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Spontaneous-fission decay constant of ^{238}U measured by nuclear track techniques without neutron irradiation

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A value of the decay constant for spontaneous fission of ^{238}U, λ_f, determined by particle-track detector techniques, is presented. The main source of systematic error in fission-track measurements of λ_f, the so called "neutron dosimetry", has been avoided. Instead of calibrating the neutron source and the usual mica detector through tracks of ^{235}U fission induced by thermal neutrons, spontaneous-fission tracks and alpha-particle tracks of ^{238}U and ^{242}Pu were used. A value of $\lambda_f = (8.66\pm0.38)\cdot10^{-17}$ y^{-1} has been obtained.

Introduction

Besides measuring track densities of ^{238}U spontaneous fission in the mineral sample itself and track density on an external detector coming from ^{235}U fission induced in the sample by thermal neutrons, in Fission-Track Dating (FTD), it is also needed to measure fission track density on a detector (muscovite mica), which had been coupled usually to a so called "dosimeter", normally a U-doped reference glass. In most laboratories, the calibration of the dosimeter is performed through the ζ-calibration method.[1,2] To a standard sample, whose age is known by other dating methods, the fission-track age equation is applied, solved for ζ a parameter including physical constants and the information about the thermal neutron fluence. This procedure has been recommended in 1990, by the Subcommission on Geochronology of the International Union of Geological Sciences,[3] owing to inaccuracy in neutron fluence metrology. By that time, neutron fluence determinations had been carried out through Au and Co pills or foils as activation monitors, which were considered as the principal source of systematic errors in both the FTD and the determinations of λ_f, the decay constant for spontaneous fission of ^{238}U. The first determination of λ_f by FT measurement[4] resulted in a value of $(6.85\pm0.20)\cdot10^{-17}$ y^{-1}. Most of the subsequent FT determinations have found values around $7\cdot10^{-17}$ y^{-1}, wich remarkably differs from most of the values found by other methods, $\sim8.5\cdot10^{-17}$ y^{-1}. More comprehensive discussions on the fission decay constant can be found elsewhere.[5–7]

Even though the use of ζ-calibration to avoid the need of explicitly knowing the thermal neutron fluence ϕ and λ_f, it makes the FTD dependent on other dating methods.

In order to make FTD an independent and reliable dating method, two conditions have to be fulfilled: (1) to elaborate an accurate (rather than precise) neutron metrology and (2) to attain a consensual value of λ_f, preferably, to determine it by particle-track techniques, without neutron irradiation. The first point has been approached by some researchers, improving the metal activation techniques[8,9] or developing alternative techniques as the use of the ^{235}U(n,f) reaction through thin films.[10,11] This work aims to contribute to the latter (2) point by developing a novel λ_f determination. Thermal neutron irradiation is replaced by a ^{242}Pu-doped fission source for the purpose of calibrating the fission-track detector, wich is a muscovite mica sheet. Just as in the case of FTD, it is supposed that the ranges of fission fragments are the same, independently of whether they are originated from ^{242}Pu or ^{238}U atoms. The value of λ_f will be obtained using the well determined value of the decay constant for spontaneous fission of ^{242}Pu and the measured track density ratio of spontaneous fission of ^{242}Pu and ^{238}U. The ratio of atomic numbers in a unit volume of ^{242}Pu and ^{238}U is determined with the help of the alpha-particle track density ratio of the same isotopes. This procedure provides a new evidence to help deciding between the λ_f values quoted above. The general considerations and the detailed description of the method were previously published in Reference 12.

Method

Suppose two thick fission-fragment sources, i.e., the thickness is greater than the fission-fragment range. The bulk material is the U_3O_8 (uranium in natural isotopic composition), but one of them is spiked with ^{242}Pu.

