Oklo Phenomenon and Nuclear Data

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Abstract

Oklo natural reactor phenomenon provides unique information on possible change of the fine structure constant α and on how the buried nuclear waste behave in geological formations. Nuclear data are indispensable in the Oklo analyses but not all of them are known or have required precision. We draw attention to neutron capture cross sections needed in the application of the ^{176}Lu/^{175}Lu thermometry for solving the existing uncertainty in operating temperatures of the most referenced Oklo reactor zones RZ2 and RZ10. In particular we propose to improve measurements of the branching ratio $B_g = \sigma_g/(\sigma_g + \sigma_m)$ in ^{175}Lu thermal neutron capture leading to the ^{176m}Lu isomer and ^{176g}Lu ground states. For Oklo reactor zone RZ10 we calculate not only neutron fluxes but gamma-ray fluxes as well to clarify the role of the photo excitation of the isomeric state ^{176m}Lu by the ^{176}Lu(γ,γ) fluorescence. Our values of the constant $\lambda_{\gamma,\gamma}$ for Oklo phenomenon do not support burning of ^{176}Lu through the (γ,γ) channel.

Introduction

The Oklo natural reactors (see e.g. Naudet [1]) have proven to be one of the most sensitive terrestrial testing grounds for studying time variation of the electromagnetic fine structure constant α, e.g. [2] and Refs. therein. The position of the first neutron resonance in ^{149}Sm strongly influences the effective cross section $\hat{\sigma}$ for neutron capture responsible for burning of the isotope ^{149}Sm, so from the Sm isotopic ratio data in Oklo reactor zones one can deduce the energy of the ^{149}Sm resonance in the primordial time of Oklo phenomenon – two billion years ago. The present day resonance energy E_0=97.3-meV is well known in neutron spectroscopy. A possible change in the resonance energy must then indicate change of α, because the resonance energy depends partly on the Coulomb interaction between protons in the Sm nucleus. While the majority of Oklo analyses have been consistent with no change, a positive effect continues to be argued for from astronomical observations [7].

Oklo phenomenon includes knowledge of the duration and age of the nuclear chain reaction at Oklo, neutron fluences, the Pu-239 breeding, etc. Most of nuclear data required for the analysis are in a relatively good shape. This is valid in particular to mass distributions of fission products of U-235, U-238, Pu-239, to isotopic ratio data Nd/U-238, Bi/U-238, to Sm, Gd and Nd isotopic ratios and to various decay constants. Here we draw attention only to nuclear data needed for the application of the Lutetium thermometry for Oklo reactor zones. These are the branching ratio $B_g = \sigma_g/(\sigma_g + \sigma_m)$ in ^{175}Lu thermal neutron

capture leading to the ^{176m}Lu isomer and ^{176g}Lu ground states, and the values of the ^{176}Lu burning constants $\lambda_{\gamma,\gamma}$ through the (γ,γ) channel at different γ-energies.

Problem of Oklo reactors temperature

In Oklo studies, the "effective" capture cross section $\hat{\sigma}$ is introduced as

$$\hat{\sigma} = \int_0^\infty n(v) v\, \sigma(v)\, dv / \int_0^\infty n(v)\, v_0\, dv. \qquad (1)$$

Here $n = \int_0^\infty n(v) dv$ is the total neutron density, $\int_0^\infty n(v)\, v_0\, dv \equiv \hat{\Phi}$ is the "effective" neutron flux, and v_0=2200 m/sec is the velocity of a neutron at thermal energy 0.0253 eV. As an integrated quantity, $\hat{\sigma}$ is not dependent on neutron velocity v (or neutron energy E) but if some nuclide, like ^{176}Lu, has a neutron resonance close to thermal enery, $\hat{\sigma}(T_O)$ may depend on the Oklo reactor temperature T_O through a temperature dependence of the neutron density $n(v)$ in such cases. Examples of such dependence for zones RZ2 and RZ10 are given as curves of $n(E, T_O)$ in Ref.[2]. The 'effective' neutron flux $\hat{\Phi}$ is different from the 'true' flux $\Phi = \int_0^\infty n(v)\, v\, dv$, but the reaction rate $R = \hat{\sigma}\,\hat{\Phi} = \bar{\sigma}\,\Phi$ is the same since the average cross section $\bar{\sigma}$ is $\bar{\sigma} = \int_0^\infty n(v)\, \sigma(v)\, v\, dv / \int_0^\infty n(v)\, v\, dv$.

