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Solid State Track Detectors: Applications to Fission Related Studies*

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Abstract

The passage of charged particles produces linear trails of damaged material in dielectric solids. These trails are frequently visible by transmission electron microscopy or can be chemically etched and observed with an optical microscope. This paper compares track registration and track retention behaviour of crystalline and non-crystalline track detectors such as stilbite, prehnite, cellulose nitrate CR-39 and LR115. The dependence on both the annealing temperature and time of the preferential track etch ratio V_t / V_g and also the track length and track density are discussed. The etching response is studied for fission fragments, alpha particles, protons and neutrons. Several useful applications of solid state track detectors in various fields are discussed.

1. Introduction

The past decade has seen some remarkable advances in fission track studies, particularly in applications to geological and cosmological problems (Fleischer *et al.* 1964; Lal *et al.* 1969; Naeser *et al.* 1973; Koul 1979; Koul *et al.* 1981). There have also been widespread applications of solid state track detectors (SSTDs) in the registration of tracks from particles other than fission fragments; track detectors allow passive observations of diverse nuclear phenomena. For these and other reasons track detectors have themselves become the object of intense scientific and technological interest. Detector materials exhibit many unique properties, some of which, in the context of their use, make them attractive in a variety of commercial applications. There is hardly a branch of science where SSTDs do not have potential application.

This article describes the essence of what is known about SSTDs and their use. In particular, we describe our experiences with representative track detectors used during the last five years in a program on fission related studies. Common features in the various effects observed are delineated and explained in terms of a few basic concepts. Detailed studies of etching behaviour and of the thermal fading (annealing) of nuclear tracks are examined, with particular attention being given to the track registration of two minerals used for age determination in geology, stilbite (zeolite) and prehnite, a phyllosilicate mineral. The problem of the thermal stability of nuclear

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tracks recorded in organic polymers has recently assumed a new importance due to the possibility of changes in track parameters which may effect the accuracy of the charge determination of energetic heavy nuclei. The track registration, thermal stability and etching parameters of a number of organic polymers are therefore described. The paper concludes with an account in Section 5 of the use of SSTDs in many applications on the verge of being commercialized and considers future possibilities.

2. Crystalline Track Detectors

Increasing attention is being paid to the study of organic polymers as track detectors. The success of such studies, as well as the properties of these materials, has been described in many papers (see e.g. Paretzke *et al.* 1973; Koul and Chadderton 1979; Riedel and Spohr 1980; Roggenkamp *et al.* 1981). On the other hand, crystalline track detectors have received far less attention in the field of track etching. This is due partly to their anisotropic track-recording properties and partly because these materials often contain microscopic imperfections and inhomogeneities, which result from the presence of aggregates of dislocations and segregated impurities. Moreover, the bulk etch velocity V_g of crystalline detector materials such as feldspar (Khan 1977) and chabazite (Koul *et al.* 1981) has shown them to be anisotropic.

Most crystalline track detectors are however more temperature resistant than plastic or glass track detectors. A detailed understanding of the thermal stability of nuclear tracks is an important problem in a number of fields. For example, some crystalline materials have been found in a number of lunar and meteoritic samples, and these materials contain a wealth of information on cosmic rays, solar particles etc. Similarly, tracks in crystals can improve our understanding of the burial history of sedimentary basins bearing fossil fuels.

Table 1. Chemical analyses (wt. %) of samples determined by inductively coupled plasma emission spectroscopy

Compound	Prehnite	Stilbite	Trace element	Prehnite	Stilbite
SiO_2	44·8	59·3	Ba	<0·002	0·005
TiO_2	<0·01	<0·01	Sr	<0·002	0·116
Al_2O_3	23·2	15·2	La	<0·005	<0·005
Fe_2O_3[A]	1·29	0·09	Y	<0·002	<0·002
MnO	<0·01	<0·01	Ce	<0·01	<0·01
MgO	<0·01	0·20	Tl	<0·01	<0·01
CaO	25·5	7·20			
NaO	<0·01	0·40			
K_2O	0·88	0·56			
P_2O_5	0·01	0·06			
LOI[B]	4·40	17·7			

[A] All Fe is present as Fe_2O_3. [B] Loss on ignition.

