoriness of uraninite due to its young age. Arguments against these theories were submitted by Davidson (1957, p. 673 - 677).

Davidson mentions the existence of beds of ferric iron ore in the Witwatersrand as evidence against a possible non-oxidizing atmosphere. He argues that the typical oligomictic nature of the auriferous conglomerates of the Witwatersrand System is evidence against a possible glacial origin. He points out that the uraninite could not have been very young when it was deposited. The arguments for and against these theories are largely of a speculative nature, and need not be considered here in detail.

It is of interest however, that a thorough geochemical study of Precambrian iron formations led Lepp et al. (1964, p. 1025) to the conclusion that the Precambrian atmosphere lacked, or was markedly deficient in free oxygen. They are of the opinion that the critical level of free oxygen in the atmosphere which permitted a marked accelaration in plant growth and an accretion of oxygen was attained only in Late Precambrian times, at least 1,200 million years ago.

It would appear therefore that the presence of beds of ferric iron ore in the Witwatersrand cannot be regarded as evidence for an oxidizing atmosphere during the time of their formation. It may be advanced for a theory that low relief existed at the time of their formation, allowing chemical weathering to take place, whereas a fairly high relief during Upper Witwatersrand times would cause the accumulation of great thicknesses of quartzite and layers of conglomerate. Amongst the arguments forwarded by Davidson against a possible detrital origin for Witwatersrand, Dominion Reef and Blind River uraninite, the one most often repeated is that uraninite does not survive as a detrital mineral (Davidson, 1953, p. 73 and 1957, p. 670).

/Davidson.....

Davidson mentions a number of occurrences of modern placers in which uraninite is absent, and in which the radio-active minerals present are richer in Th than U, resulting in Th:U ratios of the order of 10:1, whereas in the Witwatersrand and Dominion Reef the tenor of U is usually greater than that of Th.

In intermediary igneous rocks the Th:U ratio is 4.0, and in acidic igneous rocks it is 3.4 (Rankama and Sahama, p. 634). The usual mode of occurrence of uranium is in association with Zr and Th in zircon, or associated with Th and rare earths in thorite, thorianite, monazite, xenotime, allanite, etc. The survival of these minerals in the resistates will of course result in Th: U ratios in the placers which are of the same order as that of the igneous rocks from which they were derived. In most of the examples quoted by Davidson the non-occurrence of uraninite in the placers can simply be ascribed to the absence of uraninite in the source rocks, and the high Th:U ratios in the placers to the high Th:U ratios in the resistates derived from the source rocks. However, in some instances Davidson mentioned the presence of uraninite in the source rocks (e.g. in Idaho), and even in this environment it is absent from the alluvial assemblages. Such examples illustrate the tendency of uraninite to become oxidized on exposure to weathering, with the result that alteration products are easily formed. The hardness of uraninite is also decreased on oxidation with the result that it becomes more prone to mechanical diminution.

The environment in which thorian uraninite occurs in sands of the Indus River, Pakistan (Zeschke, 1960, p. 87), comply with the conditions under which mechanical weathering will be more severe than chemical weathering. It is situated in an area of high relief, and the water in which uraninite was transported is nearly neutral. Amongst the heavy minerals associated with the uraninite, arsenopyrite, pyrargyrite, pyrite, chalcopyrite and other sulphides are recorded, all of them species which will appear in the resistates only under the somewhat special conditions where chemical weathering is at a minimum.

The arguments submitted by Davidson against the survival of uraninite in placers under modern conditions, and also his demonstration that it usually does not occur in modern placers are of value in

/explaining......

explaining several phenomena.

(i) The Dominion Reef and Witwatersrand assemblages, if they are of detrital origin, could not have been derived from normal unmineralized rocks. It is necessary to assume, as Davidson does (1961, p. 48) that "South African goldfields form a metallogenetic province rich in Au and U, with an overall content of these elements much in excess of their clarkes". However, it is here postulated that the first manifestation of the metallogenisis occurred about 3,100 million years ago (Nicolaysen et al. 1962, p.17) before the Dominion Reef sediments were deposited, and not during the "Bushveld Orogenesis" about 2,000 million years ago, as claimed by Davidson. This mineralization was possibly connected with the Archaean granite known to be present in the central Transvaal, and could have been in the form of pegmatites, and high temperature veins.

(ii) In order to explain the survival of uraninite as a detrital mineral, attention must be paid to the conditions in the zone of weathering, in the streams in which it was transported, and the environment in which it was deposited.

(a) In the zone of weathering a high rate of mechanical weathering, and the absence or a low rate of chemical weathering will release uraninite from its source rocks before it is chemically reconstituted to material which does not urvive as resistates. The possible environment under which such conditions will obtain is one where land plants are absent, where relief is high, where glacial action may contribute to weathering and where the atmosphere is non-oxidizing. It is considered that such conditions obtained when the uraniferous conglomerates of the Dominion Reef and Witwatersrand were deposited.

(b) During sedimentary transport and deposition a low rate of chemical decomposition can be obtained by

(i) neutral terrestrial water, which may be due to the absence of a land flora,

(ii) low temperatures which may prevail on the highest regions of an area of high relief, and

/(iii) brief...

(iii) brief reaction times which may be due to relatively rapid transport under conditions of high relief. The grain sizes of most of the heavy minerals occurring in the Dominion Reef are so small that even extensive mechanical action would be insufficient to cause a further diminution in size. Severe mechanical weathering would thus not have been able to cause the elimination of uraninite and the other associated heavy minerals.

