The tracks left by the spontaneous fission of traces of uranium in many minerals are clues to the minerals' age. One of the main advantages of the technique is the broad span of time it covers

by J. D. Macdougall

The ability to measure the actual age of an ancient rock formation or a bone or an artifact of human culture is a comparatively new one. It stems from the fact that the radioactive isotopes of many common elements break down spontaneously at fixed rates. A familiar example is an isotope that is present in all living matter: carbon 14. Now a new radiometric method of absolute dating has begun to revolutionize chronological investigations in such diverse disciplines as cosmology, geology and archaeology. The new method measures the spontaneous fission of certain heavy elements, in particular the fission of the most abundant isotope of uranium, U-238. Given an appropriate sample for analysis, one can determine its absolute age over an enormous span of time: from as recently as a few decades ago to as far back as the time when the solar system was formed. The method is known as fission-track dating.

Fission tracks were discovered in the late 1950's by two workers at the British Atomic Energy Research Establishment at Harwell, E. C. H. Silk and R. S. Barnes. They had exposed samples of the crystalline mineral mica to ions produced by the fission of heavy elements. The ions were slowed and finally stopped by a series of interactions with the atoms in the crystal lattice of the mineral. As they did so they left thin linear tracks that were visible at extremely high magnification. In 1959 Silk and Barnes published electron micrographs of some of those tracks in the British physical journal *Philosophical Magazine*, thus establishing a new field of investigation.

It was soon demonstrated that almost any kind of energetic heavy ion would make tracks in a wide variety of solids. Then one group of investigators, Robert L. Fleischer, P. Buford Price and Robert M. Walker, who were then working at the General Electric Research and Development Center in Schenectady, pointed out that the spontaneous fission of the small amounts of uranium present as "impurities" in many different rocks would leave such tracks in crystals of the rock. Uranium 238 fissions at a characteristic rate, and when it does so, the two halves (more or less) of its nucleus violently recoil from each other and leave a characteristic track. Fleischer, Price and Walker suggested that studies of the number of tracks in the crystals of rocks would provide clues to the absolute age of the rocks. Today's application of this kind of track analysis to a multitude of questions in such unrelated fields as cosmic-ray physics and the study of human evolution has come about mainly through the pioneering work of these three men.

One of the first problems that had to be solved before fission-track dating could become a practical tool had to do with the fact that the tracks themselves were nearly invisible. In the mica specimens produced by Silk and Barnes the region affected by each impinging ion was only about 100 angstroms across—a fiftieth of a wavelength of light. At the high electron-microscope magnifications necessary to resolve such ultrafine features it would take many weeks to scan a single square centimeter of material in search of fission tracks. Then it was discovered that the region along the fission track where the crystal lattice is disrupted is less resistant to attack by a solvent than the undisrupted area of the lattice. For example, putting mica in an acid bath for an appropriate length of time dissolves the disrupted region along the track, creating a hole 100 or more times larger than the original track. This enlargement brings the tracks up to the micrometer range, where they are readily visible under the light microscope at magnifications of 500 to 1,000 diameters. Comparatively extensive surface areas can then be scanned quickly and accurately.

Precisely how are the fission tracks formed? The most satisfactory answer at present is one suggested more than a decade ago by Fleischer, Price and Walker and embodied in what they call the ion-explosion-spike model. According to this model, the fission fragment, moving at high speed and having a strong electric charge, ionizes the atoms along its path in the target material by stripping them of some of their electrons. If the target material is a conducting solid, as metals and certain minerals are, the stripped electrons are immediately replaced by free electrons, and the crystal lattice remains undamaged. If, on the other hand, the material is an insulating solid, as most rock-forming minerals are, the stripped electrons are not replaced, and the atoms are left with a net positive charge. Their mutual electrical repulsion pushes them apart, damaging the crystal. With one significant exception, the damage persists indefinitely.

An insulating solid can record a fission track even if it is not a crystal. Amorphous solids such as volcanic glasses, man-made glasses and plastics are also damaged by fission fragments. The process of track formation in these substances involves the breaking of chemical bonds rather than the disruption of a crystal lattice, but the net result is the same: a narrow region along the path of the fission fragment is made less resistant to chemical attack than the surrounding material.

