THE USE OF GAMMA LINES OF ^{134}I AND ^{135}I FOR THE ESTIMATION OF URANIUM FISSION PRODUCTS IN COLD NEUTRON IRRADIATED SAMPLES

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Two samples of zirconia and a standard reference material SRM2710 Montana soil (~300 mg from each) were prepared and packaged for irradiation (30 and 40 minutes respectively in cold neutron beam of 2.64×10^{9} neutron cm^{-2} s^{-1}), for the determination of fission rate constant of natural uranium (^{235}U is the effective isotope), at the National Institute of Standards and Technology (NIST), USA. The gamma intensities of both ^{134}I and ^{135}I fission products, arising from fission only, were measured via their unambiguous gamma lines and their decay constants. The obtained fission rate constant in average is found to be 1.1×10^{-8} fission s^{-1} per unit cold neutron flux ppm^{-1} of natural uranium in one gram sample. This constant can be used as an important number in the field of nuclear fission induced by cold neutron beams for the estimation and prediction of the concentration of fission products in an activated sample by cold neutrons.

في البحث الحالي تم تعريض عينتين من مادة الزركونيا وماده قياسيه عيارية (SRM 2710) – كتلة كل عينة تساوى ٣٠٠ مجم تقريبا – الى سيل من النيترونات الباردة قدره ٢,٦٤ ×١٠ ٩ نيوترون / سم٢ . ث. لمدة ٣٠ دقيقة للزركونيا و ٤٠ دقيقة للعينة القياسية . بدأت عمليات قياس النشاط الإشعاعي لنظيرى الإنشطار يود-١٣٤ و يود- ١٣٥ في العينات لمشعة بعد إنتهاء عملية التشعيع مباشرة. تم تعيين ثابت معدل الإنشطار من النتائج المتحصل عليها من كل عينة عن طريق خطوط جاما المؤكدة لهذين النظيرين و ثابت الإضمحلال لكل منهما و هذا المقدار الثابت (ثابت الإنشطار لليورانيوم) والذي وجد أن قيمة المقدار الثابت(في المتوسط) هي ١.١ ×١٠ ^-٨ إنشطار /ث في عينة يحتوى الجرام منها على يورانيوم طبيعى بتركيز قدره جزء في المليون معرض لسيل قدره الوحدة من النيوترونات الباردة (مع إهمال تأثيرات إمتصاص وتشتت النيوترونات في العينات المستخدمة بسمك ٢–٣ مم والذى يعتبر رقيقا إذا قورن بمدى إنتشار النيوترونات داخل العينات) كما تم تقديره عمليا . سيكون ذلك مفيدا في عدة مجالات منها مجال التحليل بالتنشيط بالنيوترونات الباردة لعينات تحتوى على يورانيوم وخاصة عند إستخدام تقنية التحليل بالتنشيط النيوتروني لتعيين التركيزات الضئيلة والتركيزات المتناهية في الضآلة لبعض العناصر حيث يمكن بذلك تلافي الخطأ الذى ينشأ عن وجود تداخل بين بعض خطوط جاما لتلك النظائر الناشئة من تفاعلات التنشيط بالنيوترونات الباردة لبعض العناصر الموجودة أصلا في العينات وتلك التى تنبعث من النظائر الناتجة من إنشطار يورانيوم–٢٣٥ في نفس عملية التشعيع .

Keywords: Cold neutron beam, Fission rate constant, Gamma spectrometry, ^{134}I and ^{135}I, Uranium

INTRODUCTION

The determination of the radioactive analytes in biological or environmental samples is almost encountered with the difficulty in measuring low level radioactivities. Some weak peaks in the gamma ray spectrum can be masked by the background intensity in the peak area. The fundamental statistical treatment shows the proportionality of the low limit of detection with the square root of the number of background continuum counts under the peak region of interest with a factor varies with the confidence level chosen [1]. The detection limit is expressed in counts, a more interesting parameter is the minimum detectable activity (Becquerels) which defined as the smallest number of radioactive nuclide that can be determined reliably [2]. The minimum detectable activity is inversely proportional to the absolute detection efficiency at the full-energy photopeak.

