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SPONTANEOUS FISSION OF ^{238}U**

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PUBLICAÇÃO IEA N.º 269
Maio — 1972

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SÃO PAULO — BRASIL

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* Separata de "Nuclear Instruments and Methods" - North-Holland Publishing Co., 91, pags. 577-579 (1971).

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DETERMINATION OF THE DECAY CONSTANT FOR SPONTANEOUS FISSION OF ^{238}U

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Received 26 October 1970

In this paper the fission track method is used to determine the decay constant λ_F for spontaneous fission of ^{238}U . This method requires the following measurements: (a) number of spontaneous fission tracks of ^{238}U , (b) number of induced fission tracks of ^{235}U due to thermal neutrons and (c) the thermal neutron flux.

1. Introduction

Fission fragments produce in minerals like mica, glass, etc. trails of radiation damage that, once chemically etched, can be observed with an optical microscope^{1,2)}. This detection method was used by Fleischer and Price³⁾ and Roberts, Gold and Armani⁴⁾ for the determination of the spontaneous fission decay constant of the ^{238}U . As has been pointed out by these authors, an accurate value of this constant is desirable, for instance, for geochronology⁵⁾. As is seen in table I, there is no satisfactory agreement between the values obtained by several investigators using different methods.

The purpose of the present paper is to obtain the value of λ_F using the fission track method. The advantages of this method are discussed by Fleischer, Price and Walker⁵⁾.

Our method is essentially the same as that of Fleischer and Price³⁾. The density ρ_s of spontaneous fission tracks due to ^{238}U , registered in a mica sheet in

contact with natural uranium, after a time interval ($\tau \ll 10^9$ y), is given by^{3,5)}:

$$\rho_s = \lambda_F \tau N C^{238} d \varepsilon(R^{238}, d), \quad (1)$$

where N is the number of uranium atoms per unit volume, C^{238} the fraction of ^{238}U atoms in natural uranium, d the sample thickness, R^{238} the average range of the ^{238}U fission in the sample and $\varepsilon(R^{238}, d)$ the detection efficiency for the fission fragments.

The determination of N and $\varepsilon(R^{238}, d)$ can be avoided by inducing fission in the ^{235}U atoms contained in the natural uranium by irradiating the sample in a thermal neutron flux. As the ^{235}U fission cross section $\sigma^{235}(v)$ for neutron absorption, follows very closely the $1/v$ law¹³⁾ (v being the neutron velocity) the density ρ_i of induced tracks is given by:

$$\rho_i = \phi t N C^{235} d \sigma^{235}(\bar{v}) g^{235}(T) \varepsilon(R^{235}, d), \quad (2)$$

where ϕ is the thermal neutron flux, t the irradiation time, C^{235} the fraction of ^{235}U atoms in the natural

TABLE I
Values of the decay constant λ_F .

| Investigators | Method | Year | λ_F |
|-----------------------------------|---|------|-----------------|
| Perfilov ⁶⁾ | fission chamber | 1947 | 5.3 ± 0.9 |
| Segré ⁷⁾ | fission chamber | 1952 | 8.7 ± 0.3 |
| Kuroda et al. ⁸⁾ | radiochemical | 1956 | 6.7 ± 0.6 |
| Parker and Kuroda ⁹⁾ | radiochemical | 1957 | 8.7 ± 0.5 |
| Gerling et al. ¹⁰⁾ | radiochemical | 1959 | 11.9 ± 1.0 |
| Kuz'minov et al. ¹¹⁾ | | 1960 | 10.7 ± 0.5 |
| Fleischer and Price ³⁾ | SSTR | 1964 | 6.6 ± 0.8 |
| Fleischer and Price ³⁾ | ^{40}K and ^{87}Rb dating | 1964 | 6.9 |
| Rao and Kuroda ¹²⁾ | radiochemical | 1966 | 7.8 ± 0.9 |
| Roberts et al. ⁴⁾ | SSTR | 1968 | 7.03 ± 0.11 |

uranium, \bar{v} the mean velocity of the thermal neutrons, $\sigma^{235}(\bar{v})$ the fission cross section of the ^{235}U for neutrons with $v = \bar{v}$, T the characteristic temperature of the thermal neutron spectrum, $g^{235}(T)$ the correction factor for the deviation of the $\sigma^{235}(v)$ from the $1/v$ law and R^{235} the mean range of the ^{235}U fission fragments in the sample. Within the experimental error we can put $R^{235} = R^{238}$.

The thermal neutron flux is obtained by measuring the activity A induced by the neutrons in gold foils which has an activation cross section $\sigma^{\text{Au}}(v)$ obeying the $1/v$ law. The flux ϕ and the activity A are related by

$$A = \phi \sigma^{\text{Au}}(\bar{v}) g^{\text{Au}}(T) \quad (3)$$

where $g^{\text{Au}}(T)$ is the correction factor for the deviation of $\sigma^{\text{Au}}(v)$ from the $1/v$ law¹⁴.

Taking into account eqs. (1), (2) and (3) and using the relation $\sigma(v_0)v_0 = \sigma(\bar{v})\bar{v}$, which follows from the $1/v$ law (where $v_0 = 2200$ m/s), we obtain:

$$\lambda_F = \frac{\rho_i A \pi \sigma^{235}(v_0) C^{235}}{\rho_i \tau \sigma^{\text{Au}}(v_0) C^{238} g^{235}(T)} t \quad (4)$$

In this way, it is not necessary to know the neutron spectrum exactly. To obtain the correction factors $g(T)$ we have assumed the neutron spectrum to be Maxwellian. Since this spectrum was observed to have, very nearly, a Maxwellian form¹⁵) this approximation introduces in $g(T)$ a very small error (see table 2).

