Fission tracks in solids—production mechanisms and natural origins

ROBERT L. FLEISCHER

Research Professor of Geology, Union College, Schenectady, New York 12308, USA E-mail: fleischr@union.edu

Radiation-damage tracks in solids from energetic nuclear particles are caused by ionization that leads to atomic disordering along particle trajectories. The process that transforms missing electrons into atomic disorder is termed an *ion explosion spike*: The ionized region is unstable because the Coulomb repulsion of adjacent ions overcomes the local bonding. Natural tracks can be of many origins, but for samples on earth, spontaneous fission of ²³⁸U (the key to fission-track dating of minerals), is by far the dominant source. There are occasional, localized exceptions, three of which are noted. Exotic sources are listed, including those that can operate for materials that are outside the protective shielding of the earth's atmosphere. © *2004 Kluwer Academic Publishers*

1. Introduction

Energetic, charged nuclear particles produce linear regions of radiation damage in an immense variety of solids. In each of these substances the tracks can be revealed by a simple chemical treatment after which they may be studied using a conventional optical microscope. D.A. Young [1] in 1958 demonstrated that charged particle tracks exist and that they can be displayed by chemical etching of lithium fluoride. R.L. Fleischer, P.B. Price, and R.M. Walker showed that a great variety of materials store tracks [2–6] and developed a great many of the original scientific and technological uses of tracks (for reviews, see references [7] and [8]). Fig. 1 shows tracks that have been revealed by chemical etching in muscovite mica [2]—these being natural fission tracks, whose origin will be described later.

2. Nature of fission

Fission is the division of a heavy nucleus into two heavy fragments that represent most of the original nuclear mass. Whether spontaneous or induced, fission of a nucleus can be modeled by a vibration and distortion of a liquid drop that leads to a splitting of the drop [9]. For track formation, the important information is that, when ²³⁸U spontaneously fissions, the mass is divided typically slightly asymmetrically in the ratio of about 1.4 to 1; correspondingly, the kinetic energies of the two fragments are typically about 100 MeV for the lighter fragment and 70 MeV for the heavier one, which consequently has a shorter range. As an example, these energies correspond to ranges of $\sim 11 \ \mu m$ and 7 or 8 μm in muscovite mica. There is in fact a wide range of mass divisions and kinetic energies, but the sum of the energies for the pair of fission fragments is normally [10] not very different from 170 MeV Neutron-induced fission of the other major isotope of U, ²³⁵U, gives marginally lower energies; but the lengths of the etched tracks are not measurably different from those for 238 U.

It follows that individual fission tracks are in reality two tracks—joined end-to-end—produced by two particles that diverge in opposite directions. But, they are revealed by etching as single, continuous tracks with no feature that identifies where the fissioned nucleus was located.

3. Track revelation and geometry

Tracks consist of narrow line-like regions that are densely populated with misplaced atoms, vacant atomic positions, and (presumably) broken or distorted atomic bonds. Macroscopically this damage amounts to a line of disturbed material with an elevated free energy and therefore enhanced chemical reactivity. It is this reactivity that allows the simple revelation of tracks by chemical etching.

The geometry of etching in the simplest case is shown [11] by Fig. 2. A conical hole results from accelerated dissolution at a rate v_T along the track and a lesser general rate v_G on the undamaged solid elsewhere. The cone half angle θ is given by $\sin^{-1}(v_G/v_T)$, as noted by Fleischer and Price [3]. Tracks at angles less than θ to a surface are not revealed, leading to only a fraction $\cos^2 \theta$ of the tracks being displayed as etch pits. This fraction of tracks that intersect a given surface is termed the *etching efficiency* (see reference [12]).

For fission tracks, the damage is most intense near the midway point of the track (i.e., where the fission occurred) and decreases near the ends, with a corresponding decrease in the etching rate. This variation in v_T with position along the track complicates the track geometry somewhat. Real track profiles are somewhat convex or concave cones, one example being sketched in Fig. 3, which presents the profiles at different times calculated for an observed $v_T(x)$ as etching slows



Figure 1 Mica was the first substance in which ²³⁸U spontaneous fission tracks were identified by Price and Walker [2]. The tracks in this Madagascar phlogopite were revealed by etching in hydrofloric acid. From Fleischer and Price [5], reprinted by permission of Pergamon Press, Ltd.

toward the end of a track [13]. It is evident that the etched track grows indefinitely with extended etching but that the pit becomes progressively both less sharp and less distinctive once etching has removed the etchable damage.