Etching/Annealing Behaviour

Chemical etching in association with optical and scanning electron microscopy provides a means for identifying and unifying the detection of nuclear tracks and other defects in solids. In a somewhat restricted sense, it is the composition of the etchant that determines the extent of the preferential etching of fission damage of a particular crystal face. Although there have been many studies of nuclear tracks by the process of chemical etching, the generally complex etching mechanism which is dependent on the material and the etchant is not properly understood.

Nuclear track recording solids can be used to identify energetic nuclear particles because the rate of growth of the conical etch pit that develops at the intersection of the particle trajectory with the surface is an increasing function of the rate of energy loss of the particle in the solid. The track etch rate is, in principle, almost immune to the energy loss statistics that plague all other charged particle detectors because it is controlled by the energy density deposited within a few tens of Ångströms (10^{-10} m) from the centre of the particle (Alhen *et al.* 1981).

In the following we attempt to illustrate the importance of both chemical etching behaviour and annealing behaviour of crystalline track detectors in providing a microscopic and quantitative basis for understanding track registration and track retention properties of these detectors. The material used in this study was natural stilbite from Tasmania and prehnite from East Greenland. The chemical analysis was carried out using inductive coupled plasma emission spectroscopy (see Table 1). Both optical and scanning electron microscopy were combined with X-ray diffraction observations to investigate track registration in different crystal planes of stilbite and prehnite. X-ray diffraction topography was the principal method used for investigating the sample crystal perfection. Stilbite and prehnite sections were cut along the 001 and 100 planes. The blocks were then cut to obtain a number of rectangular shaped slices which were rinsed in deionized doubly distilled water and then mechanically ground and polished with fine diamond abrasives. Finally, they were ultrasonically cleaned in distilled water and dried with an alcohol rinse. After this treatment, no traces of residual surface damage or etch pits were observed by reflected light optical microscopy.

The polished sections were divided into two groups, one of which was irradiated with a ^{252}Cf source, while the other was exposed to thermal neutrons using both the Risø and HIFAR reactor facilities. Following irradiation, the stilbite and prehnite specimens were immersed in a variety of freshly prepared acidic and alkaline etchants. During the etching, the samples were manually agitated to prevent gas bubbles from sticking to the surface. Etching was stopped by dilution of the etching solution with a large amount of deionized water. The etched samples were carefully rinsed with distilled water and then dried with filtered nitrogen, a method which eliminates the pattern on the sample surface which sometimes causes difficulty in distinguishing etched tracks (for more experimental details see Koul *et al.* 1983; Koul and Cannington 1984).

The fission track etching efficiency of a crystal is defined as the fraction of the total number of fission fragments to have crossed a given surface. When the rate of the damaged region V_t is very much greater than the general rate of etching V_g, the etchant readily penetrates along the full length of the track. The tracks are then revealed as cylindrical lines and the efficiency is high. The optimum etchant for stilbite and prehnite was established by observing the degree and nature of the track etching which resulted from the use of various chemical reagents.

The effectiveness of various etchants of differing concentration was studied as a function of time and temperature. The track density on different crystal planes of stilbite and prehnite was also investigated. Optimum etchant and conditions for

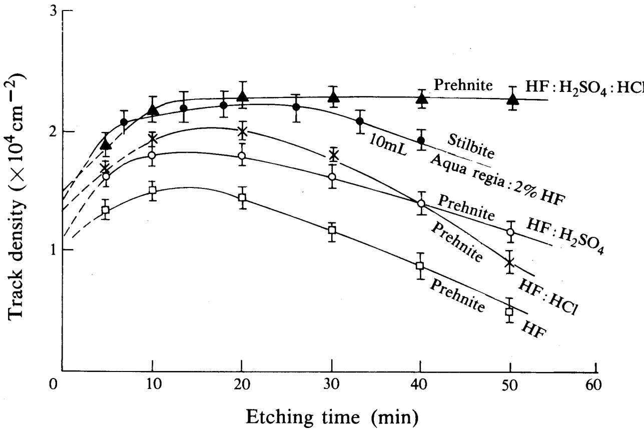

Fig. 1. Track density as a function of etching time in prehnite and stilbite track detectors, utilizing the various etchants indicated.

