product was similar to that of the original sample, but showed considerably less absorption than the original sample ($<0.2\ \mu$ size fraction). X-ray analysis indicated kaolin was still present. Either the iron-rich kaolin is a mixture of kaolin and very fine geothite or it is an Fe^{3+} kaolin which has a Mössbauer spectrum identical to goethite (which might be expected) and from which the octahedral iron can be removed by a method designed to remove "free" iron oxides.

The spectrum of siderite ($FeCO_3$) is a quadrupole doublet typical of the Fe^{2+} ion (Fig. 2). This spectrum exhibits peak broadening as a result of (i) nonuniformity of motion from the constant velocity drive, and (ii) absorption in a sample of appreciable thickness (*14*). The area of the absorption peak at +1.90 mm/sec equals that of the peak at +0.10 mm/sec, but the former is considerably more affected by non-uniformity in velocity. Both peaks are somewhat broadened because this sample was overly thick. A reanalysis of a thinner sample of siderite showed narrower peaks, but the position of the peaks is unchanged.

It should be noted that all of our peaks at about +2.0 mm/sec are somewhat broadened because of non-uniform velocity. For those clay samples of high iron content there is probably slight broadening because of absorption effects, but in no case did the clay samples have a thickness of iron as great as that of the siderite sample.

In summary, Mössbauer absorption spectrography should prove to be of considerable value for studying the sheet-structure silicates. The oxidation state of iron can easily be determined. It appears to be possible to distinguish between octahedrally and tetrahedrally coordinated iron. Minor variations in the spectra of the various minerals should prove to be significant and afford information on variations in the character of the octahedral layer.

Mössbauer absorption can be detected for iron present in amounts as low as 1 percent Fe_2O_3, but a higher concentration is required if useful spectra are to be obtained. Siderite as an impurity can usually be identified, but goethite will cause trouble. Conversely, goethite can be detected more reliably with Mössbauer equipment than with x-ray.

By increasing the count to achieve better statistics, and by minimizing the line-broadening effects noted above, it should be possible to analyze the fine structure of the complex peaks near zero velocity to supply much more information than the present preliminary study suggests.

CHARLES E. WEAVER
J. M. WAMPLER
T. E. PECUIL

Department of Earth Sciences, Georgia Institute of Technology, Atlanta 30332

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1 March 1967

Alga-Like Fossils from the Early Precambrian of South Africa

Abstract. *Micropaleontological studies of carbonaceous chert from the Fig Tree Series of South Africa (> 3.1 × 10⁹ years old) revealed the presence of spheroidal microfossils, here designated* Archaeosphaeroides barbertonensis, *interpreted as probably representing the remnants of unicellular alga-like organisms. The presumed photosynthetic nature of these primitive microorganisms seems corroborated by organic geochemical and carbon isotopic studies of the Fig Tree organic matter, and is consistent with the geologically and mineralogically indicated Early Precambrian environment. These alga-like spheroids, together with a bacterium-like organism previously described from the Fig Tree chert, are the oldest fossil organisms now known.*

Laminated stromatolitic structures, comparable in gross morphology to the biohermal deposits of modern blue-green and red algae, have long been known from sediments of Precambrian age. Although generally devoid of cellularly preserved microfossils, these structures have been regarded as presumptive evidence of early algal activity. Firm evidence for this supposition comes from the recent investigations of siliceous stromatolites of Middle Precambrian age from the Gunflint Iron Formation (1.9×10^9 years old) along the northern shore of Lake Superior in Ontario, Canada. Various microorganisms, some morphologically similar to modern blue-green algae, are structurally preserved in the organic lamellae of these black chert stromatolites (*1, 2*). Cellular microfossils are similarly preserved in laminated, primary black cherts associated with stromatolites of the genus *Collenia* in the Late Precambrian Bitter Springs Formation (about 1.0×10^9 years old) of central Australia (*3*). Morphologically, several of these billion-year-old organisms are referable to modern families of blue-green and green algae. These two microfossil assemblages and the many deposits of calcareous stromatolites and carbonaceous sediments of comparable age establish that diverse types of primitive algae were present relatively early in geologic time.