Figs 2 and 3 are drawn with θ values that are typical of silicate glasses. For most crystalline minerals and plastics, θ is small; and etched tracks are much more spike-shaped. The figures, however, are designed to clarify how track shapes come about. An extensive list of etchants for different solids is given by Fleischer *et al.* [7].

4. Mechanism of track production

Although many details of the physics of track production are not known, the ionization spike model [14] broadly fits a wide variety of observations that limit possible mechanisms. It is widely accepted as best working model for how tracks form in inorganic solids. It is a three-stage process, as described in Fig. 4.

A full-energy fission fragment moves at about 0.04 of the velocity of light, i.e., faster than the orbital velocity of many of the electrons that would be bound to the fission particle if it were at rest. Under this circumstance, these electrons are stripped off in passing



Figure 2 Track geometry for constant general dissolution rate v_G and two different track attack rates v_T . The track on the right, with the lower v_T , is smaller, shorter, and blunter. From Fleischer [11], reprinted by permission of Pergamon Press, Ltd.



Figure 3 Calculated etching sequence of a fission-fragment track with initial cone angle 26.5° in an obsidian-like glass. It was assumed that $v_{\rm G} = 0.58 \,\mu$ m/sec, $v_{\rm T} = 1.36 - 0.657 \,\mu$ m/sec, where y is the distance along the track and R (the etchable range) is 12 μ m. From Fleischer *et al.* [13], reprinted by permission of the American Physical Society.

through matter, converting the fission fragment to a multiply charged positive ion, as pictured in the upper diagram of Fig. 4. In stage 1 of track formation, the moving ion in turn removes electrons from atoms that lie close along its path, creating and leaving behind an irregular, needle-shaped region of positive ions. The track formation occurs in the second stage (Fig. 4, middle) in which the mutual repulsion of neighboring positive ions hurls them into interstitial positions, leaving a region that is permeated with vacant atomic sites. As pictured in the lowest diagram, the final stage is an elastic adjustment of the crystal. This gives rise to the long-range lattice strains that allow tracks to be imaged in the transmission electron microscope, as first seen by Silk and Barnes [15].

Note that the net result of this ionization process is damage that is atomic rather than solely electronic in nature. The expected structure is complicated when visualized on an atomic scale, and this complexity is ev-



Figure 4 The ion explosion spike mechanism for track formation in inorganic solids. The original ionization left by the passage of a charged particle (top) is unstable and ejects ions into the solid, creating vacancies and interstitials (middle). Later the stressed region relaxes elastically (bottom), straining the undamaged matrix. From Fleischer [11], reprinted by permission of Pergamon Press, Ltd.

ident in the observations of the effects of subsequent heating, which shortly will be discussed further. Close encounters at atomic and subatomic spacings between the projectile and atoms in the solid lead to statistically varying effects, which are part of the complexity just mentioned. The variability along a track is often not evident when chemical etching is used to display tracks, since the etching process usually gives a measure of average damage density. The statistical variability of the process is, however, accentuated and displayed by nuclear photographic emulsions, which superimpose the many tracks of ejected electrons, as shown in Fig. 5. Such tracks display the effects of the most energetic electrons that the track-forming ion ejects from atoms-the variable width of the track is the result of the closest, most statistically variable Coulomb collisions along the particle trajectory.

5. Predictions of the model of track formation versus experimental observations

Let us consider the experimental observations and, for each, the possible relation to the model just described [7, 14].

1. Tracks are found in dielectric substances, but with rare exceptions [16, 17] not in metals or good



Figure 5 Track of a relativistic iron nucleus in a nuclear emulsion. The irregularities in the width of the track are from the superimposed tracks of the many ejected electrons and reflect the statistical nature of the ion-electron collisions. Photograph courtesy of P.H. Fowler.

semiconductors (Table I). From Fig. 4, it is clear that if electronic conduction neutralizes the positive core before ions have time to be displaced to new sites, those displacements will be quashed. In metals, electrons promptly move to neutralize the charge; in semiconductors, the charge can dissipate directly, because it is equivalent to an array of positive current carriers (holes), which are mobile. Table I shows the correlation of track formation with electrical conductivity that is suggested by this picture. 2. From observed activation energies of from one to a few electron volts for total thermal erasure of tracks, it is inferred that the lasting damage zone of tracks consists of atomic defects, not electronic ones. Fig. 4 shows how the ionization spike mechanism does produce atomic displacements, even though the initial effects are mostly electronic.

