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Spontaneous fission decay constant of ^{238}U: measured by the fission track technique

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Abstract. The spontaneous fission decay constant for ^{238}U has been measured with the fission track technique, by the use of polycarbonate plastic and uranium metal foils. The experimental result obtained by this method is the following: $\lambda_f = (8.23 \pm 0.43) \times 10^{-17}\,\mathrm{yr}^{-1}$, which is in agreement with the majority of values found by means of this and other methods. The experimental technique is described and possible significant error sources are discussed.

Introduction

The spontaneous fission decay constant of ^{238}U was determined for the first time by Flerov and Petrzhak (1940). They measured the spontaneous half life for this uranium isotope, and found a value of ten to the sixteenth or seventeenth power (years). Since that time this constant has been remeasured by various research groups because of its importance in such diverse fields as geochronology, reactor physics etc.

In the age equation for the determination of mineral ages (crystals, glasses, meteorites etc.) all constants and parameters are known at least with 10% accuracy, and the only constant that yields an error of more than 10% is the spontaneous fission decay constant of ^{238}U. Consequently we decided to measure this constant as accurately as possible, i.e. to collect a great number of spontaneous tracks and produce a sufficient number of induced tracks by means of irradiation, and to count both of them exactly. On the other hand the thermal neutron flux in the reactor must also be known precisely.

Different authors have used diverse experimental techniques to measure λ_f, such as ionization chamber, spinner chamber, fission track method etc., the results of which are summarized in this paper (Table 2). A more complete table can be found in the work of Thiel and Herr (1976). These values are different, ranging from $(5.3 \pm 0.8) \times 10^{-17}\,\mathrm{yr}^{-1}$ measured by Perfilov (1947) to $(11.9 \pm 1.0) \times 10^{-17}\,\mathrm{yr}^{-1}$ by Gerling et al (1959). Today in the fission track dating the data 6.8–$6.9 \times 10^{-17}\,\mathrm{yr}^{-1}$ by Fleischer and Price (1964), Khan and Durrani (1973), and the values 8.3–$8.6 \times 10^{-17}\,\mathrm{yr}^{-1}$ by Segre (1952), Spadavecchia and Hahn (1967), Galliker et al. (1970), Thiel and Herr (1976) are mainly used.

The aim of this experimental work is to measure λ_f with the fission track technique, by means of plastic and natural uranium metallic foils, and compare the results with those obtained by other methods.

Experimental technique

For the purpose of measuring λ_f eight sheets of polycarbonate-plastic foils (Makrofol-E, from Bayer A.G., W. Germany) of dimensions $120 \times 15 \times 0.1$ mm were brought in close contact with four natural uranium metallic foils (from AERE, Harwell, U.K.). The four uranium foils had the same length and width as the plastic foils but a thickness of 0.18 mm. For each uranium foil we used two plastic foils, one on each side, and made a sandwich of plastic and uranium foils. Between each set of one uranium and two plastic foils a thick plexiglass sheet of the same size, is used to separate one set from the other. To prevent the oxidation of uranium foils, the whole assembly was put in an evacuated glass tube, and stored in a cool place of enviromental temperature of 5°–10°C, to avoid the annealing of tracks in plastic foils due to temperature. The time of exposure for collecting the spontaneous tracks was 426 days (6.134×10^5 minutes).

For the production of induced tracks we used four plastic foils of 11 mm diameter and put them in close contact with two natural uranium metallic foils of the same size. The two sets were positioned inside an aluminium capsule, which has 6 cm length. Between each set of two plastic and one uranium foils a thick plexiglass disk is also used. The capsule was irradiated at ASTRA-swimming pool reactor at Seibersdorf, Austria, with thermal neutron doses of $(5.57 \pm 0.21) \times 10^9$ n/cm^2 and $(5.38 \pm 0.20) \times 10^9$ n/cm^2 for each of the two sets. The thermal neutron dose determination was carried out as accurately as possible, because a reliable value for integrated neutron flux is essential for this determination. For accurate flux determination Au-monitors are positioned in the vicinity of plastic-uranium sandwiches. The irradiation time was only 22 seconds at 8 MW reactor power. After the exposures for both spontaneous and induced tracks the plastics were separated from uranium foils, and etched in (6N) NaOH for 25 minutes at 60°C. Afterwards the plastics were glued with glycerine on micro slides, and finally the tracks were counted with a Leitz Orthoplan light microscope at a magnification of 787.5× (ocular: 12.5×, objective: 63×).

Calculations and results

The results obtained from the counting of tracks in plastic foils are summarized in Table 1. We counted the tracks in 10240 fields of view of the spontaneous samples and obtained a total number of 1371 tracks of this kind. On

Table 1.
Experimentally obtained track densities for spontaneous p_s and induced tracks p_i. For induced tracks there exist two different thermal neutron doses. In the last column is given statistical weighting factor g_i.