The basic method of dating by fission-track analysis is to count the number of tracks per unit area of the sample; in general the more tracks there are, the more fissions have occurred and the older the sample is. The method may seem somewhat different from other radiometric dating techniques, such as measuring the relative abundance of radioactive potassium and its argon daughter isotope, or of radioactive rubidium and its strontium daughter isotope. Actually the two kinds of dating are closely analogous; the only fundamental difference is that the one technique measures a product of decay and the other an effect of decay. Spontaneous fission is, however, a much rarer event than other kinds of radioactive decay. For example, hen one counts the tracks produced by anium fission, their number may be in e hundreds, with each track recording e fission of one atom of uranium 238. ompare this with a high-sensitivity pottsium-argon measurement, requiring out a ten-millionth of a cubic centieter of argon 40. The argon sample presents the spontaneous decay not of few hundred atoms but of roughly ree trillion. The far smaller number of cays that provide the basis for fissionack analysis accounts for one of the eat advantages of the method: very nall samples can yield useful data. Unr favorable circumstances the analysis of even a single microscopic crystal or a fragment of volcanic glass can determine the age of the specimen.

The average concentration of uranium in the rocks of the earth's crust is no greater than a few parts per million. Even at this low concentration the passing of a million years or so sees enough spontaneous fissions to leave a measurable number of fission tracks in some crystals. Furthermore, most of the crystalline rocks of the earth's crust are far more than a million years old, and so their crystals have accumulated a large number of tracks even though they contain little more than trace amounts of uranium.

Uranium 238 is the only significant producer of tracks in terrestrial rocks and in natural and man-made glasses. Other spontaneously fissioning elements exist, but they are rare and their rate of fission is slow. For example, it is estimated that the tracks produced by the spontaneous fission of another isotope of uranium, U-235, and thorium 232 combined represent less than .5 percent of the total number of tracks in the large majority of samples. As we shall see, however, the fact that even minute quantities of U-235 are present in glass-

SSION TRACKS in a crystal of zircon appear in this light microaph. The magnification is 3,100 diameters. Visibility of the tracks of each track from about 100 angstroms to a micrometer or more. Each track records the spontaneous fission of an atom of uranium

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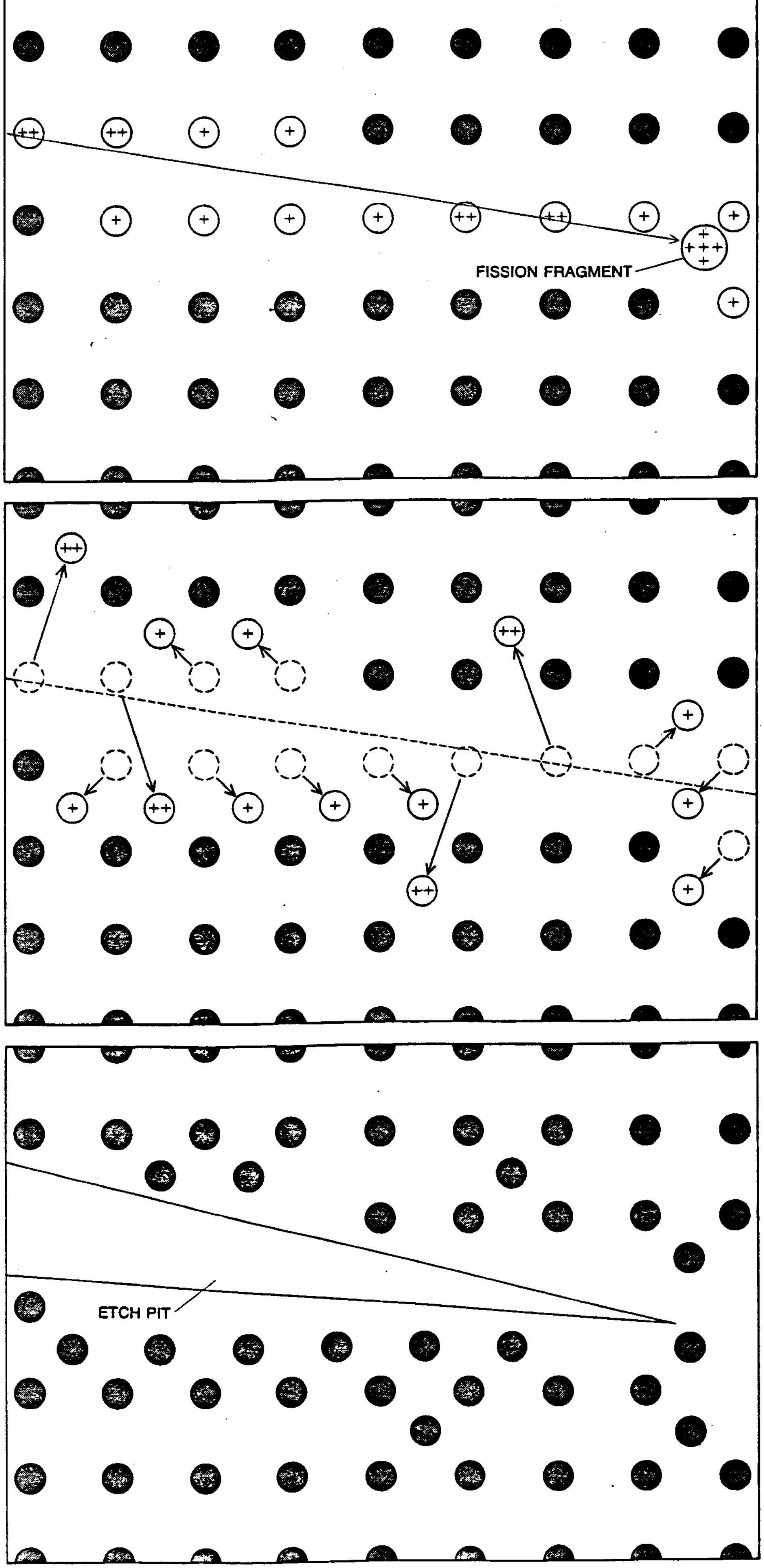
In the span of geologic time uranium 238 is transformed in two different ways. By far the commoner of the two is decay by the emission of an alpha particle. This emission transforms the uranium atom into an atom of thorium 234, a radioactive isotope that itself decays by the emission of an alpha particle. Alpha particles themselves are not, however, sufficiently massive or energetic to make tracks in common minerals.