3. The observed thresholds for track production [11, 18] (Fig. 6) fit a primary ionization criterion for registration (which will be described here interchangeably

TABLE I Relation of track formation to electrical resistivity

	Material	Resisitivity range (ohm-cm)
I. TRACK-FORMING		
Insulators	Silicate minerals Alkali halides Insulating glasses Polymers	$10^{6} - 10^{20}$
Poor insulator	MoS_2	3,000-25,000
Semiconductor	V ₂ O ₅ glass	2,000-20,000
II. NON-TRACK-FORMING	-	
Semiconductors	Germanium Silicon	10-20,000
Metals	Aluminum Copper Gold Platinum Tungsten Zinc	10 ⁻⁶ -10 ⁻⁴

From Fleischer et al. [14].

as an ionization threshold or a registration threshold) and disagree with several other proposed criteria [11] energy loss, restricted energy loss, and delta-ray energy loss. It is the primary ionizations—the core ionizations per unit path length—that provide [or do not) sufficient energy locally to induce atomic disordering (see Fig. 4, top).

4. As seen in Fig. 6A, different materials may have very different ionization thresholds. Although the details of the model based on the ion spike will not be given here, Table II shows that a quantity called the *stress ratio* correlates rather well with the groupings of ionization thresholds. The stress ratio represents, for a given ionization, the electrostatic forces tending to displace atoms divided into the bonding forces tending to hold atoms in place [14]. The stress ratio depends only on fundamental solid properties, i.e., interatomic spacings, elastic moduli, and dielectric constants. A more detailed model would need to consider specific crystal structures, but no proposals have been made as to how to do so systematically.

5. Etchable tracks are shorter than the full distance between the points where the two fission fragments come to rest after a fission event. The range deficit, as illustrated in Fig. 6B, is about 1 μ m for mica, 3.5 μ m diopside, and 4 μ m for hypersthene *at each end* of the fission track [18]. The nature of the ionization curves for fission fragments in Fig. 6 (represented by the ions Kr or Br and I as light and heavy fission fragments, respectively is that there is decreasing ionization toward low velocity for fission fragments; it follows that near the end of the range of a fission fragment there may be a space where no etchable track is formed. In part B of the figure the low-energy region of relevance to fission fragments (below 1 MeV per mass unit) is magnified to demonstrate how the ionization rate decreases toward zero energy.

6. What determines track formation in oxides is not clear, although progress has been made. Fission tracks are apparently not formed in Al_2O_3 , MgO, ThO₂, UO₂, Y_2O_3 , and ZnO, but are in Al_2MgO_4 , $Ca_2Ta_2O_7$, GeO₂,

SPECIAL SECTION IN HONOR OF ROBERT W. CAHN

TABLE II Relation of registration thresholds to calculated stress ratio

Detector class	Detector groups of similar registration thresholds (highest threshold at top)	Average stress ratio
Crystals	Hypersthene Olivine	3.3
	Laboradorite Zircon	1.4
	Diopside Augite	1.3
	Oligoclase Bytownite Orthoclase Quartz Micas	0.5
Inorganic glasses	Silica Flint Tektite	0.7
	Soda lime Phosphate	0.5
Plastics	Group of 14 plastics	0.01

From Fleischer [11].

The stress ratio is $E\varepsilon a^4/10e^2$, where E = Young's modulus, $\varepsilon =$ dielectric constant, a = lattice constant, and e = charge of the electron.

PbWO₄, SiO₂, and the related structures CaF₂, LiF, and NaCl. The picture is further complicated in that more intensely ionizing particles than fission fragments do produce tracks in superconducting oxides [19]. Correlations with thermal diffusivity [11, 20] led to a variant of the ion spike that allows de-excitation by thermal conduction. This model also is consistent with a wide range of sputtering data [21]. Touloukian *et al.*'s collection of thermal properties [22] shows that the correlation with thermal diffusivity is imperfect amongst the oxides, i.e., there is no clear threshold above which all solids register tracks and below which none do. A complete description of criteria for track formation is yet to be given.

Some of the cases in which fission tracks are not formed may exist because the solids have very high ionization thresholds, rather than inherent nonregistration. In some materials where fission tracks are not formed, tracks are nevertheless produced by accelerated much heavier ions than fission fragments, for example Pb and U, or by C_{60} (fullerine) molecules. For a fuller discussion see my review of tracks in intermetallic compounds [17].

5.1. Statistical effects

Unheated tracks of particles that ionize at a rate that is well above the registration threshold of a given mineral consist of essentially continuous damage on an atomic scale; hence, continuous etching can occur along the track. In this circumstance there is no visible effect of the statistical nature of ionization and atomic disorder along a track. On the other hand, strong effects exist for particles that ionize just above threshold or for tracks that have been annealed sufficiently that the damage becomes discontinuous or lumpy.