The oldest known stromatolites are apparently those described and figured by A. M. Macgregor (*4*) from Early Precambrian limestones near Turk Mine, 53 km north-northeast of Bulawayo, Rhodesia. Columnar, dentate, and domical structures have been noted in the deposit; the domical forms have been compared with *Collenia* (*4*). Comparison of the stable isotopes C^{12} and C^{13} in reduced organic carbon, present as finely bedded graphitic laminations, and in oxidized inorganic carbon, present in the carbonate matrix, demonstrated an isotopic fractionation probably of photosynthetic origin (*5*). Microfossils have not been reported in the calcareous Bulawayan stromatolites, but the gross morphology, carbon isotopic composition, and presence of organic matter are consistent with an algal origin. The Bulawayan limestones have been correlated, by lithologic similarity and stratigraphic setting (*6*), with sediments occurring in the Bikita tin field, about 300 km east of Bulawayo (*6, 7*). A pegmatite dike, intrusive in these sediments near Bikita, has been dated at 2640 ± 40 million years (*7–9*). The Bulawayan stromatolites have therefore been regarded as older than about 2.6 $\times 10^9$ years; some authors suggest an age approaching 3 billion years (*10*).

Several investigators have examined other Early Precambrian sediments for evidence of biological activity (*11*). Of particular interest are shales and cherts from the Fig Tree Series of the Upper Swaziland System, eastern Transvaal, South Africa (*12–14, 21*). The Fig Tree sediments comprise a thick sequence of graywackes, slates, and shales with interbedded well-developed horizons of chert, jasper, and ironstone. These sediments overlie pyroclastic deposits and dolomitic limestones of the Lower Swaziland System, and they immediately underlie sediments of the Moodies System (*15*). This Early Precambrian sedimentary sequence occurs in an area actively mined for gold; the geology, stratigraphy, and structural relationships of these deposits have been extensively studied (*15, 16*).

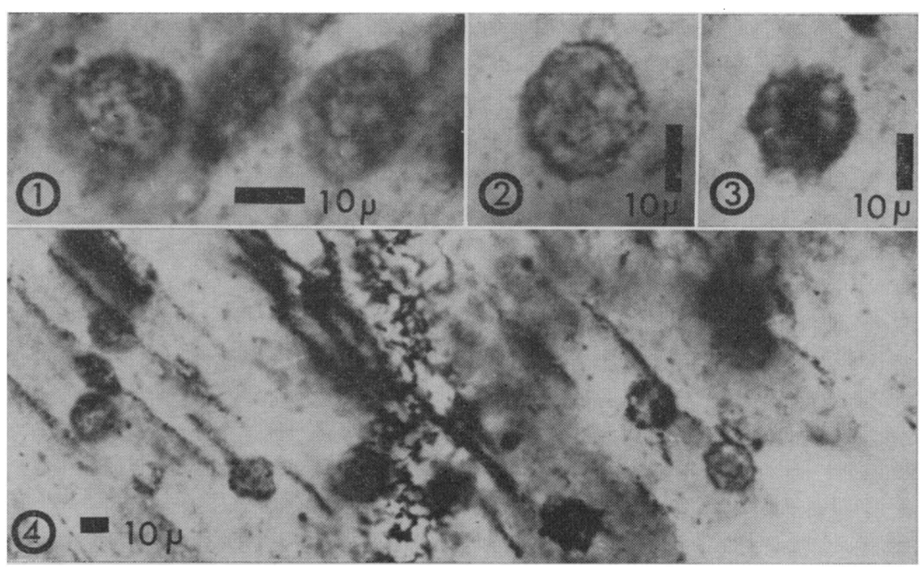
The age of the Swaziland System has been the object of considerable geochronological investigation (*9, 17*). Results have consistently indicated an age of deposition greater than 3 billion years, and a granite intruding the Swaziland System has a tentative rubidium-strontium age of 3440 ± 300 million years (*17*). Recent measurements on shales and graywackes from the Fig Tree Series, with the rubidium-strontium whole-rock method, indicate a minimum age of 3.1×10^9 years for the last period of strontium isotopic homogenization of these sediments (*18*).