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If muscovite mica sheets are coupled with each of the sources for suitable exposure time, the track densities due to spontaneous fissions can be obtained after etching, and measured under an optical microscope. They are given by:

$$\left(\frac{\rho_S}{\Delta t_S}\right)=\frac{R_f}{2}\varepsilon_f C_{238} N_U \lambda_f \quad (1)$$

and

$$\left(\frac{\rho_{S242}}{\Delta t_{S242}}\right)=\frac{R_f}{2}\varepsilon_f\left(N_{242}\lambda_{f242}+C_{238}N'_U\lambda_f\right) \quad (2)$$

In the above equations, ε_f is the detection efficiency of muscovite mica for fission fragments, C_{238} is the isotopic concentration of ^{238}U in natural U, λ_{f242} stands for the decay constant for spontaneous fission of the ^{242}Pu, N_{242} is the number of ^{242}Pu and N'_U is the number of U atoms per unity volume in the spiked source. N_U is the number of U atoms in the non-doped source. The symbols ρ_{s242} and ρ_s represent the fission-track densities obtained on detectors coupled with the doped and with the non-doped source. Finally, Δt_{s242} and Δt_s are for the exposure times of the detectors to spiked and non-spiked sources, respectively.

The ratio of track densities, when solved for λ_f, is:[12]

$$\lambda_f=\frac{\lambda_{f242}N_{242}}{C_{238}N_U}\left[\frac{(\rho_{S242}/\Delta t_{S242})}{(\rho_S/\Delta t_S)}-\frac{N'_U}{N_U}\right]^{-1} \quad (3)$$

If the U_3O_8 disks are thick enough compared to the fission-fragment range and manufactured under the same conditions, it can be stated that the number of atoms per unity volume will be the same in doped (uranium and plutonium atoms) and non-doped (uranium atoms) sources or mathematically:

$$N_U=N_{242}+N'_U \quad (4)$$

However, it is more convenient to set the following constraint:

$$\frac{N_{242}}{N_U}<<1\Rightarrow\frac{N'_U}{N_U}\approx 1 \quad (5)$$

In this way, besides λ_f, only the ratio N_{242}/N_U remains unknown in Eq. (3). It can be determined by coupling alpha particle track detectors, CR-39, with the same doped and non-doped sources for known exposure times. After etching, the alpha track densities can be determined under an optical microscope and are given by:

$$\left(\frac{\rho_\alpha}{\Delta t_\alpha}\right)=\frac{1}{4}\varepsilon_\alpha N_U\sum_{i=234,235,238}R_i C_i\lambda_{\alpha i} \quad (6)$$

and

$$\left(\frac{\rho_{\alpha242}}{\Delta t_{\alpha242}}\right)=\frac{1}{4}\varepsilon_\alpha\left(N_{242}R_{242}\lambda_{\alpha242}+N'_U\sum_{i=234,235,238}R_i C_i\lambda_{\alpha i}\right) \quad (7)$$

where ε_α is the detection efficiency of CR-39 for alpha-particles, R_{242} is the mean range of the ^{242}Pu alpha-particles and R_i is the mean range of the ^{i}U alpha-particles in the U_3O_8, C_i is the isotopic composition of the ^{i}U in natural uranium, $\lambda_{\alpha242}$ and $\lambda_{\alpha i}$ are the decay constants for alpha-decay of the ^{242}Pu and ^{i}U. The $\rho_{\alpha242}$ and ρ_α are the alpha-track densities on the CR-39 detector surface coupled with the spiked and with the the non-spiked sources. The $\Delta t_{\alpha242}$ and Δt_α values are the exposure times of the alpha-particle detectors to the doped and non-doped sources, respectively. The ratio of Eqs (6) and (7) is:

$$\frac{N_{242}}{N_U}=\frac{\left(\sum_{i=235,235,238}R_i C_i\lambda_{\alpha i}\right)}{R_{242}\lambda_{\alpha242}}\left[\frac{(\rho_{\alpha242}/\Delta t_{\alpha242})}{(\rho_\alpha/\Delta t_\alpha)}-1\right] \quad (8)$$

For convenience, the dimensionless quantity $r_{242}=R_{242}/R_U$ is introduced, with R_U defined as:

$$R_U=\frac{\sum_{i=234,235,238}R_i C_i\lambda_{\alpha i}}{\sum_{i=234,235,238}C_i\lambda_{\alpha i}} \quad (9)$$

