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Determination of the ^{238}U spontaneous fission decay constant without neutron irradiation

S. Guedes,[1]* J. C. Hadler,[1]* P. J. Iunes,[1] A. K. Burke,[1] M. H. Kakazu,[2] J. E. S. Sarkis,[2] S. R. Paulo,[3] C. A. Tello[4]

[1] *Instituto de Física "Gleb Wataghin", Universidade Estadual de Campinas, UNICAMP, 13083-970 Campinas, SP, Brazil*
[2] *Divisão de Caracterização Química, Instituto de Pesquisas Energéticas e Nucleares, IPEN/CNEN, 05508-900 São Paulo, SP, Brazil*
[3] *Departamento de Física, Instituto de Ciências Exatas e da Terra, Universidade Federal de Mato Grosso, UFMT, 78060-900, Cuiaba, MT, Brazil*
[4] *Instituto de Geociências e Ciências Exatas, Universidade Estadual Paulista, UNESP, 13506- 900, Rio Claro, SP, Brazil*

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We have developed a methodology for measuring the decay constant of the spontaneous fission of ^{238}U, λ_f, using nuclear particle track detectors where thermal neutron irradiation is unnecessary. This methodology is based on the fact that the radiation damage caused by spontaneous fission of trans-uranium elements bearing a mass number close to 238 are similar to ^{238}U spontaneous-fission ones. Loading a thick source of uranium (thickness greater than the fission fragment range) with a small amount of a suitable trans-uranium element (for instance, ^{242}Pu, which presents a spontaneous fission half-life of $6.75 \cdot 10^{10}$ y), it is possible to determine the observation efficiency of a particle-track detector for fission fragments. Procedures concerning our thick source manufacture and uniformity tests of the trans-uranium distribution are also presented. These results make it possible for the exposure of thick uranium sources (without trans-uranium element) to lead to a λ_f value.

Introduction

The decay constant for spontaneous fission of ^{238}U, λ_f, is a key parameter in the fission-track-age equation which was presented in a simplified form by FLEISCHER and PRICE:[4]

$$T = \left(\frac{\rho_S}{\rho_I} \right) \frac{n \sigma I}{\lambda_f} \qquad (1)$$

where ρ_S and ρ_I are the superficial densities of spontaneous and induced-fission tracks, respectively, that can be measured on a suitable fission-track detector or on the sample to be dated, after suitable chemical etching, T is the exposure time or the sample age, n is the fluence of thermal neutrons, σ the cross section for thermal fission of ^{235}U, and $I = C_{235}/C_{238}$ the isotopic ratio between ^{235}U and ^{238}U in natural uranium.

Many λ_f measurements have been made by fission-track workers since the FLEISCHER and PRICE one,[4] the majority of these have been grouped around $7.0 \cdot 10^{-17}$ y^{-1}. However, when all techniques are considered, the λ_f results have been placed around two central values: $7.0 \cdot 10^{-17}$ y^{-1},[4,8,12–14] and $8.5 \cdot 10^{-17}$ y^{-1},[3,6,14,15] which present a disagreement of about 20%. Fission-track results were obtained by applying Eq. (1) to the uranium samples or to artifacts whose ages were known a priori. This procedure involves neutron dosimetry. Neutron-fluence measurements were carried out using metal-activation monitors (or uranium-doped glasses calibrated by metal activation) (for instance, References 8 and 16, or directly by U (n, fission) reaction.[3,8,16] The determination of neutron fluences has been pointed out as the main source of systematic error in fission-track dating and λ_f values obtained with this method.[1,10] In order to overcome difficulties arising from neutron-dose determination, fission-track researchers have avoided the use of a λ_f value along with an absolute neutron-dose measurement in dating geological samples. Two calibration systems have been adopted: (1) use of a λ_f value obtained under the same irradiation conditions and measurement procedures to be used in dating and (2) irradiation of a standard sample, whose age is well known by means of another radiometric method of dating, together with the sample to be dated. This last technique, the ζ-calibration, was first outlined by FLEISHER and HART[5] and has been the most used procedure in dating geological mineral samples.

Thermal neutron irradiation yields fission tracks through ^{235}U induced-fission and makes it possible to determine the detector efficiency multiplied by the uranium content. Here, we have implicitly assumed the same detection efficiency for both ^{235}U and ^{238}U fission fragments. Such a hypothesis is reasonable since the mass number and energy distributions of fission fragments are very similar in ^{238}U spontaneous and ^{235}U induced-fission processes. On the other hand, the same mass and energy distributions are observed in the spontaneous-fission-decay process of trans-uranium isotopes.[2] Therefore, trans-uranium elements could be used as fission-fragment sources for obtaining the

* E-mail: guedes@ifi.unicamp.br; hadler@ifi.unicamp.br

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detector efficiency. In this paper, we present the methodological, as well as the experimental bases, for carrying out a determination of the decay constant for spontaneous fission for ^{238}U without neutron irradiation. Thus, we have eliminated the troubles linked to neutron-fluence measurements.