Fig. 7 shows track densities and track lengths measured in heated muscovite mica samples [23]. Track



Figure 6 Ionization versus velocity for different charged particles. Part A shows that each detector has an ionization threshold, below which no tracks are etched and above which all particles create tracks. The solid dots and open circles are data for Lexan polycarbonate detectors. Solid dots indicate that each ion forms an etchable track; open circles mean that none do. Thresholds for other detectors are also indicated. CELL.NIT. is cellulose nitrate. From Fleischer [11], reprinted by permission of Pergamon Press, Ltd. and the American Physical Society. Part B expands the scale to show how ionization rates decrease toward zero energy below 1 MeV per mass unit. From Price *et al.* [18].

lengths begin to decrease well before track densities do, an effect that is attributable to a few very short sections along tracks healing totally, probably due to local downward fluctuations in the original ionization density. In muscovite the general etching rate v_G , normal to the layer planes is nearly zero, so that gaps in a track prevent the full track from being etched. Thus the etched length of tracks decreases but observed numbers of tracks are stable, provided the gaps occupy a negligible fraction of the original etchable length. This behavior is likely to occur in other layer minerals, and very likely to occur in a less pronounced manner in some other minerals, i.e., using the minimum etching times that will normally display most of the tracks. The reason for this behavior is that when $v_{\rm G}$ is not too much less than $v_{\rm T}$, gaps will merely decrease the effective etching rate along tracks. Nearly the full length is still etchable, but longer etching times are needed than for unheated tracks, and track densities and lengths are more sensitive to the duration of etching.

Dartyge *et al.* [24] examined the effects of annealing on track etching rates in minerals. From small-angle X-ray scattering, they inferred the distribution of gaps that have low $v_T^{(L)}$ and what they term *extended defects* (regions of continuous damage) of higher $v_T^{(H)}$. The track length versus etching time is then calculated from the relative fractions of $v_T^{(L)}$ and $v_T^{(H)}$. Their success is especially important to the identification of



Figure 7 Effect of annealing for 1 hour at a temperature T on the average length l and density N of etched tracks in muscovite from Renfrew County, Canada. In the graph l and N are normalized to the average length and density measured with no heat treatment. From Fleischer *et al.* [23], copyright by the American Association for the Advancement of Science.

heavy cosmic-ray tracks in extraterrestrial minerals and might become useful in quantifying thermal events via the etching of heat-affected fission tracks. Tombrello *et al.* [25] present a variation on the ionization spike model for production of extended defects: Deep-shell ionization results in multiple Auger electron emission and consequent ion-spike Coulomb displacements [8].

6. A major use of fission fragment tracks: Fission track dating

Finding of natural tracks in micas by Price and Walker [26] provided scientific excitement that led to a new method for measuring geological ages-fission-track dating. The critical step was their recognition [27] that these tracks were unambiguously from spontaneous fission and thus would allow radiometric dating of minerals if tracks were stored for geological times. Fig. 1 is one of the first published optical photos of known ²³⁸U spontaneous fission tracks in a mineral. The early dating experiments [5, 6, 12] confirmed for several minerals and natural glasses that fission-track dating could give ages that agreed with established ages and thereby empirically established the spontaneous fission origin of the natural tracks. The range of useful minerals for track dating has expanded [28], and methods of recognizing and quantifying heating effects have strengthened the reliability and usefulness of dating results. Understanding of occasional differences from ages determined by other radiometric techniques has led to new qualitative and quantitative information of geological, geophysical, technological, and economic importance, as was recently summarized in Chapter 4 of reference [8].

Here I note the identified sources, and conceivable sources, of natural charged particle tracks in minerals and review how we infer that spontaneous fission of ²³⁸U is the only source of importance for terrestrial minerals—except in very special circumstances. Response of tracks to heating is a prime consideration, because much of the information that comes uniquely from track dating is derived from annealing effects.

7. Sources of natural tracks

How in general does one know that natural tracks found in terrestrial minerals and glasses result from spontaneous fission of 238 U? There are two routes. The simplest, most direct, and most widely convincing one comes from empiricism. We assume 238 U spontaneous fission is the sole contributor, measure ages, and find no evidence for other tracks. In short, fission-track ages that are too high are in general absent. Fig. 8 (from Fleischer *et al.* [7]) is an example: Of about 60 fissiontrack ages given here, none are meaningfully high relative to ages established by other methods.

