Neutron-based analysis of fission rates and ultra-trace concentrations of ^{235}U using gamma spectrometry and CR-39 (plastic track detector)

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Fission rates of ^{235}U in SRM2710 Montana Soil, zirconia, coal and coal ash samples were determined using nuclear track technique. Each sample (in the form of a disk) were sandwiched between two pieces, several mm^2 of CR-39. The sample packages were then placed in a cold neutron beam at the NIST Center for Neutron Research reactor. Almost all fission events in these samples resulting from the irradiation were due to $^{235}U(n,f)$ reactions. The induced fission were measured by track counting. The uranium content of the zirconia determined relative to the SRM2710 by both the fission track density and gamma activity of ^{239}Np, was found to be (354±18) ppm and (339±10.7) respectively. For comparison, instrumental neutron activation analysis (INAA) followed by gamma spectrometry was also used to determine the uranium content of the all test samples. The fission track density rates of SRM2710 and zirconia were found to be (3±0.15), (51.7±2.6) fission cm^{-2} s^{-1}, respectively, while the fission track registration sensitivities of CR-39 for both samples are $(6.33\pm0.32)\times10^{-9}$ and $(7.68\pm0.38)\times10^{-9}$ fission track cm^{-2} s^{-1} ppm^{-1} per unit neutron flux, respectively. This technique has been applied for the detection of trace and ultra trace ^{235}U bearing inclusions in two environmental samples, coal ash and coal, in which the ^{235}U contents have been found to be (77 ± 4) ppb and (4.18±0.2) ppb, respectively, while the related experimental fission track registration sensitivities of CR-39 were determined to be $(3.26\pm0.16)\times10^{-9}$ and $(4.7\pm0.24)\times10^{-9}$ fission track $cm^{-2}.s^{-1}$ per neutron per ppm ^{235}U respectively.

يمكن التعرف على تركيزات اليورانيوم الطبيعي أو يورانيوم – ٢٣٥. عن طريق قياس معدلات الإنشطار الحادثة في عينة ما معرضة لسيل من النيوترونات الباردة معلومة الشدة ومعلوم أيضا زمن التشعيع ويمكن تقدير معدلات الانشطار لكل عينة عن طريق قياس كثافة المسارات النووية لنواتج الإنشطار على سطح الكاشف النووى CR-39 بعد معالجتـــه كيميائيـا تحـت ظروف قياسية معلومة يتم إتباعها عند كل إجراء. لقياس كثافة المسارات الإنشطارية على سطح CR-39 الملتصق بجانبي كل عينة. في هذا البحث، تم تعريض عينة قياسية لسيل من النيوترونات الباردة، وكذا عينة زركونيا (ســبق قيــاس تركــيز اليورانيوم عن طريق التحلل بالتنشيط النيتروني ٣,١٢ × ١٠^{٣} نيوترون سم–٢ ث–١) مع عينات قياسية معدة خصيصا لهذا الغرض حيث وجد أن تركيز اليورانيوم بها عن طريق قياسات كاشف مسارات الإنشطارات وأشعة جاما لنـــاتج التنشـيط النيوتروني لليورانيوم – ٢٣٨ (نيبتونيوم –٢٣٩) هى على الترتيب: ٣٥٤ ±١٨ و ٣٣٩ ±١٠,٧ جزء في المليون. كتطبيـق لهذه التقنية لتقدير تركيز اليورانيوم – ٢٣٥ عن طريق قياس معدلات الإنشطار في العينات باستخدام النيترونات الباردة فقـد تم تقدير التركيزات الضئيلة والضئيلة جدا في تراب الفحم والفحم حيث وجدت (٧٧ ±٤, ١٨٠ ،٠,٣١ ±٠,٣١) جزء في البليـون على الترتيب محسوبة من معدلات الانشطار المقاسة عمليا لهاتين المادتين مقارنة بمعدل الإنشطار الحادث في المادة القياسية. وذلك بناء على قيم متوسطات مدى نواتج الانشطار المحسوبة في مادتي تراب الفحم والفحم على الترتيب.