2. Measurements and results

The samples of uranium and mica and the Au foils with, approximately 96 mg/cm² and a diameter of 8 mm, were irradiated* in pairs, with and without cadmium covering to eliminate the epithermal neutron influence¹³).

The induced activities of the Au foils were measured in a $4\pi\beta-\gamma$ coincidence system¹⁶⁻¹⁸).

The fission tracks registered in the mica sheets after two hours of chemical etching by HF (49%) at 23°C, were counted using an optical microscope with projection screen of 130 × enlargement.

As the density of fission tracks due to cosmic radiation is expected to be negligible we assumed that the background track density ρ_b is due only to the fossil tracks.

Using eq. (4) and taking into account the values for C^{235}/C^{238} , $\sigma^{\text{Au}}(v_0)$, $\sigma^{235}(v_0)$, $g^{\text{Au}}(T)$, T , τ and A given

*The irradiation was done in the beam hole B.H.-10 of the Institute of Atomic Energy of S. Paulo.

TABLE 2

| |
|---|
| $C^{235}/C^{238} = 7.25 \times 10^{-3}$ ¹⁹) |
| $\sigma^{\text{Au}}(v_0) = (98.8 \pm 0.3) \text{ b}^{20})$ |
| $\sigma^{235}(v_0) = (577.1 \pm 0.9) \text{ b}^{20})$ |
| $g^{\text{Au}}(T) = 1.008 \pm 0.001$ ¹⁴) |
| $g^{235}(T) = 0.964 \pm 0.003$ ¹⁴) |
| $T = (351 \pm 8) \text{ K}$ ¹⁵) |
| $\tau = (4.216 \pm 0.001) \text{ y}$ |
| $A = (1.1460 \pm 0.0007) \times 10^{-14} \text{ d pm/atom}$ |

TABLE 3
Track densities.

(The areas scanned for the tracks counting were about 2.5 cm². The induced track density ρ_i was normalized to one minute of irradiation.)

| | |
|--|--|
| Spontaneous fissions (ρ_s) (tracks/cm ²) | 2086 ± 30 |
| Induced fissions (ρ_i) (tracks/cm ² min) | Bare 3157 ± 17 Cd covered 180 ± 2 |
| Background (ρ_b) (tracks/cm ²) | 112 ± 11 |

in table 2 and the values for ρ_s , ρ_i and ρ_b given in table 3 we obtain:

$$\lambda_F = (7.30 \pm 0.16) 10^{-17} \text{ y}^{-1}.$$

As is seen in table 1, there is a good agreement between our result and the results of Kuroda et al.^{8,12}), Fleischer and Price³) and Roberts et al.⁴).

We are indebted to Dr. R. Fulfaro for his data of temperature and spectrum of the neutrons and to Dr. L. P. Moura for her assistance in the activity measurements.

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ABSTRACT

In this paper the fission track method is used to determine the decay constant, λ_F , for spontaneous fission of ^{238}U . This method requires the following measurements: a) number of spontaneous fission tracks of ^{238}U , b) number of induced fission tracks of ^{235}U due to thermal neutrons and c) the thermal neutron flux. The direct neutron flux determination is avoided by measuring instead the activity induced by this flux in a material with cross section that obeys the $1/v$ law. Samples of natural uranium in contact with mica sheets remained sealed for about four years. The decay constant was found to be $\lambda_F = (7.30 \pm 0.16)10^{-17} \text{ yr}^{-1}$.

RESUMO

Neste trabalho, o método dos traços de fissão em sólidos é utilizado na determinação da constante de decaimento, λ_F , para a fissão espontânea do ^{238}U . Este método requer as seguintes medidas: a) número de traços de fissão espontânea do ^{238}U ; b) número de traços de fissão induzida no ^{235}U por neutrons térmicos e, c) o fluxo de neutrons térmicos. A determinação direta do fluxo de neutrons, entretanto, foi evitada através da medida da atividade por ele induzida em um material cuja seção de choque obedece a lei $1/v$. Amostras de urânio natural em contato com folhas de mica permaneceram seladas durante 4 anos. Obteve-se para a constante de decaimento o valor $\lambda_F = (7.30 \pm 0.16)10^{-17} \text{ a}^{-1}$.

RÉSUMÉ

Dans ce travail, on utilise la méthode des traces de fission dans les corps solides pour déterminer la constante radioactive, λ_F , pour la fission spontanée du ^{238}U .

Cette méthode fait nécessaires quelques mensurations: a) numéro des traces de fission spontanée du ^{238}U ; b) numéro des traces de fission induite dans le ^{235}U par des neutrons thermiques et c) le flux des neutrons thermiques. Cependant, la détermination directe du flux des neutrons a été évitée par la mesure de l'activité que le même induit dans un matériau pour lequel la section efficace obéit à la loi du $1/v$. Échantillons d'uranium naturel en contact avec des lames de mica ont demeurés scellés durant 4 années. Pour la constante radioactive on a obtenu la valeur $\lambda_F = (7.30 \pm 0.16)10^{-17} \text{ a}^{-1}$.