There are, however, occasional special situations where excess tracks are expected and are seen. For this reason the alternative route must also be taken of enumerating the mechanisms for producing natural tracks, deciding which of these might be confused with ²³⁸U fission tracks, and listing the special situations where those tracks might be present. The troubling pitfall in this course is that our imagination maybe incomplete in listing possibilities. However, no existing observations demand a presently unrecognized mechanism for producing tracks. We therefore have some confidence that we are not overlooking a major, frequently occurring alternative.

We now list possible sources. By restricting our dating work to terrestrial samples, we eliminate the sources that are related to cosmic rays, most of which are shielded from the surface of the earth by the atmosphere. In extraterrestrial samples several types of tracks may be present [29], but they can usually be separated by measuring lengths or angular distributions [30, 31].

7.1. Listing of potential track sources

In the paper [27] in which Price and Walker proposed a procedure for fission track dating, they listed the following potential track sources in addition to spontaneous fission of 238 U:



Figure 8 Fission-track dating has been shown to give correct ages over time spans ranging from 0.5 year to more than 10^9 years. The graph compares fission-track ages, measured by counting natural tracks formed by spontaneous fission of 238 U, with ages known by other means, either documented ages for man-made samples (which appear in the lower left) or ages measured by other radioactive decay techniques for the geological samples indicated in the upper right. Fission-track ages of some natural samples are lowered by thermal effects, allowing the fission-track technique to serve as a thermometer for past events. The lack of fission-track ages significantly older than "known" ages implies that 238 U spontaneous fission is the sole contributor of fossil tracks. From Fleischer *et al.* [7], reprinted by permission of the University of California Press.

1. Spontaneous fission of other natural existing heavy elements or nuclides (²³⁵U, Th, Bi, Pb, and lighter elements). Extensive searches have produced no evidence of spontaneous fission of any element that is lighter than uranium.

2. Fission of heavy elements induced by natural α , β , or γ activity.

3. Fission of ²³⁵U induced by thermalized neutrons from spontaneous fission, (γ, n) reactions, and (α, n) reactions.

4. Fission of other heavy nuclides induced by fast neutrons from spontaneous fission, (γ, n) , and (α, n) reactions.

5. Fission induced by cosmic ray primaries.

6. Fission induced by cosmic ray secondaries, including neutrons and μ -mesons.

7. Spallation caused by cosmic ray secondaries.

In addition Fleischer *et al.* [29] considered these possibilities:

8 Spallation caused by primary cosmic rays.

- 9 Tracks of slowed cosmic ray primaries.
- 10 Spontaneous fission of ²⁴⁴Pu.
- 11 Meson jets.
- 12 Magnetic monopoles.

Other possibilities:

13 Recoiling nuclei from alpha decay, Huang and Walker [32]

14 Nuclei from which alpha particles have scattered, Crozaz *et al.*[33]

15 Nuclei with which alpha particles have undergone nuclear interactions, Crozaz *et al.* [33], Price and Salamon [34].

A number of these candidates can be dismissed immediately as sources of confusion with fission tracks on earth. Items 7, 8, 11 (if such tracks exist), 13, 14, and 15 are much shorter than fission tracks; item 12 (if such particles exist) would give tracks that are much

longer than fission tracks, and they would occur in minute numbers. Item 9 (heavy primary cosmic ray particles) would give some tracks that are much too long for fission events; but, regardless, all such nuclei are destroyed high in the atmosphere, thereby eliminating item 5 also. All 15 of the potential sources listed above can be eliminated on a quantitative basis by considering normal abundance's, decay rates, and cross sections [27, 30] with certain pathological exceptions.

7.2. Exceptional track sources

Since track sources other than fission can usually be distinguished from fission tracks by their lengths, orientations, or phase distributions, the exceptional track sources than can most easily be confused with ²³⁸U fission tracks are also from fission. As examples, fission tracks will be in the mineral phases in proportion to the uranium concentrations, while cosmic rays will cross from one phase into another, giving roughly equal abundances of tracks in adjacent mineral grains.

1. The best known case where an other type of fission is present occurs at the Oklo Mine in Gabon, a uraniium deposit that 2×10^9 years ago was so enriched in ²³⁵U (²³⁵U/²³⁸U~3%) that it operated as a natural, watermoderated reactor [35]. Neutrons released at such a reactor induce extra fission tracks that can be used to measure the ancient, local neutron dose [36].

2. Similarly, for a mineral grain imbedded in uranium ore, fission that is induced by the neutron background could be rapid enough to be comparable to spontaneous fission [27]. Such tracks have not been reported. For example Weiland [37] and Cunningham *et al.* [38] did not find any excessively high ages when track dating uranium mineralizations at Marysvale, Utah.