Keywords: Analysis of ^{235}U. Cold neutron induced fission. CR-39. Fission rate

1. Introduction

Nuclear track techniques have been used since the early 1962 for the detection of uranium and thorium, and for the determination of their concentrations [1-3]. The technique has also been used for dating of ancient artifacts and geological unknown samples relative to known standards [4].

The track densities from the spontaneous fission of ^{235}U in a mineral and the induced fission in the same mineral can be compared to estimate the age of that mineral [5-7]. The applications of track detector technique extend also to the measurement of atmospheric neutrons by the registration of the recoil protons onto the surface of the plastic track detector [8].

For radiation dosimetry, this technique has been applied for the determination of exposure rates to neutrons. This is done by placing hydrogen rich-material in contact with the plastic detector then counting the induced recoil proton tracks onto the surface of the detector [7,8]. Alternatively, samples containing fissile material such as uranium or thorium can be placed in contact with the plastic detector,. and then the induced fission tracks are counted [9-11].

One of the advantages of fission track technique, compared to NAA is that this method provides an indication of the distribution of fissile elements in the material [12]. The present work is an important application of nuclear track techniques for the determination of fission rates in ^{235}U or any other fissionable element, the technique can also be used non-destructively, since the residual radiation is usually negligible. This is then very useful in the field of quantitative analysis of ultra trace uranium.

Fission in ^{235}U nuclei can be induced by exposure to a cold neutron flux. For thick samples in close contact with a plastic detector, the fission tracks can be revealed by the chemical treatment of the exposed plastic detector. When unknown and known samples are irradiated by the same neutron flux, the track densities arising from both samples can be compared to determine the concentration of uranium in the unknown sample [4]. An example is the measuring of combustion waste products for uranium content, which becomes increasingly important with the greater use of coal as a source of energy.

The major objective of this study is the estimation of the low level fission rates and the evaluation of ultra trace concentrations of ^{235}U in samples.

2. About cold neutron research facility

The use of cold neutron beam in this work has been preferred because the fission reaction cross-sections of ^{238}U and ^{232}Th with cold neutrons are negligible compared with that of ^{235}U. Therefore, we will be sure that all fission are, almost, produced only from the fission of ^{235}U [13].

The Cold Neutron Research Facility (CNRF) represents the first major enhancement of reactor-based neutron scattering instrumentation in the United States. The heart of CNRF is a cold neutron moderator, or cold source, a block of D_2o- H_2o ice cooled to 35K, situated adjacent to the core of the 20- MW, National Institute of Standard and Technology (NIST) research reactor. In the cold source, neutrons are slowed down from the moderate, enhancing the population of lower-energy and longer wavelength neutrons [14], which are particularly useful for different applications.

3. Experimental procedures

3.1. Sample preparation

Two disks of nominal mass 1 g and 0.1 g were formed from each of the two materials, zirconia and SRM2710 Montana Soil, using a Perkin- Elmer Kbr press at 9 tons pressure. Each of the 1 g samples of SRM2710 and zirconia thick samples were sandwiched between two small CR-39 pieces of several mm^2 area and of 1000 μm thickness from Pershore Mouldings Ltd. U.K, and then encapsulated in clean polyethylene film for irradiation in the cold neutron beam.

3.2 Instrumental neutron activation analysis (INAA):

3.2.1. Irradiation and gamma spectrometry

The 0.1g samples were irradiated with thermal neutrons in the NIST reactor for INAA analysis of the uranium content. All test sample materials were packaged together for the irradiation in (3.12×10^{13}) n cm^{-2} s^{-1} neutron flux. 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 with the container inverted by 180 degrees at the mid-point of the total 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 with one

hour counting time for the samples and more than 24 hours for the standards.

3.2.2 Spectrum and data evaluation

Gamma spectrum data were converted to concentration values for the identified activation products using the Canberra / Nuclear Data software routines installed on the NMG VAX computer. A command file steers the quantitative evaluation by first calculating specific count rates arising from ^{238}U (n, γ) ^{239}Np reaction based on the irradiated standards. Count rates of the ^{239}Np in the samples are compared to those of the standards for calculation of concentrations. All count rates are corrected for radioactive decay, spectral interference, and pile up. The INAA comparator method using the gamma lines arising from ^{239}Np (99.5, 166.1, 228.2 and 277.6 keV) was used to determine the uranium content of the zirconia relative to SRM2710.

