GEOSCIENCE

Dynamics of the Continental Lithosphere

Fission-track Thermochronology

Methodology and Applications to Geology

Coordinated by
Marc Jolivet

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Contents

Introduction

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The age, duration and speed of processes are essential parameters for most Earth science studies. For example, to describe the growth and subsequent dismantling of a mountain range, we need to date the emplacement of magmas, determine the age and speed of metamorphic processes, and measure the slip rates of faults and erosion.

The first attempts to determine the Earth's age date back to antiquity, with Aristotle (384–322 BCE) declaring that the Earth is eternal. This thesis lasted until the end of the Middle Ages, before giving way to attempts at calculation based on the Bible. The Irish archbishop J. Ussher (1581–1656) declared that the Earth was formed at Creation, at the beginning of the night before October 23 in the year 4004 of the Julian calendar. It was not until the beginning of the 20th century, following Henri Becquerel's discovery of radioactivity in 1896, that modern geochronology developed. Using radiogenic helium produced by the decay of radium, J.W. Strutt (Lord Rayleigh) was the first to provide an age greater than 1 billion years. However, his approach was largely biased by the fact that radiogenic helium is produced not only by the radioactive decay of radium, but also by that of many other elements such as thorium and uranium. In the 1950s, technical advances made during the Second World War, notably in mass spectrometry, enabled Patterson (1956) to show that meteorites and the Earth formed at the same time 4.55 billion years ago (± 70 million years) (Figure I.1).

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Figure I.1. *Portraits of Aristotle, James Ussher (by P. Lely), John William Strutt (Lord Rayleigh) and Clair C. Patterson. For a color version of this figure, see www.iste.co.uk/jolivet/fission.zip*

From this period onwards, a whole series of geochronological methods were developed based on the "closure temperature" principle (see Chapter 1). Every geochronological system consists of a mineral and a radioactive element–radiogenic element pair. The closure temperature corresponds to the temperature below which diffusion of the radioactive and radiogenic isotopes in question is no longer sufficient to allow exchanges with the outside of the mineral (Dodson 1973). Geochronological methods therefore exist to date a wide range of geological phenomena over a wide range of temperatures, providing information in the form of an age associated with a temperature (Figure I.2). The combined use of several geochronometers on a sample of the same rock enables us to follow the cooling history of this rock

"in dotted lines", highlighting periods of accelerated cooling, synonymous with tectonic movements (Figure I.3).