No. of samples	Absolute number of counted tracks P_s	P_i	Track density (in cm^{-2}) $p_s(\times 10^2)$	$p_i(\times 10^4)$	Ratio p_s/p_i $(\times 10^{-2})$	Thermal neutron dose (n) $(\times 10^9$ cm$^{-2})$	$(p_s/p_i)\cdot n$ $(\times 10^7$ cm$^{-2})$	Statistical weighting factor g_i
VL-1	318	2116	(4.73 ± 0.26)	(13.11 ± 0.28)	(0.361 ± 0.021)	(5.57 ± 0.21)	(2.012 ± 0.140)	0.275
VL-2	344	2390	(5.08 ± 0.27)	(14.81 ± 0.31)	(0.343 ± 0.020)	(5.57 ± 0.21)	(1.911 ± 0.131)	0.309
VL-3	367	1386	(4.86 ± 0.25)	(8.59 ± 0.23)	(0.566 ± 0.033)	(5.38 ± 0.20)	(3.047 ± 0.211)	0.198
VL-4	342	1582	(4.47 ± 0.24)	(9.80 ± 0.25)	(0.456 ± 0.027)	(5.38 ± 0.20)	(2.455 ± 0.173)	0.218

the other hand counting the tracks of 120 fields of view of the induced samples yielded a total result of 7474 induced tracks.

The formula applied for this calculation is the simplified age relationship. After Wagner (1969) for ages less than 10^8 years, it follows:

$$t = \frac{p_s}{p_i} \frac{n\sigma_f I}{\lambda_f} \tag{1}$$

In this formula t is the age of sample (in this specific case the time duration for collecting spontaneous tracks in years), p_s and p_i are the spontaneous and induced track density (tracks/cm^2), n is the thermal neutron dose (n/cm^2), σ_f is the cross section for fission of ^{235}U by thermal neutrons equal to (580.2 ± 1.8) barn, Hanna et al. (1969), and I the natural isotopic abundance ratio of $^{235}U/^{238}U$ equal to $(7.259 \pm 0.104) \times 10^{-3}$ De Wet et al. (1968).

By substituting the weighted average value:

$$\overline{\left(\frac{p_s}{p_i} \cdot n\right)} = \frac{\sum_{i=1}^{4} g_i \left(\frac{p_s}{p_i} \cdot n\right)_i}{\sum_{i=1}^{4} g_i} = (2.282 \pm 0.079) \times 10^7 \text{ cm}^{-2} \tag{2}$$

and other constants given above, namely σ_f, I and the measured value of t in

Table 2
Values for λ_f measured by different authors. The results indicated by asterisk are corresponding within statistical error with the present work.

Author and year	Technique applied	$\lambda_f (\times 10^{-17}\, yr^{-1})$
Perfilov (1947)	track in photographic emulsion	(5.3 ± 0.8)
Segre (1952)	ionization chamber	(8.60 ± 0.29) (*)
Parker and Kuroda (1958)	$^{99}Mo/^{238}U$ equilibrium ratio	(8.7 ± 0.5) (*)
Gerling et al. (1959)	radiochemical analysis	(11.9 ± 1.0)
Fleischer and Price (1964)	fission track method	(6.9 ± 0.2)
Rao and Kuroda (1966)	$^{132}Te/^{238}U$ equilibrium ratio	(7.8 ± 0.9) (*)
Spadavecchia and Hahn (1967)	spinner chamber	(8.42 ± 0.10) (*)
Roberts et al. (1968)	fission track method	(7.03 ± 0.11)
Von Gunten (1969)	fission product from ^{238}U	(8.66 ± 0.22) (*)
Galliker et al. (1970)	spinner chamber	(8.46 ± 0.06) (*)
Kleeman and Lovering (1971)	fission track method	(6.8 ± 0.6)
Khan and Durrani (1973)	fission track method	(6.82 ± 0.55)
Wagner et al. (1975)	fission track method	(8.7 ± 0.6) (*)
Thiel and Herr (1976)	fission track method	(8.57 ± 0.42) (*)
Märk et al. (1977)	fission track method	(10.1 ± 0.6)
Halder et al. (1981)	fission track method	(8.6 ± 0.4) (*)
present work (1983)	fission track method	(8.23 ± 0.43) (*)

equation (1), it follows for the spontaneous fission decay constant of ^{238}U:

$$\lambda_f = (8.23 \pm 0.43) \times 10^{-17}\ \mathrm{yr}^{-1}$$

This result is in good agreement, within statistical and experimental errors with most other results obtained by other or the same method as shown in Table 2.

The spontaneous fission half life of ^{238}U calculated from the numerical value obtained in this work is the following:

$$T_{1/2}(\mathrm{s.f.}) = (8.42 \pm 0.44) \times 10^{15}\ \mathrm{yr}$$

Discussion

In this work, the main sources of error consist in the counting of p_s and p_i, and the resultant error of their weighted ratio: $\overline{(p_s/p_i)} = (0.417 \pm 0.012) \times 10^{-2}$ which is about 3%. This error is relatively small, because a high number of tracks has been counted. Another significant source of error is due to the measurement of thermal neutron dose n, which was measured as accurately as experimentally possible (Table 1), and amounts to about 3.8%. The combined error of p_s/p_i and n are given in equation (2) which is about 3.5%. Together with other sources of error (σ_f and I) the total error of these results amounts to about 5%. In conclusion the result presented in this work confirms the data found by most of authors, which are given by asterisk in Table 2.

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