3. In a mineral that is low in uranium (and therefore has little spontaneous fission), induced fission of other heavy elements by energetic cosmic ray secondary particles could be dominant. However, a portion of these high-energy fission tracks would be distinctively V-shaped, rather than straight, because of the momentum imparted by the cosmic ray that induced fission. V-tracks caused by natural cosmic rays have been observed in a high-lead glass that was exposed on the lunar surface on the Surveyor III spacecraft and recovered during the Apollo 12 mission [39]. Such tracks are rare in terrestrial samples. C.W Naeser and R.L. Fleischer (1974, unpublished) could not find V-tracks in a search of Pb-rich, high-altitude (4,300 m) feldspar crystals from Pikes Peak, Colorado, USA. In the one case where V-tracks were actually seen on earth, they were rare, and found in lead-rich, man-made glasses. These V-tracks were first thought to be due to rare superheavy elements [40, 41], but induced fission by highenergy, low-mass cosmic ray secondaries is now agreed to be the origin [41, 42].

4. Tracks from now extinct ²⁴⁴Pu which existed in the early solar system have been identified in meteorites [43, 44]. The short half-life (relative to the age of the solar system) of 82×10^6 years for ²⁴⁴Pu insures that such tracks can be present in significant numbers only in materials like the meteorites that have been cool and undisturbed since times near the beginning of the solar system. Consequently, such tracks have never been identified in terrestrial material, where they would lead to mysteriously high fission-track ages.

5. Nuclear explosions produce high local neutron fluences that can induce fission [45]. These are not regarded as natural tracks, but they are expected to be present in certain locales [46]. Tracks in glass from near to ground zero at Hiroshima have been identified in a decorative glass button [47] and in glazes on several porcelains [48].

8. Response to thermal treatments

Heat tends to heal radiation damage tracks atom-byatom; and, given sufficient time and temperature, heating can totally erase tracks. Earlier we noted that activation energies for total healing tell us that tracks consist of displaced atoms. Tracks that are only partially affected by heating have specific, slowed etching behavior that also helps to describe the radiation damage; and recognition of heat-affected samples is a key first to interpreting (or in some cases correcting ages) and second to characterizing geological thermal processes quantitatively, both as to the timing and extent of heating. Past heating can be dated, and where not to waste vast sums by drilling for oil where there is none as described briefly in Chapter 4 of reference [8].

Changes in track size that were caused by heating were identified by Berzina *et al.* [49], and Storzer and Wagner [50] showed how they could be used to infer what the track densities in a heat-affected sample would have been in the absence of heating. This was a critical step towards improving fission-track dating; it also allows a test of our understanding of the relation between track geometry and etching efficiency.

Storzer and Wagner's data [50] on an Australian tektite in Fig. 9 show that regardless of the combination of



Figure 9 Etch pit diameter versus track density for an Australian tektite as determined in laboratory annealing experiments, Storzer and Wagner [50]. The mean track diameter *D* in an annealed sample is plotted against the track density ρ for that sample; the data are normalized to the mean track diameter D_0 and track density ρ_0 for an unannealed sample. The solid curve is smoothed through the points. The dashed curve was calculated using parameters measured on a synthetic tektite glass by Fleischer and Hart [51] furnished by D.R. Chapman. From Fleischer *et al.* [7], reprinted by permission of the University of California Press.

heating time and temperature chosen, a single relation governs the decreases in the diameters and densities of etched tracks. As shown by the calculated dashed line [51], this relation can be understood as a lowered tracketching rate leading to a decreased etching efficiency and larger values of the track cone angle θ . In other work Sandhu and Westgate [52, 53] have shown that the diameter-density relationship in heated glasses is sensitive to the duration of etching; using etching times that produced mean induced track diameters between about 6 and 8 μ m, they obtained a nearly linear relationship between the decrease in track diameter and track density in heated glasses.

In summary of dating possibilities, fission tracks in natural terrestrial minerals and glasses are almost exclusively from ²³⁸U spontaneous fission. The track dater need only be aware of possible exceptions that can occur near natural or man-made nuclear activity—at sites of uranium ore or nuclear explosions. For extraterrestrial materials the situation is far more complex, and the reader is referred to other literature [7, 29, 30].

9. Overall summary

For terrestrial samples virtually all of the natural fission tracks arise from the spontaneous fission of ²³⁸U, the process that makes fission-track dating possible. Only in certain rare, pathological cases is there a background of other fission origin: Near natural reactors and nuclear explosions such tracks have been seen; in or near rich uranium ore they are likely, but not yet observed. Other natural tracks are distinctively different. One example is the rare V-shaped fission induced near sea level by cosmic ray secondary particles. In extraterrestrial matter numerous complications may arise. These extraterrestrial sources have been listed here and are analyzed carefully in Chapter 6 of Fleischer *et al.* [7].