Using these definitions, Eq. (8) becomes:

$$\frac{N_{242}}{N_U}=\frac{\left(\sum_{i=235,235,238}C_i\lambda_{\alpha i}\right)}{r_{242}\lambda_{\alpha242}}\left[\frac{(\rho_{\alpha242}/\Delta t_{\alpha242})}{(\rho_\alpha/\Delta t_\alpha)}-1\right] \quad (10)$$

The above equation is valid if the ^{242}Pu is the only spiking isotope. However, it will be shown in the next section, that the solution used for preparing the spiked sources contained not only ^{242}Pu, but ^{239}Pu, ^{241}Am and ^{241}Pu. The latter three isotopes do not contribute with fission tracks, but ^{239}Pu and ^{241}Am are very active alpha-emitters. To take into account their contribution, Eq. (10) has to be modified to:

$$\frac{N_{242}}{N_U}=\frac{\left(\sum_{i=235,235,238}C_i\lambda_{\alpha i}\right)}{\lambda_{\alpha242}\sum_{j=trans}k_j r_j}\left[\frac{(\rho_{\alpha T}/\Delta t_{\alpha T})}{(\rho_\alpha/\Delta t_\alpha)}-1\right] \quad (11)$$

where k_j is the ratio between the alpha-activity of the isotope j (j standing for ^{239}Pu, ^{241}Am and ^{242}Pu) and the ^{242}Pu one and r_j is the ratio R_j/R_U. The quantity k_j has been defined in order to couple the alpha-activity of the transuranic isotopes present in the spiked sources with the ^{242}Pu one. As it will be shown next, the k_j values can be determined by alpha-spectrometry. The subindex "242" has been replaced by "T" on the symbols for alpha-track density and exposure time to take into account that other transuranic isotopes are also contributing when CR-39 detectors are coupled with spiked sources.

By means of Eqs (3) and (11), λ_f follows from fission track counting on muscovite mica and alpha-track counting on CR-39 detector sheets.

Experimental

Four non-spiked uranium disks, numbered 62, 64, 65 and 67, were manufactured. Following the same procedure,[12] two Pu-spiked uranium sources (P1-I and P1-II) were manufactured using a nitrate solution containing ^{241}Am, ^{239}Pu, ^{241}Pu and ^{242}Pu. The relative alpha-activities of these isotopes in the solution, determined by alpha-spectrometry (Fig. 1), as well as their physical properties are given in Table 1. Deviations in k_j values are inferred by multi-peak fitting of the histogram in Fig. 1b. The sources are composed of U_3O_8 powder deposited on cylindric aluminum plates (2 cm in diameter and 2 mm in high). The thickness of the deposited powder is around or greater than 14 mg/cm^2 (see the second column of Tables 2 and 3). In Reference 12, it is shown that if the thickness of the manufactured source is higher than 10 mg/cm^2 , it can be considered as infinite.

In Table 1, R_j values leading to r_j's have been obtained by fitting data taken from Reference 13. The following expression has been obtained:

$$R_j = 0.37109 + 0.81849 E_j + 0.13516 E_j^2 \qquad (12)$$

where E_j is the energy associated to the alpha-particles coming from the isotope j. As seen in Table 1, although the alpha-activity of the former atoms contribute importantly to the total alpha-activity, their spontaneous-fission activity is negligible when compared to that of the ^{242}Pu. The presence of the ^{241}Pu is inferred by assuming that all ^{241}Am in the solution is a product of the ^{241}Pu beta-decay. Actually, the presence of this isotope is innocuous since it does not decay by alpha-particle emission and its amount of spontaneous fission can be neglected.

