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Spontaneous fission decay constant of ^{238}U determined by SSNTD method using CR-39 and DAP plates

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Abstract

This paper presents a simple method for determining the spontaneous fission decay constant (λ_f) of ^{238}U using a combination of CR-39 and diallyl phthalate (DAP) plates as solid-state nuclear track detectors (SSNTD). In this method, thin U-sources were prepared by depositing natural uranium onto 10-cm^2 stainless steel plates. Alpha-particle tracks originated from ^{238}U were detected in the 2π geometry of CR-39 detectors that had been attached to U-sources for 120–426 min. Spontaneous fission tracks, on the other hand, were detected using DAP detectors in the same geometry that had been attached to U-sources for 50–238 days. Based on precise measurements of bulk-etching and track-etching rates of the two detectors, track registration efficiencies and critical angles were determined to be 0.740 ± 0.072 and 15.1° for CR-39, and 0.995 ± 0.035 and 0.26° for DAP, respectively. Using these correction values and specific activities (or track densities) measured for each detector, a mean value of λ_f was calculated to be $(8.51 \pm 0.18) \times 10^{-17}\,\mathrm{yr}^{-1}$ in proportion to the alpha emission decay constant of $1.55 \pm 1.25 \times 10^{-10}\,\mathrm{yr}^{-1}$.

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1. Introduction

In studies using solid-state nuclear track detectors (SSNTD), the determination of spontaneous fission decay constant (λ_f) of ^{238}U has been a controversial issue [1]. Since the 1940s, nearly 60 measurement results have been reported which have been carried out utilizing various methods and detectors (see Refs. [1–5]), and one-third of these reports had been made by the SSNTD method. The debate was mainly centered around the fact that large deviations existed in the reported values of λ_f, from 7×10^{-17} to $12 \times 10^{-17}\,\mathrm{yr}^{-1}$, and that fission-track dating researchers accepted the following two values: (1) approximately $7.0 \times 10^{-17}\,\mathrm{yr}^{-1}$ and (2) $8.5 \times 10^{-17}\,\mathrm{yr}^{-1}$. The former value was supported by the results of SSNTD measurements using mainly the mica-uranium sandwich method (for example, Ref. [6]), and the latter by the results of direct determinations using the ionization-chamber or bubble-chamber methods (for example, Ref. [7]). Recently, the International Union of Pure and Applied Chemistry (IUPAC) recommended the evaluated value of $(8.5 \pm 0.1) \times 10^{-17}\,\mathrm{yr}^{-1}$ based on a series of determinations performed since 1950, excluding all results of measurements by the SSNTD method [2].

In this study, we have developed a simple method for determining the λ_f value of ^{238}U using a thin U-source and two types of SSNTD. The aim of the present study is to describe the principles of this method and to report on the λ_f value obtained by it.

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2. Principle of the method

2.1. Basic concept

In the SSNTD technique, λ_f is normally determined by using a detector-uranium sandwich and counting the number of tracks produced on the detector by fissions occurring in the U-source adjacent to it within a given time period (Fig. 1(a)). In other words, a detector is fixed onto the U-source for a given period of time to accumulate spontaneous fission tracks. Then, a different set of detector-uranium sandwich is subjected to thermal neutron irradiation for the detection of induced fission tracks of ^{235}U. Based on these results, the fission-track age equation [8] can be applied to the detector-uranium sandwich samples. However, this method requires the measurement of thermal neutron fluence for the determination of the λ_f value. At the present, reactor specialists can determine neutron fluences to errors of $<2\%$, including both statistical and systematic errors [9–11]. Another option is to use dosimeter standard-glasses pre-irradiated with well-determined fluences [12,13].

In contrast to the above, we propose here a new approach for determining the λ_f-value which will not require the process of thermal neutron irradiation. In this method, alpha-particle tracks produced by alpha emission and nuclear-fission fragment tracks produced by spontaneous fission of ^{238}U in the thin U-source were detected on two types of plastic detectors, each specialized for detecting a single type of emission (Fig. 1(b)). This method is based on a simple principle that alpha decay and spontaneous fission decay constants are proportional to the specific activities of alpha decay and spontaneous fission decay, respectively. Thus, the fission-decay rate may be determined relative to the alpha-decay rate, and so the track registration efficiency of the detector replaces the neutron fluence measurement as the most important factor in this method.