Methodology

Consider a thick uranium disk whose thickness is greater than the uranium (^{238}U, ^{235}U and ^{234}U) alpha-particle ranges and consequently, greater than the fission-fragment range. If this source, named hereafter infinite source, is coupled with a muscovite mica sheet, the superficial density of the spontaneous-fission tracks per unit of exposure time, $\rho_S/\Delta t_S$, after suitable chemical etching, is:

$$\left(\frac{\rho_S}{\Delta t_S}\right) = \frac{R_f}{4}\varepsilon_f C_{238} N_U \lambda_f \qquad (2)$$

where R_f is the range of fission fragments in the U_3O_8 bulk, ε_f is the detection efficiency of muscovite mica for fission fragments, N_U is the number of uranium atoms per unit of volume and C_{238} is the ^{238}U isotopic abundance in natural uranium.

Now, consider a thick uranium disk with the same geometry and manufacture but spiked with some trans-uranium isotope presenting a short spontaneous-fission half-life (shorter than the ^{238}U one), so that the necessary trans-uranium amount is as low as possible. Thus, one can assure that the disk-bulk density will not be affected by a possible different density of the trans-uranium isotope. The superficial density of the spontaneous-fission tracks per unit of exposure time, $\rho_{ST}/\Delta t_{ST}$ on a muscovite mica coupled with this disk is:

$$\left(\frac{\rho_{ST}}{\Delta t_{ST}}\right) = \frac{R_f}{4}\varepsilon_f (N_T \lambda_{fT} + C_{238} N'_U \lambda_f) \qquad (3)$$

where N_{UT} is the number of trans-uranium atoms per unit of volume, λ_{fT} is the decay constant for spontaneous fission for the trans-uranium isotope and N'_U is the number of uranium atoms in the trans-uranium-bearing disk.

Furthermore, it can be stated that:

$$N_U = N_T + N'_U \qquad (4)$$

Equation (4) will be valid if we are using the same procedures in both disks considered above and if we are dealing with infinite sources. Just under these conditions, we can assure that the volume in each disk contributing with fission fragments is the same.

Dividing Eq. (3) by Eq. (2), we obtain:

$$\lambda_f = \frac{\lambda_{fT} N_T}{C_{238} N_U}\left[\frac{(\rho_{ST}/\Delta t_{ST})}{(\rho_S/\Delta t_S)} - \frac{N'_U}{N_U}\right]^{-1} \qquad (5)$$

In Eq. (5) only the ratios involving the spontaneously fissile elements (N_T/N_U and N'_U/N_U) are unknown, but they can be measured by repeating the above procedure and replacing the fission-track detector with an alpha-particle-track detector. In our case, CR-39.

Consider the same infinite sources we have handled up to now. The superficial density of the alpha-particle tracks per unit of exposure time on the surface of the CR-39 coupled with the disk of natural uranium is:

$$\left(\frac{\rho_\alpha}{\Delta t_\alpha}\right) = \qquad (6)$$

$$= \frac{1}{8}\varepsilon_\alpha N_U (R_{238} C_{238} \lambda_{\alpha 238} + R_{235} C_{235} \lambda_{\alpha 235} + R_{234} C_{234} \lambda_{\alpha 234})$$

where ε_α is the CR-39 detection efficiency for alpha-particles, C_i is the ^{i}U isotopic abundance in natural uranium, $\lambda_{\alpha i}$ is the alpha-decay constant for ^{i}U and R_i is the range of the alpha-particles arising from ^{i}U in a U_3O_8 bulk.

For the uranium disk spiked with a trans-uranium isotope, we have:

$$\left(\frac{\rho_{\alpha T}}{\Delta t_{\alpha T}}\right) =$$

$$= \frac{1}{8}\varepsilon_\alpha [N_T R_T \lambda_{\alpha T} + N'_U \qquad (7)$$

$$(R_{238} C_{238} \lambda_{\alpha 238} + R_{235} C_{235} \lambda_{\alpha 235} + R_{234} C_{234} \lambda_{\alpha 234})]$$

where R_T is the range of alpha-particles arising from the trans-uranium isotope in the U_3O_8 bulk, $\lambda_{\alpha T}$ is the alpha-decay constant for the trans-uranium isotope.