Biohermal stromatolites have not been reported from the Fig Tree sediments. Ramsey (*15*), however, has figured apparently deformed, concentrically laminated "oolites" which he suggests may be of algal origin. These curious structures occur in graywackes of the sequence, and are stratigraphically and lithologically unrelated to the carbonaceous cherts which we investigated for possible microfossil content.

Chalcedonic black cherts, interpreted as primary in origin (*12*), are particularly abundant in the upper third of the Fig Tree Series. The physical organization of the organic matter indigenous to these cherts was studied in thin sections and in hydrofluoric acid macerations by optical microscopy, and in surface replicas and in macerations by electron microscopy. Much of the carbonaceous material occurs as irregularly shaped, minute, dispersed organic bodies predominantly aligned parallel to bedding planes, and, as seen in the optical microscope, it is generally devoid of evident biogenic morphology. However, electron microscopy revealed a minute, rod-shaped bacterium-like organism, *Eobacterium isolatum*, structurally and organically preserved in this Early Precambrian sediment (*12*). Also present in thin sections and in surface replicas of the chert are irregular filamentous organic structures morphologically comparable to degraded plant material (*12*); their probable biological origin, however, cannot be established by morphology alone.

Spheroidal, dark-colored organic bodies, comparable in general morphology to certain modern unicellular algae, have now been discovered in the same locality and black chert facies of the Fig Tree Series as *E. isolatum* (*19*). Figures 1–4 show several of these bodies photographed in thin sections in transmitted light. Both well-defined spheroids (Figs. 1–4) and distorted, partially flattened cell-like remnants (Figs. 1 and 4) are present. Measurements of 28 spheroids show that they range in diameter between about 15.6 μ and 23.3 μ, with an average of 18.7 μ (Fig. 5). The cell-like spheroidal bodies, which often exhibit a reticulate surface texture (Fig. 1), are three-dimensionally preserved in the chalcedonic matrix. Although sufficient organic matter is present to outline the shape of these spheroids, the walls are often rather poorly preserved. In better-preserved spheroids the wall is about 1 μ thick (Fig. 2). Irregular masses of organic material occasionally present within the spheroids (Fig. 3) appear to represent the coalesced and "coalified" remnants of the original internal contents, a feature rather commonly observed in fossil algae preserved in cherts (for example, *20*, plate 44). The organic bodies are two to six times larger than the chalcedony grains of the chert matrix in which they are embedded, and the boundaries of the chalcedony grains pass through the organic structures without deforming their spheroidal shape. They are demonstrably not organic coatings of mineral grains, and clearly not mineral artifacts. Similar organic spheroids, interpreted as being of biological origin, from the Fig Tree sediments were reported by H. Pflug (*21*).

In the paleontological assessment of



Figs. 1–4. Spheroidal, organic alga-like microfossils (*Archaeosphaeroides barbertonensis* n. gen., n. sp.) in black chert of the Early Precambrian Fig Tree Series (> 3.1×10^9 years old), near Barberton, South Africa. All structures shown in thin sections photographed in transmitted light. Fig. 1. Spheroidal and somewhat distorted alga-like fossils showing the typical, irregularly reticulate surface texture. Fig. 2. Median optical section of the type specimen of *A. barbertonensis*, showing the continuity and somewhat variable thickness of the cell wall. Fig. 3. Alga-like spheroid containing "coalified" organic material interpreted as representing the coalesced remnants of the original cytoplasmic cellular contents. Fig. 4. Organic spheroidal microfossils exhibiting varying degrees of completeness of preservation. The diagonally oriented lamellae are composed of organic particles aligned parallel to the bedding planes; the vertically oriented irregular dark material is anthraxolitic.

