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Some aspects of the genesis of heavy mineral assemblages in Lower Proterozoic uranium-gold conglomerates

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ABSTRACT. Some genetic models for Lower Proterozoic gold- and uranium-bearing pyritic conglomerates favour a modified placer origin in which low levels of atmospheric oxygen are used to account for the survival of uraninite and pyrite. There are many difficulties with such models—for example magnetite is absent in the ore-bearing horizons although it is stable in anoxic conditions, while it is abundant in over- and under-lying strata. Compact and porous pyrite grains are not in hydraulic equivalence and the deposits lack a normal detrital heavy-mineral assemblage. Moreover uranium and gold show evidence of diagenetic remobilization, the uranium becoming associated with secondary titaniferous phases and uranium and gold being enriched in reduzate facies sediments.

New evidence concerning the genesis of the deposits is derived from a clast of ferric iron clay thought to represent a precursor sediment of the Witwatersrand Basin. Reworking of such clays and transport of a magnetite and ferric clay assemblage with subsequent sulphidation, could account for the porous pyrites, the absence of magnetite and the lack of hydraulic equivalence of the mineral grains in the conglomerates. The presence of oxygen, as indicated by the ferric iron clasts, would account for the evidence of mobility of uranium and of gold and would enhance their extraction from source rocks; particularly the release of gold from a precursor auriferous iron formation source. It is suggested that some aspects of the genesis of uranium deposits of the Witswatersrand and Elliot Lake may be similar to those of the Phanerozoic 'Roll Front' ores involving interaction between oxidizing uraniferous groundwaters and previously sulphidized and reduzate facies sediments.

THE Lower Proterozoic pyritic conglomerates of the Witwatersrand in South Africa (Au-U) and the Elliot Lake region in Canada (U) provide 70% of the western world's gold supply and represent 25% of the economic uranium reserve. Other occurrences are known from Russia, Brazil, India, North America, Northern Africa, and Australia. Such sequences were formed in the timespan between 3 and 2 Gyr.

The conglomerates, together with sequences of quartzite and shale, represent the first stable cratonic cover sediments and were probably derived from erosion of high-level Archaean rocks such as greenstone belt mafics, granite intrusives and associated ore deposits. The conglomerates consist predominantly of quartz and chert pebbles in a sericite/chlorite matrix; they are thus mineralogically mature although texturally immature. Pyrite is the dominant heavy mineral, forming an abundant interstitial phase to the conglomerate clasts. Thin seams of carbonaceous material, carbonaceous muds and patches and specks of carbon are common, and may be associated with high values of uranium and gold (Simpson and Bowles, 1981; de Kock, 1964).

Interpretations of the sedimentary facies of the sequence are disputed but a fluviatile piedmont model is generally accepted (Pretorius, 1975; Schumm, 1978). The occurrence of rounded apparently waterworn (Hallbauer and Utter, 1977) grains of uraninite, with gold and pyrite along basal scour planes, on foresets, in the lowermost coarse fraction of graded beds and aligned in 'pay streaks' corresponding to ancient channels has been used to support a fossil placer origin for the deposits (Pretorious, 1975; Minter, 1976). Thus the 'yellow sand' of the Proterozoic has been equated with the 'black sand' of recent placer concentrations (Roscoe, 1969, 1973).

Investigations of the morphology and hydraulic equivalence of some of the ore particles and comparison with modern water-worn grains is also consistent with a placer origin (Hallbauer and Utter, 1977; Utter, 1980). However, some of the gold and pyrite textures are those of replacement (Davidson, 1957; Ramdohr, 1958; Saager, 1970) with uranium commonly occurring in brannerite; this is clearly not detrital and a 'modified placer' hypothesis (Liebenberg, 1955) which allows for limited mobility of elements during diagenesis and metamorphism has been developed to explain some

of these features. The suggestion that pyrite and uraninite would be unlikely to withstand weathering and transport lead to the proposal that the atmosphere was non-oxygenic (Liebenberg, 1955). Recently however the non-oxygenic early-atmosphere model has been questioned (Dimroth and Kimberley, 1976; Simpson and Bowles, 1977, 1981; Clemmey and Badham, 1981) since much of the biological and geological evidence cited in support of a non-oxygenic atmosphere has been shown to be of doubtful validity.

Problems with the placer hypothesis. One of the greatest difficulties of the placer hypothesis is the lack of black sand components and in particular the lack of magnetite in the ore-bearing horizons. Magnetite is unstable in an oxidizing atmosphere. However since it breaks down only slowly magnetite is one of the commonest heavy minerals in sediments and placer deposits. Magnetite is absent from the mineralized conglomerates of the Upper Witwatersrand but it is sufficiently abundant in the Lower Witwatersrand sediments that magnetite rich beds are used in geophysical prospecting (Borchors, 1964). Pyrite and uraninite do not occur in such sediments or in Archaean clastic sediments generally.

