

ages indicated were determined by ^{14}C dating or historically. The error bars indicate the uncertainty in age, based on other methods of dating such as fission track dating and on circumstantial evidence. Bones believed to be older on the basis of their stratigraphic location gave equal or larger archeological doses in many cases.

The annual dose was about 0.1 to 0.2 rad per year for cave formations such as stalactites and stalagmites (4); this dose rate also seems appropriate for bones excavated from calcite caves. A dose rate of about 1 rad per year fits the data for bones embedded in sandy soil or open sites. However, it is known that uranium and other elements accumulate in these bones, and in some cases uranium concentrations of 10 to 100 parts per million have been measured in old bones. The annual dose rate should be higher than 1 rad per year on the basis of the energies of α , β , and γ radiation from the elements in the ^{238}U and ^{232}Th disintegration series, assuming that the parents and daughters are in equilibrium. It should be noted that fluorination of apatite in bone proceeds continuously, and therefore the production rate of radicals, presumably of the O^- type in hydroxyapatite, diminishes considerably with time. Thus our finding that bones from exposed sites can be dated from the archeological dose by using an apparent annual dose of 1 rad per year might indicate an equilibrium between an enhancement due to radiation-induced defect or trap formation and a reduction due to impurity accumulation or fluorination of hydroxyapatite.

We conclude that ESR dating of bones and similar biological materials should be useful in archeology and anthropology, especially when thermoluminescence dating is difficult. Ages ranging from hundreds to millions of years can be deduced without difficulty. Carbon-14 dating is limited to about 50,000 years. However, application to geological materials (6) might be difficult, as the ESR signal, which is increased by artificial radiation, is saturated at high-dose regions (about 10^6 to 10^7 rads). Thermal stability data for radiation-induced defects and traps at room temperature indicate that they should be stable for millions of years, at least in bones. Only a few geological materials with low concentrations of radioactive elements and with high stability of defects and traps may be dated by ESR, as demonstrated for apatite crystals (6).

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Isotopic Disequilibrium of Uranium: Alpha-Recoil Damage and Preferential Solution Effects

Abstract. *Preferential loss of uranium-234 relative to uranium-238 from rocks into solutions has long been attributed to recoiling alpha-emitting nuclei. Direct evidence has been obtained for two mechanisms, first, recoil ejection from grains, and now release by natural etching of alpha-recoil tracks. The observations have implications for radon emanation and for the storage of alpha-emitting radioactive waste.*

Radioactive disequilibrium is seen in nature under many conditions among the members of the uranium and thorium decay series, in each of which alpha decay plays a prominent role. Earlier experiments on a specialized material demonstrated the presence of at least two distinct mechanisms for such separations of isotopes from their parents. Both are the result of recoiling, alpha-decaying nuclei (1). One of these, direct ejection of the recoiling nucleus from a grain (1, 2), is physical, depends largely on the stopping power of the surroundings, and hence is insensitive to the detailed geochemical environment. It was expected that the second, radiation damage followed by chemical attack, would depend on the chemistry of pore fluids. This mechanism has now been tested and observed in a series of sensitive experiments in which implanted recoil nuclei were located by the activation of fission tracks after different minerals were exposed to various chemical solutions.

Because the natural isotopic abundances of individual elements are normally constant, the striking variability that has been observed in $^{234}\text{U}/^{238}\text{U}$ ratios on earth (3) is of special interest. Uranium is of interest economically with respect to nuclear energy and also geochemically because its radioactivity can be used to measure geological time by a variety of techniques in which ratios such as $^{206}\text{Pb}/^{238}\text{U}$, $^{230}\text{Th}/^{238}\text{U}$, and $^{234}\text{U}/^{238}\text{U}$ are important.

Similarly, disequilibrium between chemically different nuclides in a radioactive chain is useful both in understanding the chemical or physical environment in which the decays are occurring and in learning how to locate ore deposits or

even how to predict earthquakes. As an example of the possible applications to ore discovery, the sequence of escape and migration of ^{222}Rn (a gaseous member of the ^{238}U decay chain) has been suggested as a means of locating subsurface uranium deposits (4), and the solution and transport of ^{226}Ra have been thought to form diffuse halos around (5) or displaced from (6) uranium deposits. In the above-described cases alpha-recoiling nuclei have frequently been invoked as the cause of disequilibrium, but direct experimental evidence on the detailed mechanism or mechanisms has been largely lacking. Recoiling nuclei from alpha emission could also affect the ease of loss of radioisotopes from nuclear waste that contains transuranic nuclides, since they are commonly alpha-emitters.