The already installed low level gamma-ray spectrometry with a non-contaminated lead shield and a low activity detector helps and increases the detection efficiency

The present work revealed a number, which is constant for ^{235}U fissions, induced with cold neutron beam. The absolute activities of the selected radioactive nuclides, which are related to certified concentrations of some elements in SRM2710 Montana soil, were calculated. These calculations depend mainly on good chosen, unambiguous, reaction cross-sections for the considered gamma lines. The fission products and their fission yields are found in several literatures for the fissionable nuclides [3-5].

Several works were done in the field of fast fission yields (fast neutron- induced fission) [6]. It is well known that the detection of fission events using nuclear track detectors is very useful, but its accuracy is less than that achieved by gamma activity measurements [3] . The problem in the latter technique is the required higher lower limit of fissionable element concentration, than that in the case of using solid state nuclear track detectors [7-11].

The aim of this work is to solve the problem of unavailability of estimation of the induced fission rates in environmental matrices (soil, water and air) and the accumulation of fission products especially those of long half-lives, due to the long-term exposure to cosmic rays and particles, especially neutrons. Few works were done for the estimation of atmospheric neutrons, their spatial and energy distribution in atmosphere [12-13], however the published data in this field will help.

From the average neutron fluxes jointly with the obtained fission rate constant, in the present work, one can say something about the accumulation of a specific radioactive nuclide in the environment and also can predict and estimate, by calculations, the produced ultra trace fission fragments and their gamma intensities.

The importance of the produced fission products is the accumulation of the radioactive nuclides in the environment due to their secular exposure to the cosmic rays especially cold neutrons, then the transfer of these fragments to the human bodies.

The obtained number (fission rate constant of natural uranium depending on the content of ^{235}U in cold neutron beam) is also useful in the field of reactors such as the fuel performance monitoring and monitoring of the radioactivity transport in and out of the reactor and control [14-15]. The fission reaction yields of ^{232}Th, ^{234}U and ^{238}U with cold neutrons can be neglected related to that of ^{235}U.

The good monitors of the fission reaction rate, in this wok, are ^{134}I and ^{135}I which produced, only, from fissions. The obtained results give a good agreement with those obtained from the related measurements by CR-39 solid state nuclear track detector [16].

EXPERIMENTAL PROCEDURE

Sample Irradiations

Zirconia and standard samples were packaged and irradiated individually, zirconia for 30 minutes and SRM 2710 for 40 minutes in the center of a cold neutron beam. The irradiation position was at the exit of the primary shutter of the curved neutron guide NG-0. The fluence rate of 2.64×10^{9} n cm^{-2} $s^{-1} \pm 0.3$ % was determined by the irradiation of a gold foil at the same position. The timing uncertainty is±0.5 s, which is primarily due to the uncertainty of the shutter arriving to the full open position Figure 1.

Gamma Spectrometry

It is very important and necessary to choose the suitable fission product on which the calculation of fission rates has been depending.

The more intense and measurable gamma lines arising from fission products are those lines of ^{134}I and ^{135}I, which have been produced, only, from fissions. In this work the fissionable nuclei are almost those of ^{235}U. Shortly after the irradiation, the samples Zirconia and the standard were removed from the polyethylene bags and the pellets were sealed in unirradiated polyethylene. The sealed pellets of test sample and standard are then individually placed in counting containers and transferred for gamma ray spectra measurements.

The efficiency of the detector is determined by SRM 4275-49, a mixed isotope calibration standard. Gamma spectrum data were converted to concentration values for the identified activation products. Each sample was placed directly on top of the intrinsic germanium coaxial detector, of efficiency 47.4 % (full energy peak relative to that of 3″×3″ NaI 25cm source-to-detector distance), resolution 1.02 keV FWHM, diameter 62mm, length 60mm, nominal active volume 169cm^3, model number IGC40190 from Princeton Gamma-Tech. (PGT) at National Institute of

Standards and Technology (NIST), USA. Peaks were integrated using both a peak search routines and channel-by-channel peak summation routines. Count rates of the elements (especially uranium isotopes in the sample) were compared to those of the standard for calculation of concentrations. All count rates are corrected for radioactive decay, spectral interference, pile up and peak detection efficiency for each peak for activity measurements.