Track formation in inorganic solids is attributed to ionization followed by Coulomb repulsion to produce atomic disorder. The statistical nature of ionization leads to discontinuous tracks for particles that are near the ionization threshold or for tracks that are partially annealed. These heat-affected tracks can be recognized and in many cases used to infer thermal histories.

Acknowledgments

The author thanks N. D. Naeser for helpful comments, P.H. Fowler for supplying Fig. 5, and the American Physical Society, Pergamon Press, *Science*, and The University of California Press for permission to reprint the other figures.

Dedication

This work is a tribute to Robert Cahn whose scientific excellence, versatility, and energy have earned our widespread admiration as a scientist for all seasons.

References

- 1. D. A. YOUNG, Nature 182 (1958) 375.
- 2. P. B. PRICE and R. M. WALKER, J. Appl. Phys. 33 (1962) 3407.
- 3. R. L. FLEISCHER and P. B. PRICE, Science 40 (1963) 1221.

- 4. Idem., J. Appl. Phys. 34 (1963) 2903.
- 5. Idem., Geochim. Cosmchim. Acta 28 (1964) 1705.
- 6. M. MAURETTE, P. PELLAS and R. M. WALKER, Bull. Soc. Franc. Min. Crist. 87 (1964) 6.
- R. L. FLEISCHER, P. B. PRICE and R. M. WALKER, "Nuclear Tracks in Solids—Principles and Applications" (University of California Press, Berkeley, 1975).
- R. L. FLEISCHER, "Tracks to Innovation—Nuclear Tracks in Science and Technology" (Springer-Verlag, New York, 1998).
- 9. L. MEITNER and O. R. FRISCH, Nature 143 (1939) 239.
- E. K. HYDE, "The Nuclear Properties of the Heavy Elements III: Fission Phenomena" (Prentice-Hall, Englewood Cliffs, NJ, 1964).
- R. L. FLEISCHER, Nuclear Track Production in Solids, Chapter 4 in *Progress in Materials Science*, "Chalmers Anniversary Volume" 97–123 (1981).
- 12. R. L. FLEISCHER and P. B. PRICE, J. Geophys. Res. 69 (1964) 331.
- 13. R. L. FLEISCHER, P. B. PRICE and R. T. WOODS, *Phys. Rev.* 88 (1969) 563.
- 14. R. L. FLEISCHER, P. B. PRICE and R. M. WALKER, J. Appl. Phys. 36 (1965) 3645.
- 15. E. C. H. SILK and R. S. BARNES, Phil. Mag. 4 (1959) 970.
- 16. A. BARBU, H. DAMMAK, A. DUNLOP and D. LESUEUR, *MRS Bull.* **20**(12) (1995) 29.
- R. L. FLEISCHER, Ion Tracks in Intermetallics, in "Intermetallic Compounds—Principles and Practice" vol. 3 (Progress), (J. Wiley, Ltd., Chichester, 2002) Chap. 14, p. 263.
- P. B. PRICE, R. L. FLEISCHER and C. D. MOAK, *Phys. Rev.* 167 (1968) 277.
- J. PROVOST, CH. SIMON, M. HERVIEU, D. GROULT, V. HARDY, F. STUDER and M. TOULEMONDE, MRS Bull. 20(12) (1995) 22.
- 20. A. SIGRIST and R. BALZER, Helv. Phys. Acta 50 (1977) 49.
- 21. L. E SEIBERLING, J. E. GRIFFITH and T. A. TOMBRELLO, *Rad. Effects* 52 (1980) 201.
- Y. S. TOULOUKIAN, R. W. POWELL, C. Y. HO and M. C. NICOLAOU, "Thermal Diffusivity" (IFI/Plenum, New York, 1973).
- 23. R. L. FLEISCHER, P. B. PRICE, E. M. SYMES and D. S. MILLER, *Science* 143 (1964) 349.
- 24. E. DARTYGE, J. D. DURAND, Y. LONGEVIN and M. MAURETTE, *Phys Rev. B* 23 (1981) 5213.
- 25. T. A. TOMBRELLO, C. R. WIE, N. ITOH and T. NAKAYAMO, *Phys. Lett.* A **100** (1984) 42.
- 26. P. B. PRICE and R. M. WALKER, Nature 196 (1962) 732.
- 27. Idem., J. Geophys. Res. 68 (1963) 4847.
- G. A. WAGNER and P. VAN DEN HAUTE, "Fission Track Dating" (Enke, Stuttgart, 1992).
- 29. R. L. FLEISCHER, P. B. PRICE, R.M. WALKER and M. MAURETTE, J. Geophys. Res. 72 (1967) 333.
- R. L. FLEISCHER, P. B. PRICE, R. M. WALKER, M. MAURETTE and G. MORGAN, *ibid.* 72 (1967) 355.
- 31. R. L. FLEISCHER, P. B. PRICE and R. M. WALKER, Charged Particle Tracks—Tools for Geochronology and Meteorite Studies in "Radiometric Dating for Geologists," edited by E. Hamilton and R. M. Farquhar (Wiley-Interscience, London, 1968) p. 417.
- 32. W. H. HUANG and R. M. WALKER, Science 155 (1967) 1103.
- 33. G. CROZAZ, M. HAIR, M. MAURETTE and R. M. WALKER, Nuclear Interaction Tracks in Minerals and Their Implications for Extraterrestrial Materials, in Proceedings International Conference Nuclear Track Registration in Insulating Solids (Clermont-Ferrand, France, 1969) Section VII, p. 41.
- 34. P. B. PRICE and M. H. SALAMON, Nature 320 (1986) 427.
- 35. R. BODU, H. BOUZIGUES, N. MORIN and J. P. PFIFFELMAN, *Compt. Rend., Paris* 275 (1972) 1731.
- 36. M. MAURETTE, Ann. Rev. Nucl. Sci. 26 (1976) 319.
- E. K. WEILAND, Fission Track Dating Applied to Uranium Mineralization: Thesis, Colorado School of Mines, Golden, Colorado, 1979, p. 74.
- 38. C. G. CUNNINGHAM, K. R. LUDWIG, C. W. NAESER, E. K. WEILAND, H. H. MEHNERT, T. A. STEVEN and J. D. RASMUSSEN, *Economic Geology* 77 (1983) 453.