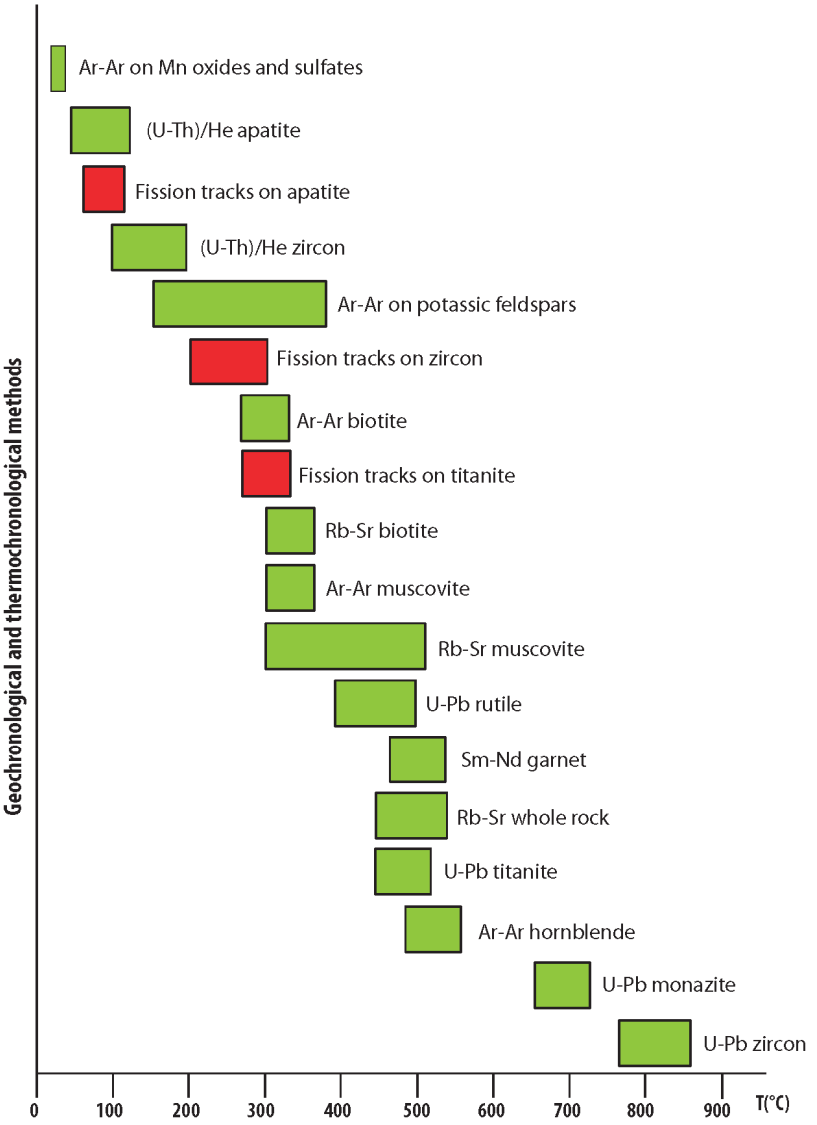

Figure I.2. *Main geochronometers and thermochronometers used and their temperature of application. Methods based on fission tracks are shown in red. For a color version of this figure, see www.iste.co.uk/jolivet/fission.zip*

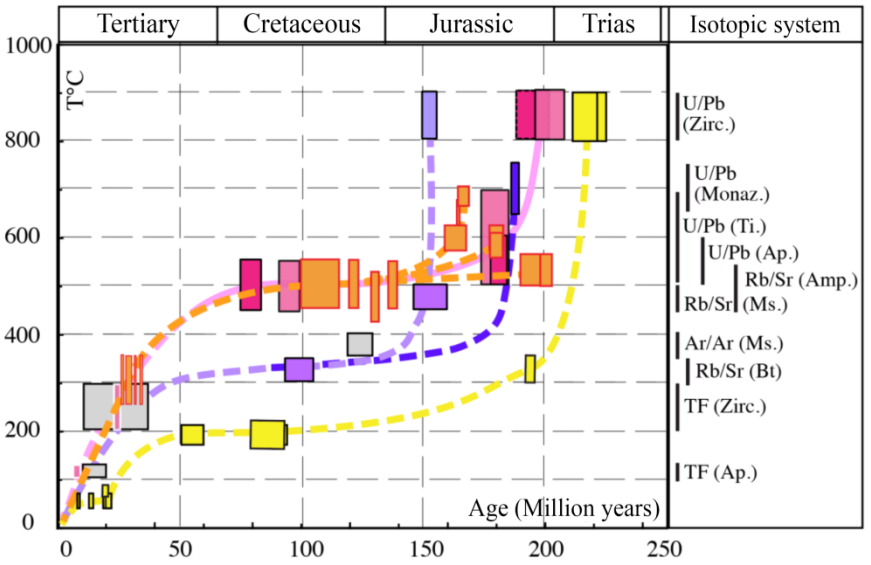

Figure I.3. *Example of a cooling curve drawn by assembling temperature-age points obtained from several geochronometers applied to the same sample. The thermal history obtained is discontinuous. Modified from Roger et al. (2011). For a color version of this figure, see www.iste.co.uk/jolivet/fission.zip*