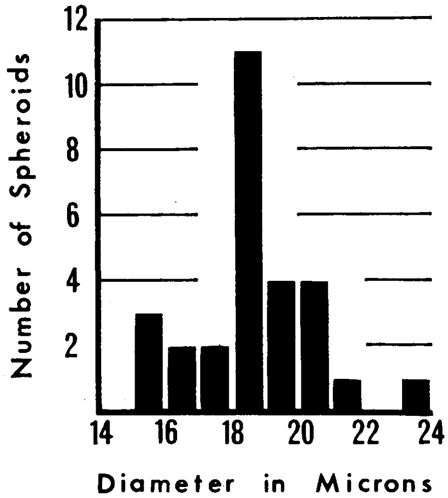


Fig. 5. Size distribution of 28 well-defined representatives of *Archaeosphaeroides barbertonensis* n. gen., n. sp., in chert from Fig Tree Series. The diameters of the alga-like bodies were measured from photomicrographs showing the organisms in thin sections of the chert.

minute structures of Precambrian age and of possible biological origin, it is often difficult to differentiate between inorganically produced pseudofossils and partially degraded remnants of primitive microorganisms. This is particularly true in the interpretation of structures from sediments such as those of the approximately 3 billion-year-old Fig Tree Series, for which there are no known fossils of equivalent age for morphological comparison. Nevertheless, the Fig Tree spheroids are almost certainly of biological origin, probably representing the remnants of single-celled alga-like microorganisms. Their organic composition, morphological consistency, limited size range, mode of preservation, and morphological similarity to known spheroidal algae, both modern and fossil, support this interpretation. Their physical relationship to grains of the chalcedonic matrix seems to render an inorganic origin untenable, and they are demonstrably indigenous to the sediment and are not laboratory contaminants.

In size, shape, and general organization, the Fig Tree spheroids are comparable to certain members of the modern blue-green algal group Chroococcales. They are morphologically dissimilar to known bacteria. In general appearance they seem quite similar to reticulate, spheroidal alga-like microorganisms from the Middle Precambrian Gunflint chert (Figs. 6 and 7), and they are morphologically comparable to well-preserved blue-green algae from the Late Precambrian Bitter Springs Formation (Fig. 8). However, perhaps because of their relatively poor preservation, the Fig Tree organisms cannot be assigned with certainty to any extant algal group. Moreover, in view of the extraordinary antiquity of these organisms, and the intervening 3 billion years of evolutionary history, there is no reason to expect that modern morphological counterparts exist.

For purposes of reference, it seems desirable to propose a taxonomic designation for these spheroidal microfossils and, in view of their somewhat uncertain biological affinities, to designate them by a binomial having morphological and geological, rather than phyletic, significance or implication.

Archaeosphaeroides, new genus.

Diagnosis: Cells small, spherical, spheroidal, or ellipsoidal, not angular; solitary, unicellular, noncolonial; more or less circular in cross section. External wall texture varies from psilate or nearly psilate to coarsely reticulate. Cell walls membranous, thin, may be ruptured and distorted. Internal organic matter may be present.

Etymology: With reference to Early Precambrian age and spheroidal form of type species.

Type species: *Archaeosphaeroides barbertonensis,* new species.

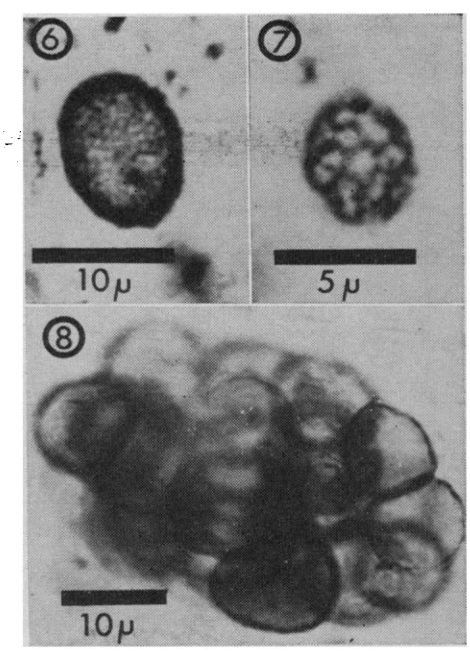
Diagnosis: Unicellular, isolated organic spheroids. Cross-sectional diameter varies between about 15 and 24 μ, with an average diameter of approximately 19 μ. Cell wall delicate, thin, commonly of somewhat variable thickness, often coarsely and irregularly reticulate. Internal organic matter occasionally present, clumped in an irregular mass near the center of the cell.