When a nucleus that is within about 200 Å of a solid surface undergoes alpha decay, the residual nucleus may recoil directly from a grain. Such behavior was first shown unambiguously by Kigoshi (2), who dispersed ZrSiO_4 crystals in various solutions and observed the build-up in the liquid of the ^{234}Th which results from the decay of ^{238}U (in turn, it decays rather quickly to ^{234}U). For wet rocks or soils this mechanism would enrich the water in ^{234}U relative to its parent ^{238}U , which tends to remain in the solid. The same process of direct recoil ejection has also been observed by a process in which a detector in vacuum is used to catch ejected ^{235}U nuclei from the decay of ^{239}Pu in PuO_2 spherules. The study established that the number of such recoils is what would be expected from the number of decays that occur in the near-surface regions of the spherules (1).

A second mechanism that was suggested as possibly important in nature (1) arises from the fact that many of the recoiling nuclei that are ejected from mineral grains become embedded in adjacent grains, producing alpha-recoil tracks (7) which extend from the surface to the position where the nucleus comes to rest. Subsequent track etching by natural solutions may release the recoil nuclei. Evidence for this process (1) was given for an unusual, synthetic material, but until now this process has not been demonstrated for natural minerals.

A sensitive test of this mechanism can

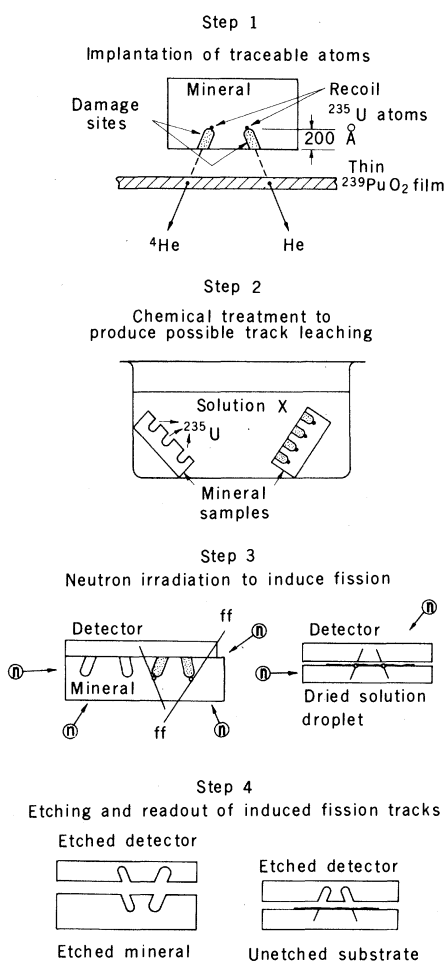


Fig. 1. Technique for implanting ^{235}U in minerals and for locating the nuclei after treatment of the mineral with a solution. The ^{235}U nuclei that recoil from the alpha decay of ^{239}Pu are implanted (step 1), and uranium nuclei are found at the end of recoil tracks (just as ^{234}U is located in nature). The implanted samples are separately exposed to different solutions (step 2), which may release ^{235}U . The mineral is then (step 3) placed next to a track detector, and the residue from a dried droplet of the solution is similarly mounted and both assemblies are exposed to thermal neutrons, which induce fission of ^{235}U nuclei; encircled n , neutrons; ff , fission fragments. In step 4, the detectors are etched and the induced fission tracks counted to measure how much ^{235}U was retained in the mineral and how much went into the solution.

be carried out if one follows the sequence of steps sketched in Fig. 1. A $^{239}\text{PuO}_2$ source has the special quality of giving ^{235}U as recoiling nuclei. These particles will leave tracks similar to the recoils from ^{238}U decay, and the implanted nuclei are chemically the same as the ^{234}U that results in nature. Because ^{235}U nuclei fission with an unusually high cross section, they may be sensitively detected by a process consisting of neutron irradiation followed by etching of the induced particle tracks (8). The ^{235}U nuclei are implanted (9) in a mineral, it is exposed to a solution, and both it and the solution are tested for their ^{235}U contents.