Another difficulty is the apparent lack of hydraulic equivalence among pyrite grains. Certain varieties of the so-called 'porous pyrite' are many times larger than the 'compact' varieties in co-existing assemblages (fig. 1); and the porous pyrites are clearly detrital and are not replacement phenomena. Whilst some difference in density can be accounted for by the porous nature of the pyrite the magnitude of the difference in size of the two types of pyrite provides a problem.

Finally the Upper Witwatersrand strata lack a typical placer heavy mineral assemblage. Only the Dominion Reef, and the Elliot Lake conglomerates of the sequences contain a normal suite of heavy minerals such as monazite, garnet and ilmenite (or its altered equivalents) (fig. 8*a*), and the lack of

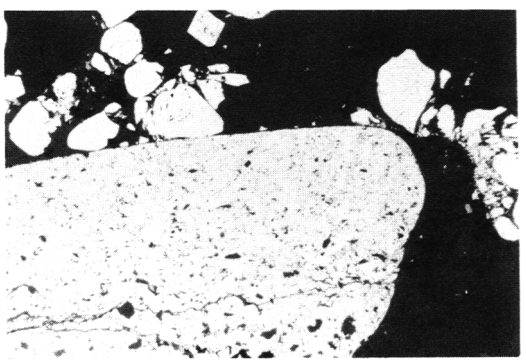

FIG. 1. Reflected light photograph illustrating the lack of hydraulic equivalence referred to in the text.

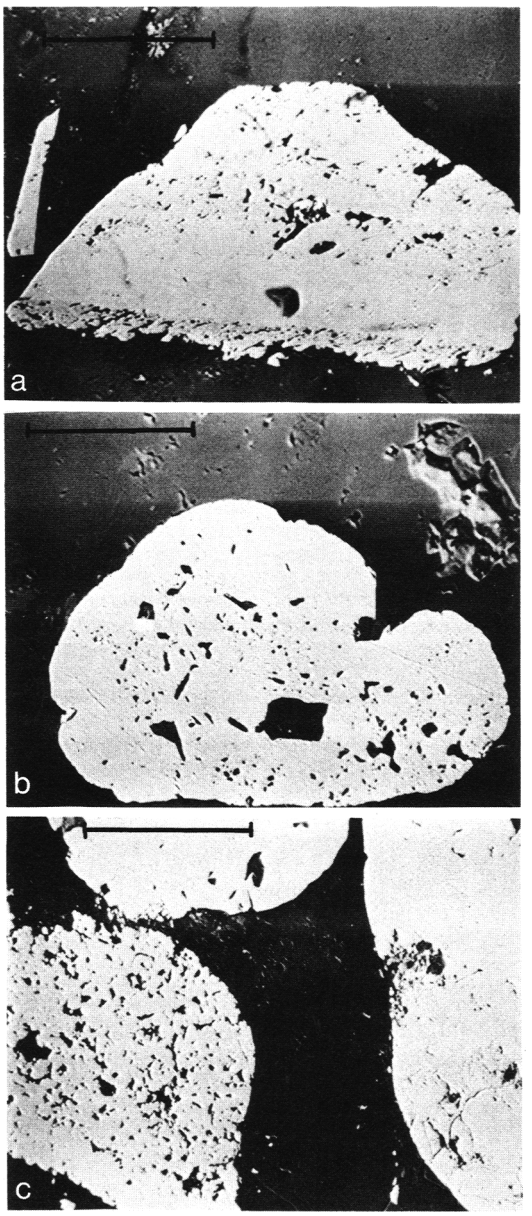

FIG. 2. Back-scattered electron images of types of pyrite from Witwatersrand conglomerates. (*a*) Sulphidized banded iron formation. Note the cleavage which relates to the pre-sedimentation phase. 3974b. (*b*) Botryoidal clast reminiscent of limonite or hematite precursor. 3974c. (*c*) Lower left; typical porous pyrite with phyllosilicate inclusions. Fabrics vary from granular to shreddy. 3974c. Scale bars 0.1 mm.

zircon, tourmaline, kyanite, corundum, and pyroxenes is unusual (Davidson 1957, Dimroth 1979).

Pyrite, sulphidation events and precursor minerals. The pyrites of the Lower Proterozoic conglomerates and those of the Rand in particular have been discussed by several authors (Ramdohr, 1958; Saager and Mihalik, 1967; Saager, 1970; Simpson and Bowles, 1977, 1981; Utter, 1977; Kimberley, 1978; Dimroth, 1979). Dimroth, and Simpson and Bowles proposed a diagenetic origin involving sulphate-reducing bacteria for the so called 'porous pyrite' of the Witwatersrand, the sulphate being derived from the oxidation of detrital sulphides or volcanic sulphur and thus indicating an oxygenic atmosphere. Bedded or framboidal pyrites can also be found in volcanogenic exhalative orebodies or in sediments through which waters enriched in reduced volcanic sulphur have percolated and Kimberley preferred a diagenetic/volcanogenic origin for the pyrites as in recent deposits at Sabatini in Italy. Saager suggested iron sulphide gels and Utter suggested iron sulphide muds as possible sources. Ramdohr differentiated between primary pyrite and pyrite formed by sulphidation