About one out of every two million transformations in uranium 238 is by fission rather than by alpha decay. The process divides the uranium nucleus into two heavy and energetic fragments that differ only slightly in mass; at the same time several neutrons are emitted. The two heavy fragments fly apart in exactly opposite directions; the damage they do to the crystal lattice is thus in the form of a single straight track, approximately half of its length being created by each fragment. The overall length of the track varies, depending on the mineral involved; in general it is from 10 to 20 micrometers.

The usual procedure for studying tracks begins by embedding the specimen in a matrix such as an epoxy resin for convenience in handling. The exposed face of the crystal is then ground flat and carefully polished before the specimen is immersed in the etching bath. After etching, the crystal is observed under the microscope. The etching will of course reveal only the tracks that intersect the polished surface.

Since the rate of decay of uranium 238 by spontaneous fission is known, only two measurements are necessary to calculate the age of a specimen on the basis of the tracks visible after etching: a count of the number of tracks per square centimeter of surface and an assay of the U-238 content of the specimen. Since track densities are expressed



FORMATION OF TRACKS is shown schematically, following the model proposed by Robert L. Fleischer, P. Buford Price and Robert M. Walker. In the top diagram one of the two positively ionized fragments produced by the fission of a U-238 atom passes through an idealized crystal lattice; as it does so it strips electrons from the atoms along its path, thereby ionizing the atoms. Next (*middle*) the ionized atoms of the lattice are displaced, disrupting the structure of the lattice. When the crystal is exposed to a solvent (*bottom*), the disrupted portion of the lattice is more susceptible to etching than the undisrupted portion. Only insulating minerals record fission tracks; the free electrons in noninsulating solids restore the electrical neutrality of the momentarily ionized atoms before disruption of the lattice of the crystal can take place.