Figure 2 gives an example of the results. Here natural quartz samples that had been implanted with ^{235}U over a 28-day period were treated with water and various concentrations of acidic or basic solutions for 24 hours, and the solutions and the quartz were irradiated with 10^{16} thermal neutrons per square centimeter. Two control samples were included: one received no solution treatment, and the other was given a 5-second rinse in distilled water. The results of the two treatments agree, showing that the ^{235}U is embedded rather than superficially located on the quartz. All the solution-treated quartz had lost ^{235}U , the average loss being about 40 percent. The solutions were found to contain most of the lost ^{235}U , providing a consistency check. About 15 percent is missing and may have become attached to the walls of the container and hence lost from solution. One solution shows contamination, giving a reading that was higher than the amount of ^{235}U that was originally present. Control solutions contained negligible uranium relative to the signal observed. The quartz contains less than 10^{-7} part per million of uranium (by weight) (10). The results of similar experiments on other silicate structures are also clear. Fractions of recoil nuclei removed with 24-hour treatments range from 50 to 75 percent for orthoclase (a feldspar mineral), 0 to 40 percent for muscovite mica (a layer mineral), and 25 to 80 percent for obsidian (an amorphous mineral) (11).

The variation with time of exposure to the solutions needs to be studied carefully. In preliminary experiments, the average loss of ^{235}U from muscovite increased from 37 percent after a 1-day exposure to the solutions to 50 percent after a week's exposure. Even if this loss is logarithmic with time, the process of removal from surface layers would be essentially complete after only 200 years, geologically a short time.

A striking aspect of the results for quartz is that the strength of the solutions is apparently unimportant. Similar results were found for orthoclase. Obsidian and mica, however, did give larger effects for the strongest solutions but clear effects even for the weak ones.

The overall conclusion arising from a study of four major minerals—a mica, a feldspar, quartz, and a natural glass—is, therefore, that virtually any solution will remove a significant fraction of alpha-recoil nuclei that have crossed the exposed surface of the mineral. If this behavior is general to uranium-bearing minerals, it is adequate to cause the observed $^{234}\text{U}/^{238}\text{U}$ anomalies (3) which appear over geological times. Even if minerals that are uranium-rich fail to show the effect, the fact that in rocks these accessory minerals are adjacent to the common low-uranium minerals such as have now been tested ensures that recoil nuclei will be injected into the abundant minerals where they can be removed by solutions. Removal of implanted recoil nuclei by exposure to solutions is consistent with the experimental finding that in permanently dry environments, such as on the moon, there is equilibrium between ^{238}U and ^{234}U (12).

The observed behavior has obvious implications for the storage of radioactive waste that contains alpha emitters. Whether these nuclides are present in a glass or a crystalline material, exposure of the surface to water is likely to lead to removal of radionuclides. A surface coating that is impervious to water

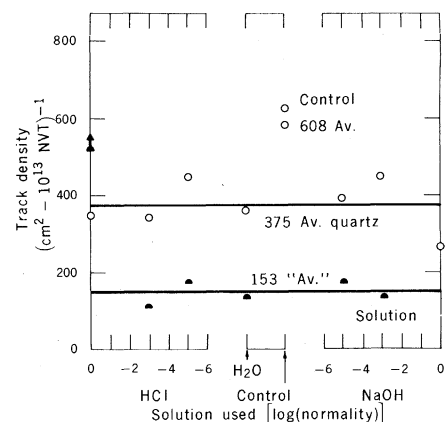


Fig. 2. Tracks per area of ^{235}U -implanted surface for quartz (open circles) and for solutions exposed to quartz (closed half circles); NVT represents the neutron dose in neutrons per square centimeter. The implantation period was 28 days, and the neutron exposure was 10^{16} neutrons per square centimeter. Separate average readings are given for control samples and those that were exposed to the solutions. For the solutions the average excludes the sample with obvious contamination, indicated as a lower limit.

could be useful in preventing such losses.

The radionuclide ^{222}Rn , the daughter of ^{226}Ra by alpha emission, should also be subject to release by the exposure of minerals to water. Thus injection of water into formerly dry rock or soil could release a pulse of stored radon. Earth strains prior to earthquakes together with water redistribution which such strains may produce could therefore be responsible for altered radon concentrations associated with earthquakes (13). Similarly, the recognition of mechanisms for radon release has implications with respect to the management of uranium tailings.