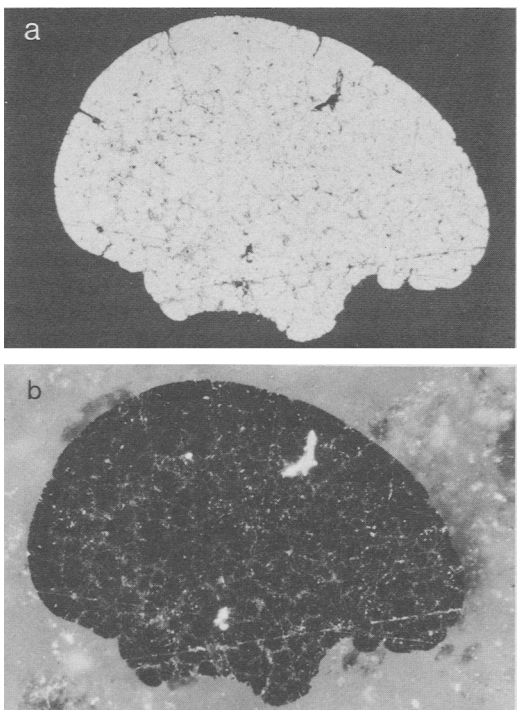

FIG. 3. (*a*) Reflected light photomicrograph of syngenetic pyrite clast. Note the peripheral radial shrinkage cracks indicative of dewatering. (*b*) Internal reflections under crossed nicols emphasize grain boundaries between framboidal aggregates.

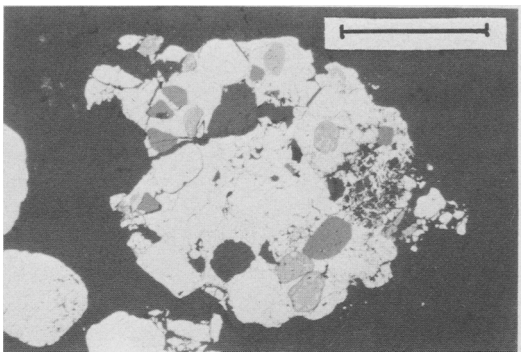

FIG. 4. Reflected light photomicrograph of a clast of precursor Rand sediment. Pyrite sub-grains occur with ferro-titanium phases and zircon in a pyrite matrix. The clast is a sulphisized heavy mineral black sand assemblage (cf. fig. 1) in a sulphidized ferric clay matrix. Tectonic extension or compaction has weakened the grain. 3974A. Scale Bar 0.5 mm.

of magnetite, banded ironstone clasts, limonite, and 'sediments'.

The sulphidation of magnetite would account for the lack of the black sand components discussed earlier. Textural evidence obtained by electron microscope analyses with backscatter images (fig. 2) is compatible with Ramdohr's suggestions if this is modified to incorporate intrabasinal 'syngenetic' pyrite muds (fig. 3) formed by sulphate-reducing bacteria (Dimroth 1979, Simpson and Bowles 1981) or of volcanic/diagenetic origin (Kimberley 1978).

Ferric iron clay in the Rand sediments. A detailed study of the clast illustrated in fig. 4 provides evidence for the origin of the spongy pyrite clasts with abundant phyllosilicate inclusions (figs. 2 and 5), an explanation of the lack of hydraulic equivalence and the time relations of the sulphidation event(s). The clast, reworked from an earlier phase of sedimentation, consists of well-rounded pyrite sub-grains and a 'normal' placer heavy mineral assemblage of $FeTiO_2$ phases and zircon in a pyrite matrix. Tectonic extension has weakened the clast and the pyrite sub-grains are clearly visible. The pyrite matrix contrasts with the compact nature of the sub-grains and is of the 'porous' variety containing phyllosilicate phases (fig. 6*a*). The iron-titanium phases within the clast have outer zones enriched in iron and cores of titanium oxide (fig. 6*b*). These textures are probably the result of diagenetic alteration of ilmenite, titanomagnetite, titanohematite, and rutile, the iron released in this reaction being represented by pyrite which has replaced iron hydroxide. Prior to sulphidation the cement was probably a ferric-iron clay which would account for the phyllosilicate inclusions in the pyrites. The porous pyrites (fig. 2) and the composite