Gamma intensities of ^{134}I and ^{135}I, which are the fission products of most interest with lines 884 and 1072.6 keV for ^{134}I and 1131.5, 1260.4 and 1678 keV for ^{135}I were measured. From these gamma intensities, the fission rate in Zirconia and SRM2710 has been calculated.

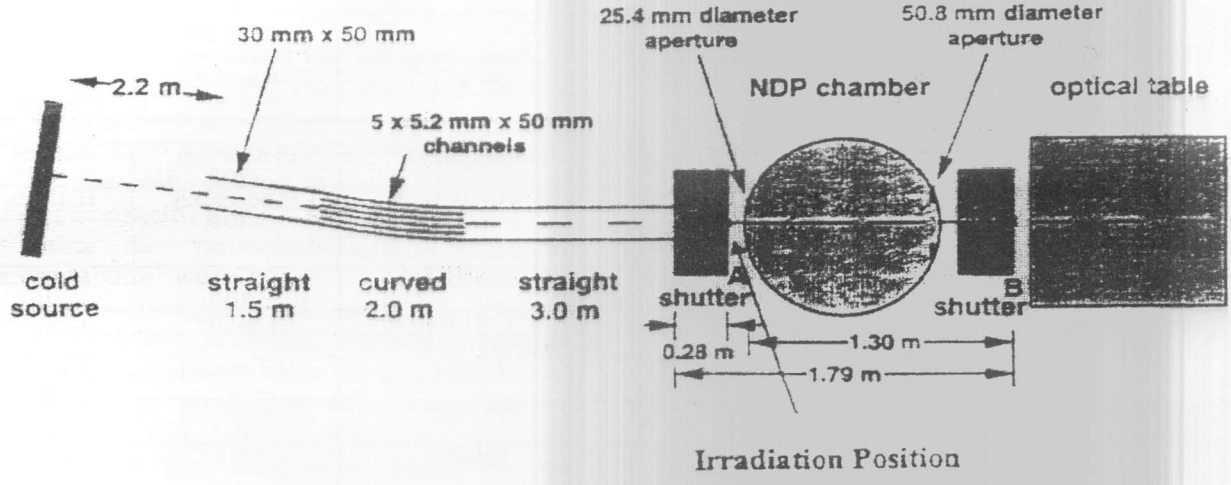

Figure 1 Samples irradiation in the cold neutron beam (At NIST, "National Institute of Standards and Technology Center for Neutron Research Reactor", USA)

RESULTS

Determination of Uranium Concentration in Zirconia by INAA:

The 0.1g samples were irradiated with cold neutrons in the NIST reactor for INAA analysis of the uranium content. Zirconia sample 1069-03 material and SRM2710 were packaged together for the irradiation in (3.12×10^{13}) n cm^{-2} s^{-1}. These disks were placed parallel to the irradiation container axes at the center of the container. The container was irradiated for 5 minutes + 5 minutes in RT-4 with the container inverted by 180 degrees at the mid-point of the irradiation time. Shortly after the irradiation, the samples were removed from the polyethylene bags and placed in clean unirradiated polyethylene bags. The bags were sealed and placed individually into counting containers. After a decay period of 5 days, gamma ray spectra were collected about one hour counting time for the samples and more than 24 hours for the standards (due to the short irradiation time)

Fission Products and Fission Yields

SRM2710 soil was activated and counted under the same conditions for the estimation of uranium content in zirconia by NAA comparator method using the gamma lines arising from ^{239}Np (99.52, 106.13, 228.18, 277.6 keV). It was found to be 340-ppm uranium. The cumulative yield of ^{134}I or ^{135}I is the fraction of ^{235}U fissions that directly yield that nuclide and its radioactive decay precursors in the constant-mass fission product chain. Many of the fission products have such short half-lives that no accurate

measure of their direct yields as primary fission products is available. Our choice of ^{134}I and ^{135}I is due to their half-lives of 52.6m and 6.6h respectively, that enables us to measure their gamma intensities, immediately, after the irradiation process. Since these radioisotopes, ^{134}I and ^{135}I, arise only from fissions, they provide unambiguous measure of a fissionable material.