Previous Oklo analyses all reveal that the bounds on variation of α depend significantly on the assumed reactor operating temperature T_O. In Refs. [3, 4, 5] Maxwellian thermal neutron fluxes were used, while in Refs. [2, 6] several realistic models of the reactors zones were elaborated with the help of modern neutron transport codes. For zone RZ2, with indirect arguments, Damour and Dyson [5] allowed for a broad interval of T_O from 450 to 1000 C. Y. Fujii et al. [4] took the interval from 180 to 400 C for zones RZ3, RZ10, while Gould et al. [2] preferred lower values from 200 to 300 C for zone RZ10. Petrov et al. [6] argued for the value $T_O = 452 \pm 55$ C as the temperature value at which their model of the active core RZ2 became critical [1]. Three studies have attempted to bound temperatures more directly using ^{176}Lu/^{175}Lu thermometry. Holliger and Devillers [8], gave temperatures $T_O = 260$ and 280 C for RZ2 and RZ3, and recently Onegin [6] found $T_O = 182 \pm 80$ C for RZ3. However, for RZ10, one of the most well characterized zones, Hidaka and Holliger[9] succeeded in getting a result for only one sample $T_O = 380$ C while for the other three samples only a (surprisingly high) lower bound, $T_O > 1000$ C, was obtained. Obviously, the temperature T_O of Oklo reactors remains a very uncertain parameter and the results of Ref.[9] are especially puzzling.

[1] We note that the T_O parameter depends strongly on the H/U atomic ratio, which in their active core model had rather low value, H/U=5.9, as compared with H/U=15.6 in a subsequent model of Onegin [6].

^{176}Lu/^{175}Lu thermometry: cross section branching B^g

We will follow the description of the Lutetium thermometry given by Holliger and Devillers [8]. The rare earth element Lutetium has one stable isotope, ^{175}Lu, with natural abundance 97.401 %, and and a second very long-lived isotope ^{176}Lu with half-life $t_{1/2} = 37.6$ Gyr and present day natural abundance 2.60. Neither isotope is produced in the fission of Uranium or Plutonium. Introducing the atomic number densites N_i and neutron reaction rates $\lambda_i = \hat{\sigma}_i\hat{\Phi}$ (the latter play roles analogous to the roles of decay constants in the radioactive decay) and neglecting for now the β decay of the 'stable' ^{176g}Lu during the time of Oklo reactor criticality, we write the coupled differential equations for the time evolution of the number densities $N_i(t)$ of our two isotopes of interest:

$$\frac{dN_6}{dt} = -\lambda_6 N_6 + B^g\lambda_5 N_5 \quad (2)$$

$$\frac{dN_5}{dt} = -\lambda_5 N_5 \quad (3)$$

with initial conditions $N_6(0) = N_6^0\exp(\frac{Dln2}{t_{1/2}})$ and $N_5(0) = N_5^0$. In these equations, the subscripts 5 and 6 refer to ^{175}Lu and ^{176}Lu, respectively, N_6^0 and N_5^0 are the present day natural abundances of Lutetium isotopes and $D = 2$ Gyr is the date of the Oklo phenomenon.
This system of equations has an analytical solution which we write for the time duration t_1 of reactor criticality:

$$N_6(t_1) = N_6^0\exp(\frac{Dln2}{t_{1/2}})\exp(-\lambda_6 t_1) + N_5^0 B^g\frac{\lambda_5}{\lambda_6-\lambda_5}[\exp(-\lambda_5 t_1) - \exp(-\lambda_6 t_1)] \quad (4)$$

$$N_5(t_1) = N_5^0\exp(-\lambda_5 t_1). \quad (5)$$

The two terms in the right hand side of Eq.(4) represent the burnup of the initial ^{176}Lu with cross section σ_6 and its partial restitution after burnup of ^{175}Lu with the partial cross section σ_5^g. Taking the ratio and accounting for the β decay of ^{176}Lu after shut down of the reactor, we obtain the present day ratio of the lutetium isotopes in Oklo wastes as:

$$\frac{N_6}{N_5}(\text{now}) = \frac{N_6^0}{N_5^0}\exp(-(\hat{\sigma}_6-\hat{\sigma}_5)\hat{\Phi}t_1) + B^g\frac{\hat{\sigma}_5}{\hat{\sigma}_6-\hat{\sigma}_5}[1-\exp(-(\hat{\sigma}_6-\hat{\sigma}_5)\hat{\Phi}t_1)]\exp(-\frac{Dln2}{t_{1/2}}), \quad (6)$$

which depends on the temperature T_O through the temperature dependence of $\hat{\sigma}_6$.
The neutron fluence $\hat{\Phi}t_1$ is a well characterized parameter in Oklo studies. The effective cross sections σ_5 and σ_6 are calculable using known resonance cross sections of Ref.[10] together with neutron densities within realistic reactor models [2]. The calculation therefore leads to a clear prediction of the present isotopic ratio if the value of the branching ratio parameter B^g is known, which however is not the case.

^{176}Lu has an exceptionally large thermal capture cross section, $\sigma_6 = 2090 \pm 70$ b [10] due to a strong resonance at 141 meV. This leads to a strong temperature dependence

in the rate at which ^{176}Lu burns up. Following neutron capture, ^{176}Lu transforms into the stable nuclide ^{177}Hf after the β decay of the reaction product ^{177}Lu. Neutron capture by ^{175}Lu has two branches with much smaller cross sections. The dominant branch with $\sigma_5^m = 16.7 \pm 0.4$ b [10] leads to an isomeric state ^{176m}Lu which decays to ^{176}Hf with half life of 3.6 hr. The minor branch with $\sigma_5^g = 6.6 \pm 1.3$ b [10] leads directly to the ground state of ^{176}Lu. While the large cross section for neutron capture in ^{176}Lu serves to deplete the small fraction of ^{176}Lu, the minor capture branch on the much larger fraction of ^{175}Lu works to restore ^{176}Lu. Defining $\sigma_5 = \sigma_5^g + \sigma_5^m$, the important parameter determining the balance between depletion and restitution of ^{176}Lu is then $B^g = \sigma_5^g/\sigma_5$, the branching ratio parameter.

According to the evaluated cross sections [10], $B^g = 0.28 \pm 0.05$ at thermal neutron energies. The precision of the evaluated B^g value is obviously overestimated. This is a difficult quantity to measure however: its value comes from taking the difference between two cross sections, total σ_5 and activation σ_5^m, of similar magnitude. Recently two precision measurements in astrophysics were carried out [11, 12] with the results $B^g = 0.117 \pm 0.046$ at neutron energy 5 keV and $B^g = 0.143 \pm 0.020$ at 25 keV. These values are about a factor of two less than the value for thermal neutrons, 0.28. Such a big difference looks rather puzzling even taking into consideration a partial contribution of p-wave neutrons, and raises the question of whether the thermal value is actually correct. There is some experimental evidence for a smaller value. A spectroscopic study [13] of thermal neutron capture found $B^g = 0.13 \pm 0.03$. Also, the only neutron transmission measurement for which there are data with an enriched ^{175}Lu sample, (Baston et al. [14]), hints at a smaller value in the following way. The thermal capture cross section they determined, $\sigma_5 = 23 \pm 3$ b, was based on subtracting a potential scattering contribution of 5.5 b from their measured total cross section. However, the potential scattering cross section is known today to be larger, $\sigma_n = 7.2 \pm 0.4$ b. Using this new value leads to $\sigma_5 = 21.3$ b, and correspondingly a smaller thermal neutron value $B^g = 0.21 \pm 0.11$ b.

While this latter result has very large uncertainty, we believe there is reason to think the value of B^g for thermal neutrons is not that well known, and merits improved measurements. A ten percent measurement of B^g is desirable and will certainly require carrying out a high precision total cross section measurement. An accuracy of 1% has been achieved in neutron spectroscopy study at the RPI linac for other rare earth element total cross sections, and similar accuracy should be feasible with an enriched ^{175}Lu sample. The ^{176}Lu isomer activation cross section is less critical but should also be improved bearing in mind that the differences in the existing activation values considerably exceed the reported uncertainties.