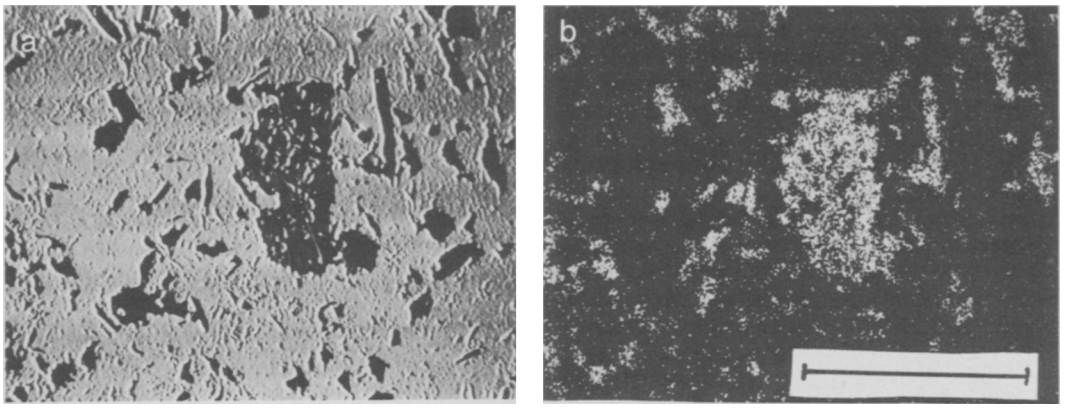

FIG. 5. (*a*) Back-scattered electron image of porous pyrite. (*b*) X-ray distribution of Al corresponding to phyllosilicates. 3974a. Scale Bar 0.1 mm.

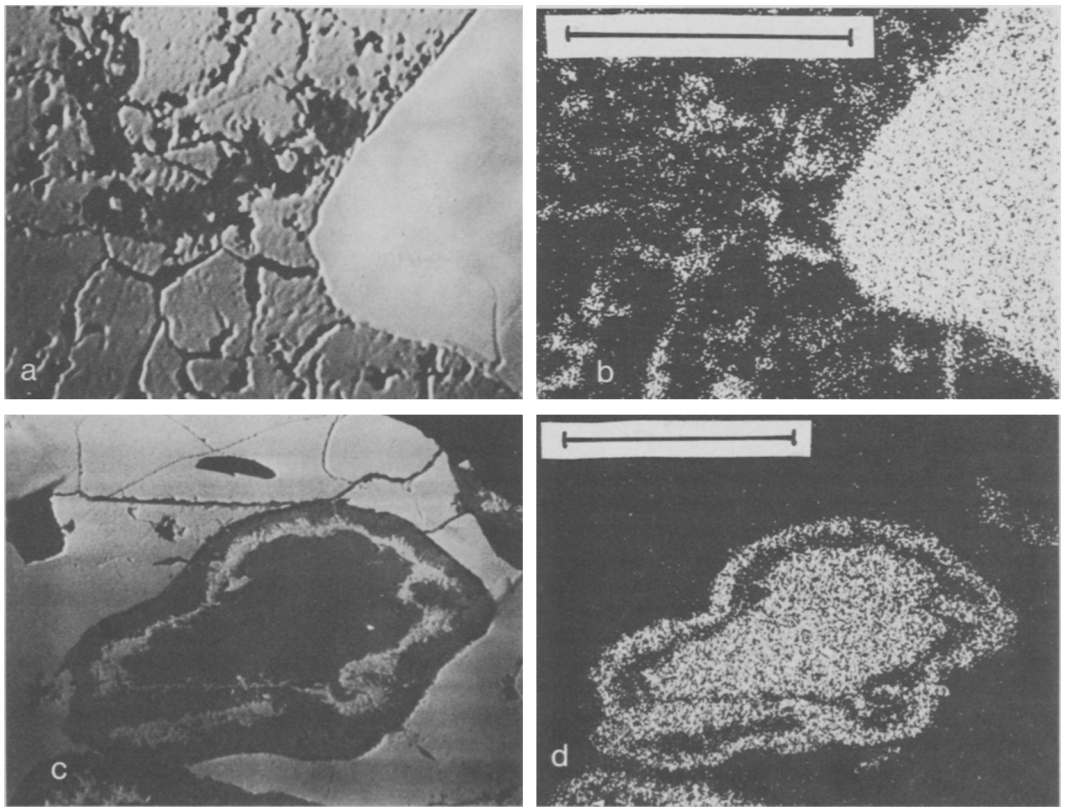

FIG. 6. (*a*) Back-scattered electron image of the matrix in the clast shown in fig. 5. The large grain is a zircon. (*b*) The X-ray distribution of Si outlines the sulphide grain boundaries. The matrix represents sulphidized ferric clay cement. 3974a. Scale Bar 0.5 mm. (*c*) Electron back-scatter image of a ferro-titanium grain from the clast shown in fig. 5. (*d*) The X-ray distribution of Ti outlines the grain. 3974a. Scale Bar 0.1 mm.

matrix of the clast (fig. 6*a*) may have a common origin, being derived from earlier cements and ferric clay, reworked from precursor sediment which were later sulphidized. This would account for their lack of hydraulic equivalence (fig. 1).