Etymology: With reference to type locality in the Barberton Mountain Land, near Barberton, South Africa.

Type locality: Black chert facies in upper third of Early Precambrian Fig Tree series, Upper Swaziland System, exposed by road cutting 100 m northwest of surface opening to Daylight Mine (Barbrook Mining Co.), 28 km east-northeast of Barberton, eastern Transvaal, South Africa.

Type specimen: Figures 1–4 show representative members of the species; Fig. 5 shows the range of measured diameters for 28 well-defined individuals; the organism shown in Fig. 2 is cited as the type specimen (thin section D/FT-1, Paleobotanical Collection, Harvard University, No. 58436).

Recent studies of the organic chemistry and carbon isotopic composition of the Fig Tree organic matter, and a consideration of the geology and mineralogy of the Fig Tree sediments, appear to provide additional evidence for the existence of photosynthetic organisms in Fig Tree time. The reactants in green-plant photosynthesis, carbon dioxide and water, were apparently present in the Fig Tree environment. The precipitation of calcite and of dolomite seems to require the availability of free carbon dioxide (*22*). Thus, the occurrence of sedimentary dolomitic limestones in the Lower Swaziland System (*15*), immediately underlying the Fig Tree sediments, may be interpreted to indicate that atmospheric CO_2 was present during this period of geologic time. Such an interpretation is consistent with the occurrence of limestones of comparable age in the Bulawayan Group of Rhodesia. The presence of liquid water is indicated by the occurrence of water-laid deposits, containing such dep-



Figs. 6–8. Spheroidal, organic alga-like microfossils in black cherts of Middle and Late Precambrian age. All structures shown in thin sections photographed in transmitted light. Fig. 6. Type specimen of *Huroniospora microreticulata* Barghoorn (*1*), from chert of the Middle Precambrian Gunflint Iron Formation (1.9×10^9 years old), Ontario, Canada. Fig. 7. Type specimen of *H. macroreticulata* Barghoorn (*1*), from the Gunflint chert, showing the morphological similarity between this organism and *Archaeosphaeroides barbertonensis.* Fig. 8. Well-preserved, spheroidal blue-green algae from chert of the Late Precambrian Bitter Springs Formation (*3*), about 1 billion years old, from central Australia.

ositional features as pillow lavas, current-bedding, ripple marks, and mud cracks, in the Swaziland and overlying Moodies System (*15*). The existence of hematite deposits in the form of banded iron-stones and ferruginous shales in the Fig Tree Series has been interpreted as indicating the presence of free oxygen (*23*), possibly produced by photosynthetic organisms. However, as Holland pointed out (*22*), ferrous iron can be oxidized in the presence of very low concentrations of atmospheric oxygen; this oxygen may conceivably reflect the abiotic, ultraviolet-induced photolysis of water, rather than the presence of photosynthesis. Thus, the geological and mineralogical evidence from the Fig Tree sediments seems consistent with, but probably does not necessarily require, the presence of a photosynthetic biota.