To solve equation (6) for different temperatures, we first evaluate (following the procedure outlined in [2]) the effective capture cross sections $\hat{\sigma}$ for ^{175}Lu and $\hat{\sigma}$ for ^{176}Lu using the T = 20 to 500 C neutron spectra derived from MCNP calculations in our earlier work [2]. For ^{175}Lu we include all resonances up to 49.4 eV along with two sub-threshold resonances and the strong resonance at 96.69 eV. For ^{176}Lu we include all resonances up to 52.13 eV. The contribution of the $E_0 = 143$-meV resonance dominates for ^{176}Lu, all other resonances contribute only a few percent. The resulting cross sections are shown in Table I.

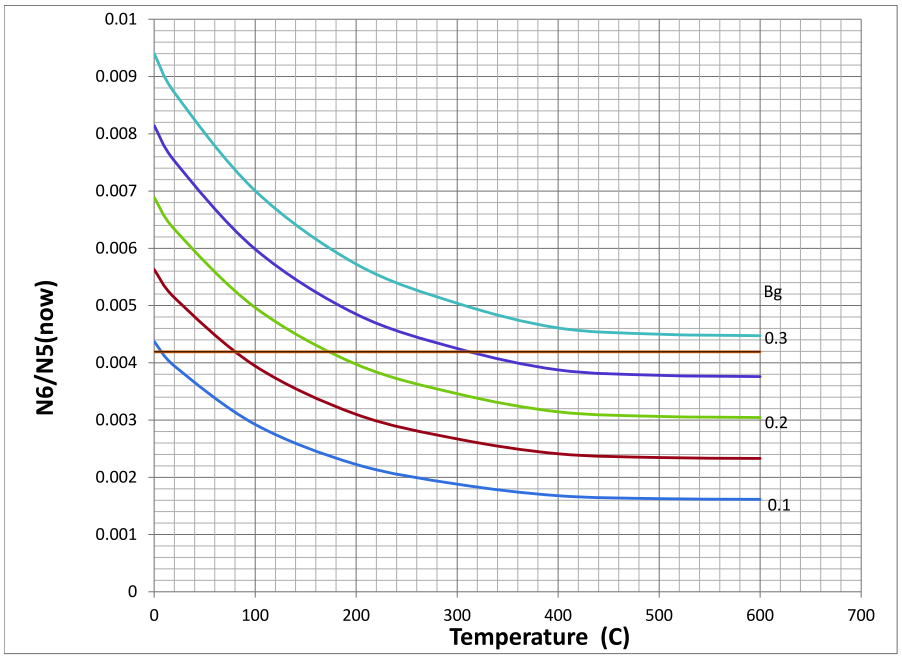

Figure 1: Solution of Eq. (6) for branching ratios B^g=0.1, 0.15, 0.2, 0.25 ans 0.3.