In 1958, D.A. Young discovered that LiF crystals coated with a uranium film and irradiated with thermal neutrons showed a number of "holes" after chemical treatment. The number of these "holes" is in perfect agreement with the theoretical number of fission fragments of uranium atoms expected for the neutron dose applied (Young 1958). In fact, the spontaneous fission of an atom of ^{238}U or ^{232}Th generates two ionizing particles which, as they pass through a solid medium, leave a "track". In a crystallized solid, this "track" corresponds to the displacement of atoms in the crystal lattice. In an organic medium such as plastic, the track materializes as chains of broken molecules (Durani and Bull 1987). In the early 1960s, R.L. Fleischer, P.B. Price and R.M. Walker developed a method for detecting these "fission tracks", adapting Young's technique for revealing tracks by chemical etching to a wide variety of materials (minerals, glass, plastics) (Fleischer et al. 1965, 1975) (Figure I.4). As the number of fission tracks in a given

mineral depends directly on the concentration of radioactive elements (mainly ^{238}U) and the time over which the mineral has accumulated tracks, it is possible to use this phenomenon as a geochronometer: measuring the concentration of ^{238}U and the density of fission tracks enables time to be calculated. Wagner (1968) demonstrated in 1968 that if minerals such as apatites or zircons are heated, the fission tracks they contain can be "erased" (the crystal lattice is reconstituted). The higher the temperature, the faster the tracks disappear. For each mineral, the temperature range in which the tracks disappear over a time scale compatible with geological time (a few million years to tens of millions of years) is called the partial annealing zone l (PAZ) of the tracks. In apatites, for example, it ranges from around 130°C to 60°C, and in zircons, it ranges from around 250°C to 180°C. Other parameters, such as pressure, have no appreciable effect on the rate at which tracks are erased (Fleischer et al. 1975). By measuring the distribution of track lengths in a mineral, it is then possible to statistically calculate a temperature-time path for each sample, making it possible to continuously monitor the thermal history of a sample in the PAZ temperature range (see Chapter 2).

Fission-track thermochronology has become a key tool in geological research, whether fundamental (tectonics, geomorphology, sedimentology) or applied (hydrocarbon prospecting). In this book, we explore the latest methodological developments that are constantly improving the results obtained using this technique. The first chapter will first provide a general reminder of the principle behind the method, before detailing its application to detrital thermochronology. The second chapter deals with the statistical modeling of the thermal history of samples. The third chapter focuses on a brand-new technique for determining the ^{238}U concentration of a sample using in situ elemental analysis. The fourth chapter develops the (U-Th-(Sm))/He thermochronology method, based on the production of radiogenic He nuclei by the radioactive decay of uranium (^{235}U and ^{238}U), thorium (^{232}Th) and samarium (^{147}Sm). This method can be applied to the same minerals as the fission-track approach (essentially zircon and apatite) and covers very complementary temperature ranges from 50°C to 200°C (Ault et al. 2019; Gautheron and Zeitler 2020). Fission-track thermochronology and (U-Th-(Sm))/He are therefore often used together in geological studies. Finally, the last chapter proposes an application to geodynamics, explaining how fission-track thermochronology has been used to describe the thermal history of the Central Asian and Tibetan ranges in the context of the collision between the Indian and Asian continents.