CR-39 sheets (Pershore, UK) were coupled with the sources for exposure times of $\Delta t_{\alpha T}$ = 1.5 minutes, for the spiked sources, and Δt_{α} = 10.0 minutes for non-spiked sources. After exposure, the detectors were etched in a 6.25N NaOH solution for 400 minutes at 70 °C. Additionally, muscovite mica sheets were coupled with the sources by exposure times of Δt_{ST} = 0.192 y, for the spiked sources and Δt_S = 0.515 y for the non-spiked sources. The micas had been pre-etched to enlarge fossil tracks and make it possible to distinguish the fresh ones. Thus, a background determination is unnecessary. Detectors were then etched at 15 °C for 90 minutes in an HF 48% solution. All muscovite mica sheets were etched together in order to assure the same etching conditions. Track counting was carried out under a Reichert optical microscope with approximately 500× nominal magnification. The background track density of alpha particles was determined to be (50±2) tracks per square centimeter. All the fission tracks were counted, and tipically 10,000 alpha-tracks were counted on every detector coupled with a doped source, and 2,500 to 10,000 alpha-tracks were counted on detectors coupled with non-doped sources to achive good counting statistics.

Results

Doped-source results

Results concerning superficial densities of alpha-particle tracks on CR-39 and of fission fragments on muscovite coupled with spiked sources are shown in Table 2. Density results are given within 1σ of the Poisson statistics.

The fourth column of Table 2 presents the results of N_{242}/N_U computed through Eq. (11). The value of $\rho_\alpha/\Delta t_\alpha$ has been extracted from Reference 12:

$$(\rho_\alpha/\delta t_\alpha) = (1.274 \pm 0.025) \cdot 10^9 \text{ cm}^{-2} \cdot \text{y}^{-1}$$

The values of r_j and k_j are given in Table 1. The needed physical constants are:[14]

$$C_{234} = 0.0054\%;$$
$$C_{235} = 0.720\%;$$
$$C_{238} = 99.2745\%;$$
$$\lambda_{\alpha 234} = 2.283 \cdot 10^{-6} \text{ y}^{-1};$$
$$\lambda_{\alpha 235} = 9.84864 \cdot 10^{-10} \text{ y}^{-1};$$
$$\lambda_{\alpha 238} = 1.55136 \cdot 10^{-10} \text{ y}^{-1};$$
$$\lambda_{\alpha 242} = 1.857 \cdot 10^{-6} \text{ y}^{-1}.$$

The mean weighted value obtained for N_{242}/N_U is $(N_{242}/N_U) = (4.03 \pm 0.28) \cdot 10^{-5}$. The error of N_{242}/N_U was calculated using the appropriate propagation formula. Note that this value fulfils the condition assumed in Eq. (5).

The mean weighted value of $(\rho_{s242}/\Delta t_{s242})$ is $(\rho_{s242}/\Delta t_{s242}) = (2.727 \pm 0.057) \cdot 10^3 \text{ cm}^{-2} \cdot \text{y}^{-1}$.

Table 1. Quantities and physical properties of the isotopes compounding the solution used for doping uranium sources. The last column shows an estimation of the contribution of each isotope compared to the ^{242}Pu contribution for the spontaneous fission activity of the solution

Isotope	Alpha-particle energy, MeV	r_j	k_j	Spontaneous fission activity compared to the ^{242}Pu one
^{241}Am	5.5	1.320 ± 0.013	37.84 ± 0.08	$3\cdot10^{-5}$
^{239}Pu	5.1	1.206 ± 0.009	6.85 ± 0.03	$4\cdot10^{-6}$
^{241}Pu	–	–	–	$4\cdot10^{-8}$
^{242}Pu	4.9	1.123 ± 0.002	1	1

Table 2. Measured and calculated data of the spiked sources

Doped source	Thickness, mg/cm^2	$\rho_{\alpha T}/\Delta t_{\alpha T}$, $\times10^{10}$ cm$^{-2}\cdot$y^{-1}	N_{242}/N_U, $\times10^{-5}$	Number of spontaneous-fission tracks counted on mica	$\rho_{s242}/\Delta t_{s242}$, $\times10^3$ cm$^{-2}\cdot$y^{-1}
I-P1	15.3	1.914 ± 0.037	4.01 ± 0.40	1172	2.758 ± 0.081
II-P1	16.4	1.925 ± 0.016	4.04 ± 0.40	1147	2.699 ± 0.080