2.2. Equations for λ_f calculation

First, a thin source is defined as one whose thickness is sufficiently smaller than the emitted particle range. Here, it is assumed that particle emission is uniform over the whole surface of the source. Note that the U-source consists of natural uranium having natural isotopic abundances. The specific activities ($cm^{-2}s^{-1}$) of alpha decay and fission decay produced by the ^{238}U in the source can be expressed as follows using decay constants (yr^{-1}):

$$A_{238\alpha} = \frac{\lambda_{238\alpha}}{365.25 \times 24 \times 60 \times 60} N_{238} \quad (1)$$

$$A_{238f} = \frac{\lambda_{238f}}{365.25 \times 24 \times 60 \times 60} N_{238} \quad (2)$$

where $A_{238\alpha}$ is the specific activity of alpha decay of ^{238}U ($cm^{-2}s^{-1}$), A_{238f} is specific activity of spontaneous fission of ^{238}U ($cm^{-2}s^{-1}$), $\lambda_{238\alpha}$ is alpha-decay constant of ^{238}U (yr^{-1}), λ_{238f} is spontaneous fission-decay constant of ^{238}U (yr^{-1}), N_{238} is number of ^{238}U atoms per unit area (cm^{-2}).

From Eqs. (1) and (2), we obtain

$$\frac{A_{238\alpha}}{\lambda_{238\alpha}} = \frac{A_{238f}}{\lambda_{238f}}. \quad (3)$$

The following relationship holds for alpha-track density measured on a detector placed in contact with the thin U-source for a given time period. Note that only a half of the alpha particles are emitted on the detector side:

$$\frac{A_\alpha}{2} t_\alpha E_\alpha = \rho_\alpha \quad (4)$$

where A_α is the specific activity of alpha decay of total uranium isotopes ($cm^{-2}s^{-1}$), t_α is irradiation time for alpha-particle detection (s), E_α is registration efficiency of alpha tracks on detector ($0 < E_\alpha \leqslant 1$), ρ_α is alpha-track density on detector (cm^{-2}).

The nuclear species which emits alpha particles for natural uranium are ^{238}U, ^{235}U, and ^{234}U. The relative isotopic abundances [14], half-lives [15] and calculated specific activities of these three species are summarized in Table 1. Using these values, the following relationship may be established between A_α and $A_{238\alpha}$:

$$A_{238\alpha} = A_\alpha \frac{4.878}{4.878 + 0.225 + 4.921} = 0.487 A_\alpha. \quad (5)$$

Using Eqs. (4) and (5), $A_{238\alpha}$ may be expressed as

$$A_{238\alpha} = \frac{0.974 \rho_\alpha}{t_\alpha E_\alpha}. \quad (6)$$

On the other hand, the following equation holds for fission-track density detected on a detector attached to a thin U-source for a given time period. Note here that,

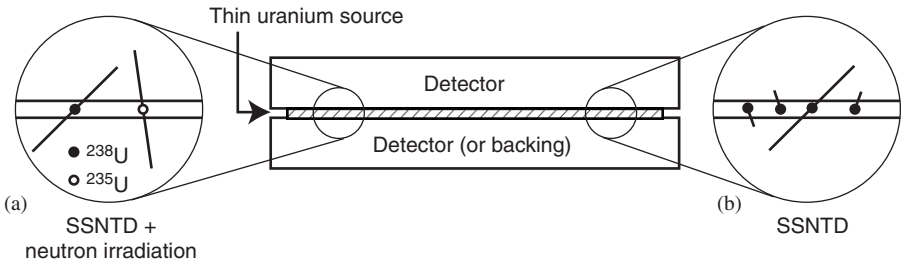

Fig. 1. Comparison of solid-state nuclear track detector (SSNTD) methods: (a) conventional uranium-detector sandwich method combined with thermal neutron irradiation to induce fission of ^{235}U, and (b) new method using plastic detectors for detecting alpha tracks and fission tracks.