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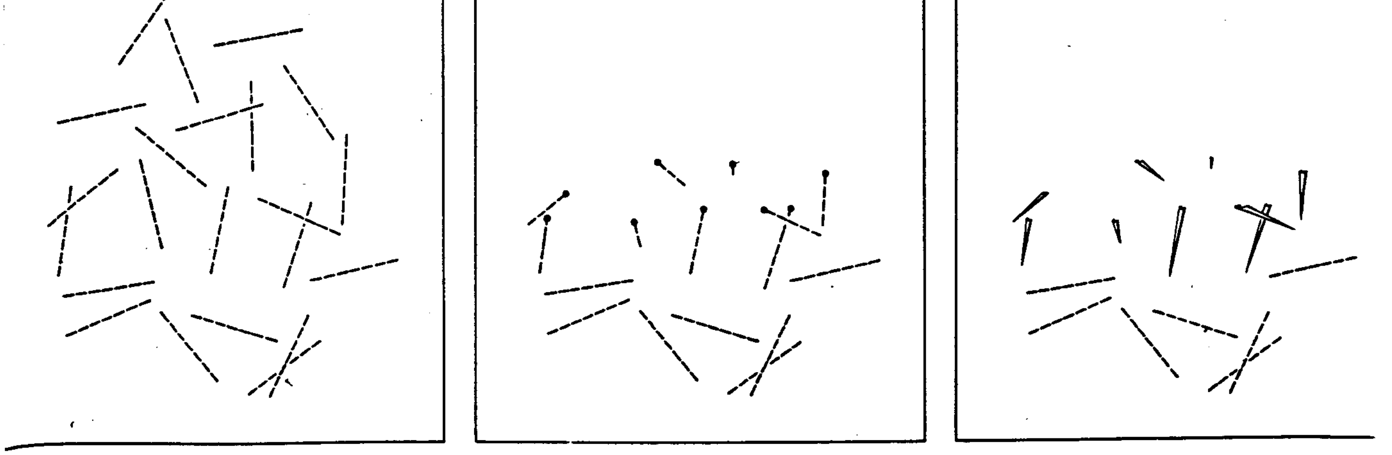

COUNT OF FISSION TRACKS requires preparation of specimen (*left*), containing tracks invisible under light-microscope magnifications, by grinding and polishing (*center*), thus exposing any tracks
that intersect the plane of polishing. The specimen is next etch
The process causes pits to develop along each exposed track; th
are large enough (*right*) to be easily seen under a light microsco

in units of surface and uranium content in units of volume, however, a geometric ambiguity exists that has the potential for introducing considerable error. There is an ingenious method for determining the uranium content of a specimen that circumvents the problem. It capitalizes on the fact that a sample with uranium in it will contain not only uranium 238 but also a small but constant fraction of uranium 235.

The atoms of U-235 can be induced to fission by exposing the specimen to slow neutrons in a nuclear reactor. The exposure adds new fission tracks to the spontaneous ones. The density of the new tracks is proportional not only to the total amount of U-235 in the specimen but also to two factors under the control of the investigator: the probability of the induced fission reaction and the number of neutrons per unit area that have passed through the sample in the reactor. The proportion of the two uranium isotopes being constant in nature, the investigator can calculate the concentration of uranium in the specimen simply by comparing the number of spontaneous tracks with the number of induced ones. The two track counts can be made with a single sample or, if it is preferred, the sample can be divided. Moreover, exposing the sample to heat will erase fission tracks by annealing, so that the sample can be annealed after the count of spontaneous tracks has been made; the U-235 tracks later induced by neutron bombardment will then be the only ones present.

The older a specimen is, the less important it is that its uranium content be large; there will be enough fission tracks for a statistically reliable count. Conversely, when a fission-track date is sought from a specimen that is substantially less than a million years old, the date can be determined only if the uranium concentration is relatively high. As an example, the mineral zircon, which is a common constituent of many rocks, can contain some hundreds of parts per million of uranium. Zircon crystals with a uranium concentration of 100 parts per million can provide an absolute age for a rock sample only 3,000 years old. As a rule of thumb, if the uranium content of the sample of crystal or glass is one part per million, the investigator can measure an absolute age as young as 300,000 years. An order-of-magnitude change in either direction with respect to uranium content brings an inversely proportional order-of-magnitude change in the youngest measurable age. Certain kinds of man-made glass contain as much as one part per 100 of uranium, and with them one can make an absolute-age determination up to as recently as 30 years ago. At the opposite extreme micas with a uranium concentration as low as a thousandth of a part per million can be used to measure absolute dates late in the Paleozoic era (300 million years ago).