Carbonaceous cherts and shales from the Fig Tree Series contain about 0.5 percent of organic matter, of which somewhat less than 1 percent is soluble in a 3 : 1 benzene : methanol solvent solution. In this soluble fraction, W. Meinschein (*24*) and T. Hoering (*13*) identified alkane hydrocarbons, ranging from about 17 to 35 carbon atoms in length and "typical of hydrocarbons formed in association with biologically produced organic matter of all ages" (*13*). The isoprenoid hydrocarbons pristane and phytane have been reported as minor constituents of the indigenous alkanes (*12, 14, 24*). Pristane and phytanes have been identified in other Precambrian deposits containing organically preserved plant microfossils (*2, 25*), and they are widely distributed in photosynthetically produced organic matter of younger geologic age (*26*). These isoprenoids are commonly regarded as geochemical or biochemical derivatives of the phytyl alcohol moiety of chlorophyll (*2, 14, 25, 27*), and for this reason their presence seems suggestive of photosynthetic activity. In the modern biota, isoprene derivatives are apparently represented in all organisms with the exception of nonphotosynthetic anaerobic bacteria (*28*). Thus, judging from extant organisms, the occurrence of fossil isoprenoids seems to require the existence of organisms different from, and probably more advanced than, nonphotosynthetic anaerobes. The reported occurrence of pristane and phytane in the Fig Tree sediments is consistent with, and seems to suggest, the presence of photosynthetic microorganisms.

In the process of photosynthesis, modern plants tend to selectively metabolize carbon dioxide containing the lighter carbon isotope C^{12} in preference to CO_2 containing the heavier isotope C^{13}. This partial fractionation results in an enrichment of C^{12} in photosynthetically produced organic matter, as compared with its concentration in the atmosphere and in precipitated carbonate sediments. Park and Epstein (*29*) have shown that in tomato plants this fractionation is biochemically complex, apparently involving both kinetic and enzymatic processes favoring the preferential incorporation of the lighter isotope in the organic matter produced. Ratios of C^{12} to C^{13} have been measured in many types of materials, of both modern and geological age, and of both biological and inorganic origin (*30*).

Hoering (*13*) determined the carbon isotopic composition of both the benzene-soluble and the insoluble kerogen fractions of organic matter indigenous to the Fig Tree sediments. The measured isotopic ratios fall well within the range exhibited by many crude oils and other organic materials of known photosynthetic origin or derivation, and they are distinctly different from values characteristic of inorganically produced carbon compounds. The carbon isotopic composition of the Fig Tree organic matter is comparable to that measured on reduced carbon from the Middle Precambrian Gunflint chert (*1, 2*), and to the isotopic composition of organic compounds from the Late Precambrian Nonesuch Shale (*25*), both of which contain pristane and phytane, as well as cellularly preserved plant microfossils.

The chemical pathways from the sedimentary deposition of organic matter to kerogen and the extractable organic components in rocks are not known. In addition, the complexities of the carbon-cycle in nature and its possible variations over geologic time are not well understood. Nevertheless, green-plant photosynthesis is the sole means of carbon fixation and isotopic fractionation of quantitative importance operating in modern environment, and the observed $C^{12} : C^{13}$ ratios seem indicative of its existence in Fig Tree time.

Together with *Eobacterium isolatum*, a bacterium-like microorganism previously described from the Fig Tree chert, the organic spheroids, here designated *Archaeosphaeroides barbertonensis*, constitute the oldest fossil organisms now known. These spheroidal microfossils probably represent the remnants of unicellular, noncolonial alga-like organisms. Possibly they are related to, or were perhaps the evolutionary precursors of, modern coccoid blue-green algae. The presumed photosynthetic nature of these primitive microorganisms seems corroborated by geological and organic geochemical considerations. The carbon isotopic composition of the Fig Tree organic matter and the reported occurrence of organic compounds related to chlorophyll suggest the presence of photosynthetic activity; the geology and mineralogy of these Early Precambrian sediments are consistent with this interpretation. The apparent existence of photosynthetic microorganisms in Fig Tree time, more than 3100 million years ago, is similarly in accord with the occurrence of laminated stromatolites of probable algal origin in Early Precambrian limestones near Bulawayo, Rhodesia. Moreover, the early appearance of the photosynthetic process provides a credible and logical explanation for the extensive occurrence of reduced carbon and organic compounds in many Early Precambrian sediments. Photosynthetic algae or alga-like organisms probably originated quite early in the evolution of biological systems, apparently during the 30 percent of the earth's history preceding the deposition of the Fig Tree Series.