Table 3. Measured data of the non-doped sources and calculated λ_f values

U_3O_8 source	Thickness, mg/cm^2	Number of counted tracks	$\rho_s/\Delta t_s$, $\times10^2$ cm$^{-2}\cdot$y^{-1}	λ_f, $\times10^{-17}\cdot$y^{-1}
62	23.1	532	4.66 ± 0.20	8.57 ± 0.76
64	14.1	547	4.79 ± 0.20	8.86 ± 0.79
65	16.8	526	4.61 ± 0.20	8.49 ± 0.75
67	16.2	540	4.73 ± 0.20	8.72 ± 0.78

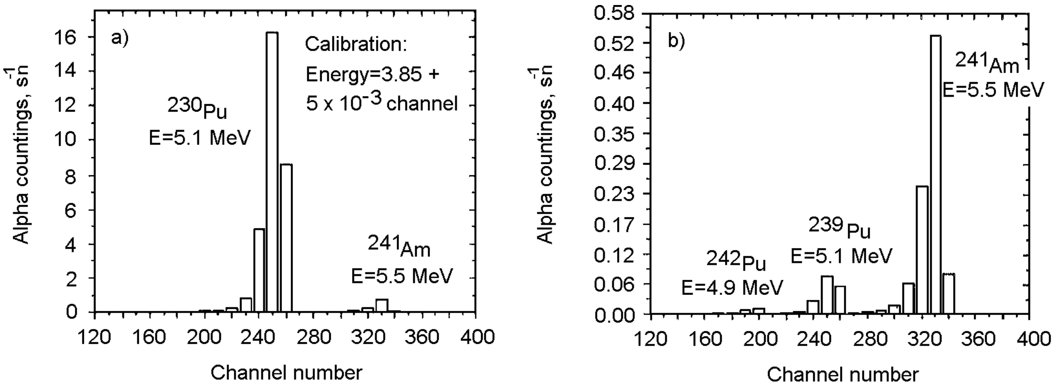

Fig. 1. Alpha-spectrometric analysis of the solution used for manufacturing doped sources. The detector was calibrated (a) through a standard source containing ^{239}Pu and ^{241}Am and (b) the energies of alpha-particles coming from the solution could be determined

Non-doped-source results and calculation of λ_f

Table 3 presents the results concerning the non-spiked sources and computed λ_f values. Track density errors are given within 1σ of the Poisson distribution. λ_f deviations are calculated using the appropriate propagation formula.

For the calculation of λ_f values in the fifth column of Table 3, the value of the decay constant for spontaneous fission of ^{242}Pu used was[7] $\lambda_{f242} = (1.024\pm0.011)\cdot10^{-11}$ y^{-1}. The weighted mean λ_f value is $\lambda_f = (8.66\pm0.38)\cdot10^{-17}$ y^{-1}.

Discussion

The error obtained for the individual values of λ_f is a consequence of the propagation of uncertainties in: (1) the characterization of the solution used for the doped sources, i.e., determination of the k_j's, (2) counting alpha-particle tracks on CR-39, (3) the relative alpha-particle ranges (r_j's), (4) counting of spontaneous-fission tracks on muscovite mica, and (5) physical constants. Each of the identified error sources is discussed in turn. In an additional subsection, (6) general comments, discuss the final obtained value.

Characterization of the solution used for doping sources

An ideal situation, in which the ^{242}Pu were the only doped isotope, could not be achieved in this work. Although it is the only significant fission-fragment source, the other doped isotopes have contributed to the alpha-activity. The presence of ^{241}Pu cannot be directly determined by alpha-spectrometry. Assuming that all ^{241}Am is a decay product, ^{241}Pu would be around 2% of the transuranic elements in the solution. Its estimated spontaneous-fission contribution, showed in the last column of Table 1, is negligible compared to ^{242}Pu.

To overcome this condition, alpha-spectrometric analysis became necessary. Although the alpha energies have been clearly distinguished (Fig. 1b), separation of energy peaks and quantification of relative alpha-activities (k_j's) are accompanied by uncertainties that must be considered. This has been the principal source increasing individual λ_f errors.