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Viscous Flow Circulation of the Solar Wind Behind Venus

Abstract. A latitudinal circulation model of solar wind flow in the near wake of Venus is presented. It is shown that solar wind fluxes entering through the polar terminator can be viscously forced to lower latitudes. The resulting motion produces a downstream elongation of the nightside polar ionosphere out to the downstream extension of the middle- and low-latitude ionopause. The geometry suggested by this flow circulation model provides a simple explanation of the ionospheric bulge inferred from the Pioneer Venus observations.

The preliminary results of the plasma probe, magnetometer, electron temperature probe, and ion mass spectrometer experiments carried out with the Pioneer Venus orbiter provided further experimental evidence in support of the deflection into the umbra of the shocked solar wind in the vicinity of the planetary terminator (1, 2). The observed orientation of the magnetic field in the near wake is suggestive of converging flows, which may be confined to the outer regions of the umbra, outside a rarefaction wake extending several Venus radii (3). The geometry of the nightside ionopause inferred from the results of the mass spectrometer experiment indicates, in addition, the presence at low latitudes of a prominent ionospheric bulge extending up to ~ 3000 km above the surface. Since the entry of solar wind fluxes into the umbra is a dynamic condition that can be described in terms of a viscous interaction between the shocked solar wind and the ionospheric material (4),

the question arises whether the presence of such a bulge is compatible with this interpretation. In this report it is shown that this feature can be explained as a result of differences in efficiency of the viscous interaction process, which may be

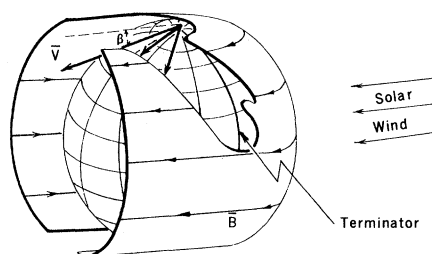


Fig. 1. Schematic diagram of the flow pattern behind Venus. Arrows indicate the motion of ionosheath fluxes entering through the polar regions and subsequently acting against the downstream extension of the middle- and low-latitude ionopause (which is represented by a cylindrical shape). The interplanetary magnetic field lines are shown draped around the dayside ionopause.

severely diminished at middle and low latitudes by the accumulation of interplanetary magnetic fluxes.

Viscous processes are believed to result from a strong dynamic interaction between the shocked solar wind and the ionospheric material. Pseudocollisions resulting from efficient wave-particle interactions should ultimately be responsible for the collective behavior of the plasma and the effective transfer of momentum across the ionopause. An important consequence of this process is a necessary influx of ionosheath particles into the umbra to satisfy conservation of mass flux. This should occur when the kinetic energy density of the local plasma is larger than the energy density of any magnetic field that may permeate the region (5). When this condition is not met, the motion of the plasma is controlled by the local magnetic geometry, and thus the plasma is inhibited from interacting dynamically with the stationary ionospheric material. Accumulation of magnetic fluxes of the ionosheath flow around the low- and middle-latitude regions of the planetary ionosphere should impose locally a flow configuration characterized by the latter condition, so that a stable and undeflected inner boundary of the ionosheath flow should exist immediately behind a large section of the terminator (magnetopause-like boundary). At polar latitudes, on the other hand, local enhancement of magnetic fluxes is expected to be significantly smaller, and there should be more efficient dynamic contact between the ionosheath flow and the ionospheric material. These general considerations indicate that the accumulation of interplanetary magnetic field lines around the ionospheric obstacle should result in a more efficient and better developed viscous boundary layer at polar latitudes than at equatorial latitudes.

The preferred access of the ionosheath flow to the umbra through the high-latitude regions of the ionopause should result in the generation of a characteristic circulation flow pattern above the nightside hemisphere. To visualize this, it must be recognized that the region of reduced plasma pressure downstream from the polar terminator is not the only space available for the viscously deviated plasma. The kinetically induced expansion should also proceed laterally into the adjacent regions located underneath the downstream extension of the undeviated ionosheath at middle and low latitudes. This effect is schematically illustrated in Fig. 1, which shows the initial stages of flow penetration from an assumed source in the polar regions. As