All of this will make it seem that fission-track dating is remarkably precise, and for the most part it is. A few cautionary remarks should nonetheless be made. A fission-track age is calculated on the basis of four assumptions. The safest of the four, which is made in all methods of radiometric dating, is that the breakdown rate of the parent isotope (uranium 238 in the case of fission-track dating) has been constant with time. Both laboratory measurements and geological comparisons indicate that this assumption is sound. The other three assumptions are less certain.

One of them is that fission tracks are produced with 100 percent efficiency. Experiments indicate that this is so. When one encounters deviations from the expected number of tracks, they appear to be caused by problems affecting

the techniques of track detection. Mo
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both spontaneous and induced fissi
tracks are examined. Even if the dete
tion technique is only 10 percent e
cient, as long as the efficiency appl
equally to the detection of spontaneo
and induced fission tracks the calcula
age will remain correct.

The third assumption is that fissi
tracks are retained with 100 percent e
ciency. Since there is always the pos
bility that tracks have been erased
natural or artificial annealing, this is
assumption that cannot always be jus
fied. It can, however, be independen
verified where other radiometric me
ods of dating are applicable. Such in
pendent assessments show that most
sion-track discrepancies fall consiste
ly toward the young end of the sca
This is particularly evident when ve
old samples are examined. Except
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fall within the range of experimental
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shown to be wrong, they are almost n
er greater than the ages arrived at
alternative techniques of absolute d
ing. The reason is not hard to find:
most minerals used for fission-track d
ing the temperatures that suffice to er
tracks by annealing are well below
temperatures that begin to bias ot
methods of radiometric dating.

Moreover, in some instances fissi
track ages that have been biased by p
tial annealing can be recognized and
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mineral has been subjected to heat
some point in its history but the heat
not entirely anneal the tracks in it, m
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bration experiments it is possible to correct the overall track-density figure for the effect of thermal shortening and arrive at a rebalanced figure for spontaneous-fission track density.

The fourth assumption presupposes that the concentration of uranium in any specimen has remained constant over the specimen's lifetime. This assumption is usually valid, but there can be exceptions. A combination of elevated temperatures and ground-water percolation can leach away a proportion of the uranium present in rock crystals. The mobility of the uranium is such that as one part of a rock formation is being impoverished another part can become abnormally enriched. Such changes can also take place at relatively low temperatures. Andrew J. W. Gleadow and John F. Lovering of the University of Melbourne have compared heavily weathered grains of apatite, a common mineral in rocks, with unweathered grains still embedded in the parent rock. The weathered grains contained approximately 25 percent less uranium than those in the parent rock and yielded anomalous age determinations.

The best way to indicate both the present usefulness and the future potential of fission-track dating is to cite some examples of its application. I shall mention only briefly that geologists have found fission-track dating particularly attractive when a chronological frame-

reason is that the system is simple, and when a large number of determinations are required, the cost per sample is low. All three major rock classes—sedimentary, metamorphic and igneous—are amenable to fission-track analysis. In igneous rocks the uranium-rich minerals that are commonly used include two I have mentioned, zircon and apatite. A third such mineral is sphene, a calcium-titanium silicate. Several varieties of mica have also been used successfully to date both igneous and metamorphic rocks.

Applications of fission-track dating in the field of prehistory have generally been confined to situations that lie outside the useful range of carbon-14 dating. Because the radioactive half-life of carbon 14 is only 5,700 years, the carbon-14 method of isotope dating becomes increasingly unreliable when the sample is older than about 30,000 years. A case in point is the discovery in 1959 of fossil hominid remains at Olduvai Gorge in Tanzania by Louis and Mary Leakey. The hominid, which the Leakeys named Zinjanthropus, was at that time one of the oldest known; fossils associated with the find suggested that it might be a million years old, an age far beyond the range of any carbon-14 determination.