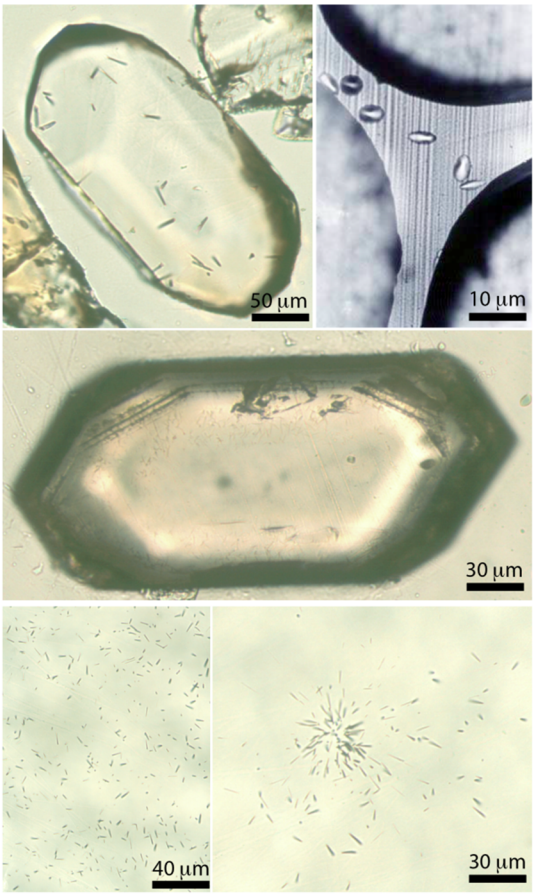

Figure I.4. *Images of fission tracks in various minerals. From top to bottom and left to right: apatite, glass, zircon (note the very small size and large number of these tracks), white micas (irradiated in contact with apatites) and white micas (irradiated in contact with an apatite containing a zircon whose very high relative uranium content has caused the high density of "star" tracks)*

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1

Introduction to Detrital Apatite and Zircon Fission-track Thermochronology

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Italienische Reise

Noch wunderlicher erschien ich diesem Begleiter, als ich auf allen seichten Stellen, deren der Fluß gar viele trocken läßt, nach Steinchen suchte und die verschiedenen Arten derselben mit mir forttrug. Ich konnte ihm abermals nicht erklären, daß man sich von einer gebirgigen Gegend nicht schneller einen Begriff machen kann, als wenn man die Gesteinsarten untersucht, die in den Bächen herabgeschoben werden, und daß hier auch die Aufgabe sei, durch Trümmer sich eine Vorstellung von jenen ewig klassischen Höhen des Erdaltertums zu verschaffen. Auch war meine Ausbeute aus diesem Flusse reich genug, ich brachte beinahe vierzig Stücke zusammen, welche sich freilich in wenige Rubriken unterordnen ließen.

Johann Wolfgang von Goethe, Palermo, 4 April 1787

1.1. Introduction

The use of detrital apatite and zircon fission-track and (U-Th)/He dating or white mica ^{40}Ar-^{39}Ar dating of modern river and beach sediments or

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ancient sandstone is a common approach in sediment provenance, source rock exhumation and basin analysis studies. In this chapter, the focus is on detrital apatite and zircon fission-track dating. Both techniques are considered as low-temperature thermochronology dating techniques, with temperature sensitivities in the range of ~ 250–180°C for zircon and ~ 130–80°C for apatite in comparison to high-temperature dating techniques such as zircon or monazite U-Pb dating (Figure 1.1).

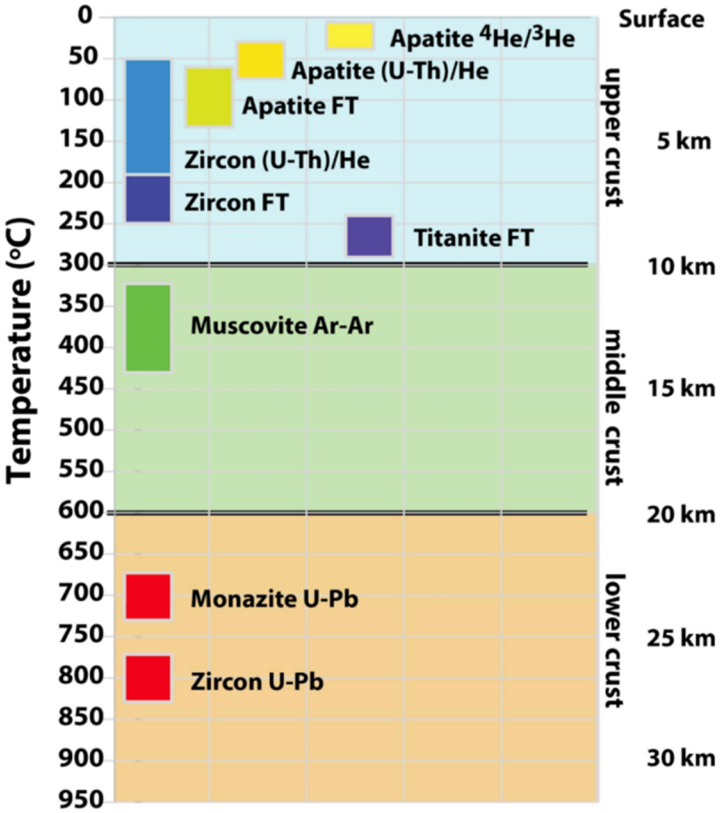

Figure 1.1. *Overview of the temperature sensitivity range of selected isotopic dating techniques. For a color version of this figure, see www.iste.co.uk/jolivet/fission.zip*