Counting of alpha-particle tracks on CR-39

CR-39 track detector sheets have been used for determining the amount of ^{242}Pu relative to uranium atoms in the sources. The U isotopes as well as the other isotopes present in the sources have alpha activities suitable for track counting. This allowed for reduced errors be attained. The overall error of the weighted mean densities is approximately 0.5% for the case of non-doped sources and 0.8% for the case of doped sources.

The value of $(\rho_\alpha/\Delta t_\alpha)$ was taken as the characteristic value of infinite sources.[12] It is worth mentioning that such a value is dependent on counting criteria, which make it observer and etching specific. In addition, it is dependent on the physical features of the sources: density and chemical composition of the bulk material. Therefore, it is manufacture-procedure specific. For that Eq. (11) could be applied, the same observer should have obtained $(\rho_\alpha/\Delta t_\alpha)$ and $(\rho_\alpha/\Delta t_{\alpha T})$ values, the same etching conditions should have been used, the same manufacture procedures should have been applied and the same bulk material should have composed doped and non-doped sources. It has been just the case in this work.

Relative alpha-particle ranges

In order to evaluate N_{242}/N_U, an r_j quantity was introduced. As explained before, it accounts for the range of alpha-particles with a specific energy traveling throughout a specific medium. The use of this ratio instead of the R_j, range, prevents the influence of scale factors, making different information sources comparable. Comparing r_j values for UO_2 found using data from Reference 13 with values found using data from Reference 15, it is noted that differences are less than 1%. In the paper by MATZKE,[15] no data is available on U_3O_8, the material used here and we were not able to find another more recent source than Reference 13. Therefore, the error of the values of r_j have not contributed importantly to the final λ_f error.

Counting of spontaneous-fission tracks on muscovite

Equation (3) presupposes the same detection efficiency of muscovite mica for spontaneous-fission fragments from both ^{238}U and ^{242}Pu. Not as for the alpha-particle case, fission fragments from ^{238}U and ^{242}Pu are expected to present the same mean range. Such hypothesis is similar to that concerning fission track dating, namely same detection efficiency for spontaneous-fission fragments from ^{238}U and induced-fission fragments from ^{235}U. The huge mass, charge and energy of the fission fragments of the isotopes used in this work indicate that the detection efficiency of the muscovite will not vary. Common chemical etching and counting criteria are also required. In this work, detectors coupled with spiked and non-spiked sources were etched together and the same observer performed the track counting.

Normally, due to its long half-life, the low number of ^{238}U spontaneous fission events is an important source of error in λ_f measurements. In this work, four non-doped sources were used to collect tracks of ^{238}U spontaneous-fission fragments. The total number of fission tracks counted was 2145 and the contribution to the total error about 2%. The collecting of fission fragment tracks from doped sources contributed also with ~2%. Two doped sources were used and 2319 fission tracks were counted.

Physical constants

All physical constants needed for the evaluation of λ_f have been well and consensually known, therefore, they are not important sources of error in this report. The highest deviation to be considered comes from λ_{f242}, ~1%.

General comments

For evaluating N_{242}/N_U and λ_f it was assumed that $N_{242} \ll N_U$. Indeed, a value of $(N_{242}/N_U) = (4.03\pm0.28)\cdot10^{-5}$ was obtained, validating the referred hypothesis.

The χ^2 test applied to the individual λ_f values results in $\chi_\nu^2 = 0.15$ (for $\nu = 3$ degrees of freedom), $P_\chi \approx 0.8$. This indicates internal coherence among data within the errors presented.

The error quoted for the final value of λ_f (~4.5%), is the variance of the weighted mean value which follows from the errors of individual values.[16]

Conclusions

Using nuclear track techniques, a λ_f determination for ^{238}U was carried out without neutron irradiation. A value of $(8.66\pm0.38)\cdot10^{-17}$ y^{-1} was obtained. This value is in agreement, within the error, with the value of $(8.5\pm0.1)\cdot10^{-17}$ y^{-1} recommended by Reference 7. However, it is not in agreement with the value $7.0\cdot10^{-17}$ y^{-1} found in most of the fission-track determinations. The result obtained in the present work supports the hypothesis that the major systematic error source in fission-track λ_f determinations is the unsufficient knowledge of the neutron fluence (and energy spectrum).

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