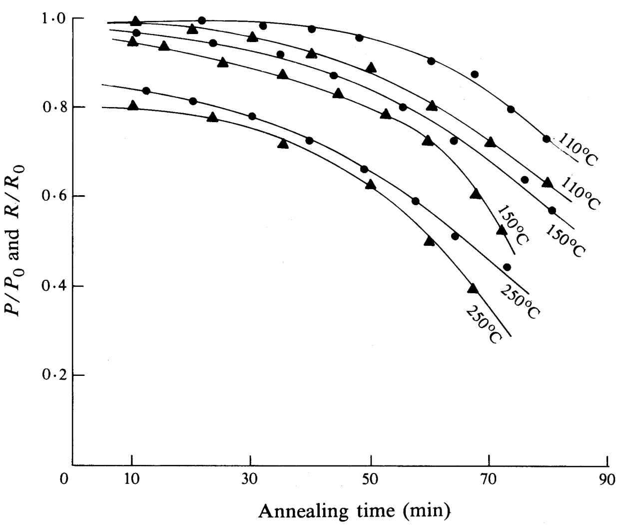

Fig. 2. Normalized track densities P/P_0 (circles) and track lengths R/R_0 (triangles) as a function of annealing time at three temperatures in stilbite.

stilbite proved to be 10 mL aqua regia : 2% HF at 23°C for 20–30 s, and for prehnite HF : H_2SO_4 : HC1 at 23°C for 15–20 min. In the case of prehnite, the etching efficiency is higher when etched in HF : H_2SO_4 : HC1 than when etched in HF or HF : HC1 or HF : H_2SO_4. The etching efficiency of stilbite in 10 mL aqua regia : 2% HF is high, but much lower than that for prehnite. The data presented in Fig. 1 and the examples chosen for illustrating the characteristic behaviour of fission tracks have been tested in ARISTAR ultra-pure reagents, and the results are probably insensitive to the presence of small amounts of impurities.

To investigate track etch properties further, we carried out experiments to find the effect of varying etching temperature with etching time on the track density and track length in stilbite. The stilbite specimens were heated to temperatures of 110, 150 and 250°C and the fractional track density and track length measured. The normalized results presented in Fig. 2 suggest that in lower concentrations the registration threshold is reduced and changes in the track length take place. In fact, however, the experiment shows the higher sensitivity of these etchants for preferential etching of stilbite and prehnite. There is an added advantage with these etchants as they reveal, at least qualitatively, the inclination of the latent tracks to the surface.

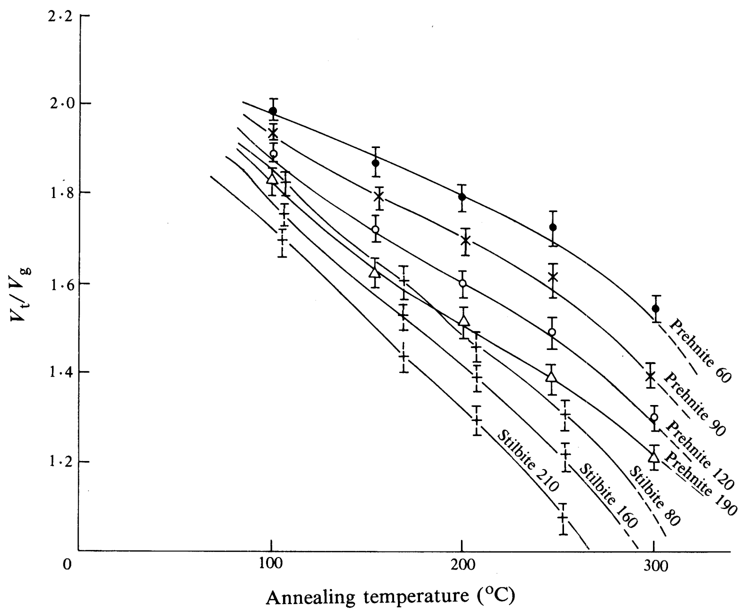

Fig. 3. Variation of the preferential track etch ratio with annealing temperatures for the annealing times indicated (minutes) in prehnite and stilbite.