3.3. CR-39 sample irradiation

Test and standard samples were packaged and irradiated individually, zirconia sample and SRM2710 were irradiated for 30 minutes and 40 minutes, respectively, while coal and coal ash samples were irradiated for 60 minutes, respectively in the center of a cold neutron beam. The irradiation position was at the exit of the primary shutter of the curved neutron guide. Because of the curvature of the guide, the number of fast neutrons remaining in the neutron beam is not significant. The cold neutron fluence rate (average flux) of 2.64×10^{9} n cm^{-2} s^{-1} ± 0.3 % was determined by the irradiation of a gold foil at the same position in order to estimate the fission track registration of the test samples zirconia and SRM2710 while the used cold neutron fluence rates of (average fluxes) (2.67×10^{9}) and (2.74×10^{9}) n cm^{-2} s^{-1} were used for the analysis of coal and coal ash samples. The timing uncertainty is 0.5 s, which is primarily due to the uncertainty in the time that it takes the shutter to come to the full open position.

3.4. Etching of CR-39 and track counting

To determine the fission track densities, CR-39 detectors were removed and chemically etched under the following conditions. The pieces were soaked in 6.25 N NaOH solution at 70°C for 45 minutes, then washed with distilled water to remove the etch chemical products. The fission track densities were evaluated using a Swift optical microscope at a magnification of 500×. The induced fission track densities in CR-39 for both zirconia and SRM 2710 were determined. The ratio of these fission rates is then used for the calculation of the concentration of ^{235}U in zirconia.

4. Calculations

This technique can be used directly to determine the ^{235}U concentration in a sample using the cold neutron reaction cross-section and known cold neutron fluence, and fission fragment ranges in samples by making use of Eq. (1, 2), given below.

4.1. Determination of ^{235}U concentration in samples

Consider a section of material containing N_5 atoms of ^{235}U per unit volume, the number n_{if} of induced fissions per unit volume produced by a cold neutron fluence F, is given by the following Eq. [4, 15, 16],

$$n_{if} = N_5 \sigma_c F. \qquad (1)$$

Where σ_c is the cold-fission cross-section for ^{235}U.

The number of induced fission tracks per unit area, ρ (i.e. fission track density) appearing on an external surface of a thick sample is related to the number (n_{if}) of fission events per unit volume by the following equation,

$$\rho = \frac{1}{2} n_{if} R_i, \qquad (2)$$

where R_i is the mean range of single fission fragment in material i (considering the critical

angle of a fission fragment $\theta_c \approx 0$ for most minerals as well as for CR-39 plastic detector). Eq. (2) will also represent the fission track density on a detector placed in close contact with the external surface of the uranium-bearing sample. The fission rate per g sample as measured by gamma lines are related to track density (in CR-39) per unit length of a sample and is given by the following equation,

Fission rate per gram $(F.\ s^{-1}.\ g^{-1})$=

$$\frac{\text{Track density rate } (\text{cm}^{-2}\ \text{s}^{-1})}{\text{Range } (\text{g cm}^{-2})}. \qquad (3)$$

The average range of fission fragments in the test materials expressed in cm can be calculated by making use the density of the test material in Eq. (3) and fission $s^{-1}\ g^{-1}$ from gamma activity arising from a specific fission product by NAA [17]. To reduce uncertainties in the estimated values, ratios of unknown to standard materials can be made using the following equation,

$$N_u = N_s \frac{\rho_u}{\rho_s}\frac{\phi_s}{\phi_s}\frac{R_s}{R_u}. \qquad (4)$$

Where:

N_s and N_u are the concentrations of uranium in standard and unknown.

ρ_s and ρ_u are the related fission track densities for standard and unknown.