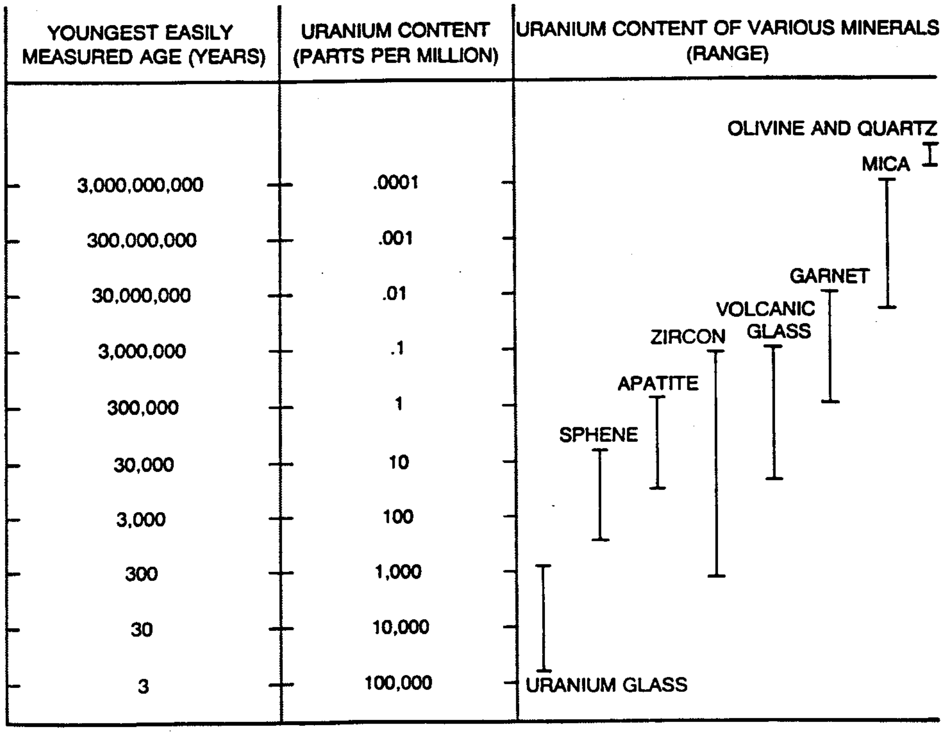
It happens, however, that the geological formation of Olduvai Gorge is a series of sedimentary rocks that includes numerous beds of volcanic tuff and ash.

so samples taken from volcanic strata generally related to the level in the gorge where the hominid remains were found were analyzed by Jack F. Evernden and Garniss H. Curtis of the University of California at Berkeley. They obtained potassium-argon determinations suggesting that the actual age of Zinjanthropus was almost twice as great as the associated fossils had indicated: about 1.75 million years.

Evernden and Curtis' finding might still be considered controversial (particularly because the mineral samples yielded a wide range of potassium-argon dates) if it were not for independent confirmation of their interpretation based on a fission-track analysis. Fleischer, Price and Walker undertook such an analysis, working with a specimen of pumice, the porous volcanic glass, from a related Olduvai stratum. The pumice was not easy to work with: the etched glass surfaces were small and irregular, and the etching solution actually dissolved away portions of the sample. Nevertheless, the investigators determined a fission-track date for the pumice: 2.0 ± .3 million years, a figure in close agreement with the potassium-argon determination of 1.75 million years.

At the other end of this anthropological spectrum one may consider certain man-made glasses that have uranium added to them in concentrations as high as one part per 100 in order to color them. Glass of this kind, which has been a standard item in Bohemia since the middle of the 19th century, can be dated by the fission-track method. Some students of Oriental art maintain that similar uranium glasses were produced in China centuries earlier. Günther A. Wagner of the University of Heidelberg has noted, however, that when a Chinese ring made of uranium glass, supposedly produced in 18th-century Ch'ing Dynasty times, was subjected to fission-track analysis, it turned out to be a 20th-century forgery, less than 70 years old.

Zircon crystals are natural components in some of the clays used to make pottery; any that are present in a piece of pottery are "reset to zero" when the pot is fired. The high temperature of the kiln erases all existing fission tracks. As a result the tracks that have been made in the time since annealing provide a precise indication of when the pottery was fired. Workers in Japan have been able to assign dates of manufacture ranging from about 300 B.C. down to 700 years ago to various pottery objects that contain zircon crystals. The dating of recently made pottery is tedious work, however; a very large number of tiny crystals must be scanned to get an accurate estimate of track density.



URANIUM CONTENTS of seven crystalline and two amorphous solids are compared. Man-made glass, colored by the addition of uranium oxide, is the material richest in uranium: from .1 to as much as 8 percent. Its date of manufacture can be calculated to within three years. Two crystals, olivine and quartz, may contain as little uranium as .1 part per billion. The age of such specimens, which may have crystallized three billion years ago, is hard to determine.