The influence of the track etch ratio V_t/V_g in different etching conditions for different detectors was also investigated. The results represented by the curves in Fig. 3 show a gradual decrease due to the surface etching rate of varying etchant concentration. The ratio decreases with both increasing temperature and time. The track etch rate of stilbite and prehnite is orientation dependent, the basal plane etching the fastest followed by the 110 and 100 planes. Even longer times are required for etching of fission fragment tracks on surfaces with other orientations. As a consequence of this variable etching rate on different crystallographic planes, it is difficult to etch all the fission tracks in all the grains of stilbite in a single operation. It is generally necessary to perform sequential etchings to obtain suitable etching of subgrains in stilbite aggregated with little or no preferred orientation. However, it may be shown that for certain materials the track density on an internal surface continues to increase with prolonged etching. With glass, for example, there is a further exposure of new tracks as the surface is progressively attacked by the etchant.

These are then added to previous tracks which remain visible with continued etching. In the case of chabazite (Koul *et al.* 1983) and stilbite, however, this effect does not occur.

A few slices of prehnite and stilbite were annealed at 490 and 460°C respectively for three hours and mechanically polished along the 100 and 110 planes in order to investigate both the possible effects of track orientation and heating on the registration efficiency. The polished slices were irradiated by a ^{252}Cf source in a 2π geometry for equal times in order to implant the same number of fission fragments. The angular track distribution was then obtained by measuring the inclination of the ends of tracks relative to a fixed, but arbitrary direction, the analysis being carried out using both optical and scanning electron microscopes. The distribution of etched tracks in the 110 plane for prehnite and in the 100 plane for stilbite for different etching times was found. The present experiment demonstrated, in addition, that the etching of different planes of prehnite and stilbite for varying times at different temperatures, however prolonged, does not improve the degree of isotropy of the etched tracks. The 100 surfaces of stilbite showed a variable distribution of fission tracks. In some relatively small regions the surface had been attacked to such an extent that the individual tracks were barely distinguishable, but in most cases they were isolated and distributed uniformly.

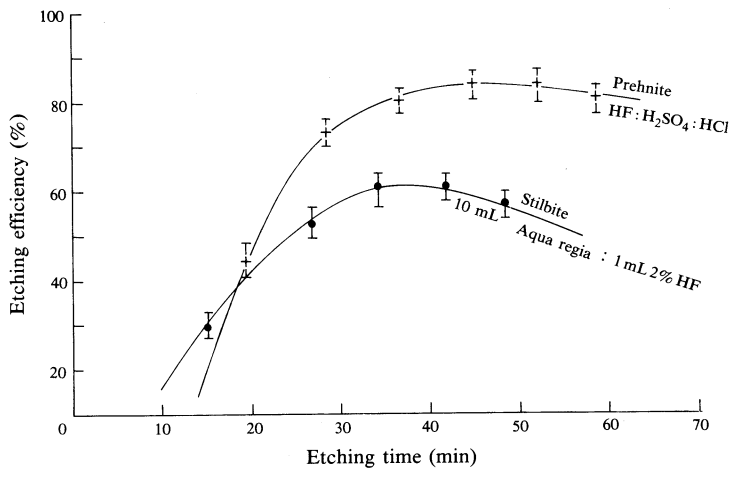

Fig. 4. Etching efficiency of prehnite and stilbite for treatment by the etchants indicated.

Fig. 4 shows a plot of etching efficiency against etching time in different etchants for stilbite and prehnite. It is interesting to note that the efficiency of stilbite shows a gradual increase followed by a slow decrease due to the progressive chemical removal of surface layers of the crystal; on the other hand, the plot for prehnite shows an abrupt initial rise to a plateau remaining constant with time. As shown, the etching efficiency of stilbite is lower than prehnite.

3. Non-crystalline Solids

Considerable research into the composition and nature of the basic properties of SSTDs has been carried out recently, the wide availability of fission track methods and X-ray diffraction methods having provided the necessary means of establishing the individuality of inorganic materials. This development in the study of organic and inorganic materials has turned out to be the most fruitful and exciting of all, and in the last few years has led to a considerable growth in radiation damage physics; here we can do no more than mention some of the more important applications by Benton (1968), Becker (1973), Fleischer *et al.* (1975) and Somogyi (1977). Although the field is wide and difficult to review, interesting advances in the fission track methods of polymers have been made recently by Cartwright *et al.* (1978) and Price *et al.* (1979). Special interest has also been shown in different types of organic polymers. Biswas *et al.* (1979) have employed organic track detectors for the detection of relativistic cosmic rays and have obtained a good record of events for the relativistic Fe group nuclei. These track detectors also provided useful results on the radiation dose received by Apollo astronauts, in particular, with the high Z component of cosmic rays (Comstock *et al.* 1972; Fleischer *et al.* 1975). These developments have provoked an increased interest in SSTDs in recent years, especially through the recognition of their growing potential for other applications in science and technology.