ϕ_s and ϕ_u are the related fluxes, used for the standard and unknown.

R_s and R_u are the average ranges of fission fragments in the standard and unknown samples, which can be evaluated by calculations depending mainly on the bulk constituents of each sample as mentioned in the next paragraph.

4.2. Evaluation of fission fragment ranges in samples by calculations

The evaluation, by calculation, of th average fission fragment rang <R> in eac sample material (mixture), of the present wor depends mainly on the mean molecular weigh <M> of the mixture and the mean number n_i c atoms of element i in the mean molecule of th mixture [18]. The calculated values of averag fission fragment ranges in the samples used i this work give agreements with the value obtained experimentally by using th measured gamma activities of ^{134}I and ^{13} fission products. The bulk constituents of th used samples are mainly as follow [19]:

SRM 2710	SiO_2 (58%), Al_2O_3 (16.5%), Fe_2O_3 (7.2%), CaO (5.1%), C (4%), K_2O (3%) and MgO (2%).
Coal	C (77%), Al_2O_3 (6.2%), SiO_2 (5.8%). H(5%), S (2.4%).
Coal Ash	SiO_2 (45%), Al_2O_3 (34%), Fe_2O_3 (16%), MgO (2.5%) .

Zirconia sample was analyzed durin measurements of gamma spectrometry Irradiated Zirconia. The bulk constituents the used sample was found to be:

Zirconia	$ZrSiO_4$ (60%), CaO (7.2%), Fe_2O_3 (8.8%) and Al_2O_3 (17.0%)

5. Results

5.1. INAA

The INAA was used to determine th amount of uranium in the zirconia samp based on comparison with SRM2710 an gamma lines arising from ^{239}Np (99.5, 166. 228.2, 277.6 keV). The amount of uranium the zirconia sample was determined to b 340±17 ppm.

Table 1. Fission track densities, and fission track per neutron per ppm 235U in samples.

Sample	Fission track density ($T.\ cm^{-2}$)	Fission track density rate ($T.\ cm^{-2}\ s^{-1}$)	(^{235}U) ppm	Total cold neutron fluence (F) $n\ cm^{-2}$	Fission track density rate per neutron ($T.\ cm^{-2}.\ n^{-1}.\ s^{-1}$)	Fission track density rate per neutron per ppm ($T.\ cm^{-2}.\ n^{-1}.\ s^{-1}.ppm^{-1}$)
SRM2710 soil	$(7.2 \pm 0.36)\times10^{3}$	3±0.15	(0.18)	6.34×10^{12} ± 0.3%	1.14×10^{-9}	$(6.33\pm 0.32)\times10^{-9}$
Zirconia	$(9.3\pm0.47)\times10^{4}$	51.7±2.6	(2.55±0.13)	4.75×10^{12} ± 0.3%	$(1.95\pm0.1)\times10^{-8}$	$(7.68\pm 0.38)\times10^{-9}$
Coal	188 ± 14	$(5.2\pm0.4)\times10^{-2}$	$(4.18\pm0.31)\times10^{-3}$	9.6×10^{12} ± 0.3%	$(1.96\pm0.15)\times10^{-11}$	$(4.7\pm0.33)\times10^{-9}$
Coal ash	$(2.48\pm0.12)\times10^{3}$	0.69±0.04	$(7.7\pm0.38)\times10^{-2}$	9.86×10^{12} ± 0.3%	$(2.52\pm0.13)\times10^{-10}$	$(3.26\pm 0.17)\times10^{-9}$

5.2. Fission track technique

About 100 fields of view were measured for each sample to obtain the average fission fragment track densities ρ which are evaluated and reported in Table 1 together with the measured uranium content of zirconia. The value obtained for the ^{235}U concentration is 2.55 ppm which leads to, 353±15 ppm of natural uranium which is in good agreement with the obtained value via INAA, 340±17 ppm. By making a measurement relative to the concentration of uranium in SRM2710, many sources of uncertainty cancel and the dominant uncertainty is the statistics of the track counting.