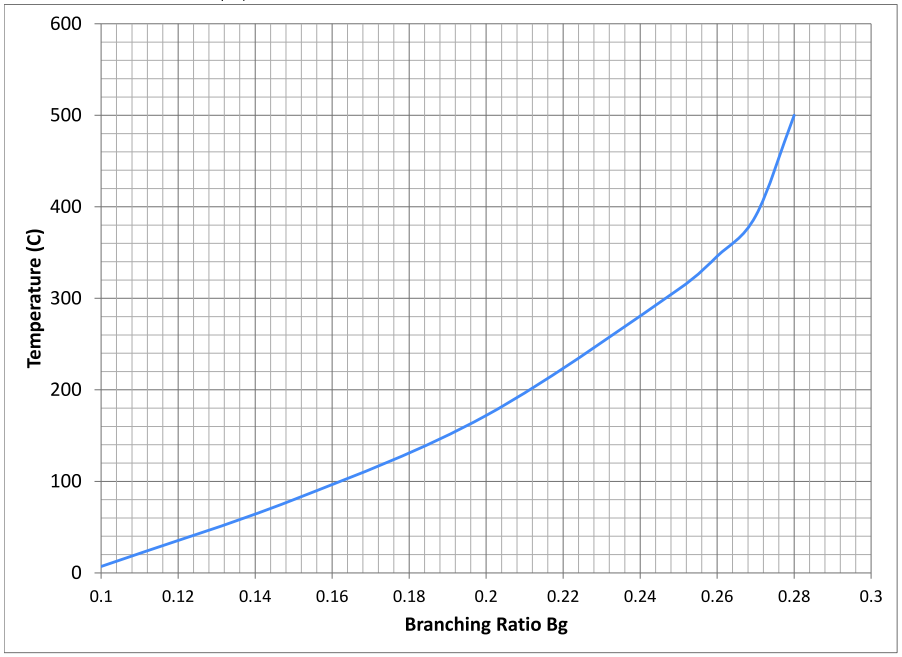

Figure 2: Oklo Reactor zone RZ10 temperature as the function of B^g.

Table 1: Lu cross sections for zone RZ10 at different temperatures.

T_O, C	0	20	100	200	300	400	500	600
$\hat{\sigma}_5$, kb	0.115	0.115	0.115	0.115	0.114	0.114	0.114	0.114
$\hat{\sigma}_6$, kb	4.22	4.42	5.36	6.31	7.01	7.54	7.71	7.75

The Lu isotopic abundance data for reactor zone RZ10 are given by Hidaka and Holliger (HH) [9]. We have found it more useful to work with a meta sample data obtained as an average over four samples that are available in [9]. This approach was shown to be successful for Sm data [2]. The meta sample ratio $N_6/N_5(\text{now}) = 0.00419$ value can be compared to the result of evaluating equation (6) for a range of B^g values and reactor temperatures. A plot of this comparison is shown in figure 1 for B^g values 0.1, 0.15, 0.2, 0.25 and 0.3, and temperatures 0 to 600 C. The intersection of the meta sample isotope ratio value with each curve yields a temperature prediction for the RZ10 reactor. A plot of these intersection values is given in figure 2 from which a temperature T_O for RZ10 can be read off if B^g is known. The evaluated thermal neutron energy value [10] $B^g = 0.28 \pm 0.05$ yields (barely) a solution $350 < T_O < 500$ C. The most accurately determined B^g ($E_n = 25$ keV) value of 0.157 ± 0.023 from the study [12] gives $T_O = 100 \pm 30$ C. A precise determination of B^g for thermal neutrons is clearly indispensable.

Prompt and delayed gamma-ray fluxes in zone RZ10

The MCNP code [16] allows to model not only the neutron transport but the energy spectra of prompt γ-ray fluxes (or fluences) also. For the latter purpose we presently run basically the same input for the Oklo zone RZ10 as elaborated in [2]. The model of an Oklo reactor zone is a flat cylinder of 70 cm height, 6 m diameter, surrounded by a 1 m thick reflector consisting of water saturated sandstone. As for any reactor, the Oklo criticality is determined, besides its geometry, by the composition of the active zone. The Oklo reactor zones include uraninite UO_2, gangue (oxides of different metals with water of crystallization) and water. Detailed composition and neutronic parameters of the RZ10 reactor zone are given in [2]. We remind only that the total density of the active core material at ancient times was about 3 g/cm^3 for RZ10 with only 30 wt. % of UO_2 in RZ10 dry ore. For this reactor the hydrogen to uranium atomic ratio in our model is $\frac{N_H}{N_U} = 13.0$. The multiplcation coefficient of a 'fresh' core was k_{eff}=1.036. The obtained *prompt* fission gamma-ray spectral flux is shown in fig. 1. This spectrum is normalized to the number of neutrons produced in RZ10 during the 15 kW pulse of 0.5 hour duration. The total gamma-ray flux is about 3x10^9 γ/cm^2 s^1. The delayed gamma-ray spectra calculations by the summation method for the decay heat, are described elsewhere [19]. An example of the *delayed* gamma spectrum after the thermal neutron fission of ^{235}U is shown in Fig. 4. Such spectra have been converted to delayed gamma fluxes which appeared to be a factor of three to five (depending on the energy) less intense than the fluxes of prompt gammas.