It is not our intention to give an exhaustive documentation of all the track detectors and the related experimental and theoretical studies. We focus here on a relatively few specific examples, pointing out the salient features of SSTDs and giving an evaluation and discussion of some of the experiments. Although some important papers are not referred to, we prefer to treat a smaller quantity of work in relatively more detail. The data presented and the examples chosen for illustrating characteristic behaviour have been taken primarily from organic polymers such as cellulose nitrate CR-39 and LR115.

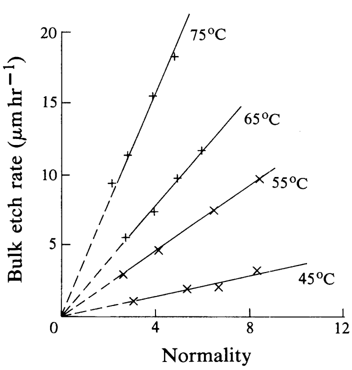

Fig. 5. Bulk etch rate of LR-115, irradiated with alpha particles, as a function of etchant normality for the different temperatures shown.

Track Etch Rate of Organic Polymers

It is well known that etch pit morphology is dependent upon a number of factors such as concentration of an etchant, temperature of an etchant and the etching conditions. Therefore, precise identification and measurement of track etch parameters of SSTDs is very important. For example, to obtain the particle energy, knowledge of the bulk etching rates of SSTDs is of considerable importance. During the process of chemical etching, a portion of track is dissolved out by the etchant, with an obvious correction required to the observed track length, the magnitude of which depends on the bulk etching rate. It is therefore essential to establish the reliability of a particular etchant for revealing tracks in a particular material.

To investigate this we exposed a number of CR-39 and LR115 track detectors to ^{252}Cf fission fragments, alpha particles, neutrons and protons at various energies. The experimental procedure used was the same as mentioned earlier. The bulk etch rate V_g was determined by the method described above (Enge *et al.* 1974; Koul *et al.* 1981). Fresh solutions were always used, so that the amount of dissolved polymer (CR-39 or LR115) which is etched off the sample is negligible compared with the volume of solution. In addition, the temperature and the etching conditions were adjusted so that the etching rate is low under such conditions. The results of our measurements of V_g as a function of the etching concentration were obtained at various temperatures using a NaOH etchant. From Fig. 5 it is evident that the behaviour of V_g has an approximate linear dependence on the normality of NaOH. Schlenk *et al.* (1975) have investigated the possible reasons for the higher V_g values obtained by some workers and found that the stirring from an open etch bath could have considerable influence on V_g. Even the environmental conditions have a profound effect on the plastic track detectors; for example, V_g increases after irradiation with gamma rays.

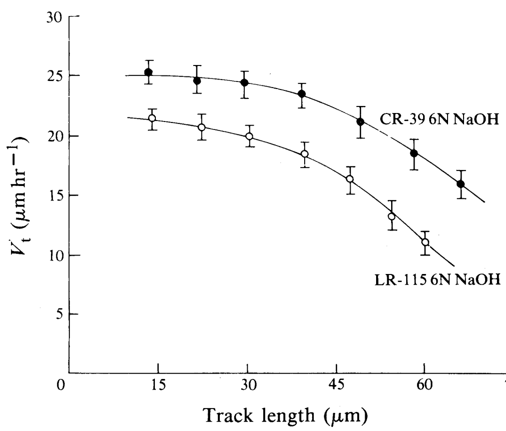

Fig. 6. Track etching rate for CR-39 and LR115 as a function of track length at a temperature of 65°C. The CR-39 samples were irradiated with collimated fission fragments at normal incidence from a ^{252}Cf source and LR115 was irradiated with α particles.

The track etch rate V_t was obtained by measuring the projected length of the tracks at various etching times. These rates decrease with etching temperature in plastic track detectors such as CR-39. Fig. 6 shows the variation of V_t with track length, when the sample was etched in 6N NaOH at 65°C, and reveals a decrease with penetration depth both in LR115 and CR-39 track detectors. Enge *et al.* (1974) found V_t/V_g to be constant between 50–80°C. The sensitivity increases in cellulose nitrate at lower temperatures. This effect was confirmed by Benton (1968) for heavy ion tracks.