It can be shown from Eqs (6) and (7) that:

$$\frac{N_T}{N_U} =$$

$$= \frac{(R_{238} C_{238} \lambda_{\alpha 238} + R_{235} C_{235} \lambda_{\alpha 235} + R_{234} C_{234} \lambda_{\alpha 234})}{R_T \lambda_{\alpha T}} \qquad (8)$$

$$\left[\frac{(\rho_{\alpha T}/\Delta t_{\alpha T})}{(\rho_\alpha/\Delta t_\alpha)} - \frac{N'_U}{N_U}\right]$$

Note that N_T/N_U is linked to N'_U/N_U through Eq. (4).

It is worth mentioning that, just as for the case of fission tracks, it was necessary to suppose the same efficiency factors that appeared in Eqs (6) and (7). However, the alpha-particle ranges cannot be canceled out in Eq. (8) because different alpha-particle energies are found for both uranium and trans-uranium isotopes. In this way, the λ_f value follows from Eqs (4), (5) and (8). So, we established a general methodology for a solid state nuclear track detector measurement of λ_f without neutron irradiation. Most of the suppositions presented above depend on the trans-uranium isotope to be used.

Therefore, the chosen isotope should have a suitable ratio between spontaneous-fission and alpha-decay half-lives for the purpose of making the alpha analysis possible and, at the same time, to yield a statistically satisfactory number of fission tracks.

Unfortunately, the access to the required type of material is very restricted due to the radiological and toxicological potential hazards associated to it. In this way, the entire experiment should be planned from the beginning to overcome these restrictions. The track sources should contain the minimum amount of material satisfying the condition of being infinite. In the next section, we present a manufacturing and analysis method for such infinite sources.

Experimental and results

Infinite sources manufacture

U_3O_8 thick films were used for this experiment. U_3O_8 powder was obtained by heating uranyl nitrate at 850 °C to guarantee that no other oxide would be present.[7] The resulting material was milled in a porcelain crucible until it became a thin powder. It was mixed with alcohol forming a homogeneous mixture (not a solution). The mixture was deposited on a substrate (aluminum or stainless steel disk 2 mm in height and 2 cm in diameter), whose mass was previously determined, and dried under an infrared lamp. When the alcohol evaporated, the oxide powder goes compactly deposited on the substrate surface. This technique is known as pulverization and compacting. Then, the entire set had its mass determined. In this way, the deposited oxide mass could be found. A detector of alpha-particle tracks, CR-39, was coupled with the source for a time interval Δt_α. In order to attain the thickness to make the source infinite, the process of depositing material, weighing sample and coupling CR-39 was repeated several times for each uranium film.

The alpha-detectors were chemically etched in a 6.25N NaOH for 400 minutes at 70 °C. Alpha-particle tracks were counted in an optical microscope under the 10×40 nominal magnification. The area of the detector containing tracks was scanned at regular intervals. Taking into account that the detector track density is proportionate to the source of activity, it can be said that this results in a source activity map. The manufacture test results are shown in Fig. 1. The errors shown in this curve are $\pm 1\sigma$, calculated using Poisson statistics.

It can be clearly seen in Fig. 1 that the alpha-track density curve reaches a plateau for thickness values greater than 10 mg/cm^2. χ^2 test carried out for the points beyond 10 mg/cm^2 resulted in $\chi^2 = 8.425$, leading to a P_χ~0.50 (for 9 degrees of freedom). This indicates a good internal coherence. Therefore, following the manufacture procedures described in this section, sources can be considered infinite if their thickness values are above that limiting value. Besides, this manufacture method yields sources with good uniformity (Fig. 2). The next step is to establish a method for spiking uranium with a trans-uranium isotope.

Spiking tests with ^{241}Am

In order to test the uranium spiking, ^{241}Am was chosen as spikier. This americium isotope has a very short half-life ($T_{1/2} = 432.2$ y) which makes it possible to analyze alpha-particle tracks on CR-39 even when the ^{241}Am-uranium amount ratio is as small as 10^{-6}. The energy of its alpha-particles is 5.5 MeV.

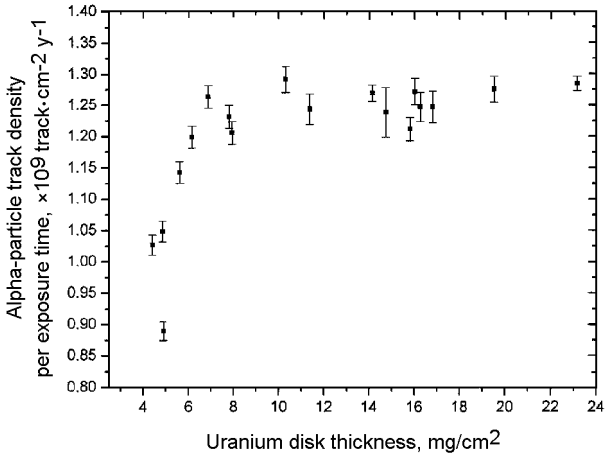

Fig. 1. Superficial density of alpha-particle tracks per exposure time vs. the source thickness for uranium sources from Table 1. The curve reaches a plateau for thickness values greater than 10 mg/cm^2. Error bars are $\pm 1\sigma$ using Poisson statistics