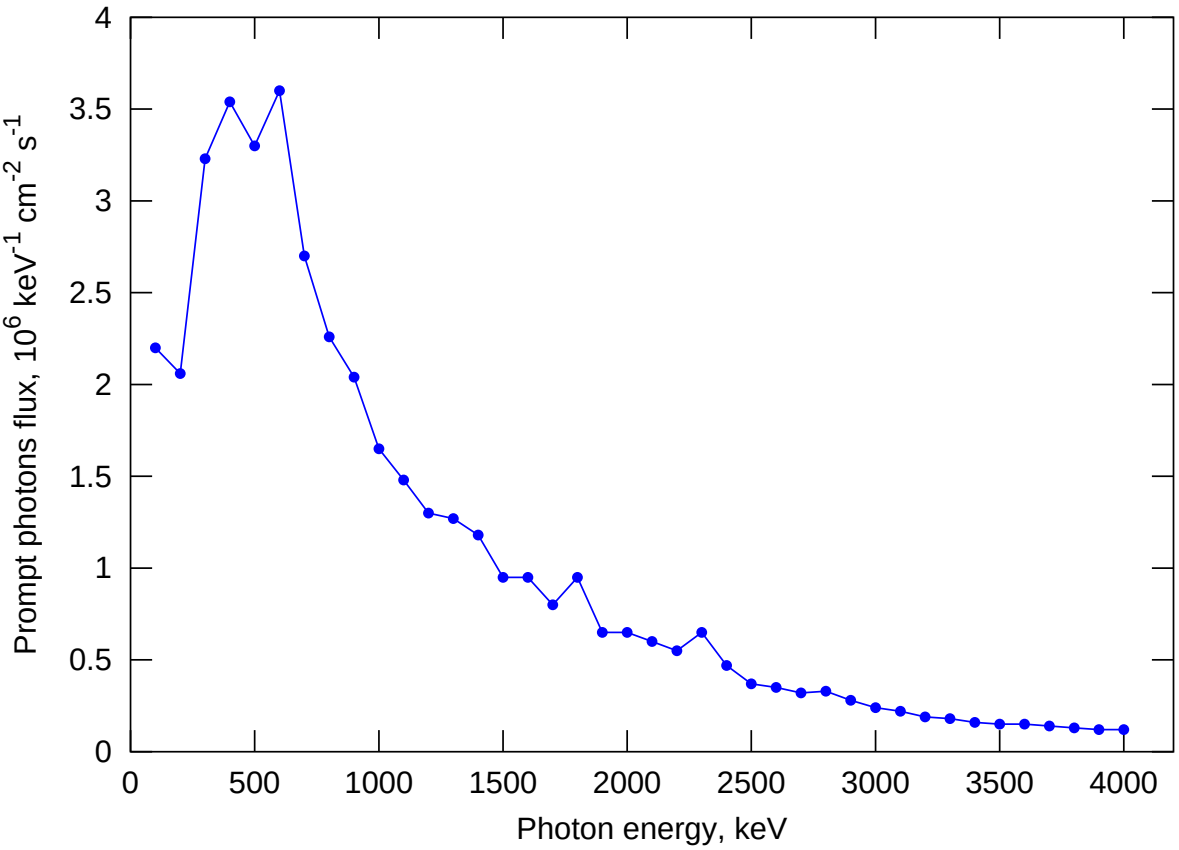

Figure 3: Prompt fission gamma-ray flux in Oklo Reactor zone RZ10.