Pyrite at Elliot Lake. The pyrite at Elliot Lake shows similarities to that of the Rand (Robertson 1976, Roscoe 1969) although not so well documented. Syngenetic pyrite is less abundant, reflecting the lack of intrabasinal reworking and diagenesis at Elliot Lake. The principal pyrite type has a sulphidized core with abundant ragged authigenic overgrowths (fig. 8), producing grains which are larger than the detrital precursor (Bottrill 1971).

Iron in the Lower Proterozoic conglomerates. Fig. 7 presents the previous discussion in schematic form, whereby a typical placer assemblage including magnetite, limonite, and ferric iron clay clasts derived from precursor sediments are altered to pyrite. This model is compatible with oxidative weathering and diagenesis and hence with sulphate reduction during diagenesis as proposed by other authors.

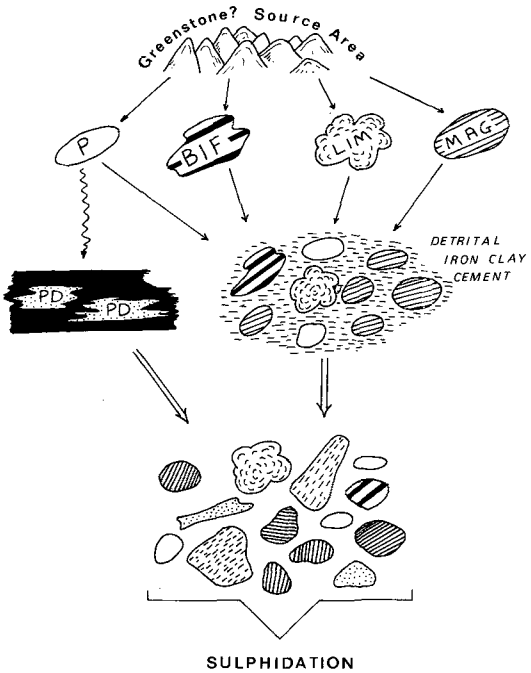

FIG. 7. A schematic model for the generation of sulphidized conglomerates in the Proterozoic, which also demonstrates the processes involved in the complex intra-basinal reworking of the Rand sediments. P, pyrite; BIF, banded iron formations; LIM, ferric iron clasts; MAG, magnetite; PD, diagenetic pyrite from intra-basin reduzate facies. Detrital iron clay cement of the first-formed sediment is derived from the breakdown of iron-bearing silicates common in the greenstone source areas.

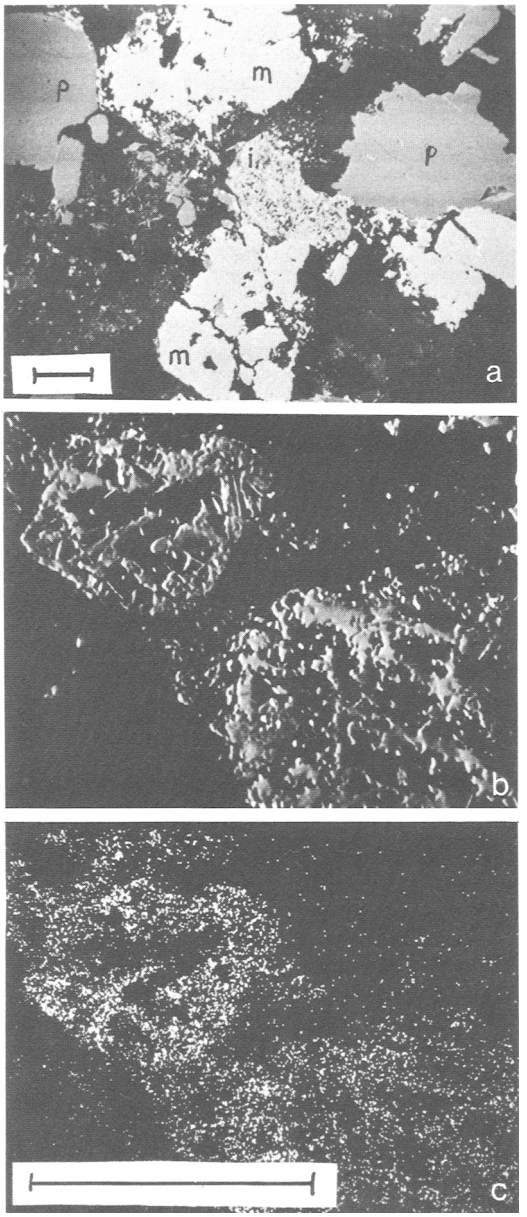

FIG. 8. (*a*) Back-scattered electron image of a typical Elliot Lake heavy mineral assemblage with monazite (m) and a titaniferous phase (i) which are both strongly altered and pyrites which have notable, ragged overgrowths. (*b*) Backscattered electron image of skeletal titaniferous grains in an advanced state of alteration. (*c*) The X-ray distribution of uranium coincides with the distribution of titaniferous grains in fig. 8*b*, 4206. Scale Bar 0.1 mm.