Apatite (density of 3.1–3.3 g/cm^3) and zircon (density of 4.5–4.6 g/cm^3) are heavy minerals in contrast to the density of quartz (2.65 g/cm^3). Despite the highly variable apatite and zircon fertility of many upper crustal plutonic, volcanic and metamorphic rocks, both apatite and zircon are relatively common accessory minerals in many sand-sized clastic sediments and sedimentary rocks (Malusà and Garzanti 2019). Although apatite is susceptible to dissolution in acid depositional environments (e.g. bogs) or soils, and abrasion during fluvial transport, zircon is considered as ultra-stable, as long as the grains have not accumulated too much α-radiation damage and are metamict (Malusà and Garzanti 2019; Malusà and Fitzgerald 2020). Both minerals are chemically stable under diagenetic conditions during burial in sedimentary basins or during basin inversion and exhumation.

Since the first detrital zircon fission-track analysis studies in the 1980s (e.g. Hurford et al. 1984; Cerveny et al. 1988), detrital apatite and zircon fission-track dating have developed into standard techniques that have been applied successfully to many different geodynamic settings, for example, the European Alps, the Himalaya, the Tibetan plateau, the Andes, the Southern Alps of New Zealand, or in Alaska, for quantifying exhumation or erosion rates, determining the timing of tectonic events, or the thermal history of sedimentary basins (see examples and references below). Today, it is possible to combine fission-track dating with U-Pb dating and even with (U-Th)/He dating for double and triple dating of single grains, as well as chemical analyses such as Sr isotopes in apatites and Lu and Hf isotopes in zircon. The combination of different techniques on single grains may provide additional valuable information for constraining more precisely sediment provenance and source rock or basin thermal histories.

This chapter provides an introduction to (A) the basics of detrital fission-track analysis using the external detector method (EDM), (B) the underlying statistics for age calculations, data interpretation and the evaluation of detrital grain age distributions, and (C) applications of detrital thermochronology with some examples.

1.2. Principals of fission-track dating

1.2.1. *Basics of single grain apatite and zircon fission-track analysis*

Fission-track dating has been developed in the 1960s by Robert L. Fleischer, P. Burford Price and Robert M. Walker, three physicists of General Electrics in Schenectady, New York, USA, who worked on nuclear defects in solids. The formation of a fission track by spontaneous fission of ^{238}U isotopes is in fact a very rare event as radioactive ^{238}U isotopes normally decay by a series of α and β-decay steps to stable ^{206}Pb (Figure 1.2(A)). The regular ^{238}U to ^{206}Pb decay is about 2 million times more common than the spontaneous fission decay of ^{238}U into two isotopes of different mass that recoil from each other and leave a damage zone in the crystal structure, which is called a latent track (Table 1.1; Figure 1.2(B)) (Fleischer et al. 1975; Wagner and Van den haute 1992). Early on Fleischer, Price and Walker realized that the formation of latent damage zones by spontaneous fission of ^{238}U isotopes in crystals of U-bearing minerals such as apatite, zircon or titanite could be of geological interest (e.g. Price and Walker 1962; Fleischer and Price 1964; Fleischer et al. 1975). The isotopes formed during the spontaneous fission events are not always the same. In fact, two of over 690 different possible isotopes, one heavier isotope with a mass number of up to 172 and a lighter isotope with a mass number as low as 66, are formed. Most of these newly formed isotopes are also radioactive and will decay over seconds, hours, days, months or years, but without causing additional fission damage. The formation of spontaneous fission-track damage in a U-bearing crystal follows a decay constant, just like regular α-decay, and the fission tracks that are formed because of the spontaneous fission decay of ^{238}U are regarded as the daughter products of this decay event (Table 1.1). Given the isotopic abundance and the spontaneous fission-decay constant of ^{238}U isotopes with respect to ^{235}U, ^{234}U and ^{232}Th, only ^{238}U is considered as important for fission-track dating, and the contributions of spontaneous fission tracks from ^{235}U, ^{234}U and ^{232}Th spontaneous fission are negligible. In dependence of U content, fission tracks accumulate in crystals over time and will be at least partially preserved, if the ambient temperatures are below the so-called closure temperature (*Tc*).