(c) In the environment in which it was deposited the absence of oxidizing conditions is essential for the survival of uraninite. If the detrital theory is correct in assigning a placer origin to the sulphides, such as pyrite, cobaltite and arsenopyrite, oxidizing conditions would have caused the formation of sulphuric acid, and the dissolution of uraninite. That oxidizing conditions did not prevail is indicated by the presence of mainly ferrous iron, and the presence of carbonaceous material in the Witwatersrand may even be cited as evidence for sapropelic conditions. Such reducing conditions may exist in sediments even though the atmosphere is oxygen rich. Many such cases exist at the present day.

(d) According to the detrital theory, gold, and probably some other minerals, were redistributed by hydrothermal action after the lithification of the sediments. According to Davidson (1955, p. 234) the survival of uraninite in an environment where gold, "the very paragon of chemical stability", was redistributed, is an anomaly that cannot be accepted.

In the samples from the Dominion Reef most uraninite grains have been corroded. In the samples in which uraninite was best preserved (1(a), 1(b) and 2) the uraninite tenor is rather high, and about 10% by volume of the uraninite was corroded. In these samples this 10% is about 10 to 50 times more than the total amount of gold present.

Under certain conditions, for instance in the presence of chloride and heavy metal base ions, gold is not so difficult to corrode.

/according...

According to Thompson (1947, p. 46) "there is no scientific reason for dissociating the idea of corrosion of a noble metal from that of a base one, since the terms are purely relative. The term "noble" applies only under the normal atmospheric conditions and in solutions which happen to be somewhat less cathodic (or "cathogenic") than gold ions themselves. In cyanide solutions of sufficient strength, gold functions more "basely" than iron ...." The corrosion of uraninite at a rate which does not greatly exceed the corrosion rate of gold could thus easily be possible under certain conditions.

From a study of the Dominion Reef samples the impression was gained that in those samples which have a low uraninite content the uraninite grains were extensively corroded, whereas in samples having a high tenor of uraninite, the mineral has been only slightly attacked. This relationship is unfortunately refractory to statistical treatment, as a satisfactory quantitative and objective measure of the degree of corrosion could not be devised. However, if this relationship does exist, it can be regarded as an indication that in samples rich in uraninite the corrosion reaction was arrested when only a small percentage of the uraninite was taken into solution, whereas in the case of the gold it was probably notterminated before most of the gold was taken into solution and deposited at some small distance from its original locality. This condition could have obtained if only a small amount of solvent was present, and if the movement of this solvent was restricted to a small area, of the order of millimeters only. Thus in the vicinity of the uraninite grains the amount of solvent could have been so small that it was able to dissolve only a small percentage of the uraninite before becoming saturated. The amount of gold on the other hand could have been so small that all of it could have been taken into solution.

This argument will be more convincing if it can be shown that the movement of solvents through the Dominion Reef samples was actually restricted at the time when corrosion took place. Evidence which may be cited in support of this argument is the following:

(a) In general the alteration products of corroded uraninite are to be observed in close proximity to the grains from which they were derived.

/(b) According.....

(b) According to Davidson (1957, p. 686) the results of isotopic analysis of two portions of a sample of galena from Sub-Nigel which were beneficiated to different degrees, yielded different results. Davidson continues: "...and one cannot avoid the surprising conclusion that this fractionating must be attributable to the differing amounts of physical beneficiation that the samples have received. It follows that the discrete particles of galena present in the reef at any one locality may vary considerably in isotopic composition, and as a corrollary to this conclusion one must accept that in the reworking of the ores the movement of the metals has been restricted to a limited radius (perhaps no more than a centimeter or two) without general diffusion throughout the reef as a whole". It must be pointed out that the conclusion above does not pertain to the Dominion Reef. However, as far as the distribution of galena is concerned the Sub-Nigel and Dominion Reef ores are very similar. That the redistribution of galena took place at the same time that uraninite was corroded also remains unproved, but it may be considered probable.

The arguments above have shown that uraninite may survive as a detrital mineral, and in Dominion Reef times probably did survive.

CONCLUSIONS

(1) The elements present in the uraniferous conglomerates of the Dominion Reefs Mine in undoubtedly detrital minerals have a high degree of correlation with each other.

(2) The elements Co and Ni, which are present in sulphides, the microscopic size and form of which correspond to that of detrital minerals also have high degrees of correlation with the elements of the detrital minerals. All the evidence therefore points to a detrital origin for the major Co and Ni minerals in the Dominion Reef.

(3) The elements Cu and Au, which show signs of hydrothermal redistribution, have low degrees of correlation with the elements from detrital minerals as well as with the chalcophile elements.

(4) U has a high. degree of correlation with the elements present in the detrital minerals, and therefore also appears to be of detrital origin.

(5) Uranium......

(5) Uranium and gold have a low degree of correlation, which was however found to be highly significant. This is the pattern to be expected if gold is also of detrital origin, but was redistributed to a limited extent.

(6) No vindication could be found for Davidson's polygenetic theory, viz. that uraninite has a groundwater origin, gold and sulphides a hydrothermal origin, and chromite, zircon, etc. a detrital origin.

(7) The pattern of element distribution indicates that the exogenic cycle in Dominion Reef times differed from that of today in that the resistates of uranium, copper, cobalt, nickel, and iron played a more dominant role. It is considered that a set of conditions under which mechanical weathering is more pronounced than today, and chemical weathering less pronounced is necessary to explain this phenomenon.

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