By calculations we can get a new factor and a constant which are useful in the field of NAA especially by cold neutron beam, this factor can be called fission or absolute fission rate sensitivity, and the obtained constant has been called fission rate constant $s^{-1}\ \phi^{-1}\ ppm^{-1}$ of natural uranium irradiated in cold neutron beam. Table 1 gives the results.

Table 1 Fission rate constant of natural uranium, and fission rate sensitivities of zirconia and SRM2710 irradiated in cold neutron beam of 2.64×10^9 n cm^{-2} s^{-1}, calculated from the most unambiguous gamma line intensities arising from ^{134}I and ^{135}I fission products (the calculations are for one gram sample).

^{135}Igamma energy (keV)	Gamma intensity %	Absolute activity (B_q)	Number of ^{135}I (atoms)	Fission rate ($s^{-1}\ g^{-1}$)	Absolute* fission rate sensitivity ($s^{-1}g^{-1}\phi^{-1}$)	Absolute fission rate Constant** ($s^{-1}\phi^{-1}$ ppm^{-1})
For Zirconia						
1131.5	22.5	31.8±2.2	(1.1±0. 07) $\times10^6$	(1.0 ±0.08)$\times10^4$	(3.8 ±0 .32) $\times10^{-6}$	(1.12 ±0.1) $\times10^{-8}$
1260.4	28.5	32.44±2.3	(1.12±0.08) $\times10^6$	(1.03±0.08)$\times10^4$	(3.88 ±0.32) $\times10^{-6}$	(1.14 ±0.1) $\times10^{-8}$
1678.1	9.5	31.3±2.2	(1.08±0.07) $\times10^6$	(9.9±0.8)$\times10^3$	(3.74 ±0.32) $\times10^{-6}$	(1.1 ±0.1) $\times10^{-8}$
Average	-	31.8	1.1×10^6	$1.0\pm\times10^4$	3.8×10^{-6}	1.12×10^{-8}
For SRM2710						
1311.1	22.5	2.35 ±0.2	(7.9±0.6) $\times10^4$	735±51	(2.79±0.26) $\times10^{-7}$	(1.12 ±0.1) $\times10^{-8}$
1260.4	28.5	2.4 ±0.2	(8.13±0.7) $\times10^4$	753±53	(2.85±0.26) $\times10^{-7}$	(1.14 ±0.1) $\times10^{-8}$
Average		2.39	8.1×10^4	750±52	2.83×10^{-7}	1.13×10^{-8}
The related results for ^{134}I						
(a) for zirconia						
884.1	65	283±18	(1.3±0.08) $\times10^6$	(1.08±0.07) $\times10^4$	(4.1±0.29) $\times10^{-6}$	(1.1±0.1) $\times10^{-8}$
1072.6	1.5	252 ±16	(1.15±0.08) $\times10^6$	(9.44±0.66) $\times10^3$	(3.53 ±0.25) $\times10^{-6}$	(1.02 ±0.1) $\times10^{-8}$
Average	-	267	1.22×10^6	1.03×10^4	3.83×10^{-6}	1.06×10^{-8}
(b) for SRM2710 Montana soil						
884.1	65	19.87 ±1.4	(9.07±0.64) $\times10^4$	763±54	(2.9±0.2) $\times10^{-7}$	(1.08±0.1) $\times10^{-8}$

* Absolute fission rate sensitivity is defined as (fissions $s^{-1}\ g^{-1}\ \phi^{-1}$) of the used samples.
** Absolute fission rate constant of natural uranium is defined as (fissions $s^{-1}\phi^{-1}\ ppm^{-1}$),
ϕ= unit cold neutron flux (n cm^{-2} s^{-1}).
ppm= part per million of natural uranium in one gram sample.

Errors in Analysis

Like other analytical techniques, the activation analysis is not entirely free from errors, especially, for ultra-trace fission product elements produced from neutron induced fission reactions of trace fissionable nuclides such as ^{235}U in natural uranium-containing samples.