Uranium in Lower Proterozoic conglomerates. Uranium mobility in the conglomerates has been discussed by several authors. Liebenberg (1955) proposed a detrital origin and accounted for the fine-grained uraninite/carbon association by the invasion and shattering of detrital phases by a hydrocarbon fluid/gas phase polymerised by radiation. Many examples can be observed of such shattering. Other authors (Davidson and Bowie, 1951; Simpson and Bowles, 1981) advocate interaction of a uraniferous fluid with organic matter. Simpson and Bowles consider such a fluid to have been derived by inter-stratal oxidation of uraninite releasing the uranyl ion in solution and point out that this is the most plausible way for the uranium to have moved in solution. They suggest that the uranium precipitated in carbon seams, thought to be of algal mat origin as a result of sorption and reduction of the uranium to the less soluble U^{4+} by the organic matter. Uranium mobility in solution is also indicated by a decrease in the thorium content from predominantly detrital sediments (Elliot Lake and Dominion Reef) to the more mature reworked types (Witwatersrand). Such processes are consistent with the contrast in ages yielded by the uraninites from the Dominion Reef ($\sim$ 3 Gyr) and uraninites associated with hydrocarbon in the Upper Witwatersrand (2.0 Gyr). Thus the evidence is compatible with a detrital placer origin for the uraninite followed by sedimentary and diagenetic reworking after the deposition of the enclosing strata (cf. active 'roll fronts' in Carboniferous sediments). Such anomalous 'young' ages are often reported from uranium deposits.

Uranium and titanium in Elliot Lake conglomerates. In the Elliot Lake sediments degraded relicts of ilmenite and titanomagnetite are common (fig. 8) and are associated with uranium (fig. 8*b*). The uranium concentration increases with the degree of alteration of the grains. Phanerozoic uranium deposits in sandstones also commonly exhibit oxidation of the titanium phases which absorb quantities of uranium from groundwater (Reynolds and Goldhaber 1978). At Elliot Lake sections of ore horizons are usually uraninite-rich and grade distally into uraninite-deficient rocks (Theis 1978). Some uraninite crystals have altered and oxidized cores, with authigenic rims. Oxidized titanium phases are the principal uranium-bearing heavy minerals in distal deposits where they are associated with monazite and hydrocarbon-rich seams and muds (fig. 9), and the whole assemblage is comparable with the Witwatersrand (Simpson and Bowles, 1977, 1981).

Uraninite has apparently been oxidized *in situ* resulting in separation of uranium and thorium as solutions bearing uranyl ions migrate until they encounter reducing conditions when uranium is precipitated to form low thorium uraninite, or is absorbed in titanium phases. Reworking of such

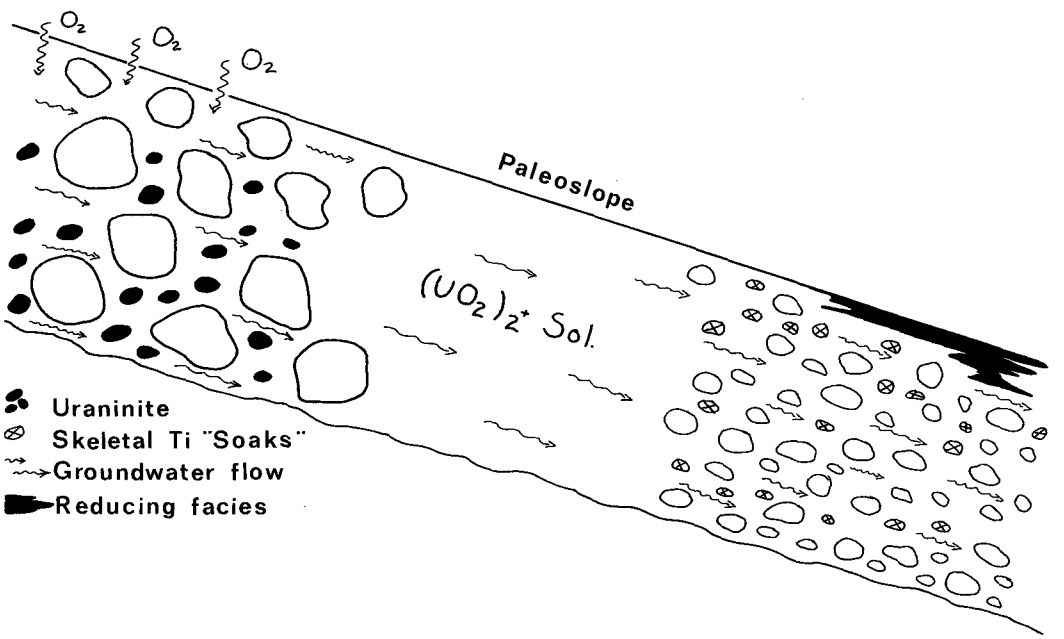

FIG. 9. A schematic model for the intrastratal oxidation of uraninite with remobilization and reprecipitation of uranium in more distal facies at Elliot Lake.

sediments would release uranium and titanium in solution, and also form fine-grained detritus of low-thorium uraninite and secondary urano-titanium minerals which would all concentrate in distal reduzate facies, as in the Witwatersrand.