6. An application for the detection of trace and ultra trace ^{235}U in environmental samples

A disk of coal and another disk of coal ash were prepared and irradiated with cold neutrons of total fluences (9.6×10^{12} and 9.86×10^{14}) ±0.3% n cm^{-2}, exactly under the same conditions, mentioned above, for SRM2710. The ultra-trace concentration of ^{235}U in coal and coal ash samples, as environmental samples, were found to be 4.18±0.2 and 77±4 ppb respectively .

7. Conclusions

The advantage of using CR-39 as a track-etch detector is the ability to determine the content of ^{235}U at low concentrations. This technique can also provide information about the homogeneity of the uranium concentration in the sample. For samples that are too large for core irradiation, track etch detectors combined with beam irradiation provide a non-destructive alternative for uranium measurements. ^{235}U concentrations in zirconia, coal ash and coal samples compared with SRM2710, were found to be 2.55± 0.13, (7.7± 0.38) $\times10^{-2}$ and (4.18±0.31)$\times10^{-3}$ ppm where the average fission fragment ranges are 4.1×10^{-3} and 5.0×10^{-3} g.cm^{-2} , in SRM2710 and zirconia samples, respectively, while the related values by calculations give agreements with the experimental results which are found to be 4.3×10^{-3} and 5.5×10^{-3} g $cm.^{-2}$. The mean fission fragment ranges are calculated to be 3.06×10^{-3} and 2.2×10^{-3} g cm^{-2} for coal and coal ash, respectively [18]. Gamma spectrometry techniques can give more accurate results, but only for higher concentrations than those which are needed for the solid state nuclear track detector technique.

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References

[1] P. B. Price and R. M. Walker , Phys. Rev. letters, 8, 217 (1962).

[2] R. L. Fleicher, P. B. Price and R. M. Walker, "Nuclear Tracks in Solids: Principles and Applications," University of California press, Berkeley (1975).

[3] A.F. Hafez, B. M. Moharram, A.M. ELKhatib and Abdel-Naby, Isotopenpraxis, 27, 185 (1991).

[4] S.A. Durrani and R.K. Bull, Solid State Nuclear Track Detection, Principles, Methods and Applications, Pergamon press, Oxford (1987).

[5] G.A. Wagner, Earth and planetary science letters 4, 411 (1968).

[6] M. Sohrabi, Nuc. Inst. and Methods, 165, 135 (1979).

[7] G. A. Wagner, G. M. Reimer, B. S Carpenter, H. Faul, R. Van Der Linden and R. Gijbels, The Spontaneous fission rate of U-238 and fission track dating, Geochemica et cosmochemica Acta, Vol. 39, 1279-1286 (1975).

[8] B. M. Moharram, Radioisotopes Vol 47, No. 9, Sep., 678 (1998).

[9] J B. Little. Proc. Nat Acad. Sci USA. 94. 5996 (1997).

[10] K. Becker Health Physics 12 769 (1966)

[11] G Somogi. Atomki Reprent E/25 Budapest. Hungary (1986)

[12] G. Bernard Harvey Introduction to nuclear physics and chemistry 2nd ed. (1969).

[13] G.R. Choppin & J Rydberg, "Nuclear chemistry theory and application" Pergamon Press (1980).

[14] NIST Center For Neutron Research, Annual Report. NISTIR 6112. National Institute of Standards and Technology (NIST). Gaithersburg. MD. USA, October 1996 through September (1997).

[15] K. Moriwaki. Y. Shimpuku and M. Makimura. Radioisotopes. 47. 611 (1998).

[16] B. S. Bajwa and H S. Virk. Environmental international Vol. 22 Suppl. 1. S 379 (1996).

[17] B. M. Moharram "The use of gamma lines of ^{41}I and ^{131}I for determination of uranium fission products in cold-neutron irradiated samples" under publication in Alexandria Engineering Journal. (AEJ)

[18] G Cecini. G Lucarini: Evaluation of Fission Fragment Ranges in Any Medium" Nucl Inst and Meth., 127 579 (1975)

[19] Standard Reference Material Catalog, National Institute of Standards and Technology (NIST). USA (1999).

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