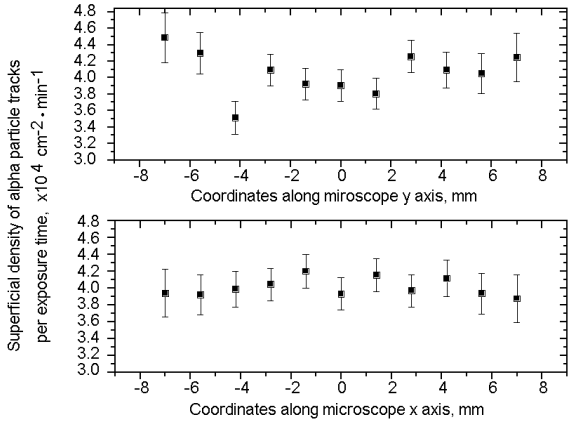

Fig. 2. Superficial density of alpha-particle tracks per exposure time unit vs. the of grid lines (y) and grid columns (x) for source AmI. Both curves show the uniformity of the trans-uranium isotope distribution in the source. Error bars are $\pm 1\sigma$ using Poisson statistics

Table 1. Alpha-track results for uranium sources spiked with ^{241}Am

Source	Track density per exposure time $\rho_\alpha^{241}/\Delta t_\alpha^{241}$ ($\pm 1\sigma$), $\times 10^4$ tracks·cm^{-2}·min^{-1}	Ratio between the number of ^{241}Am and natural uranium atoms, $\times 10^{-6}$ N_{241}/N_U ($\pm 1\sigma$)
AmI	4.03 ± 0.07	3.01 ± 0.06
AmII	3.99 ± 0.07	2.98 ± 0.06

An americium-nitrate solution whose specific activity is 874 kBq/g (6.9 μg/g) and a uranyl- nitrate solution concentrated to 150 mg U/ml were available. A 3 ml of uranyl-nitrate solution and 100 μl of americium-nitrate solution were deposited in a porcelain crucible. The resulting solution was heated at 850 °C and, in this way, an oxide powder (U_3O_8+AmO_2) was obtained. Two sources, named AmI and AmII were prepared. Pieces of CR-39 were coupled to these sources and chemically etched. Tracks were counted and the superficial densities of alpha-particle tracks were obtained. Source manufacture, CR-39 exposition, chemical etching and counting followed procedures described in the previous section. The ratio between the number of ^{241}Am and natural uranium atoms was calculated through Eqs (4) and (8). The counting results and calculations are presented in Table 1. The errors in this table are poissonian deviations ($\pm 1\sigma$). In such calculations, it was used a $(\rho_\alpha/\Delta t_\alpha)$ value that is an averaged mean of the values found on the plateau of Fig. 1, $(\rho_\alpha/\Delta t_\alpha) = (1.274 \pm 0.006)\cdot 10^9$ cm^{-2}·y^{-1}.

The counting of tracks on the detector was performed at well known positions along a quadrangular grid. If one takes the average values from the grid lines and plot them against the line coordinate, one has the mean profile of the track density. If one repeats the latter procedure for grid columns, one has a mean profile of the track density along the perpendicular direction. In Fig. 2 these profiles of alpha-track densities per exposure time interval for source AmI are presented.

Observing Table 1 and Fig. 2, we see that the spiking method presented in this section yields a uniform distribution of the spikier material.

Conclusions

In this paper, we have presented the general methodological bases for measuring λ_f through track detectors without using neutron irradiation. In choosing the spikier material we should follow definite criteria in order to satisfy the methodological requirements: to present a spontaneous-fission half-life shorter than that of ^{238}U, suitable alpha-decay half-life so that the alpha-track analysis is feasible, and alpha energies similar to those of natural uranium.

Results on manufacturing infinite U_3O_8 sources as well as on spiking these sources with a trans-uranium isotope, ^{241}Am, show that the applied method yields uniform material distribution, including the spikier distribution inside the latter.

Unfortunately, due to its not so short spontaneous-fission ($T_{1/2} = 1.0\cdot 10^{14}$ y), ^{241}Am is not a good fission-track source. For instance, we need 0.12% in mass of ^{241}Am for obtaining as many americium fission tracks as uranium ones from this kind of spiked source. This number becomes much smaller if we use ^{240}Pu or ^{242}Pu: $\approx 10^{-5}$. Besides, these two plutonium isotopes have alpha-decay half-lives that are suitable for the analysis method presented in this study.

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