Because the possibility of external contamination from (n, γ) reactions is always present, the present work depends mainly on ^{134}I and ^{135}I, which are produced only from fissions, and have been chosen to be the indicators for fission rates monitoring by gamma intensity measurement.

Another source of errors in this work has been arisen from the determination of uranium concentration in zirconia (466.12 mg of zirconia 1069). The calculated one sigma counting statistics in the present work are shown in table1. Proper precautions have been taken in this study, however, the total

error amounts to about 10% for gamma spectrometry.

CONCLUSION

It must be mentioned that ^{134}I and ^{135}I are very good indicators for the detection and estimation of fission yields and fission rates in the irradiated samples. These isotopes arise only from fission processes and have half-lives of 52.6m and 6.6h respectively, that give the capability to follow their decays. Fission rate sensitivities can be obtained by gamma spectrometry technique, via the activities of ^{134}I and ^{135}I due to neutron irradiation; especially for samples having ultra trace uranium. The obtained fission rate constant, in average, of natural uranium of [1.1×10^{-8} fissions $s^{-1}\ \varphi_c^{-1}\ ppm^{-1}$] where φ_c^{-1} is the cold neutron flux will be useful in the environmental, INAA and dating fields.

Using the published data of uranium concentration in different environmental matrices and atmospheric neutrons, one can predict the accumulation of fission products in several matrices such as soil and rocks. The chemical extraction of a chosen fission product of a long half-life gives the capability to estimate the saturation factor of the accumulated fission product, which will be useful in the field of dating.

The obtained fission constant can be used to estimate the fission events, then the atom numbers of each fission product with its fission yield as follow:

Considering fissions occur at constant rate (N_f) and considering more that the neutron-absorption reactions in the fission product and its precursors can be neglected; the number of fission events (N_f) per second (fission rate) can be given by making use of the obtained fission constant ($B_a = (1.1\pm0.1)\times 10^{-8}\ s^{-1}\ \phi^{-1}\ ppm^{-1}$ natural uranium) in Equation 1

$$N_f = B_a \cdot C_u \cdot \varphi_c \qquad (1)$$

(neglecting the effects of the absorption and scattering of neutrons in the used samples (~2mm thicknesses) which are thin relative to the diffusion lengths of neutrons in the used materials)[13].

where

C_u= the concentration of natural uranium in ppm (μg / g)

φ_c= the cold neutron flux (cold neutron $cm^{-2}\ s^{-1}$)

The activity of a specific long-lived fission product (i) after irradiation time (T_R) can be given by Equation 2

$$(N_{fp})_i \times \lambda_i = N_f \times y_i \times (1 - e^{-\lambda_i \cdot T_R}) \qquad (2)$$

where

$(N_{fp})_i$ is the number of atoms of long-lived fission product (i) produced along time of irradiation (T_R);

y_i is the cumulative fission yield of fission product (i), atoms per atom fissioned.

Then the residual activity of a specific long-lived fission product (i) after cooling for a time (T_c), in the irradiated sample (assuming irradiation is in a constant cold neutron flux) can be given by making use Equation 3:

$$(N_{fp})_i \times \lambda_i = N_f \times y_i \times (1 - e^{-\lambda_i \cdot T_R}) \times e^{-\lambda_i \cdot T_c} \qquad (3)$$

where

λ_I is the decay constant of nuclide (i), s^{-1} ;

T_R is the irradiation time, s ;

T_c is the cooling time, s

According to the secular exposure of environmental matrices such as soil, rocks, ... etc, to the atmospheric cold neutrons [17], Equation 3 can be given in the following form, where the saturation factor $(1 - e^{-\lambda_i \cdot T_R})$ equals unity;

$$(N_{fp})_i \times \lambda_i = B_a \times C_u \times \varphi_c \times y_i \times e^{-\lambda_i \cdot T_c} \qquad (4)$$

The last term $e^{-\lambda_i \cdot T_c}$ equals unity at the end of exposure (at $T_c = 0$)

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