J. WILLIAM SCHOPF
Department of Biology and *Society of Fellows, Harvard University, Cambridge, Massachusetts 02138*

ELSO S. BARGHOORN
Department of Biology and *Botanical Museum, Harvard University*

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23 January 1967

Uranium Distribution in Rocks by Fission-Track Registration in Lexan Plastic

Abstract. *When Lexan plastic is used to register fission tracks from thermal neutron-induced fission of uranium in rocks, a print of the rock texture is formed on the plastic surface after chemical etching. This print allows positive, rapid location of uranium-bearing phases in the rock and accurate determination of uranium abundances.*

The registration of fission tracks from the thermal neutron-induced fission of U^{235} in uranium-bearing materials (*1*) has been used to determine quantitatively the distribution of uranium in a number of natural materials (*1–4*). Hamilton (*3*) used mica to observe uranium distributions in thin sections of meteorite and rock, while Kleppe and Roger (*4*) used both mica and Mylar for a similar study of a thin section of a granodiorite from the Sierra Nevada batholith. However, both studies were limited by difficulties in comparing the occurrences of fission tracks in the registering material with the source phases in the rock or meteorite section. Fleischer (*5*) partially overcame this difficulty by using a thin skin of plastic which remains tightly bonded to the surface during irradiation, etching, and observation.

To study uranium distribution in rocks of deep-seated origin by the fission-track method, we prepared polished, thin sections of many of these rocks, placed a sheet of Lexan plastic (*6*) in contact with each sample, and held it there with a clip during irradiation in a predominantly thermal-neutron flux. After irradiation, the plastic was removed and etched in 6*N* NaOH at 70°C for 8 minutes. Location marks had been carefully placed on both rock surface and plastic sheet so that we could refer the occurrence of fission tracks in the plastic to individual phases in the rock section. Despite the use of a rather complicated system of these marks, it would normally be very difficult, if not impossible, to correlate small areas of fission-track aggregations with tiny uranium-bearing phases in the rock section.

However, after the Lexan plastic sheets were etched, the surface which had been in contact with the rock section during irradiation developed a detailed print of the rock surface. Grain boundaries and cracks were shown with remarkable accuracy, and each mineral phase present in the rock could be identified by its shape and characteristic effect on the plastic. The broad features of these Lexan prints are best seen under oblique lighting (Fig. 1), while detailed studies may be made with transmitted light under a microscope (Figs. 2 and 3). Fission tracks in the plastic, arising from uranium-bearing phases in the adjoining rock, are also clearly visible after etching, but the immediate advantage of using Lexan as a track register is that fission-track sources can be seen in accurate and unequivocal relation to the boundaries of the various phases in the rock.

The Lexan print (Fig. 2) of a pyroxene granulite inclusion from the deep-seated basic breccia pipe at Delegate (*7*) shows that the visible fission tracks are randomly distributed within the boundaries of a single mineral grain. Reference to the rock section readily establishes that this mineral is apatite, and the obviously homogeneous uranium content was calculated (*1*) as 2.3 parts per million. A Lexan print of an eclogitic inclusion from the breccia pipe (Fig. 3) shows that the fission tracks are confined within the boundaries of zones of secondary hydrated silicate minerals (kelyphite) which surround the garnets and also form tiny veins through the rock. These secondary phases result from reactions with hydrothermal fluids entering the inclusion from the volatile-rich basic material filling the pipe. We conclude that by far the largest proportion of the total uranium content of this eclogitic inclusion was introduced after the fragment was caught up in the pipe.

The major cause of the contrast between various minerals in the Lexan print is a variation in the density of small, shallow pits present on the plastic surface after etching (Fig. 3). Grain boundaries and cracks in minerals are also visible as the result of an increase in pit densities (Fig. 2). These shallow pits are quite distinct from the long (approximately 10 μ) linear track arising from the passage of massive, charged particles from induced U^{235} fission (Fig. 3). By analogy with the ion explosion spike theory for the origin of fission tracks (*8*), we suggest