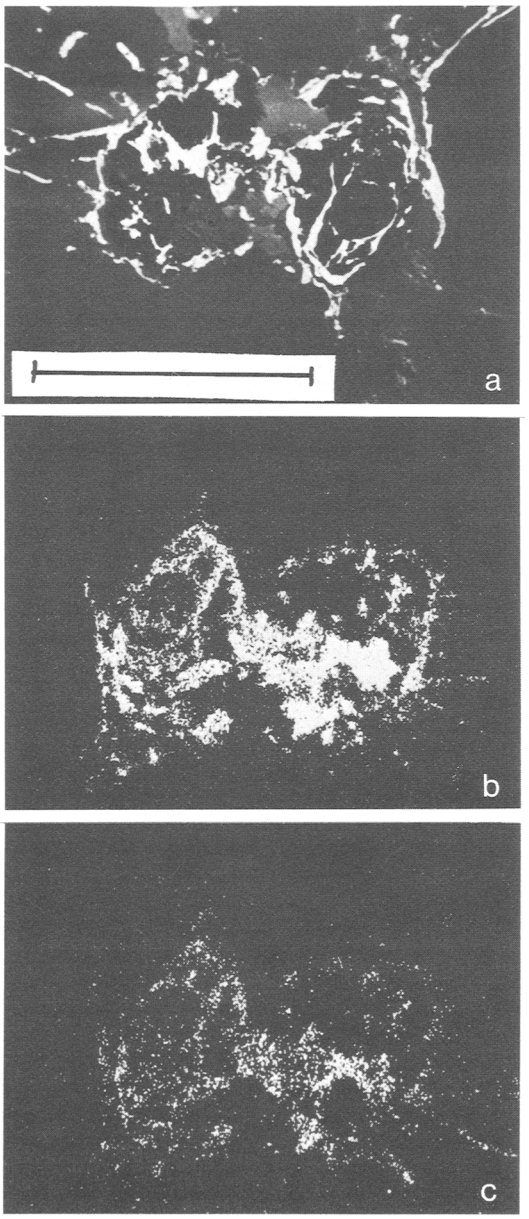

FIG. 10. (*a*) Back-scattered electron image of uraniferous phyllic minerals (light grey) nucleated around hydrocarbon specks. The X-ray distribution for titanium (b) and uranium (c) indicate brannerite. Witwatersrand. 3974c. Scale Bar 0.5 mm.

Uranium and titanium in the Witwatersrand conglomerates. Oxidized titanium-bearing phases are present, particularly in the Dominion and Vaal reefs, but Ti phases are less abundant than in black sands. Uranium is associated with titanium in phyllic minerals nucleated on specks of carbon (fig. 10) which provided a micro-reducing environment for precipitation of uraniferous and titaniferous solutions.

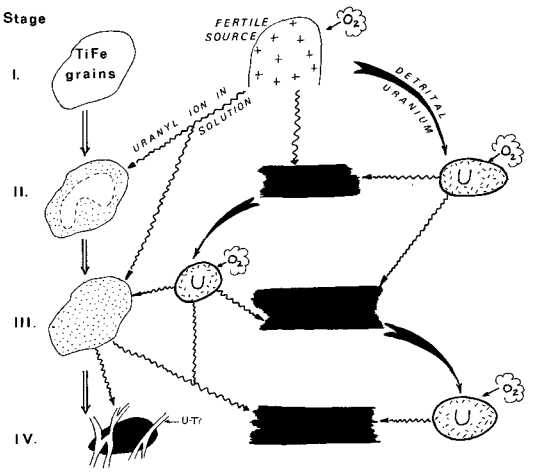

FIG. 11. Schematic model for uranium distribution in Proterozoic conglomerates. *Stage I*, derivation of uranium from source areas. *Stage II*, first sediment cf. Rand precursor grain (fig. 4). *Stage III*, reworked or diagenetically altered sediment. cf. Dominion Reef or Elliot Lake. *Stage IV*, reworked, mineralogically mature sediment cf. main Witwatersrand conglomerates. Stippling on the Ti/Fe phases indicates progressive alteration. Only small particles progress beyond stage III. The solid black arrows indicate reworking of primary and secondary uraninite and uranothorite grains; O_2 indicates oxidative diagenesis cf. fig. 9.