- 39. R. L. FLEISCHER, H. R. HART and G. M. COMSTOCK, *Science* **171** (1971) 1240.
- G. N. FLEROV and V. P. PERELYGIN, On Spontaneous Fission of Lead—Search for Very far Transuranium Elements (Joint Institute for Nuclear Research, Dubna, U.S.S.R., 1969) Preprint D7-4205, p. 8.
- 41. G. N. FLEROV, V. P. PERELYGIN and O. OTGONSUREN, Atomnaya Energiya 33 (1972) 979.
- 42. F. H. GEISLER, P. R. PHILLIPS and R. M. WALKER, *Nature* 244 (1973) 428.
- 43. R. L. FLEISCHER, P. B. PRICE and R. M. WALKER, J. Geophys. Res. 70 (1965) 2703.
- 44. R. L. FLEISCHER, P. B. PRICE and R. M. WALKER, *Geochim. Cosmochim. Acta* **32** (1968) 21.
- 45. R. M. WALKER, Characteristics and Applications of Solid State Track Detectors in Proceedings Strasbourg Conference on New Methods of Track Detection, Centre de Recherches Nucleaires, Strasbourg, France, 1963.
- 46. R. L. FLEISCHER, Health Phys. 52 (1987) 219.

- 47. R. L. FLEISCHER, S. FUJITA and M. HOSHI, *ibid.* 81 (2001) 720.
- 48. J. MACDONALD, R. L. FLEISCHER, S. FUJITA and M. HOSHI, *Health Physics* 85 (2003) 428.
- 49. I. G. BERZINA, I. B. BERMAN and I. M. ZLOTOVA, Izv. Akad. Nauk SSR, Ser. Geological 31 (1966) 10.
- 50. D. STORZER and G. A. WAGNER, *Earth Planet. Sci. Lett.* **5** (1969) 463.
- 51. R. L. FLEISCHER and H. R. HART, Jr., 1972, Fission Track Dating: Techniques and Problems, in "Calibration of Hominoid Evolution," edited by W. W. Bishop, D. A. Miller and S. Cole (Scottish Academic Press, Edinburgh, 1972) p. 135.
- 52. A. S. SANDHU and J. A. WESTGATE, *Earth Planet. Sci. Lett.* **131** (1995) 289.
- 53. A. S. SANDHU and J. A. WESTGATE, The Relationship Between Reduction in Fission Track Diameter and Areal Track Density in Natural Glasses: Influence of Etching Conditions, in "Abstracts, International Workshop on Fission-Track Dating" (Gent, Belgium, 1996) p. 97.