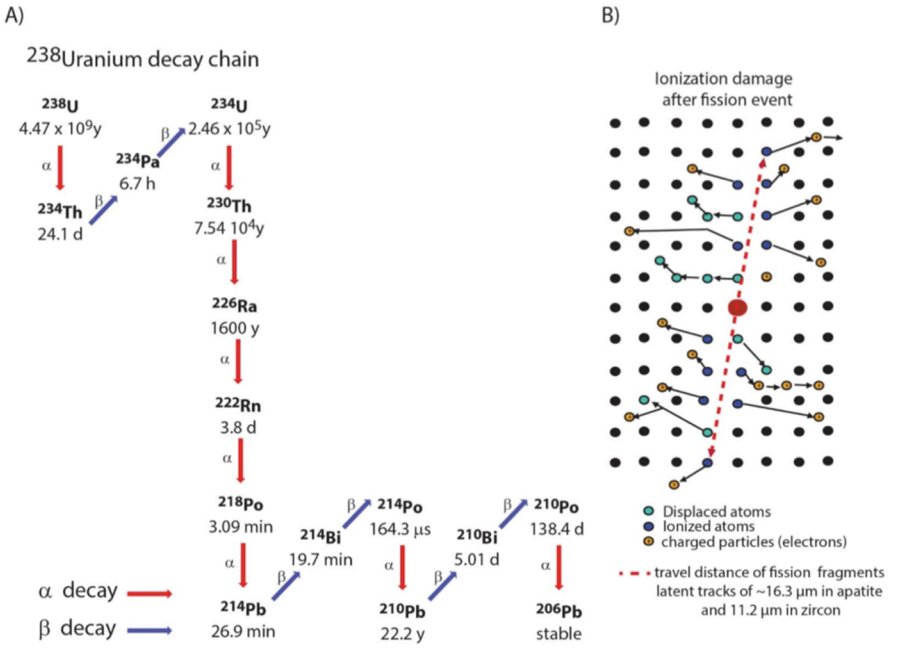

Figure 1.2. *(A) ^{238}U-^{206}Pb decay chain, with a series of eight α-decay steps and six β-decay steps and their half-lives. (B) Spontaneous fission of ^{238}U and formation of latent fission tracks. For a color version of this figure, see www.iste.co.uk/jolivet/fission.zip*

Isotope	Abundance (%)	Decay Constant (year^{-1})	Half-life (year)	Thermal neutron capture cross-section (10^{-24} cm^2)
^{238}U	99.2743	(α) 1.55×10^{-10} (s.f.) $\sim 7.5 \times 10^{-17}$	(α) 4.47×10^{9} (s.f.) $\sim 1.3 \times 10^{16}$	2.7
^{235}U	0.7200	9.85×10^{-10}	7.04×10^{8}	580
^{234}U	0.0057	2.81×10^{-6}	2.46×10^{5}	100
^{232}Th	100.0000	4.92×10^{-11}	1.41×10^{10}	7.4

Table 1.1. *Decay constants and half-lives of U and Th isotopes. Note: α for α-decay, s.f. for spontaneous fission decay (from Wagner and Van den haute (1992); Donelick et al. (2005))*