We also investigated the effect of etching and temperature on the efficiency of CR-39 and LR115 after exposing these detectors to neutrons and protons. Fig. 7 shows the efficiencies of these detectors as a function of energy. The data show a definite trend towards increased efficiency with neutron energy in CR-39, which may be caused by the increase in the etching range of recoils and the production of charged particles from different threshold reactions.

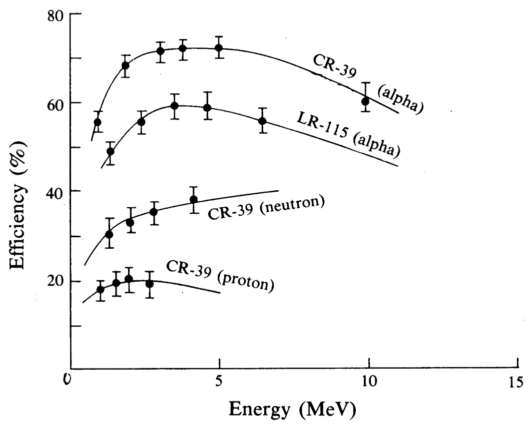

Fig. 7. Measured detection efficiency of CR-39 and LR115 track detectors as a function of energy for alpha particles, neutrons and protons.

To test the effect of temperature on the etching efficiency of CR-39, a series of slices were heated at various temperatures in the range 60–100°C. Small perturbations in temperature (+5°C) up to 70°C had no visible effects on the efficiency, whereas longer etching intervals caused a decrease in efficiency.

4. Conclusions from the Experiments

The experiments carried out on the track registration and retention characteristics of latent tracks in crystalline and non-crystalline track detectors suggest the following:

(i) The fission track method is useful for stilbite and prehnite, so these minerals should have a practical application in fission track geochronology.

(ii) The fission tracks in stilbite and prehnite are not very stable, implying a low track retention and low closing temperature. The thermal histories of low temperature zeolites and phyllosilicate minerals may provide models for the solution of important problems related to the burial histories of sedimentary basins bearing fossil fuels.

(iii) The shapes of etch pits depend not only on the crystal plane but also on the etchant.

(iv) Cellulose nitrate CR-39 and LR115 track detectors were tested for the recording of alpha particles, fission fragments, protons and neutrons. These experiments suggest that the etch induction time is an important parameter in track detection and in the resolution of ions of different mass and charge.

5. Application to Fission Related Studies

Many important aspects of SSTDs are related to technological challenges of the present and future. Among the present challenges are the design of new electron devices. The development of nuclear tracks could be used for generating metal/oxide metal point contact diodes. Point contact diodes are used as detectors, harmonic generators and mixers in the ultra high frequency range up to optical wavelengths (Fischer and Spohr 1983).

Formidable challenges for the future include geophysical applications such as the use of crystalline track detectors in evaluating the thermal histories of sedimentary rock accumulations. These experiments involve fundamental studies of the physics of fission tracks in crystalline materials (minerals) with special emphasis on the nature and distribution of tracks and their thermal behaviour. Such an experiment would certainly contribute to the understanding of the tectonic evolution of sedimentary basins bearing fossil fuels, the evolution of secondary mineral paragenses within them, and the understanding of thermal and temporal influences on coalification and hydrocarbon generation. These are important aspects in the exploration and evaluation of fuel resources, especially as the search for hydrocarbons turns towards the more subtle traps that relate to the post-depositional history of the sedimentary successions.

An important application of SSTDs occurs in the mapping of uranium/thorium in terrestrial, extraterrestrial and biological matter. The biological applications include radiation dosimetry of both manufactured and natural resources and the investigation of the distribution of fissionable material in biological tissue. Water, plants and human hair have been investigated for their distribution of fissionable materials (Koul *et al.* 1981, 1983).

Uranium is important in extraterrestrial solids because it is the parent nuclide of a number of radioactive decay schemes that enable cosmic ages to be determined, and because, being the heaviest naturally occurring element, it is significant for the different models of nucleosynthesis. Track experiments are in progress on meteorites, which have been exposed to cosmic radiation for long periods of time, to obtain information about the cosmic radiation and the history of the meteorite itself.

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