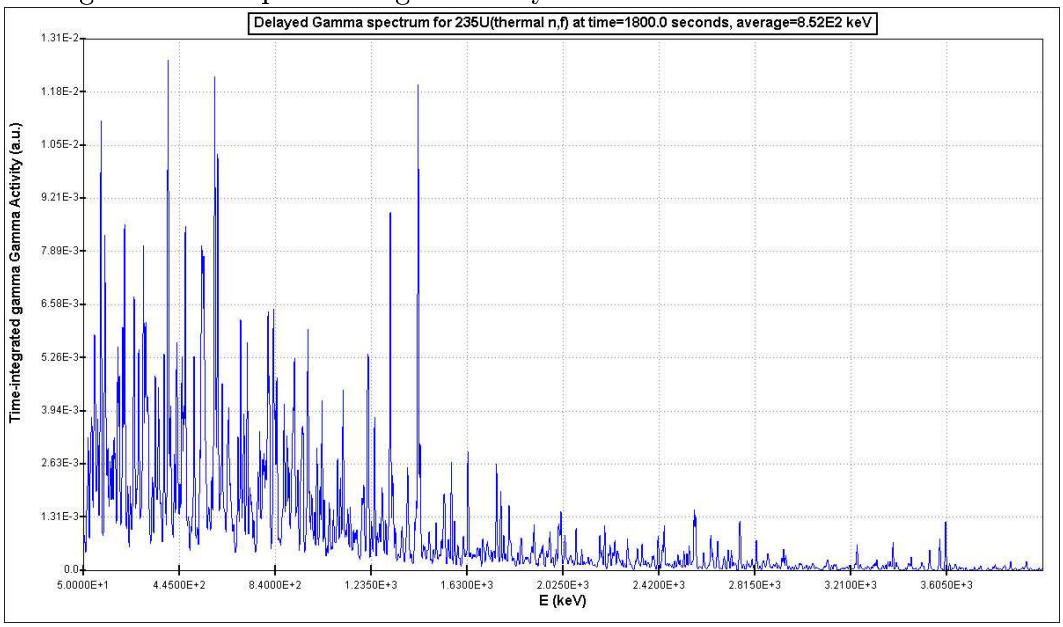

Figure 4: The delayed gamma-ray spectrum after U-235 fission by thermal neutrons.

Relation of γ-ray data to Lutetium thermometry

Analogously to the reaction rate, which is the number of reactions per one target nucleus per second, the rate $\lambda_{\gamma,\gamma'}$ of photo excitation of the isomeric state ^{176m}Lu in photon inelastic scattering through a higher lying narrow state E_i is

$$\lambda_{\gamma,\gamma'}(E_i) = \int \Phi(E)\sigma_{\gamma,\gamma'}(E,E_i)dE = \Phi(E_i)\sigma^{int}_{\gamma,\gamma'}(E_i), \tag{7}$$

where $\Phi(E_i)$ is the differential photon flux with the dimension of keV^{-1} cm^{-2} s^{-1}, and $\sigma^{int}_{\gamma,\gamma'}(E_i) = \int \sigma_{\gamma,\gamma'}(E,E_i)dE$ is the integrated cross section.
Astrophysical data suggest that a long lived ^{176}Lu in the photon bath of celestial bodies can be partially transformed into the metastable ^{176m}Lu by photons with energies closed to 839 keV and around 2.1 MeV. These particular energy windows contain excited states of ^{176}Lu with specific spins and parities which allow them (after de-excitation to lower states, including the isomeric one at E^m = 123 keV) act as intermediators for photo excitation of isomeric state. The constant $\lambda_{\gamma,\gamma}$ of Eq. (6) plays the role of the radioactive decay constant for burning ^{176}Lu through the γ,γ channel. The half-live time is then $t_{1/2} = 0.693/\lambda_{\gamma,\gamma}$.

According to Meshik et al. [18], the cycling operation of the Oklo reactors consisted of 0.5 hours pulses separated by 2.5 hours dormant periods. Photo burning of ^{176}Lu could, in principle, additionally change the Lu176/Lu175 isotopic ratio if the photon flux is high enough. Details of gamma-ray fluxes in Oklo reactors are discussed in our recent paper [19]. With the reported values of the photo-excitation cross sections $\sigma^{int}_{\gamma,\gamma'}(E_\gamma)$=33.4 mb·eV for E_γ=839 keV [15]) and σ_{int} = 140 mb·keV for $E_\gamma \simeq$2.1 MeV [20], and with our total spectral fluxes of 0.86x10^6 γ cm^{-2} s^{-1} keV^{-1} and 1.9x10^5 γ cm^{-2} s^{-1} keV^{-1} correspondly, we have obtained $\lambda_{\gamma,\gamma'} = 0.26 \cdot 10^{-22}$ /s for the first option and $\lambda_{\gamma,\gamma'} = 6.8 \cdot 10^{-20}$ /s for the second option. Because the neutron burning constant is larger by seven order of magnitude even for the second option we conclude that in Oklo reactors, in contrast to astrophysical processes, Lu176/Lu175 isotopic ratio is not influenced by the gamma radiation and therefore Lutetium thermometry is fully applicable to analyses of Oklo reactor data.

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