Summary of uranium in Proterozoic conglomerates. A model for uranium distribution in Proterozoic conglomerates during sediment diagenesis and reworking is presented in fig. 11 based on phase relations discussed in this paper. The model is compatible with oxidative weathering and diagenesis, and can be used to explain differences between the Witwatersrand and Elliot Lake conglomerates. It also accounts for the occurrence of detrital uraninite in the light of other evidence for oxidative weathering and diagenetic phenomena.

Gold in the Proterozoic conglomerates. Gold is associated with some varieties of porous pyrite in which it occurs as rims, crack and void fillings, and as disseminated blebs (Ramdohr, 1958; Simpson

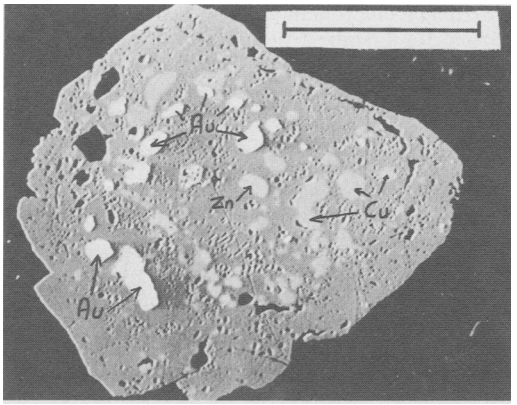

FIG. 12. Back-scattered electron image of porous pyrite with inclusions of gold, copper, and zinc sulphides. The copper and zinc are confined to the core, whereas the gold is generally disseminated. 3974. Scale Bar 0.1 m.

and Bowles, 1981). It is frequently associated with other metal sulphides (fig. 12), and is also concentrated in carbon seams (de Kock, 1964). Gold must therefore have been mobile in solution during diagenesis and deformation.

Examples of gold mobility in low-temperature surficial solutions are common especially in association with ferruginous rocks (Krauskopf, 1967). Limonite seeps often carry high gold values and gold crystals are reported from soil horizons (Warren, 1979) while replacement of plants in placer deposits and the occurrence of gold 'platelets' in the grass roots demonstrate the precipitation of gold from solution in reducing conditions. Gold may be enriched by a factor of twenty to thirty in gossans over massive sulphides in the pyrite belt in Spain and Mt. Lyell in Tasmania (Solomon, 1967). McIlveen and Stevens (1979) have shown that gold veins in Australia have undergone supergene enrichment, and gold nuggets in placers may originate from supergene extraction of gold from low bedrock values and concentration in palaeosols. Large placer fields form at the end of a geomorphological cycle, in association with deeply weathered bedrock and ferruginous gravels. For example in the Klondyke the highest values occur in quartz conglomerates containing red ferric clay cements, while the overlying gravels with white clay cement are usually barren (Hester, 1970; Spurr, 1898).

Witwatersrand and Elliot Lake conglomerates also formed at the end of a geomorphological cycle and are associated with weathered bedrock (Pienaar, 1963; Button and Tyler, 1979). The ore-bearing gravels are ferruginous and the precursor sediments may have had a ferric clay cement. Moreover the gold within pyrites is found in the type thought to have had a ferric phase precursor. The auriferous banded iron formations of the Kaapvaal Craton (Fripp, 1971) are a possible source. Saager *et al.* (1978) also suggested the iron formations as a source of the Rand gold. The Rand sediments therefore have all the characteristics of modern placer fields (Hallbauer and Utter, 1977). The high ferric iron contents of the pre-sulphidation sediment may have assisted in the movement of gold in solution during diagenesis and at least some of the gold within porous pyrite (fig. 12) may represent concentrations within the clay cement of the gravels.

Summary and conclusions. Reservations expressed recently concerning the use of pyrite and uraninite as indicators of an anoxic early Proterozoic atmosphere (Simpson and Bowles, 1977, 1981; Clemmey and Badham, 1981; Dimroth, 1979) are supported by evidence of ferric clasts and of oxidative diagenesis. A major sulphidation event which replaced magnetite and the ferric clasts would account for most of the pyrite in the sediments.

Uranium in solution as the uranyl ion was probably derived from oxidation of uraninite in the basement, from intrabasinal detrital uraninite of primary and secondary origin and also from leaching of intrabasinal tuffs. The uranium was redeposited on encountering reducing conditions such as organic matter or titanium oxide 'soaks'. The model proposed here for uranium deposits in Proterozoic conglomerates is similar to those used to account for 'red-bed, roll front' deposits in the Phanerozoic. The similarities between the ferruginous clay gravels and black sand ores of recent placer fields and their Proterozoic counterparts support a placer origin for the gold. The ore deposits are in supracrustal rocks derived from reworking of Archean crust enriched in uranium and gold (Badham, 1978).

The models presented are inconsistent with the concept of an anoxic atmosphere in the Proterozoic. Such processes as sulphidation, intrabasinal reworking, oxidative weathering, and diagenesis which best account for the features of the deposits are incompatible with a reducing or anoxic atmosphere. Furthermore they have modern analogues.

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