1.2.2. *Closure temperature concept*

The *Tc* is the temperature at which an isotopic system closes to the loss of daughter products (Dodson 1973). In case of fission-track dating, it means that fission tracks are preserved and not lost to total annealing, which occurs at elevated temperatures above the closure temperature. The *Tc* of the apatite and zircon fission-track dating systems, and also other thermochronological systems such as (U-Th)/He or $^{40}Ar/^{39}Ar$ dating, depends primarily on cooling rate (Figure 1.3(A)). Equation [1.1] shows that the *Tc* is controlled by diffusion and can be calculated as follows:

$$T_c = \frac{E_a}{R\ln\left(A\tau\frac{D_0}{a^2}\right)} \qquad [1.1]$$

where *Ea* is the activation energy, *R* is the gas constant, *A* is the cylinder shape of the crystal, τ is the characteristic time taken for the diffusivity to decrease by a factor *e* and Do/a^2 is the diffusion. Because the calculation of τ also requires a *Tc* value (see equation [1.2]), it is necessary to find an iterative solution for *Tc*. Table 1.2 shows commonly used values of activation energy and diffusion parameters for apatite and zircon fission-track dating determined by laboratory diffusion experiments (see summary in Wagner and Van den haute (1992); Reiners and Brandon (2006)).

$$\tau = -\frac{RT_c^{\,2}}{E_a\frac{\partial T}{\partial t}} \qquad [1.2]$$

Equation [1.2] also shows why the cooling rate, the change of temperature (δT) over time (δt), has an important influence on the *Tc*. The parameter τ is linked to diffusion, as that annealing of fission tracks, which will be discussed in more detail below, is controlled by the diffusion of atoms and electrons that were displaced in the crystal structure during the spontaneous fission event, back into place to repair or anneal the damaged area (latent track) of the crystal. The diffusion ($D_{(t)}$) equation is shown in equation [1.3]:

$$D_{(t)} = D_{(0)}e^{-t/\tau} \qquad [1.3]$$

where $D_{(0)}$ is the diffusivity constant, *t* is the time and τ is again the characteristic time given in equation [1.2]. The graphic in Figure 1.3(B) relates

the number of spontaneous tracks Ns counted on an internal crystal surface today to the time when the crystal apparently cooled below the effective *Tc* and the fission-track dating system changed from an open system, where fission track was rapidly lost to annealing, to a closed system, where all newly formed fission tracks were preserved in the crystal at lower temperatures (Braun et al. 2006). We must keep in mind that the *Tc* concept should only be applied for interpreting the thermochronological data of rocks that experienced relatively rapid (≥ 10–15°C/Myr) monotonic cooling. For detrital grains collected from modern or ancient clastic sediments, this assumption may only be justified for the youngest grain age populations (see below). Furthermore, for apatite crystals, the chemical composition of the apatites has an influence on the *Tc* and the annealing of fission tracks, as F-apatites tend to have a lower *Tc* than Cl-apatites (Figure 1.3(B)) (Donelick et al. 2005; Reiners and Brandon 2006). For zircons, it is rather the amount of accumulated α-radiation damage from regular ^{238}U, ^{235}U and ^{232}Th α-decay (α-recoil damage) that will have an influence on the *Tc*. It is important to distinguish between zero-damage zircons and α-radiation damaged zircons. Zero-damage zircons have not accumulated significant amounts of α-radiation damage before cooling below the zircon fission-track *Tc*. Such zircons must cool very rapidly from crystallization or high-grade metamorphic temperatures below the zircon fission-track *Tc* (Rahn et al. 2004; Reiners and Brandon 2006). With certain exceptions of modern river sediments derived from zones of active volcanism, or rapidly exhuming orogenic mountain belts such as in the Himalayan syntaxes, Taiwan, or the Southern Alps of New Zealand, most detrital zircons will be considered as α-radiation damaged, as only a few million years and average U concentrations are needed to damage the crystals enough to no longer be considered as zero-damage grains. α-Radiation-damaged zircons tend to have *Tc*s about 100°C lower than zero-damage zircons (Figure 1.3(A)). We can see that the zircons have accumulated α-radiation damage through their change in color from clear transparent crystals when they have no or very little radiation damage, to yellow, pink or violet-red colors when there is an increasing amount of α-radiation damage. Similar to fission tracks, α-radiation damage can be annealed, but at higher temperatures than fission tracks (Garver and Kamp 2002). Using Raman spectroscopy, it is possible to determine the impact of α-radiation damage on the crystal structure of zircons (e.g. Guenthner et al. 2013, 2017).